

# SCANNING MOBILITY PARTICLE SIZER™ (SMPS™) SPECTROMETER

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BIBLIOGRAPHY  
(2015—A4)

This bibliography lists selected publications relating to TSI's Model 3938 SMPS™ Spectrometer. TSI's Scanning Mobility Particle Sizer (SMPS™) spectrometer is widely used as the standard for measuring airborne particle size distributions. This system is also routinely used to make accurate nanoparticle size measurements of particles suspended in liquids. SMPS spectrometer sizing is a discreet technique in which number concentrations are measured directly without assuming the shape of the particle size distribution. The method is independent of the refractive index of the particle or fluid, and has a high degree of absolute sizing accuracy and measurement repeatability.

The table of contents is composed of clickable links can be used to navigate this document. The publications are sorted by their application into application specific bibliographies. Each application specific bibliography is further sorted by year. The application specific bibliographies consist of a list of sources and corresponding abstracts. The general bibliography is a comprehensive list of all the sources mentioned in the application specific bibliographies.

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## APPLICATION SPECIFIC BIBLIOGRAPHIES

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### Atmospheric Science/Outdoor Studies

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#### 2013

Von Bismarck-Osten, C., Birmili, W., Ketzel, M., Massling, A., Petäjä, T., and Weber, S., 2013, "Characterization of parameters influencing the spatio-temporal variability of urban particle number size distributions in four European cities," *Atmospheric Environment*, **77**: 415–429

Three years of aerosol measurements from four European cities were analysed to identify and quantify the parameters influencing the particle number size distribution (NSD) at different measurement sites (Copenhagen, Helsinki, Leipzig, London). A "triple-site" approach was established with data from a roadside, an urban background and a rural measurement station, which was available for each city. The NSD measurements were conducted over a diameter size range of 8-700 nm. The average annual total number concentrations varied between 1700 cm<sup>-3</sup> (Finnish rural site) and 23,000 particles cm<sup>-3</sup> (London roadside). Highest variability on the diurnal course was found for the roadside sites and on the seasonal course for the rural sites. A first identification of the influencing parameters was found using correlation analysis. The most dominating factors were then quantified using a multiple linear regression approach documenting the highest influence for daytime (45%) followed by wind direction (18%), season (12%), wind speed (11%), temperature (8%) and solar radiation (6%), on average. The variation of influence between the different cities is analysed and discussed. Subsequently NSDs were modeled using the regression approach in order to verify how well the parameters are capable of "explaining" the actual NSD, showing best performance for roadside NSD with correlation coefficients between 0.65 and 0.8 and limited performance for the rural sites (0.5-0.6).

Buonanno, G., Fuoco, F. C., Morawska, L., and Stabile, L., 2013, "Airborne particle concentrations at schools measured at different spatial scales," *Atmospheric Environment*, **67**: 38–45

Potential adverse effects on children health may result from school exposure to airborne particles. To address this issue, measurements in terms of particle number concentration, particle size distribution and black carbon (BC) concentrations were performed in three school buildings in Cassino (Italy) and its suburbs, outside and inside of the classrooms during normal occupancy and use. Additional time resolved information was gathered on ventilation condition, classroom activity, and traffic count data around the schools were obtained using a video camera. Across the three investigated school buildings, the outdoor and indoor particle number concentration monitored down to 4 nm and up to 3 mm ranged from 2.8 x 10<sup>4</sup> part cm<sup>-3</sup> to 4.7 x 10<sup>4</sup> part cm<sup>-3</sup> and from 2.0 x 10<sup>4</sup> part cm<sup>-3</sup> to 3.5 x 10<sup>4</sup> part cm<sup>-3</sup>, respectively. The total particle concentrations were usually higher outdoors than indoors, because no indoor sources were detected. I/O measured was less than 1 (varying in a relatively narrow range from 0.63 to 0.74), however one school exhibited indoor concentrations higher than outdoor during the morning rush hours. Particle size distribution at the outdoor site showed high particle concentrations in different size ranges, varying during the day; in relation to the starting and finishing of school time two modes were found. BC concentrations were 5 times higher at the urban school compared with the suburban and suburban-to-urban differences were larger than the relative differences of ultrafine particle concentrations.

Liu, C., Chu, B., Liu, Y., Ma, Q., Ma, J., He, H., Li, J., and Hao, J., 2013, "Effect of mineral dust on secondary organic aerosol yield and aerosol size in  $\alpha$ -pinene/NO<sub>x</sub> photo-oxidation," *Atmospheric Environment*, **77**: 781–789

Although it is a significant contributor to atmospheric particles, the role of mineral dust in secondary organic aerosol (SOA) formation has not been fully recognized. In this study, alumina was chosen as the surrogate to investigate the effect of mineral dust on  $\alpha$ -pinene/NO<sub>x</sub> photo-oxidation in a 2 m<sup>3</sup> smog chamber at 30 C and 50% relative humidity (RH). Results showed that alumina seeds could influence both the SOA yield and the aerosol size in the photo-oxidation process. Compared to the seed-free system, the presence of alumina seeds resulted in a slight reduction of SOA yield, and also influenced the final concentration of O<sub>3</sub> in the chamber. As an important oxidant of  $\alpha$ -pinene, the decrease in O<sub>3</sub> concentration could reduce the formation of semi-volatile compounds (SVOCs) and consequently inhibited SOA formation. In addition, the size of aerosol was closely related with the mass loading of alumina seeds. At low alumina concentration, SVOCs condensed onto the pre-existing seed surface and led to aerosol size growth. When alumina concentration exceeded about 5  $\mu\text{g m}^{-3}$ , SVOC species that condensed to each seed particle were dispersed by alumina seeds, resulting in the decrease in aerosol size.

Song, S., Wu, Y., Xu, J., Ohara, T., Hasegawa, S., Li, J., Yang, L., and Hao, J., 2013, "Black carbon at a roadside site in Beijing: Temporal variations and relationships with carbon monoxide and particle number size distribution," *Atmospheric Environment*, **77**: 213–221

Black carbon (BC), carbon monoxide (CO), and particle number size distribution were measured near a major urban expressway of Beijing during summer and winter field campaigns in 2009. BC was also observed at urban and rural sites. The temporal variations of BC and its relationships with CO and particle number size distribution were analyzed. The average BC concentrations at the roadside site were 12.3 and 17.9  $\mu\text{g m}^{-3}$  during the summer and winter campaigns, respectively. BC concentrations ranked in the order of roadside > urban > rural. A general diurnal pattern at all sites showed that the higher BC levels were observed at night. The diurnal pattern of summertime BC at the roadside site followed the variations of heavy-duty diesel vehicles (HDDVs). The increased proportion of HDDVs at night contributed to high DBC/DCO ratios. This study suggests that HDDVs are an important contributor to nighttime BC and particle number concentrations of both Aitken and accumulation modes near major roadways in Beijing, especially in summer.

Yue, D. L., Hu, M., Wang, Z. B., Wen, M. T., Guo, S., Zhong, L. J., Wiedensohler, A., and Zhang, Y. H., 2013, "Comparison of particle number size distributions and new particle formation between the urban and rural sites in the PRD region, China," *Atmospheric Environment*, **76**: 181–188

Particle number size distributions were simultaneously measured at the Guangzhou (GZ) urban site (23.13N, 113.26E) and the Back-garden (BG) rural site (23.5N, 113.03E) in the Pearl River Delta (PRD) region in July, 2006. It provided new findings into the evolution of particle number size distribution and new particle formation (NPF) in two different environments. Number concentration of particles (20 nm–10  $\mu\text{m}$  diameter) at GZ was about 70% higher than at BG and significantly affected by traffic emission. However, number concentrations of the regional aerosols (100–660 nm) were  $6 \pm 3 \times 10^3 \text{ cm}^{-3}$  at both sites. At BG, the diurnal variation of particle number size distributions showed an obvious particle growth process beginning at about 9:00 (LT), probably caused by NPF. In contrast, particle number concentrations in the size ranges of 20–45 nm, 45–100 nm, and 100–660 nm showed similar trends with two main peaks at about 12:00 (LT) and 19:00 (LT) at GZ. NPF events were observed at both sites, but the occurrence frequency at GZ was about 50% lower than at BG. Regional NPF events at both sites probably in the same air mass were simultaneously observed with similar growth rates, concentrations and production rates of the condensable vapors, and condensational sinks on July 6. On the whole, decreasing traffic emission will improve air quality efficiently in the aspect of particle number concentration and fine particulate pollution in the summer of PRD should be controlled in a regional scale, especially with stagnant air mass from South China Sea.

## 2012

Patidar, V., Tripathi, S. N., Bharti, P. K., and Gupta, T., 2012, "First Surface Measurement of Cloud Condensation Nuclei over Kanpur, IGP: Role of Long Range Transport," *Aerosol Science and Technology*, **46**(9): 973–982

Measurements have been carried out for cloud condensation nuclei (NCCN, number concentration at 0.38% average depleted supersaturation, SS) and submicron aerosol (NCN), using a CCN (cloud condensation nuclei) counter (Droplet Measurement Technology) and Scanning Mobility Particle Sizer (TSI), respectively, for a large number of days in each season of the year 2008 and 2009 at Kanpur, North India. Aerosol chemical composition was also measured for 3 days and 3 nights during November–December 2009. NCCN was generally much higher than observed at similar environments elsewhere

except in Chinese cities. Due to higher loading of CCN the supersaturation depletion correction is applied. Significant intraseasonal variability was observed in NCCN and CCN/CN ratio (NCCN /NCN), due to different air masses coming from north-west, east, and central parts of India. The CCN concentrations at 0.38% and CCN/CN ratio for the year 2008 varied between 10,043–12,107 cm<sup>-3</sup> and 0.12–0.30 in winter season and 5942–7184 cm<sup>-3</sup> and 0.07–0.15 in premonsoon season, respectively. For 2009, it varied between 10,518–13,029 cm<sup>-3</sup> and 0.28–0.53 in winter season and 3596–8040 cm<sup>-3</sup> and 0.20–0.28 in postmonsoon season, respectively. Higher CCN/CN ratio was observed during winter season when the air mass came from north-west, central, and eastern landmass of India. This was most likely due to relatively high accumulation mode particle concentration and large number of forest fires observed in those regions. As expected, polluted continental air masses lead to a significant increase in CCN concentrations over the winter months, most likely due to increased anthropogenic activities, i.e., increased fuel usage, large biomass burning coupled with lower mixed boundary layers. A closure study was performed by application of Kohler theory, utilizing chemical composition, and size distribution measured by SMPS. CCN concentrations were predicted for 3 days and 3 nights and these values were compared with measured CCN values at 0.13, 0.33, and 0.64% SS. In the present closure study, CCN values were slightly overpredicted to the extent of 21% ± 18%.

Rutter, a. P., Shakya, K. M., Lehr, R., Schauer, J. J., and Griffin, R. J., 2012, "Oxidation of gaseous elemental mercury in the presence of secondary organic aerosols," *Atmospheric Environment*, **59**: 86–92

Gaseous elemental mercury (GEM; Hg<sup>0</sup> (g)) was oxidized by ozone and secondary hydroxyl radicals generated by the chemistry associated with the formation of secondary organic aerosols. The reaction was investigated in a 9-m<sup>3</sup> Teflon batch reactor. The losses of GEM in ozone-only experiments compared well with numerical model predictions based on published reaction rates, and a second order rate analysis gave a reaction rate of  $(7.4 \pm 0.5) \times 10^{-19}$  cm<sup>3</sup> molecules<sup>-1</sup> s<sup>-1</sup>, which was statistically indistinct from recent publications. Furthermore, the net oxidation of GEM observed in the SOA reaction system agreed well with a numerical model based on the GEM-ozone reaction rate determined in this study and a published GEM-OH oxidation rate. Recent modeling studies of mercury atmospheric cycling have found that use of laboratory-based GEM-ozone reaction rate coefficients caused overestimation of GEM oxidation, while theoretical studies cast doubt over the viability of the GEM-ozone oxidation reaction in the real atmosphere. The results presented here suggest that the reaction is viable in the atmosphere and that recent published reaction rates for GEM and ozone are pertinent for use in atmospheric models. An average of GEM-ozone rates determined during this and recent studies was  $6.9 \pm 0.9 \times 10^{-19}$  cm<sup>3</sup> molecules<sup>-1</sup> s<sup>-1</sup>. This value is recommended for use in future modeling studies.

Siingh, D., Chate, D. M., and Ali, K., 2012, "Time-elapsd evolution of aerosol size distributions by snow particles after the passage of blizzards over the Maitri (Antarctica)," *International Journal of Remote Sensing*, **33**(4): 962–978

Measurements of the aerosol size distribution in the diameter range 0.003–20 μm were made by a Scanning Mobility Particle Sizer (SMPS) spectrometer and an Aerodynamic Particle Sizer<sup>®</sup> (APS<sup>™</sup>) spectrometer at Maitri (70° 45' 52" S, 11° 44' 03" E, Antarctica) during the 24th Indian Antarctic Expedition, which took place in January and February 2005. Time-elapsd evolutions of the aerosol size distributions on 30 January and 17 and 20 February 2005, after the passage of blizzards, were selected for estimation of the scavenging coefficient by considering the aerosol size distribution changes with snowfall over Antarctica. Scavenging efficiencies of snow particles and ice crystals collecting aerosols are obtained from the observed scavenging coefficients for 0.002–10 μm aerosols. Also, theoretical snow scavenging efficiencies of snow particles and ice crystals in the diameter range 0.02–10 mm for 0.002–10 μm aerosols are determined and, thereupon, scavenging coefficients are calculated by assuming snowfall rates of 0.25 and 4 mm h<sup>-1</sup> (in view of the reported decadal mean annual Antarctica snowfall accumulation rate of 180 mm yr<sup>-1</sup>). Snow scavenging efficiencies based on the scavenging coefficients found by considering the aerosol size distribution changes with snowfall after the occurrences of blizzards are higher than theoretical scavenging efficiencies and those of laboratory experiments for planar and columnar ice crystals. Also, the scavenging coefficients found are higher by two to three orders of magnitude than theoretical scavenging coefficients. Scavenging coefficients found from aerosol size distribution changes by snowfall after the passage of blizzards on 30 January and 17 and 20 February 2005 over Antarctica suggest that the orders of magnitude of their best estimation and the corresponding scavenging efficiencies can be viewed as being on a par with the natural snow scavenging of aerosols over this remote region.

Thompson, J. E., Hayes, P. L., Jimenez, J. L., Adachi, K., Zhang, X., Liu, J., Weber, R. J., and Buseck, P. R., 2012, "Aerosol optical properties at Pasadena, CA during CalNex 2010," *Atmospheric Environment*, **55**: 190–200

Aerosol optical properties measured at the Pasadena, CA site during the CalNex field campaign in May–June 2010 are summarized. Average measurements of PM<sub>2.5</sub> aerosol extinction, scattering, absorption coefficients, and single scattering albedo (bext, bscat, babs and SSA) at λ = 532 nm were 62 Mm<sup>-1</sup>, 58 Mm<sup>-1</sup>, 4 Mm<sup>-1</sup>, and 0.92, respectively. The aerosol optical densities were 5 times lower than during the

SCAQ5 study in 1987, highlighting major progress in PM control in the Los Angeles area in the last two decades. The period May 30 - June 8 2010 was characterized by exceptionally high aerosol loading (bext up to  $250 \text{ Mm}^{-1}$ ). During this period, bext, bscat, and SSA tended to peak during the mid-morning. Correlation of PM<sub>2.5</sub> bext, bscat with mass concentration data yielded mass scattering and mass extinction coefficients of  $3.5 - 5.1 \text{ m}^2 \text{ g}^{-1}$  for 532 nm. Aerosol babs were compared directly to mass concentration of elemental carbon (EC) yielding a campaign average mass absorption cross section (M.A.C.) of  $5.7 \pm 1.8 \text{ m}^2 \text{ g}^{-1}$ . TEM analysis of particles suggests soot was often internally mixed or adhering to sulfate and/or organics. Total non-refractory PM<sub>1</sub> mass was a good quantitative indicator of coated soot fraction. Alteration of M.A.C. with mixing/coating state was not detected, however, increases in M.A.C. were linked to the presence of light absorbing, water-soluble organic carbon (WSOC) suggesting a possible role of this material invisible light absorption in the LA basin.

## 2011

Bari, M. A., Baumbach, G., Brodbeck, J., Struschka, M., Kuch, B., Dreher, W., and Scheffknecht, G., 2011, "Characterisation of particulates and carcinogenic polycyclic aromatic hydrocarbons in wintertime wood-fired heating in residential areas," *Atmospheric Environment*, **45**(40): 7627–7634

Wood as a renewable and worldwide available fuel is used for residential heating in small-scale firings during winter. This wood combustion can cause very high emissions of inhalable particles resulting in short and long-term health effects. The target of this study was to characterise particulate matter, emissions of polycyclic aromatic hydrocarbons (PAHs) and their size distribution and to show that those emissions can be found in the ambient air of residential areas with wood-fired heating. Emission samples were collected from pellet stove and log wood boiler under different combustion conditions. Ambient PM<sub>10</sub> sampling was performed during two winter seasons at two rural residential areas near Stuttgart in Germany. Samples were extracted using toluene with ultrasonic bath and analysed by gas chromatography mass spectrometry (GC-MS). Twenty-one PAH compounds including nine carcinogenic ones were detected and quantified. It was found that emission concentrations of carcinogenic PAHs were higher during incomplete combustion compared to complete combustion. Significant amounts of ambient PAHs were found in the residential villages, where the contribution of carcinogenic PAHs was 44% of total PAHs in the ambient air during winter 2009. The morphology and elemental analysis of ambient particles were also investigated. The findings indicate a rising concern to reduce emissions from wood-fired heating during winter in residential areas and underline the importance of using good wood combustion technologies to improve the air quality.

Buonanno, G., Stabile, L., Avino, P., and Belluso, E., 2011, "Chemical, dimensional and morphological ultrafine particle characterization from a waste-to-energy plant," *Waste management (New York, N.Y.)*, **31**(11):2253-62

Waste combustion processes are responsible of particles and gaseous emissions. Referring to the particle emission, in the last years specific attention was paid to ultrafine particles (UFPs, diameter less than  $0.1 \mu\text{m}$ ), mainly emitted by combustion processes. In fact, recent findings of toxicological and epidemiological studies indicate that fine and ultrafine particles could represent a risk for health and environment. Therefore, it is necessary to quantify particle emissions from incinerators also to perform an exposure assessment for the human populations living in their surrounding areas. To these purposes, in the present work an experimental campaign aimed to monitor UFPs was carried out at the incineration plant in San Vittore del Lazio (Italy). Particle size distributions and total concentrations were measured both at the stack and before the fabric filter inlet in order to evaluate the removal efficiency of the filter in terms of UFPs. A chemical characterization of UFPs in terms of heavy metal concentration was performed through a nuclear method, i.e., Instrumental Neutron Activation Analysis (INAA), as well as a mineralogical investigation was carried out through a Transmission Electron Microscope (TEM) equipped with an Energy Dispersive Spectrometer (EDS) in order to evaluate shape, crystalline state and mineral compound of sampled particles. Maximum values of  $2.7 \times 10^7 \text{ part. cm}^{-3}$  and  $2.0 \times 10^3 \text{ part. cm}^{-3}$  were found, respectively, for number concentration before and after the fabric filter showing a very high efficiency in particle removing by the fabric filter. With regard to heavy metal concentrations, the elements with higher boiling temperature present higher concentrations at lower diameters showing a not complete evaporation in the combustion section and the consequent condensation of semi-volatile compounds on solid nuclei. In terms of mineralogical and morphological analysis, the most abundant compounds found in samples collected before the fabric filter are Na-K-Pb oxides followed by phyllosilicates, otherwise, different oxides of comparable abundance were detected in the samples collected at the stack.

Cheng, Y.-H., and Li, Y.-S., 2011, "Influences of traffic volumes and wind speeds on ambient ultrafine particle levels—Observations at a highway electronic toll collection (ETC) lane," *Atmospheric Environment*, **45**(1): 117–122

The levels of ultrafine particles (UFPs) and their size distributions on a highway electronic toll collection (ETC) lane were measured from October 30 to November 1 and November 5 to November 6, 2008. The hourly UFP levels measured at the highway ETC lane were  $1.3 \times 10^4$  -  $1.9 \times 10^5$  particles  $\text{cm}^{-3}$  (mean= $9.4 \times 10^4$  particles  $\text{cm}^{-3}$ ). Compared with urban UFP levels, average UFP levels at the highway ETC lane were about 5-10 times higher than those previously measured in urban areas, indicating that a considerable amount of UFPs were exhausted from vehicles. At the highway ETC lane, the average UFP number size distribution had a dominant mode at about 10 nm and a minor mode at about 33 nm. Measurement results indicate that nucleation mode particles remained at relatively high levels compared to Aitken mode and accumulation mode particles at the highway ETC lane. This study identifies the impacts of traffic volumes and wind speeds on ambient UFP levels. Measurement results show that the elevated UFP levels resulting from traffic volume decreased exponentially as wind speed increased on the highway when wind speed was  $<2.0\text{ms}^{-1}$ . However, the elevated UFP levels resulting from traffic volume increased slightly when wind speed was  $>2.0\text{ms}^{-1}$  due to local turbulent mixing causes UFPs at high wind speeds. According to measurement results, high levels of UFP were observed at low wind speeds and under high traffic volumes.

Gómez-Moreno, F. J., Pujadas, M., Plaza, J., Rodríguez-Maroto, J. J., Martínez-Lozano, P., and Artíñano, B., 2011, "Influence of seasonal factors on the atmospheric particle number concentration and size distribution in Madrid," *Atmospheric Environment*, **45**(18): 3169–3180

The ambient particle number concentration and size distribution have been measured in an urban background site in Madrid, a continental Mediterranean area, over more than two years (Oct 2006-Dec 2008). The objective was to study the sources and processes affecting or contributing to fine and ultrafine particles in this area. They have been measured with a TSI-SMPS (15-600 nm) instrument and with a modified Vienna type DMA (3-80 nm) and a CPC 3025 (TSI) during 6 months. The average particle number concentration was lower than in other sites as it is an urban background site and because of its location in the Mediterranean area. The particle number concentrations have shown a clear seasonal influence: maximum values were observed every year in the period November-January, coinciding with atmospheric stagnant conditions and pollution episodes, while minimum values were measured in springtime, a period in which wind speed produced high atmospheric dilution. The Aitken and accumulation modes have shown similar seasonal behavior, with two maxima related to vehicle emissions. The nucleation mode had a third maximum observed at noon during spring and summer. The size distributions were bimodal during most of the time: the first mode was centered on 20e50 nm and was associated with fresh particles related to vehicle exhaust emission; the second mode, between 50 and 160 nm, mainly corresponded to the evolution of the first mode. The evolution of the size distributions reveals a marked annual cycle related to the season, with an increase of median diameters during summer and a decrease during winter. Different evolutions of particle size distribution corresponding to different meteorological and seasonal scenarios were identified. The influence of higher wind speeds on particle size distribution has been confirmed to cause a decrease in the particle number concentration and in the size distribution mode. Particle nucleation is not a frequent phenomenon in this measurement site, where 63 events per year have been observed. They mainly occurred during spring and summer periods, with the minimum number during winter. This suggests that insolation and temperature are important variables in nucleation. Class Ia nucleation events mainly occurred during spring and summer. High wind speeds were important during class II events, as the particles suffered low growth or lost their semivolatile compounds.

Hallar, a. G., Lowenthal, D. H., Chirokova, G., Borys, R. D., and Wiedinmyer, C., 2011, "Persistent daily new particle formation at a mountain-top location," *Atmospheric Environment*, **45**(24): 4111–4115

Frequent new particle formation (NPF) has been observed regularly at Storm Peak Laboratory, a high elevation mountain-top observatory in Colorado. These events occurred during 52% of the 474 measurement days from 2001 to 2009, consistently during the mid-afternoon throughout the spring, summer and winter months. Average growth rates, condensation sinks, and formation rates are presented. The strongest correlation factor with NPF is ultraviolet radiation. The events are not related to increased ozone concentration or pre-existing aerosol surface area, implying that nucleation can occur anywhere in the free troposphere.

Rattanavaraha, W., Rosen, E., Zhang, H., Li, Q., Pantong, K., and Kamens, R. M., 2011, "The reactive oxidant potential of different types of aged atmospheric particles: An outdoor chamber study," *Atmospheric Environment*, **45**(23): 3848–3855

The reactive oxygen species (ROS) potential of aged diesel exhaust particulate matter (PM) and other aged aerosol systems in the presence and absence of an urban hydrocarbon environment was assessed.

Experiments were performed in a 274 m<sup>3</sup> dual outdoor Teflon film chamber. Filter samples were taken to assess the oxidant generation associated with PM by an optimized dithiothreitol (DTT) method. Diesel exhaust PM had a higher ROS response when it was in the presence of an urban hydrocarbon mixture and was associated with significant O<sub>3</sub> production. For all the aged dilute diesel systems, ROS expression increased by a factor of 2-4 over fresh diesel particles. Other particle systems were also investigated. A low ROS was observed in most of the nighttime experiments, including the nighttime aerosols from SO<sub>2</sub> with O<sub>3</sub> and SO<sub>2</sub> aged by itself. However, when all the systems were compared, aged diesel exhaust tended to express very high ROS potentials, with secondary organic aerosols from an  $\alpha$ -pinene + toluene + an urban HC mixture giving the highest ROS response.

Sorribas, M., de la Morena, B. a., Wehner, B., López, J. F., Prats, N., Mogo, S., Wiedensohler, A., and Cachorro, V. E., 2011, "On the sub-micron aerosol size distribution in a coastal-rural site at El Arenosillo Station (SW - Spain)," *Atmospheric Chemistry and Physics*, **11**(21): 11185–11206

This study focuses on the analysis of the submicron aerosol characteristics at El Arenosillo Station, a rural and coastal environment in South-western Spain between 1 August 2004 and 31 July 2006 (594 days). The mean total concentration (NT) was 8660cm<sup>-3</sup> and the mean concentrations in the nucleation (NNUC), Aitken (NAIT) and accumulation (NACC) particle size ranges were 2830cm<sup>-3</sup>, 4110cm<sup>-3</sup> and 1720cm<sup>-3</sup>, respectively. Median size distribution was characterised by a single-modal fit, with a geometric diameter, median number concentration and geometric standard deviation of 60 nm, 5390cm<sup>-3</sup> and 2.31, respectively. Characterisation of primary emissions, secondary particle formation, changes to meteorology and long-term transport has been necessary to understand the seasonal and annual variability of the total and modal particle concentration. Number concentrations exhibited a diurnal pattern with maximum concentrations around noon. This was governed by the concentrations of the nucleation and Aitken modes during the warm seasons and only by the nucleation mode during the cold seasons. Similar monthly mean total concentrations were observed throughout the year due to a clear inverse variation between the monthly mean NNUC and NACC. It was related to the impact of desert dust and continental air masses on the monthly mean particle levels. These air masses were associated with high values of NACC which suppressed the new particle formation (decreasing NNUC). Each day was classified according to a land breeze flow or a synoptic pattern influence. The median size distribution for desert dust and continental aerosol was dominated by the Aitken and accumulation modes, and marine air masses were dominated by the nucleation and Aitken modes. Particles moved off-shore due to the land breeze and had an impact on the particle burden at noon, especially when the wind was blowing from the NW sector in the morning during summer time. This increased NNUC and NAIT by factors of 3.1 and 2.4, respectively. Nucleation events with the typical "banana" shape were characterised by a mean particle nucleation rate of 0.74cm<sup>-3</sup> s<sup>-1</sup>, a mean growth rate of 1.96nm h<sup>-1</sup> and a mean total duration of 9.25 h (starting at 10:55GMT and ending at 20:10GMT). They were observed for 48 days. Other nucleation events were identified as those produced by the emissions from the industrial areas located at a distance of 35 km. They were observed for 42 days. Both nucleation events were strongly linked to the marine air mass origin.

## 2010

Chen, S.-C., Tsai, C.-J., Chou, C. C.-K., Roam, G.-D., Cheng, S.-S., and Wang, Y.-N., 2010, "Ultrafine particles at three different sampling locations in Taiwan," *Atmospheric Environment*, **44**(4): 533–540

Atmospheric ultrafine particles (UPs or PM<sub>0.1</sub>) were investigated at the roadside of Syuefu road in Hsinchu city, in the Syueshan highway tunnel in Taipei and in the NTU Experimental Forest in Nantou, Taiwan. A SMPS (TSI 3936) and three MOUDIs (MSP 110) were collocated to determine the number and mass concentrations of the PM<sub>0.1</sub> simultaneously. The filter samples were further analyzed for organic carbon (OC), element carbon (EC), water-soluble ions and trace elements. Taking into account the OC artifact of PM<sub>0.1</sub>, good chemical mass closure (ratio of the reconstructed chemical mass to the gravimetric mass of PMs) was obtained with an unknown percentage of 10.6, 26.2 and 37.2% at the roadside, tunnel and forest, respectively. The unexplained mass was attributed to aerosol water in this study. The artifact at the roadside, tunnel and the forest PM<sub>0.1</sub> mass was found to be as high as 51.6 ± 10.7%, 20.0 ± 5.4% and 85.6 ± 18.4%, respectively. Finally, the effective density of the roadside, tunnel and forest PM<sub>0.1</sub> was calculated based on the results of chemical speciation and found to be 1.45, 1.29 and 1.22 g cm<sup>-3</sup>, respectively, which was in good agreement with that obtained by using the method of Spencer et al. (2007). Based on these results, it is foreseeable that the number concentration of the SMPS can be converted using the effective density determined by Spencer et al. (2007) for the real time measurement of the PM<sub>0.1</sub> concentration.

Hagler, G. S. W., Thoma, E. D., and Baldauf, R. W., 2010, "High-Resolution Mobile Monitoring of Carbon Monoxide and Ultrafine Particle Concentrations in a Near-Road Environment," *Journal of the Air & Waste Management Association*, **60**(3): 328–336

Assessment of near-road air quality is challenging in urban environments that have roadside structures, elevated road sections, or depressed roads that may impact the dispersion of traffic emissions. Vehicles traveling on arterial roadways may also contribute to air pollution spatial variability in urban areas. To characterize the nature of near-road air quality in a complex urban environment, an instrumented all-electric vehicle was deployed to perform high spatial- and temporal-resolution mapping of ultrafine particles (UFPs, particle diameter <100 nm) and carbon monoxide (CO). Sampling was conducted in areas surrounding a highway in Durham, NC, with multiple repeats of the driving route accomplished within a morning or evening commute time frame. Six different near road transects were driven, which included features such as noise barriers, vegetation, frontage roads, and densely built houses. Under downwind conditions, median UFP and CO levels in near-road areas located 20–150 m from the highway were a factor of 1.8 and 1.2 higher, respectively, than in areas characterized as urban background. Sampling in multiple near-road neighborhoods during downwind conditions revealed significant variability in absolute UFP and CO concentrations as well as in the rate of concentration attenuation with increasing distance from the highway. During low-speed meandering winds, regional UFP and CO concentrations nearly doubled relative to crosswind conditions; however, near-road UFP levels were still higher than urban background levels by a factor of 1.2, whereas near-road CO concentrations were not significantly different than the urban background.

Ling, X., Jayaratne, R., and Morawska, L., 2010, "Air ion concentrations in various urban outdoor environments," *Atmospheric Environment*, **44**(18): 2186–2193

Atmospheric ions are produced by many natural and anthropogenic sources and their concentrations vary widely between different environments. There is very little information on their concentrations in different types of urban environments, how they compare across these environments and their dominant sources. In this study, we measured airborne concentrations of small ions, particles and net particle charge at 32 different outdoor sites in and around a major city in Australia and identified the main ion sources. Sites were classified into seven groups as follows: park, woodland, city centre, residential, freeway, power lines and power substation. Generally, parks were situated away from ion sources and represented the urban background value of about 270 ions cm<sup>-3</sup>. Median concentrations at all other groups were significantly higher than in the parks. We show that motor vehicles and power transmission systems are two major ion sources in urban areas. Power lines and substations constituted strong unipolar sources, while motor vehicle exhaust constituted strong bipolar sources. The small ion concentration in urban residential areas was about 960 cm<sup>-3</sup>. At sites where ion sources were co-located with particle sources, ion concentrations were inhibited due to the ion-particle attachment process. These results improved our understanding on air ion distribution and its interaction with particles in the urban outdoor environment.

Rizzo, L. V., Artaxo, P., Karl, T., Guenther, A. B., and Greenberg, J., 2010, "Aerosol properties, in-canopy gradients, turbulent fluxes and VOC concentrations at a pristine forest site in Amazonia," *Atmospheric Environment*, **44**(4): 503–511

Aerosol physical and chemical properties were measured in a forest site in central Amazonia (Cuieiras reservation, 2.61S; 60.21W) during the dry season of 2004 (Aug-Oct). Aerosol light scattering and absorption, mass concentration, elemental composition and size distributions were measured at three tower levels (Ground: 2 m; Canopy: 28 m, and Top: 40 m). For the first time, simultaneous eddy covariance fluxes of fine mode particles and volatile organic compounds (VOC) were measured above the Amazonian forest canopy. Aerosol fluxes were measured by eddy covariance using a Condensation Particle Counter (CPC) and a sonic anemometer. VOC fluxes were measured by disjunct eddy covariance using a Proton Transfer Reaction Mass Spectrometer (PTR-MS). At nighttime, a strong vertical gradient of phosphorus and potassium in the aerosol coarse mode was observed, with higher concentrations at Ground level. This suggests a source of primary biogenic particles below the canopy. Equivalent black carbon measurements indicate the presence of light-absorbing aerosols from biogenic origin. Aerosol number size distributions typically consisted of superimposed Aitken (76nm) and accumulation modes (144nm), without clear events of new particle formation. Isoprene and monoterpene fluxes reached respectively 7.4 and 0.82 mg m<sup>-2</sup> s<sup>-1</sup> around noon. An average fine particle flux of 0.05 ± 0.10 10<sup>6</sup> m<sup>-2</sup> s<sup>-1</sup> was calculated, denoting an equilibrium between emission and deposition fluxes of fine mode particles at daytime. No significant correlations were found between VOC and fine mode aerosol concentrations or fluxes.



Zelenyuk, A., Ezell, M. J., Perraud, V., Johnson, S. N., Bruns, E. a., Yu, Y., Imre, D., Alexander, M. L., and Finlayson-Pitts, B. J., 2010, "Characterization of organic coatings on hygroscopic salt particles and their atmospheric impacts," *Atmospheric Environment*, **44**(9): 1209–1218

The photooxidation of  $\alpha$ -pinene in the presence of NO<sub>2</sub>, with and without added NaNO<sub>3</sub> seed particles, has been studied in a large-diameter flow tube. Particles formed by homogeneous nucleation and by condensation on the pre-existing seeds were sampled at various stages of the reaction, dried using four diffusion dryers, size selected at different mobility diameters (dm) using a differential mobility analyzer (DMA), and characterized with a single particle mass spectrometer (SPLAT II). It was found that homogeneously nucleated particles are spherical, have a density ( $\rho$ ) of  $1.25 \pm 0.02 \text{ g cm}^{-3}$  ( $\pm 2\sigma$ ) and contain a significant amount of organic nitrates. The mass spectra of the low volatility products condensed on the NaNO<sub>3</sub> seed particles were found to be virtually the same as in the case of homogeneous nucleation. The data show that the presence of even a submonolayer of organics on the NaNO<sub>3</sub> particles causes water retention that leads to a decrease in particle density and that the amount of water retained increases with organic coating thickness. Thicker coatings appear to inhibit water evaporation from the particle seeds altogether. This suggests that in the atmosphere, where low volatility organics are plentiful, some hygroscopic salts will retain water and have different densities and refractive indices than expected in the absence of the organic coating. This water retention combined with the organic shell on the particles can potentially impact light scattering by these particles and activity as cloud condensation nuclei (CCN), as well as heterogeneous chemistry and photochemistry on the particles.

## 2009

Gietl, J. K., and Klemm, O., 2009, "Analysis of Traffic and Meteorology on Airborne Particulate Matter in Münster, Northwest Germany," *Journal of the Air & Waste Management Association*, **59**(7): 809–818

The importance of street traffic and meteorological conditions on the concentrations of particulate matter (PM) with an aerodynamic diameter smaller than 10  $\mu\text{m}$  (PM<sub>10</sub>) was studied in the city of Münster in northwest Germany. The database consisted of meteorological data, data of PM<sub>10</sub> mass concentrations and fine particle number (6– 225 nm diameter) concentrations, and traffic intensity data as counted with tally hand counters at a four- to six-lane road. On working days, a significant correlation could be found between the diurnal mean PM<sub>10</sub> mass concentration and vehicle number. The lower number of heavy-duty vehicles compared with passenger cars contributed more to the particle number concentration on working days than on weekend days. On weekends, when the vehicle number was very low, the correlation between PM<sub>10</sub> mass concentration and vehicle number changed completely. Other sources of PM and the meteorology dominated the PM concentration. Independent of the weekday, by decreasing the traffic by approximately 99% during late-night hours, the PM<sub>10</sub> concentration was reduced by 12% of the daily mean value. A correlation between PM<sub>10</sub> and the particle number concentration was found for each weekday. In this study, meteorological parameters, including the atmospheric stability of the boundary layer, were also accounted for. The authors deployed artificial neural networks to achieve more information on the influence of various meteorological parameters, traffic, and the day of the week. A multilayer perceptron network showed the best results for predicting the PM<sub>10</sub> concentration, with the correlation coefficient being 0.72. The influence of relative humidity, temperature, and wind was strong, whereas the influence of atmospheric stability and the traffic parameters was weak. Although traffic contributes a constant amount of particles in a daily and weekly cycle, it is the meteorology that drives most of the variability.

Lu, Z., Hao, J., Takekawa, H., Hu, L., and Li, J., 2009, "Effect of high concentrations of inorganic seed aerosols on secondary organic aerosol formation in the m-xylene/NO<sub>x</sub> photooxidation system," *Atmospheric Environment*, **43**(4): 897–904

High concentrations ( $>15 \mu\text{m}^3 \text{ cm}^{-3}$ ) of CaSO<sub>4</sub>, Ca(NO<sub>3</sub>)<sub>2</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> were selected as surrogates of dry neutral, aqueous neutral and dry acidic inorganic seed aerosols, respectively, to study the effects of inorganic seeds on secondary organic aerosol (SOA) formation in irradiated m-xylene/NO<sub>x</sub> photooxidation systems. The results indicate that neither ozone formation nor SOA formation is significantly affected by the presence of neutral aerosols (both dry CaSO<sub>4</sub> and aqueous Ca(NO<sub>3</sub>)<sub>2</sub>), even at elevated concentrations. The presence of high concentrations of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> aerosols (dry acidic) has no obvious effect on ozone formation, but it does enhance SOA generation and increase SOA yields. In addition, the effect of dry (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> on SOA yield is found to be positively correlated with the (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> surface concentration, and the effect is pronounced only when the surface concentration reaches a threshold value. Further, it is proposed that the SOA generation enhancement is achieved by particle-phase heterogeneous reactions induced and catalyzed by the acidity of dry (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> seed aerosols.

Murugavel, P., and Chate, D. M., 2009, "Generation and growth of aerosols over Pune, India," *Atmospheric Environment*, **43**(4): 820–828

The measured physical size distributions of sub-micron particles during cold season at Pune, India are analyzed to explore the characteristics of nucleation and growth properties. Preliminary analysis of aerosol size distribution in time-series shows large increase in number concentration due to nucleation events between 0800 h and 1030 h at this location. The observable quantities such as condensable vapor concentration (C), its source rate (Q), growth rate (GR) and condensable sink (CS) are estimated from the time-series evolutions of aerosol size distributions. The concentration of vapor and its source rate were about  $19.8 \pm 2.15 \times 10^7$  molecules  $\text{cm}^{-3}$  and  $1.28 \pm 0.084 \times 10^7$   $\text{cm}^{-3} \text{ s}^{-1}$  respectively. The average condensation sink and growth rate were  $7.1 \pm 0.4 \times 10^{-2} \text{ s}^{-1}$  and  $16.95 \pm 1.86 \text{ nm h}^{-1}$  respectively during the growth period. The values are high enough to trigger the nucleation bursts and enhance subsequent growth rates of nucleation mode particles at this location. The magnitudes are in the range of those observed at New Delhi, India and much higher than those of European cities. The ratio of apparent to real nucleation rate is found to be a measure of number concentration of freshly produced particles by photochemical nucleation. The predicted number concentrations corresponding to measured distributions of mid-point diameter increases with the size for both 1 nm nucleated clusters and 3 nm particles. The database of all the possible event days and the event characteristics forms the basis for future works into the causes and implications of atmospheric particle formation at this location

Zhu, Y., Pudota, J., Collins, D., Allen, D., Clements, A., DenBleyker, A., Fraser, M., Jia, Y., McDonald-Buller, E., and Michel, E., 2009, "Air pollutant concentrations near three Texas roadways, Part I: Ultrafine particles," *Atmospheric Environment*, **43**(30): 4513–4522

Vehicular emitted air pollutant concentrations were studied near three types of roadways in Austin, Texas: (1) State Highway 71 (SH-71), a heavily traveled arterial highway dominated by passenger vehicles; (2) Interstate 35 (I-35), a limited access highway north of Austin in Georgetown; and (3) Farm to Market Road 973 (FM-973), a heavily traveled surface roadway dominated by truck traffic. Air pollutants examined include carbon monoxide (CO), oxides of nitrogen (NO<sub>x</sub>), and carbonyl species in the gas-phase. In the particle phase, ultrafine particle (UFP) concentrations (diameter < 100 nm), fine particulate matter (PM<sub>2.5</sub>, diameter < 2.5  $\mu\text{m}$ ) mass and carbon content and several particle-bound organics were examined. All roadways had an upwind stationary sampling location, one or two fixed downwind sample locations and a mobile monitoring platform that characterized pollutant concentrations fall-off with increased distance from the roadways. Data reported in this paper focus on UFP while other pollutants and near-roadway chemical processes are examined in a companion paper. Traffic volume, especially heavy-duty traffic, wind speed, and proximity to the road were found to be the most important factors determining UFP concentrations near the roadways. Since wind directions were not consistent during the sampling periods, distances along wind trajectories from the roadway to the sampling points were used to study the decay characteristics of UFPs. Under perpendicular wind conditions, for all studied roadway types, particle number concentrations increased dramatically moving from the upwind side to the downwind side. The elevated particle number concentrations decay exponentially with increasing distances from the roadway with sharp concentration gradients observed within 100–150 m, similar to previously reported studies. A single exponential decay curve was found to fit the data collected from all three roadways very well under perpendicular wind conditions. No consistent pattern was observed for UFPs under parallel wind conditions. However, regardless of wind conditions, particle concentrations returned to background levels within a few hundred meters of the roadway. Within measured UFP size ranges, smaller particles (6–25 nm) decayed faster than larger ones (100–300 nm). Similar decay rates were observed among UFP number, surface, and volume.

## 2008

Fukuhara, N., Suzuki, K., Takeda, K., and Nihei, Y., 2008, "Characterization of environmental nanoparticles," *Applied Surface Science*, **255**(4): 1538–1540

In this study, we attempted to characterize environmental nanoparticles and particle diameter distributions in the atmospheric environment neighboring a traffic route by using a Scanning Mobility Particle Sizer (SMPS) spectrometer. The composition of the environmental nanoparticles was analyzed using time-of-flight secondary ion mass spectrometry (TOF-SIMS). It was observed that the environmental nanoparticles showed peaks at 20 nm and 100 nm. The secondary ions C<sup>+</sup>, O<sup>+</sup>, Si<sup>+</sup>, and SiH<sub>3</sub><sup>+</sup> were strongly detected in the environmental nanoparticles with a peak at 20 nm. On the other hand, the secondary ions NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and Ca<sup>+</sup> were detected in the environmental nanoparticles with a peak at 100 nm. Moreover, it was found that the secondary ion spectral patterns of the organic compounds were different for each particle diameter. Hence, we concluded that the combination of the SMPS with TOF-SIMS is a powerful technique to characterize environmental nanoparticles.

Hatch, C. D., Gierlus, K. M., Schuttlefield, J. D., and Grassian, V. H., 2008, "Water adsorption and cloud condensation nuclei activity of calcite and calcite coated with model humic and fulvic acids," *Atmospheric Environment*, **42**(22): 5672–5684

Recent studies have shown that organics can alter the water adsorption and cloud condensation nuclei (CCN) activity of common deliquescent species in the Earth's atmosphere. However, very little is known about the effect of organics on water adsorption and CCN activity of insoluble nuclei, such as mineral dust aerosol. A large fraction of unidentified organic material in aerosol particles is composed of poly-acidic compounds resembling humic substances. The presence of these humic-like substances (HULIS) can alter the water adsorption and CCN activity of mineral dust aerosol. We have measured the CCN activity of model humic and fulvic acids and of mineral dust particles coated with these substances in the laboratory. We find that coatings of humic and fulvic acids on calcite particles significantly increases water adsorption compared to uncoated particles. CCN measurements indicate that humic- or fulvic acid-coated calcite particles are more CCN active than uncoated calcite particles. Additionally, thicker coatings of humic or fulvic acids appear to result in more efficient CCN activity. Thus, mineral dust particles coated with high molecular weight organic materials will take up more water and become more efficient CCN in the atmosphere than uncoated mineral dust particles, potentially altering the effect of mineral dust on the Earth's climate. In addition to the experimental results, we have explored a newly modified Kohler theory for predicting the CCN activity of insoluble, wettable particles based on multi layer water adsorption measurements of calcite.

## 2007

Spencer, Matthew T., Shields, Laura G., Prather, Kimberly A., 2007, "Simultaneous Measurement of the Effective Density and Chemical Composition of Ambient Aerosol Particles," *Environmental Science & Technology*, ASAP Article 10.1021/es061425+ S0013-936X(06)01425-8

Simultaneous measurements of the effective density and chemical composition of individual ambient particles were made in Riverside, California by coupling a differential mobility analyzer (DMA) with an ultrafine aerosol time-of-flight mass spectrometer (UF-ATOFMS). In the summer, chemically diverse particle types (i.e., aged-OC, vanadium-OC-sulfate-nitrate, biomass) all had similar effective densities when measured during the same time period. This result suggests that during the summer study the majority of particle mass for the different particle types was dominated by secondary species (OC, sulfates, nitrates) of the same density, while only a small fraction of the total particle mass is accounted for by the primary particle cores. Also shown herein, the effective density is a dynamic characteristic of the Riverside, CA ambient aerosol, changing by as much as 40% within 16 h. During the summer measurement period, changes in the ambient atmospheric water content correlated with changes in the measured effective densities which ranged from ~1.0 to 1.5 g·cm<sup>-3</sup>. This correlation is potentially due to evaporation of water from particles in the aerodynamic lens. In contrast, in the fall during a Santa Ana meteorological event, ambient particles with a mobility diameter of 450 nm showed three distinct effective densities, each related to a chemically unique particle class. Particles with effective densities of ~0.27 g·cm<sup>-3</sup>, 0.87 g·cm<sup>-3</sup>, and 0.93 g·cm<sup>-3</sup> were composed mostly of elemental carbon, lubricating oil, and aged organic carbon, respectively. It is interesting to contrast the seasonal differences where in the summer, particle density and mass were determined by high amounts of secondary species, whereas in the fall, relatively clean and dry Santa Ana conditions resulted in freshly emitted particles which retained their distinct source chemistries and densities.

## 2006

Geller, Michael; Biswas, Subhasis; Sioutas, Constantinos, 2006, "Determination of Particle Effective Density in Urban Environments with a Differential Mobility Analyzer and Aerosol Particle Mass Analyzer," *Aerosol Science & Technology*, **40**(9):709–723

Effective densities of atmospheric aerosols in various locations of the Los Angeles Basin were determined by a DMA-APM technique. Effective density was calculated by comparing voltage distributions of sampled atmospheric aerosols with PSL particles of known density. The five sites chosen for field experiments were: (1) Interstate-710 Freeway, impacted by heavy-duty diesel vehicles; (2) State Route CA-110, open only to gasoline vehicles; (3) Riverside, a receptor site known for secondary particle formation; (4) University of Southern California, a typical urban and industrial environment; and (5) Coast for marine aerosol. The size range selected for this study was from 50 nm to 414 nm. While 50 nm particles exhibited a single effective density multiple effective densities were measured for each of the other particle sizes as significant fractions of these particles are transported from background sources. Regardless of location, 322–414 nm particle effective densities were considerably lower than unity. The lowest effective densities (~ 0.1 g cm<sup>-3</sup>) were reported for I-710, confirming that diesel combustion aerosols are rich in chain agglomerates with large void spaces. Riverside exhibited high effective densities (~ 1.2–1.5 g cm<sup>-3</sup>) for 50–202 nm particles, which we hypothesize is due to transformations that occur during advection from Los Angeles. Measurements of diurnal variation of effective density at Riverside support this

hypothesis. Overall, our results suggest that effective density declines as the particle mobility diameter increases irrespective of location. Fractal dimensions calculated from average effective densities were lowest for I-710 ( $D_f = 2.41$ ) and CA-110 ( $D_f = 2.54$ ) aerosols, presumably due to the influence of vehicular combustion emission on these sites. By contrast, average fractal dimensions at USC, Riverside and Coast were found to be 2.79, 2.83, and 2.92, respectively. High fractal dimensions at these sites may be the effects of aging, moisture absorption and/or organic vapor condensation on the particles, which fills void space and makes particles more spherical.

Hogrefe, O.; Lala, G. G.; Frank, B. P.; Schwab, J. J.; Demerjian, K. L., 2006, "Field Evaluation of a TSI Model 3034 Scanning Mobility Particle Sizer in New York City: Winter 2004 Intensive Campaign," *Aerosol Science and Technology*, **40**(10):753–762

A new "single box" Scanning Mobility Particle Sizer (TSI SMPS Model 3034) was deployed and operated during a period of four weeks as a part of the PMTACS-NY Winter 2004 intensive study in Queens College, New York City. The SMPS 3034 is an alternative to a conventional multi-component TSI SMPS and houses a Differential Mobility Analyzer and butanol-based Condensation Particle Counter in one cabinet. The SMPS 3034 operates at a fixed 1 L/min sample flow rate (4 L/min sheath flow rate) and measures size distributions within a 10–487 nm size range. One size scan is produced every 3 minutes. Four other measurement systems (a conventional TSI SMPS with a Nano Differential Mobility Analyzer, an Aerodynamic Particle Sizer, a stand-alone Condensation Particle Counter, and an R&P Inc. Filter Dynamic Measurement System (FDMS) TEOM mass monitor) were operated side-by-side with the SMPS 3034. It is shown that total particle number concentrations measured by the SMPS 3034 are highly correlated with those from the conventional Nano SMPS, the Condensation Particle Counter and the FDMS TEOM monitor, and that the number median diameters measured by the SMPS 3034 and the Nano SMPS agree within 3 nm.

Zhu Y.; Kuhn T.; Mayo P.; Hinds W.C., 2006, "Comparison of daytime and nighttime concentration profiles and size distributions of ultrafine particles near a major highway," *Environmental Science and Technology*, **40**(8):2531-2536

Previously we have conducted systematic measurements of the concentration and size distribution of ultrafine particles in the vicinity of major highways during daytime in Los Angeles. The present study compares these with similar measurements made at night. Particle number concentration was measured by a condensation particle counter (CPC) and size distributions in the size range from 7 to 300 nm were measured by a Scanning Mobility Particle Sizer (SMPS) spectrometer. Measurements were taken at 30, 60, 90, 150, and 300 m upwind and downwind from the center of the I-405 freeway. Average traffic flow at night was about 25% of that observed during the day. Particle number concentration measured at 30 m downwind from the freeway was 80% of previous daytime measurements. This discrepancy between changes in traffic counts and particle number concentrations is apparently due to the decreased temperature, increased relative humidity, and lower wind speed at night. Particle size distributions do not change as dramatically as they did during the daytime. Particle number concentration decays exponentially downwind from the freeway similarly to what was observed during the day, but at a slower rate. No particle number concentration gradient has been observed for the upwind side of the freeway. No PM<sub>2.5</sub> and very weak PM<sub>10</sub> concentration gradients were observed downwind of the freeway at night. Ultrafine particle number concentration measured at 300 m downwind from the freeway was still distinguishably higher than upwind background concentration at night. These data may be used to help estimate exposure to ultrafine particles in the vicinity of major highways for epidemiology studies.

## 2005

Bottenheim, Jan W.; Brickell, Peter; Brook, Jeffrey R.; Hayden, Katherine; Jayne, John T.; Leitch, W. Richard; Li, Shao-Meng; Lohmann, Ulrike; Lu, Gang; Rupakheti, Maheswar; Toom-Sauntry, Desiree; Vet, Robert; Worsnop, Douglas R, 2005, "An intensive study of the size and composition of submicron atmospheric aerosols at a rural site in Ontario, Canada," *Aerosol Science and Technology*, **39**(8):722–736

Atmospheric sampling was conducted at a rural site near Egbert, about 70 km north of Toronto, Ontario, Canada from March 27 to May 8, 2003 to characterize the physical and chemical properties of the ambient aerosol in near real-time. The instrumentation included a tapered element oscillating microbalance (TEOM), an ultrafine condensation particle counter (UCPC), a Scanning Mobility Particle Sizer (SMPS) spectrometer, an Aerodynamic Particle Sizer (APS) spectrometer, an aerosol mass spectrometer (AMS), and a particulate nitrate monitor (R&P 8400N) for aerosol measurements. Gas-phase non-methane hydrocarbon compounds (NMHCs) were measured by gas chromatograph-flame ionization detection (GC-FID). Filter samples were also collected for analysis of inorganic ions by ion chromatography (IC). Aerosol properties varied considerably depending upon meteorological conditions and airmass histories. For example, urban and industrial emissions advected from the south strongly influenced the site occasionally, resulting in higher particulate mass with the higher fractions of nitrate and organics. Cleaner northwesterly winds carried aerosols with relatively higher fractions of organics and sulfate. The AMS

derived mass size distributions showed that the inorganic species in the particles with vacuum aerodynamic diameters between about 60 nm and 600 nm had mass modal vacuum aerodynamic diameters around 400-500 nm. The particulate organics often exhibited two modes at about 100 nm and 425 nm, more noticeable during fresh pollution events. The small organic mode was well correlated with gas-phase nonmethane hydrocarbons such as ethylbenzene, toluene, and propene, suggesting that the likely sources of small organic particles were combustion related emissions. The particulate nitrate exhibited a diurnal variation with higher concentrations during dark hours and minima in the afternoon. Particulate sulfate and organics showed evidence of photochemical processing with higher levels of sulfate and oxygenated organics in the afternoon. Reasonable agreement among all of the co-located measurements is found, provided the upper size limit of the AMS is considered.

Fine, Philip M.; Fruin, Scott; Sax, Todd; Sioutas, Constantinos; Westerdahl, Dane, 2005, "Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles," *Atmospheric Environment*, **39**(20) 3597-3610

Recent health studies have reported that ultrafine particles (UFP) (<0.1  $\mu\text{m}$  in diameter) may be responsible for some of the adverse health effects broadly attributed to particulate matter. In urban areas, UFP are produced by combustion sources, such as vehicle exhaust, and by secondary formation in the atmosphere. While UFP can be monitored, few studies have explored the impact of local primary sources in urban areas (including mobile sources on freeways) on the temporal and spatial distribution of UFP. This paper describes the integration of multiple monitoring technologies on a mobile platform designed to characterize UFP and associated pollutants, and the application of this platform in a study of UFP number concentrations and size distributions in Los Angeles. Monitoring technologies included two condensation particle counters (TSI Model 3007 and TSI 3022A) and Scanning Mobility Particle Sizers for UFP. Real-time measurements made of  $\text{NO}_x$  (by chemiluminescence), black carbon (BC) (by light absorption), particulate matter-phase PAH (by UV ionization), and particle length (by diffusional charging) showed high correlations with UFP numbers, ( $r^2=0.78$  for  $\text{NO}$ , 0.76 for BC, 0.69 for PAH, and 0.88 for particle length). Average concentrations of UFP and related pollutants varied strongly by location, road type, and truck traffic volumes, suggesting a relationship between these concentrations and truck traffic density.

Johnson, Jason P.; Kittelson, David B.; Watts, Winthrop F., 2005, "Source apportionment of diesel and spark ignition exhaust aerosol using on-road data from the Minneapolis metropolitan area," *Atmospheric Environment*, **39**(11): 2111-2121

Air quality measurements were made on interstate highways in the Minneapolis metropolitan area. Gas and aerosol concentrations were measured on weekdays and weekends. By exploiting the difference in the relative volumes of heavy duty (HD) diesel and light duty (LD) spark ignition (SI) vehicles on weekdays and weekends, we were able to estimate apportioned fuel specific emissions. The on-road, apportioned, fuel specific particle number emissions factors, estimated from condensation particle counter (CPC) measurements were  $1.34 \pm 0.2 \cdot 10^{16}$  /particles  $\text{kg}^{-1}$  for diesels and  $7.1 \pm 1.6 \cdot 10^{15}$  particles  $\text{kg}^{-1}$  for spark ignition vehicles. Estimates from the Scanning Mobility Particle Sizer (SMPS) spectrometer measurements were  $2.1 \pm 0.3 \cdot 10^{15}$  particles  $\text{kg}^{-1}$  for diesels and  $3.9 \pm 0.6 \cdot 10^{14}$  particles  $\text{kg}^{-1}$  for SI vehicles. The difference between CPC and SMPS measurements is mainly due to different lower size detection limits of the instruments, similar to 3 and similar to 10 nm, respectively. On a weekly weighted basis and on weekdays, the majority of particle number was attributed to HD diesel traffic. Weekend production of particles can be primarily attributed to light duty SI automobiles. On a per vehicle basis, HD vehicles produced substantially greater numbers of particles. On a fuel specific basis, HD vehicles produce slightly higher concentrations of particles than light duty vehicles. The relative contribution of LD vehicles to particle number emissions increased as particle size decreased. The HD apportioned size distributions were similar to size distributions measured during other on-road and laboratory studies. The LD apportioned size distribution was bounded by laboratory and on-road size distributions. Our work is representative of summer, highway cruise conditions. It is likely that under cold start and high load operating conditions LD emissions will increase relative to HD emissions.

Kuhn, Thomas; Biswas, Subhasis; Fine, Philip M.; Geller, Michael; Sioutas, Constantinos, 2005, "Physical and Chemical Characteristics and Volatility of PM in the Proximity of a Light-Duty Vehicle Freeway," *Aerosol Science & Technology*, **39**(4):347-357

Volatility properties of ultrafine particles were analyzed next to State Route 110 (Pasadena freeway CA), a light-duty vehicle freeway where heavy-duty traffic is prohibited. In addition, mass concentration and chemical composition of particulate matter (PM) were measured in coarse, accumulation, and ultrafine modes. On weekdays from 17 May to 4 June 2004, measurements were performed in two locations, one very close to the freeway (within 2.5 m from the curb) and one at a distance of about 50 m from the freeway. For measurement of mass and chemical composition, the study employed in each location a micro-orifice uniform deposit impactor (MOUDI) and a modified high-volume sampler. Both instruments sampled with the same size cutpoints: a coarse mode from 2.5 to 10  $\mu\text{m}$ , an accumulation mode from 0.18

to 2.5  $\mu\text{m}$ , and an ultrafine mode of particles less than 0.18  $\mu\text{m}$  in aerodynamic diameter. Alternately, a tandem differential mobility analyzer (TDMA) was used at the two sites. A heater between the two DMAs evaporated volatile material from the monodisperse aerosol, size selected by the first DMA. The second DMA analyzed the losses of volatile components. The ultrafine number concentrations next to the freeway were 46,000  $\text{cm}^{-3}$  on average during the sampling period. The MOUDI ultrafine mass concentration, nitrate, and EC were higher next to the freeway than at the background site farther from the freeway. The other components analyzed in the ultrafine mode had similar concentrations next to the freeway and at the background site. Volatility ranged from about 65% volume losses of 120 nm particles heated to 110°C to 95% of 20 nm particles. The 20 nm aerosol was only internally mixed, whereas increasing nonvolatile fractions were found for 40 nm (6% next to the freeway), 80 nm (20%), and 120 nm (28%) aerosols.

Lawless, Philip A.; Noble, Christopher A.; Rodes, Charles E., 2005, "A sampling approach for evaluating particle loss during continuous field measurement of particulate matter," *Particle and Particle Systems Characterization*, **22**(2):99–106

A method for evaluating sample bias in field measurements is presented. Experiments were performed in the field and laboratory to quantify the bias as a function of particle size for the Scanning Mobility Particle Sizer and the aerodynamic particle sizer. Sources of bias and sample loss considered in this work were sampling line loss, instrumental differences and inlet efficiencies. Measurement of the bias and sample loss allow for correction of the data acquired in the field, so as to obtain more representative samples of atmospheric concentrations. Substantial losses of fine and ultrafine particle count were observed, with sampling line losses ranging from 10-50 %, dependent on particle size. Only minor line losses were observed for coarse particles (approximately 5 %) because the sampling line was oriented vertically.

Rupakheti, Maheswar; Leaitch, W.; Lohmann, Ulrike; Hayden, Katherine; Brickell, Peter; Lu, Gang; Li, Shao-Meng; Toom-Sauntry, Desiree; Bottenheim, Jan; Brook, Jeffrey; Vet, Robert; Jayne, John; Worsnop, Douglas, 2005, "An Intensive Study of the Size and Composition of Submicron Atmospheric Aerosols at a Rural Site in Ontario, Canada," *Aerosol Science and Technology*, **39**(8):722–736

Atmospheric sampling was conducted at a rural site near Egbert, about 70 km north of Toronto, Ontario, Canada from March 27 to May 8, 2003 to characterize the physical and chemical properties of the ambient aerosol in near real-time. The instrumentation included a tapered element oscillating microbalance (TEOM), an ultrafine condensation particle counter (UCPC), a Scanning Mobility Particle Sizer (SMPS) spectrometer, an Aerodynamic Particle Sizer (APS) spectrometer, an aerosol mass spectrometer (AMS), and a particulate nitrate monitor (R&P 8400N) for aerosol measurements. Gas-phase non-methane hydrocarbon compounds (NMHCs) were measured by gas chromatograph-flame ionization detection (GC-FID). Filter samples were also collected for analysis of inorganic ions by ion chromatography (IC). Aerosol properties varied considerably depending upon meteorological conditions and air mass histories. For example, urban and industrial emissions advected from the south strongly influenced the site occasionally, resulting in higher particulate mass with the higher fractions of nitrate and organics. Cleaner northwesterly winds carried aerosols with relatively higher fractions of organics and sulfate. The AMS derived mass size distributions showed that the inorganic species in the particles with vacuum aerodynamic diameters between about 60 nm and 600 nm had mass modal vacuum aerodynamic diameters around 400–500 nm. The particulate organics often exhibited two modes at about 100 nm and 425 nm, more noticeable during fresh pollution events. The small organic mode was well correlated with gas-phase nonmethane hydrocarbons such as ethylbenzene, toluene, and propene, suggesting that the likely sources of small organic particles were combustion related emissions. The particulate nitrate exhibited a diurnal variation with higher concentrations during dark hours and minima in the afternoon. Particulate sulfate and organics showed evidence of photochemical processing with higher levels of sulfate and oxygenated organics in the afternoon. Reasonable agreement among all of the co-located measurements is found, provided the upper size limit of the AMS is considered.

Westerdahl, D.; Fruin, S.; Sax, T.; Fine, P. M.; Sioutas, C., 2005, "Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles," *Atmospheric Environment (Atmos. Environ)*, **39**(20): 3597–3610

Recent health studies have reported that ultrafine particles (UFP) (<0.1  $\mu\text{m}$  in diameter) may be responsible for some of the adverse health effects broadly attributed to particulate matter. In urban areas, UFP are produced by combustion sources, such as vehicle exhaust, and by secondary formation in the atmosphere. While UFP can be monitored, few studies have explored the impact of local primary sources in urban areas (including mobile sources on freeways) on the temporal and spatial distribution of UFP. This paper describes the integration of multiple monitoring technologies on a mobile platform designed to characterize UFP and associated pollutants, and the application of this platform in a study of UFP number concentrations and size distributions in Los Angeles. Monitoring technologies included two condensation particle counters (TSI Model 3007 and TSI 3022A) and Scanning Mobility Particle Sizers for UFP. Real-time measurements made of  $\text{NO}_x$  (by chemiluminescence), black carbon (BC) (by light absorption), particulate matter-phase PAH (by UV ionization), and particle length (by diffusional

charging) showed high correlations with UFP numbers. Average concentrations of UFP and related pollutants varied strongly by location, road type, and truck traffic volumes, suggesting a relationship between these concentrations and truck traffic density.

## 2004

Abu-Allaban, M.; Gertler, A. W.; Rogers, C. F., 2004, "A Quantitative Description of Vehicle Exhaust Particle Size Distributions in a Highway Tunnel," *Journal of the Air and Waste Management Association*, **54**(3):360–366

During the period May 18–May 22, 1999, a comprehensive study was conducted in the Tuscarora Mountain Tunnel on the Pennsylvania Turnpike to measure real-world motor-vehicle emissions. As part of this study, size distributions of particle emissions were determined using a Scanning Mobility Particle Sizer spectrometer. Each measured size distribution consisted of two modes: a nucleation mode with midpoint diameter less than 20 nm and an accumulation mode with midpoint diameter less than 100 nm. The nucleation and accumulation components in some distributions also exhibited second maxima, which implies that such particle size distributions are superpositions of two particle size distributions. This hypothesis was utilized in fitting the particle size distributions that exhibited second maxima with four lognormal distributions, two for the nucleation mode and two for the accumulation mode. The fitting assumed that the observed particle size distribution was a combination of two bimodal log-normal distributions, one attributed to the heavy-duty diesel (HDD) vehicles and another attributed either to a different class of HDD vehicles or to the light-duty spark ignition vehicles. Based on this method, estimated particle production rates were  $1.8 \times 10^{13}$  and  $2.8 \times 10^{14}$  particles/vehicle-km for light-duty spark ignition and HDD vehicles, respectively, which agreed with independently obtained estimates.

Chan, W. R.; Khlystov, A. Y.; Mandiro, M.; Pandis, S. N.; Stanier, C. O., 2004, "A method for the in situ measurement of fine aerosol water content of ambient aerosols: The dry-ambient aerosol size spectrometer (DAASS)," *Aerosol Science & Technology*, **38**(S1):215–228

Hygroscopic growth of atmospheric particles affects a number of environmentally important aerosol properties. Due to the hysteresis exhibited by the aerosol hygroscopic growth, the physical state of particles and the amount of aerosol water are uncertain within a wide range of relative humidities (RHs) found in the troposphere, leading to uncertainties in optical and chemical properties of the aerosol. Here we report the design and tests of an automated system that was built to assess the amount of aerosol water at atmospheric conditions. The system consists of two Scanning Mobility Particle Sizers (SMPS) spectrometer and an Aerodynamic Particle Sizer (APS) spectrometer that measure the aerosol size distribution between 3 nm and 10  $\mu$ m in diameter. The inlets of the instruments and their sheath air lines are equipped with computer-controlled valves that direct air through Nafion dryers or bypass them. The Nafion dryers dehydrate the air streams to below 30% RH at which point ambient particles are expected to lose most or all water. The switch between the dried and the ambient conditions occurs every 7 min and is synchronized with the scan times of the aerosol spectrometers. In this way the system measures alternatively dried (below 30% RH) and ambient aerosol size distributions. A comparison of the ambient RH and the dried RH size distributions and the corresponding integrated volume concentrations provides a measure of the physical state of particles and the amount of aerosol water. The aerosol water content can be treated as a growth factor or as an absolute quantity and can be calculated as a time series or as a function of RH (humidigram). When combined with aerosol composition measurements, the DAASS can be used to compare hygroscopic growth models and measurements.

Fine, Philip; Shen, Si; Sioutas, Constantinos, 2004, "Inferring the Sources of Fine and Ultrafine Particulate Matter at Downwind Receptor Sites in the Los Angeles Basin Using Multiple Continuous Measurements," *Aerosol Science and Technology*, **38**(S1):182–195

Recent studies that have found increased health effects of atmospheric ultrafine particulate matter (PM) have refocused attention on particle number rather than particle mass concentrations as a relevant measurement of PM pollution. As part of the Southern California Supersite program, ambient particle characteristics were measured over 13 months at three different sites in the eastern portion of the Los Angeles Basin: Riverside, Rubidoux, and Claremont, CA. The sites represent receptor locations that are influenced by local particle sources as well as advection from the more intense particle sources upwind closer to Downtown Los Angeles. An SMPS/APS tandem system was employed to collect continuous particle size distributions, from which particle number and mass concentrations were calculated. An aethalometer provided continuous particulate elemental carbon (EC) concentrations. Results show no meaningful correlation between particle number and mass, indicating that fine particle standards may not be effective in controlling ultrafine concentrations. Diurnal patterns show a morning traffic peak indicated by increases in particle mass, number, and EC. Afternoon periods in the warmer months are characterized by high number counts while mass and EC remain low, suggesting the formation of new particles by photochemistry. Particle mode diameters range from 30 nm up to above 100 nm, a result not seen in most other studies of particle size distributions in other urban or rural areas where mode

diameters are generally less than 50 nm. Evidence is presented that the observed ultrafine particle concentrations and size distributions are influenced by long-range advection and photochemical processes as well as vehicular emissions, which have been previously assumed to dominate day-to-day ultrafine particle levels.

Froines, J. R.; Jaques, P. A.; Sioutas, C.; Teh, H. W.; Yu, R. C., 2004, "Quality control of semi-continuous mobility size-fractionated particle number concentration data," *Atmospheric Environment*, **38**(20):3341-3348

Fine and ultrafine particles have been postulated to play an important role in the association between ambient particulate matters and adverse health effects. As part of the EPA Supersite Program, the Southern California Particle Center & Supersite has conducted a series of monitoring campaigns that contribute to a better understanding of the sources, chemical composition and physical state of ambient aerosols. The Scanning Mobility Particle Sizer (SMPS) spectrometer was deployed to semi-continuously measure mobility size-fractionated particle number concentrations. As part of the quality control efforts, we developed a two-stage graphic and statistical procedure to label and identify potentially discordant observations. The first stage considered the entire size-fractionated data by date-time as a whole to plot total concentration (TC) vs. coefficient of variation (CV), both in log scale. TC represents the magnitude of overall concentration for a size distribution; while CV represents the relative variability. This plot was used to partition all size distributions into four to five distinct regions. In each region, a generalized extreme studentized deviate (ESD) and a modified Z-score procedure were applied to identify potential discordant outliers. We have found that the majority of particle size distributions are concentrated within a 'normal' region, with TC ranging from  $10^2$  to  $10^5 \text{cm}^{-3}$  and CV varying between 20% and 200%. Size distributions that are contaminated with discordant outliers are displayed distinctly from the 'normal' region and form four to five clusters in the LogTC-LogCV plot. The pattern of clusters in the plot is consistent among the four sampling sites in this study, suggesting the robustness of this technique. The generalized ESD and modified Z-score effectively identify discordant outliers and reveal that the pattern of clustering outliers are consistent within each distinct region. It has, thus, been concluded that the new approach is a useful quality control tool to identify potential discordant outliers in SMPS data.

Hinds, W. C.; Shen, S.; Sioutas, C.; Zhu, Y., 2004, "Seasonal trends of concentration and size distribution of ultrafine particles near major highways in Los Angeles," *Aerosol Science & Technology*, **38**(S1):5-13

Ultrafine particles (diameter  $<0.1 \mu\text{m}$ ) have been suggested as a possible causative agent for the observed increases in mortality and morbidity with increases in particulate matter (PM) concentration. Zhu et al. conducted systematic measurements of the concentration and size distribution of ultrafine particles in the vicinity of Interstate 405 (mostly gasoline traffic) and Interstate 710 (large proportion of heavy-duty diesel traffic) in Los Angeles during the summer of 2001. The present study compares these measurements with those made at the same locations in the winter of 2001-2002. Particle number concentration and size distribution in the size range from 6 to 220 nm were measured by a condensation particle counter (CPC) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. Measurements were taken at five distances downwind from the freeway. At each sampling location, concentrations of carbon monoxide (CO) and black carbon (BC) were also measured by a Dasibi CO monitor and an aethalometer, respectively. Average temperatures were about 7 degrees Celsius higher in summer than in winter. Wind directions are the same for both seasons, wind speeds are slightly higher in summer. Traffic densities were not statistically different between summer and winter for both the 405 and 710 freeways. The decay rates of CO and BC are slightly greater in summer than in winter for both freeways, suggesting a weaker atmospheric dilution effect in winter. Particle number concentration in the size range of 6-12 nm is significantly higher in winter than in summer. The associated concentration in that size range decreased at a slower rate in winter than in summer. The surface area concentrations in the size range of 6-220 nm are consistently higher in summer for all sampling locations. These results suggest that wintertime conditions favor greater particle formation, possibly due to increased condensation of organic vapors. These data may be useful to estimate exposure to ultrafine particles in the vicinity of major highways for epidemiological studies and to evaluate the adverse health effects of such particles.

Khlystov, A. Y.; Pandis, S. N.; Stanier, C. O., 2004, "Ambient aerosol size distributions and number concentrations measured during the Pittsburgh Air Quality Study (PAQS)," *Atmospheric Environment*, **38**(20):3275-3284

Twelve months of aerosol size distributions from 3 to 560 nm, measured using Scanning Mobility Particle Sizer are presented with an emphasis on average number, surface, and volume distributions, and seasonal and diurnal variation. The measurements were made at the main sampling site of the Pittsburgh Air Quality Study from July 2001 to June 2002. These are supplemented with 5 months of size distribution data from 0.5 to 2.5  $\mu\text{m}$  measured with a TSI aerosol particle sizer and 2 months of size distributions measured at an upwind rural sampling site. Measurements at the main site were made continuously under both low and ambient relative humidity. The average Pittsburgh number concentration (3-500 nm) is  $22,000 \text{cm}^{-3}$  with an average mode size of 40nm. Strong diurnal patterns in number



concentrations are evident as a direct effect of the sources of particles (atmospheric nucleation, traffic, and other combustion sources). New particle formation from homogeneous nucleation is significant on 30–50% of study days and over a wide area (at least a hundred kilometers). Rural number concentrations are a factor of 2-3 lower (on average) than the urban values. Average measured distributions are different from model literature urban and rural size distributions.

Kittelson, D.; Watts, W.; Johnson, J.; Remerowki, M.; Ische, E.; Oberdörster, G.; Gelein, R.; Elder, A.; Hopke, P.; Kim, E.; Zhao, W.; Zhou, L.; Jeong, C.-H., 2004, "On-Road Exposure to Highway Aerosols. 1. Aerosol and Gas Measurements," *Inhalation Toxicology*, **16**(S1):31–39

On-road experiments were conducted to determine the sensitivities of rats to real-world aerosol. This article summarizes the on-road aerosol and gas measurements and provides background information for the companion paper on the rat exposures. Measurements were carried out over 10 days, 6 h/day, driving a route from Rochester to Buffalo. Aerosol instrumentation used in this study included two Scanning Mobility Particle Sizers (SMPS) spectrometer to determine the aerosol size distribution from 10 to 300 nm, two stand-alone condensation particle counters to determine the total aerosol number concentration, and an electrical aerosol detector to determine the aerosol length concentration. A thermal denuder (TD) was used with one of the SMPS instruments to determine the size distribution of the non-volatile fraction. Filter samples were collected and analyzed for elemental carbon, and gas analyzers measured ambient levels of CO, CO<sub>2</sub>, and NO. Average daily total aerosol number concentration ranged from 200,000 to 560,000 particles/cm<sup>3</sup>. Past studies on urban highways have measured total number concentrations ranging between 10<sup>4</sup> and 10<sup>6</sup> particles/cm<sup>3</sup>. The average daily NO concentration ranged from 0.10 to 0.24 ppm and the corresponding CO<sub>2</sub> concentration ranged from 400 to 420 ppm. The average daily geometric number mean particle size determined by the SMPS ranged from 15 to 20 nm. The TD reduced the average SMPS number concentration between 87 and 95% and the SMPS volume between 54 and 83%, suggesting that most of the particles consisted of volatile material. The TD also increased the geometric number mean diameter from 15 to 20 nm to 30 to 40 nm.

Matsunaga, S.; Mochida, M.; Kawamura, K., 2004, "Variation on the atmospheric concentrations of biogenic carbonyl compounds and their removal processes in the northern forest at Moshiri, Hokkaido Island in Japan," *Journal of Geophysical Research. D. Atmospheres*, **109**(D4)

Biogenic aldehydes, hydroxycarbonyls, and dicarbonyls in gas and particle phases were collected with an annular denuder sampling system (ADSS) in a *Quercus crispula* and *Picea glehnii* mast mixed forest. Eighty samples were collected from 22 to 29 August 2002. The size distributions of aerosols were also observed concurrently with a Scanning Mobility Particle Sizer (SMPS) spectrometer. The gaseous concentrations of these carbonyl compounds ranged from the detection limit (approximately 1 pptv) to 154 pptv (630 ng/cu m, 4-oxopentanal), and the particulate concentrations ranged from the detection limit (approximately 3 ng/cu m) to 200 ng/cu m (4-oxopentanal). Although the production processes of these compounds are different from each other, the temporal variations of the gaseous concentrations were quite similar. In addition, the variation was also similar to that of the ambient temperature. Furthermore, gas-to-particle conversion was suggested to be an important removal process of these compounds. We could evaluate the importance of the gas-to-particle conversion as a removal process of the gaseous species by an ADSS measurement. In addition, the results of our experiment indicated that the conversion includes two processes. The first is an adsorption onto the aerosols which have already existed in the atmosphere. The next is dissolution into the water phases in the aerosols. The latter process was particular to water soluble compounds. The measurement allowed us to identify the most likely removal processes of biogenic semivolatile organic compounds (SOCs). In this study, we discuss about these processes of semivolatile and biogenic carbonyls in the forest atmosphere.

Philippin, S.; Scheer, V.; Vogt, R.; Wehner, B.; Wiedensohler, A., 2004, "Variability of non-volatile fractions of atmospheric aerosol particles with traffic influence," *Atmospheric Environment*, **38**(36):6081–6090

Emissions from vehicular traffic are considered to be a major source of anthropogenic submicrometer particles in the urban environment. In this study, volatile and non-volatile number and volume fractions were distinguished in particles sampled for three consecutive days (Friday, Saturday, and Sunday) close to a highway (HW) (A4) in Germany and for one day in the urban area of Aachen, Germany. A volatility tandem differential mobility analyzer (VTDMA) and a thermal denuder (TD) combined with two Scanning Mobility Particle Sizers (SMPS) spectrometer were used to get insight into a size-resolved mixing state of volatile and non-volatile particle fractions. Operating the VTDMA at 280°C, the number size distribution of non-volatile particle cores was determined for initial particle sizes of 30, 50, 80, and 150 nm. The number size distributions from 10 to 400 nm of ambient and non-volatile aerosol particles were measured by using parallel a SMPS and TD/SMPS combination, respectively. Number size distributions measured near the HW showed a bimodal size distribution with a maximum number concentration at particle sizes between 10 and 20 nm. The TD/SMPS results for the HW site revealed that the nucleation mode disappeared after heating (completely volatile) and in total 10--20% by number of traffic-related particles were non-volatile. The VTDMA results revealed that only 22% of the 30 nm particles had a non-

volatile core above the instrument detection limit of 10 nm. With increasing particle diameter, this non-volatile number fraction increased to ca. 60% for 50 nm particles and to approximately 100% for 80 and 150 nm particles. These findings mean that each particle in the upper Aitken and lower accumulation mode range contains a non-volatile core. With increasing traffic influence the number fraction of less-volatile particles representing mainly primary soot emissions increases to 62% for 50 nm particles and 71% for 80 nm particles.

Young, Li-Hao; Keeler, G. J., 2004, "Characterization of Ultrafine Particle Number Concentration and Size Distribution During a Summer Campaign in Southwest Detroit," *Journal of the Air & Waste Management*, **54**(9)

This paper presents results from a study conducted in southwest Detroit from July 20 to July 30, 2002, to characterize ambient ultrafine particles ( $d_p < 0.1 \mu\text{m}$ ), and to examine the effect of local sources and meteorological parameters on the ultrafine number concentration and size distribution. The number concentrations of ambient particles in the size range of 0.01-0.43  $\mu\text{m}$  were obtained from a Scanning Mobility Particle Sizer (SMPS) spectrometer. Meteorological parameters including ambient temperature, relative humidity, wind speed, wind direction, rainfall, and solar radiation flux were also monitored concurrently atop a 10-m tower. On average, ultrafine particles ranged from  $1.4 \times 10^4$  to  $2.5 \times 10^4 \text{ cm}^{-3}$ , with significant diurnal and daily variations, and accounted for 89% of the total number concentration ( $0.01 < d_p < 0.43 \mu\text{m}$ ). Time-series plots of the 5-min number concentrations revealed that peak concentrations often occurred during morning rush hour and/or around solar noon when photochemical activity was at a maximum. The morning traffic-related peak coincided with the  $\text{NO}_x$  peak, whereas the photochemical-related peak correlated with solar radiation flux. On some days, the noon peak concentration was many times higher than the morning peak concentration. Although the number size distribution varied considerably over the course of the study, it typically exhibited one to three modes, with diameters around 0.01, 0.05, and 0.09  $\mu\text{m}$ . Analysis of the influence of wind direction indicated that stationary sources could be one of the contributors to elevated ultrafine particle concentration. Overall, the data indicated that fossil fuel combustion and atmospheric gas-to-particle conversion of precursor gases are the major sources of ultrafine particles in the southwest Detroit area during the summer.

Zhou, Liming; Kim, Eugene; Hopke, Philip K; Stanier, Charles; Pandis, Spyros N, 2004, "Advanced Factor Analysis on Pittsburgh Particle Size-Distribution Data," *Aerosol Science and Technology*, **38**(S1):118-132

Positive matrix factorization (PMF) method was applied to particle size-distribution data acquired during the Pittsburgh Air Quality Study (PAQS) from July 2001 to August 2001. After removing those days with nucleation events, a total of 1632 samples, each with 165 evenly-sized intervals from 0.003 to 2.5  $\mu\text{m}$ , were obtained from Scanning Mobility Particle Sizer (SMPS) spectrometer and Aerodynamic Particle Sizer (APS) spectrometer. The temporal resolution was 15 min. The values for each set of five consecutive-size bins were averaged to produce 33 new size channels. The size distributions of particle number as well as volume were analyzed with a bilinear model. Three kinds of information were used to identify the sources: the number and volume size distributions associated with the factors, the time frequency properties of the contribution of each source (Fourier analysis of source contribution values) and the correlations of the contribution values with the gas-phase data and some composition data. Through these analyses, the sources were assigned as sparse nucleation, local traffic, stationary combustion, grown particles and remote traffic, and secondary aerosol in a sequence of decreasing number concentration contributions. Conditional probability function (CPF) analysis was performed for each source so as to ascertain the likely directions in which the sources were located.

## 2003

Hirabayashi, Motohiro, 2003, "Size distribution of Ultra-Fine Particles in the Roadside Atmospheres," *JSAE Technical Paper No. 20035679*, *JSAE Autumn Conference*, September 17-19, 2003, Tokyo, Japan

Size distribution of ultra-fine particle was measured by using the Scanning Mobility Particle Sizer in the roadside of three places with different traffic conditions, in order to grasp the influence of exhaust ultra-fine particles to the atmospheric environment. The high-concentration, ultra-fine particles under the 50 nm diameter that is thought it originates in the vehicle exhaust, was observed in each roadside atmospheres. It was found that the heavy-duty vehicles have contributed largely for the particles under 50 nm diameter, and the contribution of exhaust soot is large for the particles above 50 nm diameter.

Nessler, R.; Bukowiecki, N.; Henning, S.; Weingartner, E.; Calpini, B.; Baltensperger, U., 2003, "Simultaneous dry and ambient measurements of aerosol size distributions at the Jungfraujoch," *Tellus. Series B: Chemical and Physical Meteorology*, **55B**(3):808-819

In a field campaign at the high-alpine site Jungfraujoch (JFJ, 3580 m asl), in-situ aerosol size distributions were measured simultaneously outdoor at ambient conditions (temperature  $T < -5 \text{ C}$ ) and indoor at dry conditions ( $T = 25 \text{ C}$  and relative humidity  $\text{RH} < 10$  percent) by means of two Scanning Mobility Particle Sizer (SMPS) spectrometers. In addition, measurements of hygroscopic growth factors were performed

with a hygroscopicity tandem differential mobility analyzer (H-TDMA). The measured growth factors, being a monotonic function of the relative humidity (RH), were fitted with a modified Koehler model. A comparison between dry and ambient size distributions shows two main features: First, the dry total number concentration is often considerably smaller (on average 28 percent) than the ambient total number concentration, and is most likely due to the evaporation of volatile material at the higher temperature. These particle losses mainly concern small particles (dry diameter  $D < 100$  nm), and therefore have only a minimal effect on the surface and volume concentrations. A slight correlation between ambient RH and the magnitude of particle loss was observed, but it was not possible to establish an empirical model for a quantification. Second, the dry number size distribution is shifted towards smaller particles, reflecting the hygroscopic behavior of the aerosols. To link the ambient and the dry size distributions we modeled this shift using the H-TDMA measurements and a modified Koehler model. The corrected dry surface and volume concentrations are in good agreement with the ambient measurements for the whole RH range, but the correction works best for  $RH < 80$  percent. The results indicate that size distribution data measured at indoor conditions (i.e., dry and warm) may be successfully corrected to reflect ambient conditions, which are relevant for determining the impact of aerosol on climate.

Noble, C. A.; Mukerjee, S.; Gonzales, M.; Rodes, C. E.; Lawless, P. A.; Natarajan, S.; Myers, E. A.; Norris, G. A.; Smith, L.; Oezkaynak, H.; Neas, L. M., 2003, "Continuous measurement of fine and ultrafine particulate matter, criteria pollutants and meteorological conditions in urban El Paso, Texas," *Atmospheric Environment*, **37**(6):827–840

Continuous measurements of aerosol size distributions were made in El Paso, TX, for a 21 day period in winter 1999. Size distribution measurements were performed at two urban locations in El Paso using two pairs of the Scanning Mobility Particle Sizer and the aerodynamic particle sizer. Complementary measurements also were performed for gas phase pollutants (CO, NO, NO<sub>2</sub>, O<sub>3</sub> and meteorological conditions. Throughout the study, the mean ultrafine particle (those smaller than 0.1  $\mu\text{m}$  in diameter) number concentration was 14,400 particles  $\text{cm}^{-3}$ . There was a significant correlation between CO and both ultrafine and accumulation mode (those between 0.1 and 1  $\mu\text{m}$  in diameter) particle count, with the Pearson correlation coefficient ( $r$ ) values of 0.81 and 0.87, respectively. The correlation between NO and both ultrafine and accumulation mode particle count is also significant, but not as strong as the correlation of CO and the particle concentrations. Most pollutants were found to vary on diurnal cycles and to follow one of two different trends, either vehicular traffic schedules or sunlight intensity. Wind direction was found to have an influence not only on pollutant concentrations, but also on the correlation between pollutants. With southerly winds, CO, NO and NO<sub>2</sub> concentrations were 25–140% greater than when the wind was coming from the north. Likewise, ultrafine and accumulation mode particle concentrations were approximately 100% greater for southerly than for northerly winds.

Palmgren, F.; Waahlin, P.; Kildesoe, J.; Afshari, A.; Fogh, C.L., 2003, "Characterization of particle emissions from the driving car fleet and the contribution to ambient and indoor particle concentrations," *Physics and Chemistry of the Earth, Parts A/B/C*, **28**(8):327–334

The population is mainly exposed to high air pollution concentrations in the urban environment, where motor vehicle emissions constitute the main source of fine and ultrafine particles. These particles can penetrate deep into the respiratory system, and studies indicate that the smaller the particle, the larger the health impacts. The chemical composition, surface reactivity and physical properties are also important. However, the knowledge about chemical and physical properties of particles and the temporal and spatial variability of the smallest particles is still very limited. The present study summarizes the first results of a larger project with the aims to improve the knowledge. The concentration and the emissions of ultrafine particles from petrol and diesel vehicles, respectively, have been quantified using Scanning Mobility Particle Sizer spectrometer of ultrafine particles in the size range 6–700 nm and routine monitoring data from urban streets and urban background in Denmark. The quantification was carried out using receptor modeling. The number size distributions of petrol and diesel emissions showed a maximum at 20–30 nm and non-traffic at about 100 nm. The contribution of ultrafine particles from diesel vehicles is dominating in streets. The same technique has been applied on PM<sub>10</sub>, and about 50 percent contribution from non-traffic. The technique has also been introduced in relation to elemental and organic carbon, and the first data showed strong correlation between traffic pollution and elemental carbon. The outdoor air quality has a significant effect on indoor pollution levels, and we spend most of the time indoors. Knowledge about the influence of ambient air pollution on the concentrations in the indoor environment is therefore crucial for assessment of human health effects of traffic pollution. The results of our studies will be included in air quality models for calculation of human exposure. Preliminary results from our first campaign showed, that the deposition rate of particles in the apartment is negligible in the particle size range 100–500 nm. In the size range below 100 nm the deposition rate increases with decreasing particle diameter to a value of approximately 1/h at 10 nm. The penetration efficiency shows a maximum of 60 percent at 100 nm. More detailed studies of exchange of particles in outdoor/indoor air and the transformation are planned to take place during three next campaigns.

Weijers, E. P.; Kos, G. P. A.; Jongejan, P. A. C., 2003, "Ultrafine particle emissions along motorways. Emission factors, particle numbers and size distributions," *Technical Report, Energy Research Centre of the Netherlands ECN*, Petten (Netherlands), Report No. ECN-C--03-04

The aim of this study is the determination of emission factors for ultrafine particles and size distributions along motorways in the Netherlands. In the experimental campaign particle number concentrations and size distribution in the size range from 13 to 583 nm were measured by condensation particle counters (CPC) and a Scanning Mobility Particle Sizer (SMPS) spectrometer, respectively. The measurements were taken upwind and at various distances downwind of two Dutch motorways of which one site was situated about 20 km north of the city of Amsterdam (A9-motorway) in the open field, and the other within the Amsterdam agglomeration (A4, near the junction with the A10). At each sampling day, concentrations of particulate matter, CO and NO<sub>x</sub> were measured over 20 to 30 min after which the mobile measurement was driven to the next location at another distance. In this study the experimental data collected on eight days are discussed.

## 2002

Abu-Allaban, M.; Coulomb, W.; Gertler, A.W.; Gillies, J.; Pierson, W.R.; Rogers, C.F.; Sagebiel, J.C.; Tarnay, L., 2002, "Exhaust Particle Size Distribution Measurements at the Tuscarora Mountain Tunnel," *Aerosol Science & Technology*, **36**(6):771-789

On-road particle size distributions were measured at the Tuscarora Mountain tunnel on the Pennsylvania Turnpike in May 1999. The data were obtained using a Scanning Mobility Particle Sizer spectrometer. The nucleation modes of the size distributions contained most of the particles on a number concentration basis and exhibited peak diameters ranging from 11 to 17 nm. This observation is consistent with previous calculations and measurements, indicating that significant numbers of ultrafine aerosol particles can be expected in close proximity to busy motorways. The experiment provided 4 case studies for which the tunnel inlet data could be used to correct data obtained at the outlet, allowing for estimates of particle production within the tunnel. Exhaust particle production rates per vehicle kilometer were estimated; the results are presented with the caveat that the measurements were affected by ambient dilution. The 4 case study nucleation mode sizes varied inversely with ambient temperature. The light-duty vehicle contributions to the ultrafine particle distributions were apparently dominated by the heavy-duty vehicle contributions.

Hand, Jenny L.; Kreidenweis, Sonia M., 2002, "A New Method for Retrieving Particle Refractive Index and Effective Density from Aerosol Size Distribution Data," *Aerosol Science & Technology*, **36**(10):1012-1026

As part of the Big Bend Regional Aerosol Visibility and Observation study (BRAVO) in July-October 1999, dry aerosol size distributions were measured over the size range of  $0.05 < D_p < 20 \mu\text{m}$  using a TSI differential mobility analyzer (DMA), a PMS LASAIR 1003 optical particle counter (OPC), and a TSI aerodynamic particle sizer 3320 (APS). Extensive calibrations were performed to characterize the response of the OPC and APS to particles of different size and composition. This paper describes a new method that was developed to align size distributions in the instrument overlap regions, allowing for the retrieval of aerosol real refractive index and effective density. To validate the method, retrieved particle real refractive index was compared with volume weighted model estimates based on measured PM<sub>2.5</sub> chemical composition. The study average retrieved real refractive index was  $m_r = 1.566 - 0.012$ , and the average computed PM<sub>2.5</sub> refractive index was  $m_r = 1.56 - 0.02$ ; the agreement is well within experimental uncertainties. The average value of computed PM<sub>2.5</sub> bulk density was  $1.85 - 0.14 \text{ g cm}^{-3}$ . The average value of retrieved effective density, a function of particle dynamic shape factor, was  $1.56 - 0.12 \text{ g cm}^{-3}$ . The comparisons of effective density to computed bulk density suggested an average particle dynamic shape factor of  $h = 1.2$ . Sensitivity studies showed that real refractive index could be retrieved with uncertainties on the order of 2-3%, and effective density was retrieved with uncertainties on the order of 20-30%.

Hitchins, J.; Morawska, L.; Gilbert, D.; Jamriska, M., 2002, "Dispersion of particles from vehicle emissions around high- and low-rise buildings," *Indoor Air*, **12**(1):64-71

Understanding the distribution of outdoor pollutants around a building envelope, generated by sources located in its vicinity, is important when choosing the location of building ventilation system intakes, as well as for quantifying the exposure of people living or working in the building. A systematic experimental characterization of the number concentration of submicrometer particles was undertaken around the envelope of six buildings (both low- and high-rise) at different distances from a road (the main pollution source). The concentrations were measured using two TSI Scanning Mobility Particle Sizing. PM<sub>2.5</sub> concentrations were also monitored around the low-rise buildings using two TSI DustTrak™ monitors. For the three high rise buildings the concentration of fine and ultra-fine particles decreased in most cases to about 50-60% from the approximate ground level readings (between heights of 0 to 6 m), to full building height (from 24 to 33 m above the ground). Measurements of submicrometer particle

number concentrations as well as PM<sub>2.5</sub> fraction in the envelope around low-rise isolated buildings did not show any significant trends from the front to the rear of the building. The sensitivity of PM<sub>2.5</sub> measurements to a small number of larger particles, possibly from sources other than vehicle emissions, was observed.

Sarnat, J. A.; Coull, B. A.; Koutrakis, P.; Long, C. M.; Schwartz, J.; Suh, H. H., 2002, "Using sulfur as a tracer of outdoor fine particulate matter," *Environmental Science & Technology*, **36**(24):5305–5314

Six homes in the metropolitan Boston area were sampled between 6 and 12 consecutive days for indoor and outdoor particle volume and mass concentrations, particle elemental concentrations, and air exchange rates (AERs). Indoor/outdoor (I/O) ratios of nighttime (i.e., particle non-indoor source periods) sulfur, PM<sub>2.5</sub> and the specific particle size intervals were used to provide estimates of the effective penetration efficiency. Mixed models and graphical displays were used to assess the ability of the I/O ratios for sulfur to estimate corresponding I/O ratios for PM<sub>2.5</sub> and the various particle sizes. Results from this analysis showed that particulate sulfur compounds were primarily of outdoor origin and behaved in a manner that was representative of total PM<sub>2.5</sub> in Boston, MA. These findings support the conclusion that sulfur can be used as a suitable tracer of outdoor PM<sub>2.5</sub> for the homes sampled in this study. Sulfur was more representative of particles of similar size (0.06-0.5 μm), providing evidence that the size composition of total PM<sub>2.5</sub> is an important characteristic affecting the robustness of sulfur-based estimation methods.

Shen, S.; Jaques, P. A.; Zhu, Y. F.; Geller, M. D.; Sioutas, C., 2002, "Evaluation of the SMPS-APS system as a continuous monitor for measuring PM 2.5, PM 10 and coarse (PM 2.5-10) concentrations," *Atmospheric Environment*, **36**(24):3939–3950

Respirable particulate matter (PM) has been linked to mortality and morbidity by a variety of epidemiological studies. This research has led to the creation of a new PM standard for particles with diameters <2.5 μm (PM<sub>2.5</sub>). Since the conclusion of these studies, many leaps have been made in the realm of continuous particle measurement. Because the literature is still dominated by 24-hour averaged data, the US Environmental Protection Agency still uses this time average as the basis of its federal reference method, despite the fact that PM varies on much shorter time intervals. The purpose of this work is to compare the Scanning Mobility Particle Sizer spectrometer and Aerodynamic Particle Sizer tandem (SMPS-APS) to other continuous PM measurement devices and to time-integrated mass samplers. The instruments used for comparison include the DataRAM nephelometer, Micro-Orifice Uniform Deposit Impactor (MOUDI), and Partisol Dichotomous Sampler. The data was collected over 4–5 months at various sites in the Los Angeles basin. The results show excellent agreement between the SMPS-APS and the mass based MOUDI and Partisol samplers for PM<sub>2.5</sub>. The DataRAM and SMPS-APS continuous monitors show robust correlation with each other when relative humidity <70%. The coarse fraction (PM<sub>2.5-10</sub>) measured by the Partisol, however, does not track well with the same size range measured by the APS spectrometer. Several sources of sampling error are discussed to account for this. Finally, mass concentrations collected in individual size ranges of the MOUDI were compared with those determined by the SMPS. While the size ranges from 0.32 to 10 μm agree between samplers, the size ranges <0.32 μm are significantly different from MOUDI to SMPS, probably due to the differences between the aerosol sizing principles underlying each technique.

Wiegand, A., 2002, "Dirty air Down Under," *Environmental Protection*, **13**(9):22-25

In 1992, the Environmental Aerosol Laboratory (EAL) was established within the Facility of Science at Queensland University of Technology, specializing in environmental science with particular emphasis on fine and ultrafine particles. The city of Brisbane in particular is expanding rapidly necessitating careful planning for the future. Smog chamber experiments combined with airshed (a large area over which air quality and movement can be considered) modeling are described. These are applicable in the planning of infrastructure with a view to the prevention of exposure of large populations to pollutants. Recent developments in computer technology including high-performance graphing and data analysis tools such as Origin Corp and tools for the study of the formation of ultrafine particles such as high resolution Scanning Mobility Particle Sizers (TSI Inc) have dramatically increased the capacity of researchers to conduct such modeling studies.

Zhu, Y. F.; Hinds, W. C.; Kim, S.; Shen, S.; Sioutas, C., 2002, "Study of ultrafine particles near a major highway with heavy-duty diesel traffic," *Atmospheric Environment*, **36**(27):4323–4335

An investigation to characterize the ultrafine particles emitted from heavy-duty diesel trucks, on Interstate 710 in Los Angeles, is presented. Particle number concentration and size distribution, from 6-220 nm, were measured by a condensation particle counter (CPC) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. The output data were analyzed by the Aerosol Instrument Manager Software. Measurements were taken at 17, 20, 30, 90, 150 and 300 m downwind and 200 m upwind from the centre of the freeway. Wind speed and direction were measured at 1 min intervals and logged by computer. The

black carbon (BC) concentrations were measured every 5 min with a dual beam Aethalometer and CO with a near-continuous monitor. Atmospheric dilution and coagulation are important in the changes in particle number concentration and in particle size distribution.

Zhu, Y.; Hinds, W. C.; Kim, S.; Sioutas, C., 2002, "Concentration and size distribution of ultrafine particles near a major highway," *Journal of the Air & Waste Management Association*, **52**(9):1032-1042

Motor vehicle emissions usually constitute the most significant source of ultrafine particles (diameter <0.1  $\mu\text{m}$ ) in an urban environment, yet little is known about the concentration and size distribution of ultrafine particles in the vicinity of major highways. In the present study, particle number concentration and size distribution in the size range from 6 to 220 nm were measured by a condensation particle counter (CPC) and a Scanning Mobility Particle Sizer (SMPS) spectrometer, respectively. Measurements were taken 30, 60, 90, 150, and 300 m downwind, and 300 m upwind, from Interstate 405 at the Los Angeles National Cemetery. At each sampling location, concentrations of CO, black carbon (BC), and particle mass were also measured by a Dasibi CO monitor, an aethalometer, and a DataRam, respectively. The range of average concentration of CO, BC, total particle number, and mass concentration at 30 m was 1.7-2.2 ppm, 3.4-10.0  $\mu\text{g}/\text{m}^3$ ,  $1.3-2.0 \times 10^5/\text{cm}^3$ , and 30.2-64.6  $\mu\text{g}/\text{m}^3$ , respectively. For the conditions of these measurements, relative concentrations of CO, BC, and particle number tracked each other well as distance from the freeway increased. Particle number concentration (6-220 nm) decreased exponentially with downwind distance from the freeway. Data showed that both atmospheric dispersion and coagulation contributed to the rapid decrease in particle number concentration and change in particle size distribution with increasing distance from the freeway. Average traffic flow during the sampling periods was 13,900 vehicles/hr. Ninety-three percent of vehicles were gasoline-powered cars or light trucks. The measured number concentration tracked traffic flow well. Thirty meters downwind from the freeway, three distinct ultrafine modes were observed with geometric mean diameters of 13, 27, and 65 nm. The smallest mode, with a peak concentration of  $1.6 \times 10^5/\text{cm}^3$ , disappeared at distances greater than 90 m from the freeway. Ultrafine particle number concentration measured 300 m downwind from the freeway was indistinguishable from upwind background concentration. These data may be used to estimate exposure to ultrafine particles in the vicinity of major highways.

## 2001

Lawless, P. A.; Rodes, C. E.; Evans, G.; Sheldon, L.; Creason, J., 2001, "Aerosol Concentrations During the 1999 Fresno Exposure Studies as Functions of Size, Season, and Meteorology," *Aerosol Science & Technology*, **34**(1):66-74

Two 1999 Fresno exposure studies took place in February (winter season) and April/May (spring season) for periods of four weeks each. During that time, nearly continuous measurements of outdoor aerosol concentrations were made with a TSI Scanning Mobility Particle Sizer (SMPS) spectrometer and a PMS optical particle counter (LASX). These instruments provide particle size distribution information from about 0.01 to 3  $\mu\text{m}$ . Although the primary concentration measurements were performed with integrated samplers, the SMPS/LASX combination provided useful information on the real-time variations of concentration and particle size distributions. This paper describes the agreement between the real-time instrumentation and the integrated measurements and examines some of the information available from the SMPS/LASX combination. In particular, two different types of aerosols contributing to  $\text{PM}_{2.5}$  were deduced from the instrumental data.

Morawska, L.; He, Congrong; Hitchins, J.; Gilbert, D.; Parappukaran, S., 2001, "The relationship between indoor and outdoor airborne particles in the residential environment," *Atmospheric Environment*, **35**(20):3463-3473

The relationship between indoor and outdoor airborne particles was investigated for 16 residential houses located in a suburban area of Brisbane, Australia. The submicrometer particle numbers were measured using the Scanning Mobility Particle Sizer, the larger particle numbers using the Aerodynamic Particle Sizer and an approximation of  $\text{PM}_{2.5}$  was also measured using a DustTrak™ monitor. The measurements were conducted for normal and minimum ventilation conditions using simultaneous and non-simultaneous measurement methods designed for the purpose of the study. Comparison of the ratios of indoor to outdoor particle concentrations revealed that while temporary values of the ratio vary in a broad range from 0.2 to 2.5 for both lower and higher ventilation conditions, average values of the ratios were very close to one regardless of ventilation conditions and of particle size range. The ratios were in the range from 0.78 to 1.07 for submicrometer particles, from 0.95 to 1.0 for supermicrometer particles and from 1.01 to 1.08 for  $\text{PM}_{2.5}$  fraction. Comparison of the time series of indoor to outdoor particle concentrations shows a clear positive relationship existing for many houses under normal ventilation conditions (estimated to be about and above  $2\text{h}^{-1}$ , but not under minimum ventilation conditions (estimated to be about and below  $1\text{h}^{-1}$ ). These results suggest that for normal ventilation conditions, outdoor particle concentrations could be used to predict instantaneous indoor particle concentrations but

not for minimum ventilation, unless air exchange rate is known, thus allowing for estimation of the "delay constant".

Shi, J. P.; Evans, D. E.; Khan, A. A.; Harrison, R. M., 2001, "Sources and concentration of nanoparticles (less 10 nm diameter) in the urban atmosphere," *Atmospheric Environment*, **35**(7):1193-1202

Air at four sites was monitored and the nanoparticles size and concentrations determined using condensation particle counters (particles less than 7 nm) and ultrafine condensation particle counters (particles larger than 3 nm). A Scanning Mobility Particle Sizer was used to measure particle size distribution in the range 9.47-359 nm. An epiphaniometer was used to measure the particle Fuchs surface area. SO<sub>2</sub>, NO and NO<sub>2</sub> were determined using analyzers. A mini meteorological station was used to measure wind speed, direction, temperature, relative humidity, UVA and UVB in the measuring site. Results are presented graphically, tabulated and discussed.

Vette, Alan F.; Rea, Anne W.; Lawless, Philip A.; Rodes, Charles E.; Evans, Gary; Highsmith, V. Ross; Sheldon, Linda, 2001, "Characterization of Indoor-Outdoor Aerosol Concentration Relationships during the Fresno PM Exposure Studies," *Aerosol Science & Technology*, **34**(1):118-126

Particle size distributions were measured indoors and outdoors of a single, detached residence during the Fresno particulate matter exposure studies in winter (February 1-28, 1999) and spring (April 18-May 16, 1999). Data was collected for particle sizes ranging from about 0.01 to 2.5 µm. These data were used to investigate the temporal relationships between indoor and outdoor aerosol concentrations and to determine particle deposition rates and penetration factors for discrete particle sizes. Indoor/outdoor aerosol concentration ratios for particle sizes > 1 µm were diurnally variable with highest ratios occurring during daytime (6:00-18:00) due to resuspension from indoor activities. Daytime and nighttime (19:00-7:00) aerosol concentration ratios were very similar for particle sizes < 1 µm. Particle deposition rates were determined by measuring the decay in indoor aerosol concentrations after indoor levels were elevated by infiltration of native ambient aerosols. Deposition rates varied depending on particle size and were consistent with model results up to about 0.4 µm. The experimentally determined deposition rates were considerably higher than model results at larger particle sizes, suggesting the possibility of an additional indoor loss mechanism. Penetration factors were determined to be less than unity for all particle sizes and ranged from 0.5 to 0.9.

## 2000

Abt, E.; Suh, H. H.; Catalano, P.; Koutrakis, P., 2000, "Relative contribution of outdoor and indoor particle sources to indoor concentrations," *Environmental Science and Technology*, **34**(17):3579-3587

The effect of indoor particle sources on indoor particle size distributions and concentrations was previously investigated using real-time indoor and outdoor particle size distribution data collected in four homes in Boston in 1996. These data demonstrated the importance of indoor sources (i.e., cooking, cleaning, and movement of people) and air exchange rates on observed indoor concentrations. As part of the continued analyses of these data, a simple physical model was used to determine the source emission and infiltration rates for specific particle sizes. Decay rates were also estimated. Cooking, cleaning, and indoor work (characterized by movement of people) significantly increased PM<sub>(0.7-10)</sub> concentrations by 0.27, 0.27, and 0.25 µm<sup>3</sup> cm<sup>-3</sup> min<sup>-1</sup>, respectively. Cooking was the only variable significantly associated with generation of particles less than 0.5 µm in diameter. Outdoor particles (0.02-0.5 and 0.7-10 µm) were found to contribute significantly to indoor particle levels. Effective penetration efficiencies ranged from 0.38 to 0.94 for 0.02-0.5 µm particles and from 0.12 to 0.53 for 0.7-10 µm particles. Estimates for 0.7-10 µm particles decreased with increasing particle size, reflecting the influence of deposition losses from gravitational settling. The real-time particle size distribution data in conjunction with time-activity information provides valuable information on the origin and fate of indoor particles.

Abu-Allaban, M.; Gertler, A. W.; Rogers, C. F.; Gillies, J. A., 2000, "Measurement of ultrafine particle emissions in a highway tunnel," Recent advances in the science and management of air toxics : proceedings of the joint international specialty conference, exhibition and workshop of the Air and Waste Management Association, Abstract only; PP. 15, 300 pages, *Published by Air and Waste Management Association*, Edmonton, AB (Canada)

In May 1999, a six day experiment was conducted at the Tuscarora Mountain tunnel on the Pennsylvania Turnpike to measure particle number-size distributions and size-segregated particle chemistry at the entrance and exit of the tunnel. A Scanning Mobility Particle Sizer (SMPS) spectrometer was used to measure the particle size distributions over a particle diameter range from 10 to 300 nanometers. The ultrafine particle chemistry was determined from afterfilter measurements using a Davis Rotating-drum Universal-size-cut Monitoring sampler. Heavy-duty diesel vehicles were found to contribute to the overall fleet composition by a fraction of 12 to 85 per cent. The SMPS data suggested that the aerosol was a mixture of three or more components. Diluted mobile source exhaust was usually present as a result of both heavy- and light-duty vehicle traffic. A non-mobile background aerosol was also observed. SMPS

data from the tunnel exit showed characteristic size spectra for fresh vehicle exhaust for the heavy and light duty fractions. The size spectra demonstrate dominant nucleation modes, smaller accumulation modes and elevated particle number concentrations. It was hypothesized that the nucleation mode peak diameter decreases somewhat with a decrease in the heavy-duty vehicle fraction. Sulfur was the most abundant element in the particles having diameters of 70 nanometers or less.

Bukowiecki, N.; Henning, S.; Hoffer, A.; Weingartner, E.; Baltensperger, U., 2000, "Cloud and aerosol characterization experiment in the free troposphere (CLACE) - a field experiment at the Jungfraujoch (3580 m ASL)," *Journal of Aerosol Science*, **31**(S1):S11-S12

The CLACE (cloud and aerosol characterization experiment in the free troposphere) aims to: examine cloud formation processes under different meteorological conditions; characterize organic aerosol compounds; compare particle size distributions measured under different ambient conditions; and determine hygroscopic properties of aerosols. Particle size distributions of the total and interstitial aerosol were measured with three different Scanning Mobility Particle Sizer (SMPS) systems operated with a closed loop.

Henning, S.; Weingartner, E.; Baltensperger, U.; Gaggeler, H. W., 2000, "Investigation of the interaction between aerosol and clouds at the Jungfraujoch (3580 m ASL)," *Journal of Aerosol Science*, **31**(S1):S13-S14

Two cloud and aerosol characterization field campaigns were conducted at the Jungfraujoch to study the effects of aerosol properties on cloud formation. A Scanning Mobility Particle Sizer was used to measure aerosol number size distributions of the total aerosol, and interstitial aerosol in two size classes (TSP and PM<sub>1</sub>). Assuming a pure ammonium sulfate particles with supersaturation equals 0.4% is needed to activate particles, a 50 nm diameter was observed for 50% scavenging DP<sub>50</sub>.

Hitchins, J.; Morawska, L.; Wolff, R.; Gilbert, D., 2000, "Concentrations of submicrometer particles from vehicle emissions near a major road, *Atmospheric Environment*, **34**(1):51-59

As part of a program of study to assess the exposure risks related to particulate matter in the outdoor environment, number concentrations of particles from vehicle emissions were measured at increasing distances from a major road. Particles in the size range from 0.015 to 0.697  $\mu\text{m}$  were measured with the Scanning Mobility Particle Sizer (SMPS) spectrometer and in the size range from 0.5 to 20  $\mu\text{m}$ , with the Aerodynamic Particle Sizer (APS) spectrometer. In addition to number concentration measurements, an approximation of PM<sub>2.5</sub> fraction was obtained using a DustTrak™ monitor (simple photometer). The measurements conducted at distances from the road ranging from 15 to 375 m showed, that for conditions where the wind is blowing directly from the road, the concentration of fine and ultrafine particles decays to around half of the maximum (measured at the closest point to the road) at a distance of approximately 100-150 m from the road. For the wind blowing parallel to the road, the reduction to half of the concentration occurs at 50-100 m. There is no effect on total particle number concentration at a distance greater than 15 m from the road when the wind is blowing towards the road and away from the sampling points. Total number concentrations of larger particles measured were not significantly higher than the average values for the urban environment, and decrease with distance from the road, reaching about 60% at 150 m from the road for wind from the road. PM<sub>2.5</sub> levels also decrease with distance to around 75% for wind from the road and to 65% for wind parallel to the road, at a distance of 375m.

Hung, H. F.; Wang, C. S., 2000, "Characterization of ultrafine particles in Taipei aerosols," *Journal of Aerosol Science*, **31**(S1):S248-S249

The mass and number concentrations, and chemical compositions of atmospheric ultrafine particles in an urban environment were measured. Samples of ultrafine particles were taken from a third-floor balcony, a courtyard, the vicinity of a bus stop, and in an underpass. Results indicate that samples collected at the courtyard, which were mainly from vehicular exhausts, has higher mass concentrations of ultrafine particles and the main carbonaceous material was organic carbon. The results imply that the vehicular exhaust is the major source of ultrafine particles in the City of Taipei.

Roberts, G. C.; Artaxo, P.; Andreae, M. O., 2000, "Chemistry and role of cloud condensation nuclei in the Amazon Basin," *Journal of Aerosol Science*, **31**(S1):S62-S63

The cloud condensation nuclei (CCN) interactions for cloud formation in the Amazon Basin is discussed. The aerosol measurements are carried out to study the chemical and physical properties of the CCN. Warm precipitating clouds in the Amazon Basin have been observed. The effect of organic material on CCN activity and the growth of cloud droplets are discussed. The number and mass size distributions for CCN activation are measured using a Scanning Mobility Particle Sizer and a multiple stage cascade impactor. The factors affecting the heat flux in the rainforests are presented. The modifications of CCN concentrations contribute to meteorological cycle and affect the climate in Amazon Basin.



Harrison, R. M.; Jones, M.; Collins, G., 1999, "Measurements of the physical properties of particles in the urban atmosphere," *Atmospheric Environment*, **33**(2):309-321

Measurements of the physical properties of particles in the atmosphere of a UK urban area have been made, including particle number count by condensation nucleus counters with different lower particle size cut-offs; particle size distributions using a Scanning Mobility Particle Sizer; total particle Fuchs surface area using an epiphaniometer and particle mass using Tapered Element Oscillating Micro-balance (TEOM) instruments with size selective (PM<sub>10</sub> and PM<sub>2.5</sub>) inlets. Mean particle number counts at three sites range from  $2.86 \times 10^4$  to  $9.60 \times 10^4 \text{ cm}^{-3}$ . A traffic-influenced location showed a substantially higher ratio of particle number to PM<sub>10</sub> mass than a nearby background location despite being some 70 m from the roadway. Operating two condensation nucleus counters in tandem to determine particles in the 3-7 nm size range by difference showed significant numbers of particles in this range, apparently related to homogeneous nucleation processes. Measurements with the Scanning Mobility Particle Sizer showed a clear difference between roadside size distributions and those at a nearby background location with an additional mode in the roadside samples below 10 nm diameter. Particle number counts were found to show a significant linear correlation with PM<sub>10</sub> mass ( $r^2 = 0.44$ ;  $n = 44$  for 24 h data at an urban background location), although during one period of high pollution a curvilinear relationship was found. Measurements of the diurnal variation in PM<sub>10</sub> mass, particle number count and Fuchs surface area show the same general pattern of behavior of the three variables, explicable in terms of vehicle emission source strength and atmospheric dispersion, although the surface area growth was out of phase with the particle number and mass. It appears that particle number gives the clearest indication of recent road traffic emissions.

Holub, R. F.; Reimer, G. M.; Hopke, P. K.; Hovorka, J.; Krcmar, B.; Smrz, P. K., 1999, "Geo-aerosols: their origin, transport and paradoxical behavior: a challenge to aerosol science," *Journal of Aerosol Science*, **30**(S1):S111-S112

Aerosols in the range 3 to 200 nm in a cave and in an experimental mine were measured using Diffusion Particle Sizer (DPS) and Scanning Mobility Particle Sizer (SMPS) spectrometer. The areas were also sampled for volatile organics by means of TENEX adsorbers, and for the filterable inorganics and organics by means of porous membrane filters. Some of the filters were measured by means of electron microscope and gas chromatography to determine the concentration and composition of the inert aerosols. The concentration and composition of the radioactive aerosols was measured by means of standard nuclear counting equipment. Simultaneously, the outdoor inert and radioactive aerosols were also sampled. The results for both DPS (cave) and SMPS (mine) were basically the same.

Ji P.; Khan, A. A.; Harrison, R. M.; Harrison, R. M.; Hamilton, R. S., 1999, "Measurements of ultrafine particle concentration and size distribution in the urban atmosphere," *Science of the Total Environment*, **235**(1-3):51-64

Particle size distributions were measured at three adjacent sites in Birmingham: a busy roadside (A38); 30 m away from the road and a nearby urban background site. Two Scanning Mobility Particle Sizers, (SMPS) spectrometer, an electrical low pressure impactor (ELPI), a condensation particle counter and a thermophoretic precipitator were employed to measure and collect particles. Excellent agreement on the number weighted size distribution was found between the SMPS and ELPI, as well as with sizes measured by transmission electron microscopy. The average number concentration at roadside measured on four separate days was between  $1.6$  and  $1.9 \times 10^5 \text{ cm}^{-3}$  with similar size distributions and more than half of measured particles smaller than 30 nm. Traffic was the main source of ultrafine particles at the roadside. A 24-h average of  $2.68 \times 10^4$  plus or minus  $1.29 \times 10^4 / \text{cm}^3$  was measured, which is close to that in Hughes et al. (Physical and chemical characterization of atmospheric ultrafine particles in the Los Angeles area. *Environ Sci Technol* 1998; **32**:1153-1161) in Pasadena, CA, USA. Total particle number concentration declined downwind of the traffic, faster than the mass concentration. Model calculations show that dilution with background air is the main mechanism for the rapid drop in particle number concentration and change in particle size distribution when moving away from traffic.

Morawska, L.; Thomas, S.; Gilbert, D.; Greenaway, C.; Rijnders, E., 1999, "A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road," *Atmospheric Environment*, **33**(8):1261-1274

Epidemiological studies are consistently reporting an association between fine particulate pollution and ill-health. Motor vehicle emissions are considered to be the main source of fine particles in ambient urban air of cities which are not directly influenced by industrial emissions. The aim of this work was to assess the influence of a major arterial road on concentration levels of airborne fine particles in its vicinity. Measurements of over 500 particle size distributions in the particle size range 16-626 nm, were made using two Scanning Mobility Particle Sizer (SMPS) spectrometer. A subsequent comparison of the recorded values from differing locations is discussed, with reference made to topographic and climatic

influences. Both horizontal and vertical profile measurements of fine particle number size distributions are described; the combination of the two yielding information as to the relative exposures of occupants of buildings in the vicinity of a major arterial route. With the exception of measurements in close proximity to the freeway (about 15 m), the horizontal profile measurements did not provide any evidence of a statistically significant difference in fine particle number concentration with respect to distance at ground level up to a distance of 200 m within the study area. The vertical profile measurements also revealed no significant correlation between particle concentration and height. However, for buildings in the immediate proximity to the arterial road (about 15 m) concentrations around the building envelope are very high, comparable to those in the immediate vicinity of the road, indicating undiluted concentrations drawn directly from the freeway. This finding has a significant implication for management of indoor air quality in the buildings located in the immediate vicinity of major roads.

Morawska, L.; Thomas, S.; Jamriska, M.; Johnson, G., 1999, "The modality of particle size distributions of environmental aerosols," *Atmospheric Environment*, **33**(27):4401–4411

Epidemiological studies are consistently reporting an association between fine particulate pollution and ill-health. Motor vehicle emissions are considered to be the main source of fine particles in ambient urban air of cities which are not directly influenced by industrial emissions. The aim of this work was to assess the influence of a major arterial road on concentration levels of airborne fine particles in its vicinity. Measurements of over 500 particle size distributions in the particle size range 16–626 nm, were made using two Scanning Mobility Particle Sizer (SMPS) spectrometers. A subsequent comparison of the recorded values from differing locations is discussed, with reference made to topographic and climatic influences. Both horizontal and vertical profile measurements of fine particle number size distributions are described; the combination of the two yielding information as to the relative exposures of occupants of buildings in the vicinity of a major arterial route. With the exception of measurements in close proximity to the freeway (about 15 m), the horizontal profile measurements did not provide any evidence of a statistically significant difference in fine particle number concentration with respect to distance at ground level up to a distance of 200 m within the study area. The vertical profile measurements also revealed no significant correlation between particle concentration and height. However, for buildings in the immediate proximity to the arterial road (about 15 m) concentrations around the building envelope are very high, comparable to those in the immediate vicinity of the road, indicating undiluted concentrations drawn directly from the freeway. This finding has a significant implication for management of indoor air quality in the buildings located in the immediate vicinity of major roads.

## 1997

Lee, W.-M. G.; Shih, P.-M.; Wang, C.-S., 1997, "The influence of relative humidity on the size of atmospheric aerosol," *J. Environ. Sci. Health, Part A: Environ. Sci. Eng. Toxic Hazard. Subst. Control*, **A32**(4):1085–1097

The relative humidity of ambient air is very often over 75% throughout the year in Taiwan. Thus, the influence of relative humidity on the growth of hygroscopic aerosols in the atmosphere is particularly important. In this study, a TDMA system (Tandem Differential Mobility Analyzer) equipped with newly developed a SMPS (Scanning Mobility Particle Sizer) was used to observe the sizes of submicron ammonium sulfate aerosols, from 63.8nm to 224.7nm, at five controlled relative humidities, 50%, 75%, 81%, 85% and 89%. It is the first time that the hygroscopic growths and deliquescence points of submicron aerosols of ammonium sulfate have ever been closely observed by use of the SMPS. The experimental results showed that the abrupt size changes of aerosols at their deliquescence points can be more precisely observed than in previous studies. It was also shown that there was no apparent growth in an ammonium sulfate aerosol until relative humidities was over 75%. At relative humidities between 81% and 89%, the growth ratio of size was about a factor of 1.3 to 1.6 and the water content of ammonium sulfate aerosols by mass was about 47% to 66%.

## 1995

Weingartner E.; Keller C.; Stahel W.A.; Baltensperger U.; Burtscher H., 1995, "Field Study of Vehicle Emission Factors in a Road Tunnel," *Journal of Aerosol Science*, **26**(S1):669–670

Within the scope of measurements in the Gubrist tunnel, a 3'250 m long highway tunnel near Zurich, Switzerland, from 20.9 to 26.9.1993, continuous measurements of aerosol emissions were performed. These measurements were part of the Genemis-project, an EUROTRAK sub project.

## 1994

Zhang, Xinqiu; Turpin, Barbara J.; McMurray, Peter H.; Hering, Susanne V.; Stolzenburg, Mark R., 1994, "Mie Theory Evaluation of Species Contributions to 1990 Wintertime Visibility Reduction in the Grand Canyon," *Air & Waste Management Association*, **44**:153-162

Contributions of the major fine particle species to light scattering were evaluated using Mie theory from measurements of size-resolved chemistry and particle hygroscopicity obtained during the 1990 NGS Visibility Study at Hopi Point, Grand Canyon, from January 9, 1990 through March 31, 1990. It was found that scattering efficiencies of particulate carbon mass ranged from 1.5 to 8 m<sup>2</sup> per gram of carbon particle mass (assumed equal to 1.4 times carbon mass), with an average value of 5.4 ± 1.5 m<sup>2</sup>/g. Sulfur size distributions, which are available for the entire 80-day study, show that sulfate scattering efficiencies depend on both relative humidity and on median particle size, and that 'dry' (RH <60 percent) sulfur scattering efficiencies range from about 1.5 to 4.5 m<sup>2</sup> per gram of ammonium sulfate. This range reflects the wide variability in median sulfur particle size (0.07 to 0.66 μm) that was observed during the study. For the 20-day period during which size distribution data for the major fine particle species are available, the contributions of soil dust, carbon, sulfates, and nitrates to scattering are 0.38, 5.0, 4.8 and 0.7 Mm<sup>-1</sup> respectively, and average daily percentage contributions are 4.2, 50.1, 38.5, and 6.9 percent. Sulfur mass median diameters >0.3 μm were only found during periods of high relative humidity, but not all high humidity periods had large sulfur mass median diameters. Sulfate scattering efficiencies for >60 percent RH were smaller for instances of direct transport of <36 hr from Navajo Generating Station to Hopi Point than for the regional background. Scattering efficiencies for longer transport times resembled the regional background.

## 1993

Covert, David S.; Heintzenberg, Jost, 1993, "Size Distributions and Chemical Properties of Aerosol at Ny Ålesund, Svalbard," *Atmospheric Environment*, **27A**(17/18):2989-2997

Physical and chemical parameters of the arctic aerosol were investigated at Ny Ålesund, Svalbard, in March and April 1989 in connections with the third Arctic Gas and Aerosol Project (AGASP III). The number size distribution of the particles was measured over the range of 0.02-1.0 μm. Filter samples were analyzed for elemental composition and two integral chemical properties, hygroscopic growth and volatility, were measured. Along with the latter measurements, the distribution of these properties at specific particle sizes, i.e., the degree of internal mixing was determined. Both clean, marine conditions and 'arctic haze' episodes were included in the series of measurements. The number size distribution indicated that the aerosol was well aged based on its narrowness and the relative low concentration of nuclei mode particles. It had a number mode at 0.22 μm diameter and geometric standard deviation of 1.4. Generally the particles exhibited uniform gyroscopic growth properties, i.e., they were largely internally mixed. The growth factor was 1.45 at 90% relative humidity. A volatile fraction varied from particle to particle, i.e. the particles were externally mixed with respect to volatility.

Li, Chih-Shan; Lin, Wen-Hai; Jenq, Fu-Tein, 1993, "Characterization of Outdoor Submicron Particles and Selected Combustion Sources of Indoor Particles," *Atmospheric Environment*, **27B**(4):413-424

Environmental tobacco smoke, mosquito-coil smoke, and joss stick smoke are the major indoor combustion sources in Asian countries. Field evaluations of the size distributions of outdoor submicron particles and selected combustion sources of indoor particles were conducted in an apartment in Taipei urban area. The size distributions of submicron aerosols were determined by a high resolution particle sizer, which could measure the particles in the size range of 0.017-0.886 μm. The particle sizer contains a differential mobility analyzer (TSI 3071) and a condensation particle counter (TSI 3022). The number concentrations of the indoor and outdoor submicron particles varied from 14,000 to 150,000 cm<sup>-3</sup> and from 10,000 to 45,000 cm<sup>-3</sup>, respectively. The changes of the size distributions and the number concentrations of submicron aerosols before, during, and after the aerosol generations were compared. The average number median diameters of environmental tobacco smoke, smoldering cigarettes, mosquito-coil smoke, joss stick smoke, the indoor typical conditions, and the outdoor typical conditions were 0.090, 0.085, 0.094, 0.084, 0.091 and 0.054 μm respectively. Regarding the surface area-weighted size distributions, the average surface median diameters of these conditions were 0.229, 0.219, 0.282, 0.188, 0.224 and 0.221 μm, respectively. In addition, the average volume median diameters were 0.338, 0.332, 0.398, 0.289, 0.330 and 0.340 μm, respectively. These indoor combustion sources did generate a significant number of the ultrafine and submicron particle which have higher deposition probabilities in the respiratory tract. Further health evaluations of the submicron particles from these combustion sources are needed.

Hoppel, William A.; Larson, Reginald; Vietti, Michael A., 1984, "Aerosol Size Distributions at a Site on the East Coast of the United States," *Atmospheric Environment*, **18**(8):1613-1621

Aerosol size distributions covering the range from 0.006 to 2.2  $\mu\text{m}$  radius were measured with the NRL differential mobility analyzer and an optical particle counter during a 12-day period in June of 1979 at Wallops Island, VA. Changes in the size distribution were associated primarily with the history of the air mass as indicated by synoptic conditions. The coastal location made it possible to contrast continental and marine conditions. On one occasion a high pressure system moved from over the Great Lakes, across New England, and out to sea. Circulation around this high first brought to the site air which had moved down the east coast of the U.S., and later air from the east with increasingly longer trajectories over water. As the length of the trajectory over water increased, a decrease in small particles was clearly evident.

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## Combustion Source Emissions

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### 2013

Asimakopoulou, A., Daskalos, E., Lewinski, N., Riediker, M., Papaioannou, E., and Konstandopoulos, A. G., 2013, "Development of a dose-controlled multiculture cell exposure chamber for efficient delivery of airborne and engineered nanoparticles," *Journal of Physics: Conference Series*, **429**(012023):

In order to study the various health influencing parameters related to engineered nanoparticles as well as to soot emitted by Diesel engines, there is an urgent need for appropriate sampling devices and methods for cell exposure studies that simulate the respiratory system and facilitate associated biological and toxicological tests. The objective of the present work was the further advancement of a Multiculture Exposure Chamber (MEC) into a dose-controlled system for efficient delivery of nanoparticles to cells. It was validated with various types of nanoparticles (Diesel engine soot aggregates, engineered nanoparticles for various applications) and with state-of-the-art nanoparticle measurement instrumentation to assess the local deposition of nanoparticles on the cell cultures. The dose of nanoparticles to which cell cultures are being exposed was evaluated in the normal operation of the in vitro cell culture exposure chamber based on measurements of the size specific nanoparticle collection efficiency of a cell free device. The average efficiency in delivering nanoparticles in the MEC was approximately 82%. The nanoparticle deposition was demonstrated by Transmission Electron Microscopy (TEM). Analysis and design of the MEC employs Computational Fluid Dynamics (CFD) and true to geometry representations of nanoparticles with the aim to assess the uniformity of nanoparticle deposition among the culture wells. Final testing of the dose-controlled cell exposure system was performed by exposing A549 lung cell cultures to fluorescently labeled nanoparticles. Delivery of aerosolized nanoparticles was demonstrated by visualization of the nanoparticle fluorescence in the cell cultures following exposure. Also monitored was the potential of the aerosolized nanoparticles to generate reactive oxygen species (ROS) (e.g. free radicals and peroxides generation), thus expressing the oxidative stress of the cells which can cause extensive cellular damage or damage on DNA.

### 2012

Gómez, a, Armas, O., Lilik, G. K., and Boehman, A., 2012, "Estimation of volatile organic emission based on diesel particle size distributions," *Measurement Science and Technology*, **23**(10): 105305

In this work, an experimental method for the estimation of the volatile organic emission (VOE) of diesel particles is proposed. This method involves a minitunnel (MT) diluted exhaust sample, which is sent through a thermal denuder (TD) to a Scanning Mobility Particle Sizer (SMPS). VOE was estimated by the difference in particle mass concentration when using the TD at two temperatures, 30 and 300 C. The results were obtained by testing an engine at two operating modes with three different fuels: an ultra low sulfur diesel fuel and two synthetic fuels derived from low and high temperature Fischer-Tropsch processes. Although the absolute values are clearly different in comparison with traditional techniques, a good correlation, between the VOE experimentally determined by TGA analysis and the VOE estimated by means of MT-TD-SMPS, has been obtained.

Kinsey, J. S., Timko, M. T., Herndon, S. C., Wood, E. C., Yu, Z., Miake-Lye, R. C., Lobo, P., Whitefield, P., Hagen, D., Wey, C., Anderson, B. E., Beyersdorf, A. J., Hudgins, C. H., Thornhill, K. L., Winstead, E., Howard, R., Bulzan, D. I., Tacina, K. B., and Knighton, W. B., 2012, "Determination of the emissions from an aircraft auxiliary power unit (APU) during the Alternative Aviation Fuel Experiment (AAFEX)," *Journal of the Air & Waste Management Association*, **62**(4): 420-430

The emissions from a Garrett-AiResearch (now Honeywell) Model GTCP85-98CK auxiliary power unit (APU) were determined as part of the National Aeronautics and Space Administration's (NASA's) Alternative Aviation Fuel Experiment (AAFEX) using both JP-8 and a coal-derived Fischer Tropsch fuel (FT-2). Measurements were conducted by multiple research organizations for sulfur dioxide (SO<sub>2</sub>), total hydrocarbons (THC), carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), speciated gas-phase emissions, particulate matter (PM) mass and number, black carbon, and speciated PM. In addition, particle size distribution (PSD), number-based geometric mean particle diameter (GMD), and smoke number were also determined from the data collected. The results of the research showed PM mass emission indices (EIs) in the range of 20 to 700 mg/kg fuel and PM number EIs ranging from 0.5 x 10<sup>15</sup> to 5 x 10<sup>15</sup> particles/kg fuel depending on engine load and fuel type. In addition, significant reductions in both the SO<sub>2</sub> and PM EIs were observed for the use of the FT fuel. These reductions were on the order of

~90% for SO<sub>2</sub> and particle mass EIs and ~60% for the particle number EI, with similar decreases observed for black carbon. Also, the size of the particles generated by JP-8 combustion are noticeably larger than those emitted by the APU burning the FT fuel with the geometric mean diameters ranging from 20 to 50 nm depending on engine load and fuel type. Finally, both particle-bound sulfate and organics were reduced during FT-2 combustion. The PM sulfate was reduced by nearly 100% due to lack of sulfur in the fuel, with the PM organics reduced by a factor of ~5 as compared with JP-8.

Park, S. H., Cha, J., Kim, H. J., and Lee, C. S., 2012, "Effect of early injection strategy on spray atomization and emission reduction characteristics in bioethanol blended diesel fueled engine," *Energy*, **39**(1): 375–387

This study is to investigate the emission reduction characteristics of bioethanol blended diesel fuel at early injection condition including spray, atomization and evaporation characteristics. The spray atomization and evaporation characteristics were investigated using spray visualization system and KIVA-3V code, respectively. In this work, the effect of ethanol blending on the spray behavior is more evident at early injection condition. In the calculation results, the droplet size of bioethanol blended fuel was smaller than that of diesel, and bioethanol blended diesel droplets firstly evaporated by its volatility and superior atomization characteristics. In early injection condition, the bioethanol blending caused an increase in indicated mean effective pressure with an extension of the ignition delay. The cooling effect of the bioethanol fuel reduced NO<sub>x</sub>. The HC emission increased and the CO emission decreased because of the ethanol blending. The geometry mean diameter and total number density increased as a result of ethanol blending, the particle number in the nuclei mode decreased, and the particle number in the accumulation mode increased in early injection condition.

## 2011

Buonanno, G., Stabile, L., Avino, P., and Belluso, E., 2011, "Chemical, dimensional and morphological ultrafine particle characterization from a waste-to-energy plant," *Waste management (New York, N.Y.)*, **31**(11): 2253-62

Waste combustion processes are responsible of particles and gaseous emissions. Referring to the particle emission, in the last years specific attention was paid to ultrafine particles (UFPs, diameter less than 0.1 μm), mainly emitted by combustion processes. In fact, recent findings of toxicological and epidemiological studies indicate that fine and ultrafine particles could represent a risk for health and environment. Therefore, it is necessary to quantify particle emissions from incinerators also to perform an exposure assessment for the human populations living in their surrounding areas. To these purposes, in the present work an experimental campaign aimed to monitor UFPs was carried out at the incineration plant in San Vittore del Lazio (Italy). Particle size distributions and total concentrations were measured both at the stack and before the fabric filter inlet in order to evaluate the removal efficiency of the filter in terms of UFPs. A chemical characterization of UFPs in terms of heavy metal concentration was performed through a nuclear method, i.e., Instrumental Neutron Activation Analysis (INAA), as well as a mineralogical investigation was carried out through a Transmission Electron Microscope (TEM) equipped with an Energy Dispersive Spectrometer (EDS) in order to evaluate shape, crystalline state and mineral compound of sampled particles. Maximum values of  $2.7 \times 10^7$  part. cm<sup>-3</sup> and  $2.0 \times 10^3$  part. cm<sup>-3</sup> were found, respectively, for number concentration before and after the fabric filter showing a very high efficiency in particle removing by the fabric filter. With regard to heavy metal concentrations, the elements with higher boiling temperature present higher concentrations at lower diameters showing a not complete evaporation in the combustion section and the consequent condensation of semi-volatile compounds on solid nuclei. In terms of mineralogical and morphological analysis, the most abundant compounds found in samples collected before the fabric filter are Na-K-Pb oxides followed by phyllosilicates, otherwise, different oxides of comparable abundance were detected in the samples collected at the stack.

Cheng, Y.-H., and Li, Y.-S., 2011, "Influences of traffic volumes and wind speeds on ambient ultrafine particle levels—Observations at a highway electronic toll collection (ETC) lane," *Atmospheric Environment*, **45**(1): 117–122

The levels of ultrafine particles (UFPs) and their size distributions on a highway electronic toll collection (ETC) lane were measured from October 30 to November 1 and November 5 to November 6, 2008. The hourly UFP levels measured at the highway ETC lane were  $1.3 \times 10^4$  -  $1.9 \times 10^5$  particles cm<sup>-3</sup> (mean =  $9.4 \times 10^4$  particles cm<sup>-3</sup>). Compared with urban UFP levels, average UFP levels at the highway ETC lane were about 5-10 times higher than those previously measured in urban areas, indicating that a considerable amount of UFPs were exhausted from vehicles. At the highway ETC lane, the average UFP number size distribution had a dominant mode at about 10 nm and a minor mode at about 33 nm. Measurement results indicate that nucleation mode particles remained at relatively high levels compared to Aitken mode and accumulation mode particles at the highway ETC lane. This study identifies the impacts of traffic volumes and wind speeds on ambient UFP levels. Measurement results show that the elevated UFP

levels resulting from traffic volume decreased exponentially as wind speed increased on the highway when wind speed was  $<2.0\text{ms}^{-1}$ . However, the elevated UFP levels resulting from traffic volume increased slightly when wind speed was  $>2.0\text{ms}^{-1}$  due to local turbulent mixing causes UFPs at high wind speeds. According to measurement results, high levels of UFP were observed at low wind speeds and under high traffic volumes.

Rattanavaraha, W., Rosen, E., Zhang, H., Li, Q., Pantong, K., and Kamens, R. M., 2011, "The reactive oxidant potential of different types of aged atmospheric particles: An outdoor chamber study," *Atmospheric Environment*, **45**(23): 3848–3855

The reactive oxygen species (ROS) potential of aged diesel exhaust particulate matter (PM) and other aged aerosol systems in the presence and absence of an urban hydrocarbon environment was assessed. Experiments were performed in a 274m<sup>3</sup> dual outdoor Teflon film chamber. Filter samples were taken to assess the oxidant generation associated with PM by an optimized dithiothreitol (DTT) method. Diesel exhaust PM had a higher ROS response when it was in the presence of an urban hydrocarbon mixture and was associated with significant O<sub>3</sub> production. For all the aged dilute diesel systems, ROS expression increased by a factor of 2-4 over fresh diesel particles. Other particle systems were also investigated. A low ROS was observed in most of the nighttime experiments, including the nighttime aerosols from SO<sub>2</sub> with O<sub>3</sub> and SO<sub>2</sub> aged by itself. However, when all the systems were compared, aged diesel exhaust tended to express very high ROS potentials, with secondary organic aerosols from an  $\alpha$ -pinene + toluene + an urban HC mixture giving the highest ROS response.

Samy, S., Zielinska, B., Sagebiel, J. C., and McDonald, J. D., 2011, "Application for a Newly Developed High-Capacity NO<sub>x</sub> Denuder: Low-NO<sub>x</sub> Diesel Transformation Experiments," *Journal of the Air & Waste Management Association*, **61**(3): 319–323

To conduct low oxides of nitrogen (NO<sub>x</sub>) chamber experiments with modern diesel emissions (DE), a high-capacity NO<sub>x</sub> denuder was developed and used at the European Photoreactor (EUPHORE) outdoor simulation chamber. The denuder displayed a sufficient NO<sub>x</sub> storage capacity for use with DE, and efficient removal of NO<sub>x</sub> during injections of DE was achieved ( $>98\%$ ). Degradation of the denuder performance after repeated regeneration by heating (400 °C) and flushing with an air/oxygen ratio of 2:1 was not observed for a total of nine experiments. Evaluation of dark (with chamber cover closed) experiments (four in total) with and without the denuder in-line revealed some reduction (22%) of diesel particulate matter (DPM) with use of the denuder, most likely a result of impaction or settling of DPM during DE transit. However, DPM reduction may have also been a result of reductions in effective load of the engine-dyno system during the DE injections. Extensive chemical characterization of DPM revealed no significant perturbation of major compound groups associated with denuder use, except for nitrated polyaromatic hydrocarbon (NPAH) concentrations. The implications of high-NO<sub>x</sub> experiments without the use of a NO<sub>x</sub> denuder are discussed.

## 2010

Barone, T. L., Storey, J. M. E., and Domingo, N., 2010, "An Analysis of Field-Aged Diesel Particulate Filter Performance: Particle Emissions before, during, and after Regeneration," *Journal of the Air & Waste Management Association*, **60**(8): 968–976

A field-aged, passive diesel particulate filter (DPF) used in a school bus retrofit program was evaluated for emissions of particle mass and number concentration before, during, and after regeneration. For the particle mass measurements, filter samples were collected for gravimetric analysis with a partial flow sampling system, which sampled proportionally to the exhaust flow. A condensation particle counter and Scanning Mobility Particle Sizer measured total number concentration and number-size distributions, respectively. The results of the evaluation show that the number concentration emissions decreased as the DPF became loaded with soot. However, after soot removal by regeneration, the number concentration emissions were approximately 20 times greater, which suggests the importance of the soot layer in helping to trap particles. Contrary to the number concentration results, particle mass emissions decreased from  $6 \pm 1$  mg/hp-hr before regeneration to  $3 \pm 2$  mg/hp-hr after regeneration. This indicates that nanoparticles with diameters less than 50 nm may have been emitted after regeneration because these particles contribute little to the total mass. Overall, average particle emission reductions of 95% by mass and 10,000-fold by number concentration after 4 yr of use provided evidence of the durability of a field-aged DPF. In contrast to previous reports for new DPFs in which elevated number concentrations occurred during the first 200 sec of a transient cycle, the number concentration emissions were elevated during the second half of the heavy-duty Federal Test Procedure (FTP) when high speed was sustained. This information is relevant for the analysis of mechanisms by which particles are emitted from field-aged DPFs.

Bugarski, A. D., Cauda, E. G., Janisko, S. J., Hummer, J. a., and Patts, L. D., 2010, "Aerosols Emitted in Underground Mine Air by Diesel Engine Fueled with Biodiesel," *Journal of the Air & Waste Management Association*, **60**(2): 237-244

Using biodiesel in place of petroleum diesel is considered by several underground metal and nonmetal mine operators to be a viable strategy for reducing the exposure of miners to diesel particulate matter. This study was conducted in an underground experimental mine to evaluate the effects of soy methyl ester biodiesel on the concentrations and size distributions of diesel aerosols and nitric oxides in mine air. The objective was to compare the effects of neat and blended biodiesel fuels with those of ultralow sulfur petroleum diesel. The evaluation was performed using a mechanically controlled, naturally aspirated diesel engine equipped with a muffler and a diesel oxidation catalyst. The effects of biodiesel fuels on size distributions and number and total aerosol mass concentrations were found to be strongly dependent on engine operating conditions. When fueled with biodiesel fuels, the engine contributed less to elemental carbon concentrations for all engine operating modes and exhaust configurations. The substantial increases in number concentrations and fraction of organic carbon (OC) in total carbon over the baseline were observed when the engine was fueled with biodiesel fuels and operated at light-load operating conditions. Size distributions for all test conditions were found to be single modal and strongly affected by engine operating conditions, fuel type, and exhaust configuration. The peak and total number concentrations as well as median diameter decreased with an increase in the fraction of biodiesel in the fuels, particularly for high-load operating conditions. The effects of the diesel oxidation catalyst, commonly deployed to counteract the potential increase in OC emissions due to use of biodiesel, were found to vary depending upon fuel formulation and engine operating conditions. The catalyst was relatively effective in reducing aerosol number and mass concentrations, particularly at light-load conditions, but also showed the potential for an increase in nitrogen dioxide concentrations at high-load modes.

Giechaskiel, B., Chirico, R., Decarlo, P. F., Clairotte, M., Adam, T., Martini, G., Heringa, M. F., Richter, R., Prevot, a S. H., Baltensperger, U., and Astorga, C., 2010, "Evaluation of the particle measurement programme (PMP) protocol to remove the vehicles' exhaust aerosol volatile phase," *The Science of the total environment*, **408**(21): 5106-16

European regulation for Euro 5/6 light duty emissions introduced the measurement of non-volatile particles with diameter >23 nm. The volatile phase is removed by using a heated dilution stage (150 degrees C) and a heated tube (at 300-400 degrees C). We investigated experimentally the removal efficiency for volatile species of the specific protocol by conducting measurements with two Euro 3 diesel light duty vehicles, a Euro 2 moped, and a Euro III heavy duty vehicle with the system's heaters on and off. The particle number distributions were measured with a Scanning Mobility Particle Sizer (SMPS) and a Fast Mobility Particle Sizer (FMPS). An Aerosol Mass Spectrometer (AMS) was used to identify the non-refractory chemical composition of the particles. A Multi-Angle Absorption Photometer (MAAP) was used to measure the black carbon concentration. The results showed that the condensed material in the accumulation mode (defined here as particles in the diameter range of approximately 50-500 nm) was removed with an efficiency of 50-90%. The (volatile) nucleation mode was also completely evaporated or was decreased to sizes <23 nm; thus these particles wouldn't be counted from the particle counter, indicating the robustness of the protocol.

## 2009

Giechaskiel, B., Ntziachristos, L., and Samaras, Z., 2009, "Effect of ejector dilutors on measurements of automotive exhaust gas aerosol size distributions," *Measurement Science and Technology*, **20**(4): 045703

Ejector dilutors have long been used for automotive exhaust particle sampling, as they can offer a low-cost option for stable dilution. In an ejector dilutor, pressurized air expanding in the periphery of a nozzle draws in and mixes with an exhaust sample which is then led to analytical equipment. The combination of processes involved may lead to particle losses which can affect the measurement. This study examines the losses of diesel exhaust particles of different characteristics (nucleation mode, non-volatile accumulation mode, internally and externally mixed accumulation mode) when these are sampled through an ejector dilutor. A Scanning Mobility Particle Sizer (SMPS) spectrometer, an electrical low-pressure impactor and a diffusion charger were used as analytical equipment to characterize losses with different instruments. Particle losses were found negligible for all practical applications of diesel exhaust aerosol sampling. Also, the sampling outlet and the operating pressure of the ejector dilutor were found to have a non-measurable effect on the distribution shape. Some variation of the labile nucleation mode particles was attributed to evaporation within the SMPS rather than an ejector effect, and this was confirmed by sampling solid NaCl particles in the same size range. The study further confirms the usability of ejector dilutors for exhaust particle sampling and dilution.



Li, Q., Wyatt, A., and Kamens, R. M., 2009, "Oxidant generation and toxicity enhancement of aged-diesel exhaust," *Atmospheric Environment*, **43**(5): 1037–1042

Diesel exhaust related airborne Particulate Matter (PM) has been linked to a myriad of adverse health outcomes, ranging from cancer to cardiopulmonary disease. The underlying toxicological mechanisms are of great scientific interest. A hypothesis under investigation is that many of the adverse health effects may derive from oxidative stress, initiated by the formation of reactive oxygen species (ROS) within affected cells. In this study, the main objective was to determine whether aged-diesel exhaust PM has a higher oxidant generation and toxicity than fresh diesel exhaust PM. The diesel exhaust PM was generated from a 1980 Mercedes-Benz model 300SD, and a dual 270 m<sup>3</sup> Teflon film chamber was utilized to generate two test atmospheres. One side of the chamber is used to produce ozone–diesel exhaust PM system, and another side of the chamber was used to produce diesel exhaust PM only system. A newly optimized dithiothreitol (DTT) method was used to assess their oxidant generation and toxicity. The results of this study showed: (1) both fresh and aged-diesel exhaust PM had high oxidant generation and toxicity; (2) ozone–diesel exhaust PM had a higher toxicity response than diesel exhaust PM only; (3) the diesel exhaust PM toxicity increased with time; (4) the optimized DTT method could be used as a good quantitative chemical assay for oxidant generation and toxicity measurement.

## 2008

Chapman, E. M., and Boehman, A. L., 2008, "Pilot ignited premixed combustion of dimethyl ether in a turbodiesel engine," *Fuel Processing Technology*, **89**(12): 1262–1271

This paper describes combustion studies of dimethyl ether in a common rail turbodiesel engine wherein the dimethyl ether was fumigated into the intake air and the conventional diesel injection was used with the intention of igniting the premixed DME-air charge. This combustion process is referred to here as a "mixed mode" process and is similar in some respects to what is commonly referred to as "dual fuel" combustion. In contrast to "dual fuel" combustion, however, in which the gaseous fuel is often natural gas or biogas, in this process with DME the gaseous charge ignites largely independently of the diesel injection. The diesel injection was accomplished with a single, main injection. The engine was operated at a single speed and load. Gaseous and particulate emissions were monitored and heat release analysis was performed to examine how the fuels burn and the impact on emissions formation at various levels of substitution of diesel fuel with fumigated DME, at as high as 44% of the fuel energy from DME. Reductions in NO<sub>x</sub> emissions and increases in particulate matter emissions are observed with DME fumigation. The increase in PM emissions is attributed to enrichment of the diesel fuel spray, due to displacement of intake oxygen by the fumigated DME, despite the widely observed soot suppressing effect of DME.

Chung, a., Lall, A. a., and Paulson, S. E., 2008, "Particulate emissions by a small non-road diesel engine: Biodiesel and diesel characterization and mass measurements using the extended idealized aggregates theory," *Atmospheric Environment*, **42**(9): 2129–2140

Particulate emissions from a 4.8-kW diesel generator running on ultra-low sulfur diesel and biodiesel fuels are characterized as a function of engine load. Number distributions measured by a Scanning Mobility Particle Sizer (SMPS) show that particle mobility diameters rise with increasing engine loads. The elemental carbon (EC) to organic carbon (OC) ratio, measured by thermo-optical transmission evolved gas analysis, with careful attention to avoid OC sampling artifacts, increases from about 0.5 at idle load to 3.8 at 100% load when using diesel fuel. Transmission electron microscopy (TEM) images of the particles showed that at idle, the particles were liquid droplets together with a few aggregates. When a load was applied, the droplets were replaced by chain aggregates, which had a mean primary particle size of 29±9 nm at 100% load. Fractal dimension averaged 1.63±0.13, consistent with much larger diesel engines emissions reported in the literature. The use of biofuel (B100) results in emissions of particles that are compact, irregular, and lack the clearly defined primary particles of diesel aggregates, and yet at maximum load they have similar EC and OC content as diesel particles. The accuracy of the idealized aggregate (IA) theory correction and its extension to the transition regime [Lall, A.A., Friedlander, S.K., 2006. On-line measurement of ultrafine aggregate surface area and volume distributions by electrical mobility analysis: 1. Theoretical analysis. *Journal of Aerosol Science* 37, 260–271] was tested as a method to obtain mass distributions for diesel aggregates using and SMPS. The total mass concentrations calculated from the SMPS measurements using the extended IA theory are in good agreement with the mass concentrations obtained from gravimetric and EC/OC measurements. The loss of aggregates in the TSI SMPS inlet impactor is also discussed.

Corporan, E., Quick, A., DeWitt, M., Lee, C., Serre, S., Zhao, Y., Lee, S., Hastings, T., Chin, P., Ollis, D., Qian, J., Ferro, A., Fowler, K., Oh, H., Annamalai, K., Sweeten, J., Stokke, J., Mazyck, D., Corbett, J., Winebrake, J., Tecer, L., Ren, P., Alagha, O., Karaca, F., Tuncel, G., Sheya, S., Glowacki, C., Chang, M.-C., Chow, J., Watson, J., Lin, C.-H., Ge'go, E., Gilliland, A., Godowitch, J., Rao, S., Porter, P., and Hogrefe, C., 2008, "Characterization of Particulate Matter and Gaseous Emissions of a C-130H Aircraft," *Journal of the Air & Waste Management Association*, **58**(4): 474–483

The gaseous and nonvolatile particulate matter (PM) emissions of two T56-A-15 turboprop engines of a C-130H aircraft stationed at the 123rd Airlift Wing in the Kentucky Air National Guard were characterized. The emissions campaign supports the Strategic Environmental Research and Development Program (SERDP) project WP1401 to determine emissions factors from military aircraft. The purpose of the project is to develop a comprehensive emissions measurement program using both conventional and advanced techniques to determine emissions factors of pollutants, and to investigate the spatial and temporal evolutions of the exhaust plumes from fixed and rotating wing military aircraft. Standard practices for the measurement of gaseous emissions from aircraft have been well established; however, there is no certified methodology for the measurement of aircraft PM emissions. In this study, several conventional instruments were used to physically characterize and quantify the PM emissions from the two turboprop engines. Emissions samples were extracted from the engine exit plane and transported to the analytical instrumentation via heated lines. Multiple sampling probes were used to assess the spatial variation and obtain a representative average of the engine emissions. Particle concentrations, size distributions, and mass emissions were measured using commercially available aerosol instruments. Engine smoke numbers were determined using established Society of Automotive Engineers (SAE) practices, and gaseous species were quantified via a Fourier-transform infrared-based gas analyzer. The engines were tested at five power settings, from idle to take-off power, to cover a wide range of operating conditions. Average corrected particle numbers (PNs) of  $(6.4\text{--}14.3) \times 10^7$  particles per  $\text{cm}^3$  and PN emission indices (EI) from  $3.5 \times 10^{15}$  to  $10.0 \times 10^{15}$  particles per kg-fuel were observed. The highest PN EI were observed for the idle power conditions. The mean particle diameter varied between 50 nm at idle to 70 nm at maximum engine power. PM mass EI ranged from 1.6 to 3.5 g/kg-fuel for the conditions tested, which are in agreement with previous T56 engine measurements using other techniques. Additional PM data, smoke numbers, and gaseous emissions will be presented and discussed.

Isella, L., Giechaskiel, B., and Drossinos, Y., 2008, "Diesel-exhaust aerosol dynamics from the tailpipe to the dilution tunnel," *Journal of Aerosol Science*, **39**(9): 737–758

We study, experimentally and theoretically, the dynamics of non-volatile particles emitted from a diesel EURO 3 light-duty vehicle along the transfer tube that conducts exhaust fumes from the tailpipe to the dilution tunnel. Particle agglomeration, diffusional and thermophoretic transport are modeled. For turbulent, but moderate, Reynolds numbers and under steady-state conditions we map the combustion-generated nanoparticle dynamics onto a one-dimensional dynamics of aerosol particles in an ageing chamber. The aggregate fractal dimension, determined self-consistently by comparing mass distributions, varied from 2 to 2.3. The relative importance of aerosol processes is estimated by defining appropriate characteristic time scales. Agglomeration and convection by the bulk motion of the fluid are the dominant processes for inlet number concentrations of the order of  $10^8$  particles/ $\text{cm}^3$  and transfer-tube lengths of 6–9 m. Thermophoretic losses are calculated to be non-negligible. For modern vehicles with particulate filters agglomeration is estimated to be negligible, whereas thermophoresis may be significant.

Lim, J., Yu, L. E., Kostetski, Y. Y., Lim, C., Ryu, J., and Kim, J., 2008, "Effects of Driving Conditions on Diesel Exhaust Particulates," *Journal of the Air & Waste Management Association*, **58**(8): 1077–1085

Four driving conditions were examined to characterize how speeds and loads of a medium-duty diesel engine affect resultant diesel exhaust particulates (DEPs) in terms of number concentrations ( $<400$  nm), size distribution, persistent free radicals, elemental carbon (EC), and organic carbon (OC). At the medium engine load (60%), DEPs surged in number concentrations at around 40–70 nm, whereas DEPs from the full engine load (100%) showed a distinctive bimodal distribution with a large population of 30–50 nm and 100–400 nm. Under the full engine load, engine speeds insignificantly affected resultant DEP number concentrations. When the engine load decreased from 100% to the medium level (60%), DEPs of ultrafine size and 100–400nm decreased at least 1.4 times (from  $5.6 \times 10^8$  to  $4 \times 10^8$  #/ $\text{cm}^3$ ) and more than 3 times (from  $2.7 \times 10^8$  to  $0.8 \times 10^8$  #/ $\text{cm}^3$ ), respectively. The same reduction in the engine load significantly decreased persistent free radicals in DEPs up to approximately 30 times (from  $123 \times 10^{16}$  to  $4 \times 10^{16}$  #spin/g). Decreasing the engine load from 100 to 60% also concurrently reduced both EC and OC in total DEPs around 2 times, from 27.3 to 13.9  $\text{mg}/\text{m}^3$ , and from 17.6 to 9.2  $\text{mg}/\text{m}^3$ , respectively. For DEPs smaller than 1  $\mu\text{m}$ , under the full engine load, EC and OC consistently peaked at 170–330 nm under an engine speed of 1800 rpm or 94–170 nm under an engine speed of 3000 rpm, reflecting processes of nucleation, cluster-cluster agglomeration, and condensation. Decreasing the engine load from 100 to 60% reduced EC and OC in DEPs (smaller than 1  $\mu\text{m}$ ) at least 3 times ( $0.6$  to  $0.2$   $\text{mg}/\text{m}^3$ ) and 2 times ( $0.4$  to  $0.2$   $\text{mg}/\text{m}^3$ ), respectively. Taken together, decreasing the full engine load to a medium (60%) level effectively

reduced the number concentrations (<400 nm), persistent free radicals, EC, and OC of total DEPs, as well as the concentration of EC and OC in ultrafine and accumulation-mode DEPs.

Paganini, E., Mariotti, G., Gasperetti, S., Vallebona, C., Predolin, L., Muré, E., Palleschi, V., Salvetti, A., and Tognoni, E., 2008, "Multi-diagnostic approach to characterize the onset of formation of nanoparticles in a premixed laminar ethylene/air flame," *Spectrochimica Acta Part B: Atomic Spectroscopy*, **63**(2): 191–201

The onset of formation of nanoparticles in a premixed laminar ethylene/air flame was studied using optical and non-optical diagnostic techniques. The optical techniques (Visible Imaging, Spectroscopic analysis of optical emission, Laser Induced Incandescence and Optical Extinction) were checked against the direct measurement of particles size distribution done with a commercial sampler. The diagnostic techniques were set up on a standard burner (McKenna). The combustion conditions were varied by tuning two experimental parameters: the equivalent ratio  $\Phi$  and the cold gases velocity  $v$ . Values of  $\Phi$  were investigated in the range 1.3 to 2.2, in combination with cold unburned gas velocities of 5, 7 and 10 cm/s. By varying the combustion conditions, a transition in the signal detected by the different techniques was observed. The transition was put in correspondence with the sensitivity of the given technique to the onset of the nanoparticles formation. A comparison of the sensitivity of the different techniques was performed. The different sensitivities of the diagnostics and the different information that they provide confirmed the need for a multi-diagnostics setup for the characterization of nanoparticle production during the combustion process.

## 2007

Chang, Yu-Chen; Lee, Hsiu-Wei; Tseng, Huan-Hsiung, 2007, "The formation of incense smoke," *Journal of Aerosol Science*, **38**(1):39–51

The formation of incense smoke generated from four different types of incense sticks, three manufactured in Taiwan and one in Japan, was investigated in a small controlled chamber. The Scanning Mobility Particle Sizer and the quartz crystal microbalance were used for particle size analyses. The count median diameter (CMD) was found to rise swiftly along the path of the incense smoke. Consequently, a representative sampling location was selected for all measurements performed thereafter. All four types of incense smoke were shown to exhibit characteristic size distributions, CMDs, and mass median aerodynamic diameters (MMADs). Electron microscopy depicted liquid and solid nature of Taiwan and Japan incense smoke, respectively. The different physical states of the particles were suspected to be a result of different smoke-generating ingredients used by different cultures. Finally, the formation mechanisms of both liquid and solid incense smoke were discussed.

Thierley, M; Grotheer, H.-H.; Aigner, M.; Yang, Z.; Abid, A.; Zhao, B.; Wang, H., 2007, "On existence of nanoparticles below the sooting threshold," *Proceedings of the Combustion Institute*, **31**(1):639–647

Nanometer-sized particles were studied by photoionisation mass spectrometry and Scanning Mobility Particle Sizer in laminar premixed ethylene flames above and below the critical sooting threshold. For sooting flames, both techniques detected a large number of particles with masses between 1 and 50 ku or diameter around a few nanometers. Neither method detected an appreciable number of particles below the sooting threshold in flames similar to those studied earlier for UV absorption and scattering of transparent soot. The absence of particle signals in both experimental techniques raises the question about the origin of UV absorption under nonsooting conditions.

Zhao, Bin; Uchikawa, Kei; Wang, Hai, 2007, "A comparative study of nanoparticles in premixed flames by Scanning Mobility Particle Sizer, small angle neutron scattering, and transmission electron microscopy," *Proceedings of the Combustion Institute*, **31**(1):851–860

Scanning Mobility Particle Sizer (SMPS) spectrometer and transmission electron microscopy (TEM) studies were conducted for TiO<sub>2</sub> and soot particles. The TiO<sub>2</sub> particles were produced from a premixed stagnation ethylene–oxygen–argon flame ( $\Phi = 0.36$ ) doped with titanium tetraisopropoxide. Soot was generated from a burner-stabilized premixed ethylene–oxygen–argon flame ( $\Phi = 2.5$ ). The close agreement among SMPS, TEM, and X-ray diffraction results for TiO<sub>2</sub> nanoparticles demonstrates that the probe sampling/mobility measurement technique is accurate for on-line analysis of the size distribution of particles as small as 3 nm in diameter. In the case of soot, notable disagreement between the SMPS and TEM sizes was found and attributable to the fact that the soot taken from the flame studied herein is liquid-like and that upon deposition on the TEM grid, the primary particles do not retain their sphericity. This interpretation is supported by measurements with photo ionization aerosol mass spectrometry, small angle neutron scattering, and thermocouple particle densitometry.

Arai M.; Amagai K.; Nakaji T.; Hayashi S., 2006, "Primary and aggregate size distributions of PM in tail pipe emissions from diesel engines," *JSME International Journal, Series B: Fluids and Thermal Engineering*, **48**(4):639-647

Particulate matter (PM) emission exhausted from diesel engine should be reduced to keep the clean air environment. PM emission was considered that it consisted of coarse and aggregate particles, and nuclei-mode particles of which diameter was less than 50nm. However the detail characteristics about these particles of the PM were still unknown and they were needed for more physically accurate measurement and more effective reduction of exhaust PM emission. In this study, the size distributions of solid particles in PM emission were reported. PMs in the tail-pipe emission were sampled from three type diesel engines. Sampled PM was chemically treated to separate the solid carbon fraction from other fractions such as soluble organic fraction (SOF). The electron microscopic and optical-manual size measurement procedures were used to determine the size distribution of primary particles those were formed through coagulation process from nuclei-mode particles and consisted in aggregate particles. The centrifugal sedimentation method was applied to measure the Stokes diameter of dry-soot. Aerodynamic diameters of nano and aggregate particles were measured with Scanning Mobility Particle Sizer (SMPS) spectrometer. The peak aggregate diameters detected by SMPS were fallen in the same size regime as the Stokes diameter of dry-soot. Both of primary and Stokes diameters of dry-soot decreased with increases of engine speed and excess air ratio. Also, the effects of fuel properties and engine types on primary and aggregate particle diameters were discussed.

Chan, Tat Leung; Dong, Gang, 2006, "Particle number and size distributions along the vehicular exhaust plume from a light-duty diesel vehicle," *Neiranji Xuebao/Transactions of CSICE (Chinese Society for Internal Combustion Engines)*, **24**(1):50-56

The characteristics of particle number and size distributions ranging from 15 nm to 30  $\mu\text{m}$  along the vehicular exhaust plume emitted from a light-duty diesel vehicle under the low and high idling conditions were examined. The sampled exhaust particles were analyzed using the Scanning Mobility Particle Sizer (SMPS) spectrometer and the Aerodynamic Particle Sizer (APS) spectrometer along the centerline of vehicular exhaust plume and under the environmental background. The results showed that the total particle number concentrations emitted from the tested diesel vehicle at high idling condition were much higher than those at low idling condition. However, the total particle number concentrations for both of idling conditions decreased with the distance departing from the exit port due to the deposition process. The results also indicated that most of particles from diesel vehicle were the fine particles less than 700 nm in diameter, and those particles would suspend longer time in the atmosphere than the large particles, resulting in the local particulate pollution.

Cheung C.S.; Liu M.A.; Lee S.C.; Pan K.Y., "Experimental study on emission characteristics of diesel engines with diesel fuel blended with dimethyl carbonate," *Clean Air*, **6**(3): 239-253

In the present study, dimethyl carbonate (DMC) was blended with ultra-low sulphur diesel oil at different proportions, up to 30% by volume, and tested on a single cylinder direct-injection diesel engine, a 4-cylinder indirect-injection diesel engine, and a light-duty diesel vehicle to assess the effect of %DMC on fuel consumption and exhaust emissions. With an increase in %DMC, there was an increase in fuel consumption or a decrease in power output. Particulate emissions were measured with a Scanning Mobility Particle Sizer spectrometer and an Aerodynamic Particle Sizer spectrometer. DMC was found to significantly reduce particulate emissions, both sub-micron and micron, but the reduction was not proportional to the %DMC in the blended fuel. Within addition to the reduction in particulate emissions, there was a slight decrease in NO<sub>x</sub> and a slight increase in CO. For HC, there was a reduction at 5%DMC but an increase at higher %DMC. A lug-down test on the light-duty diesel vehicle showed a significant reduction in smoke emission coupled with a reduction in power output of the vehicle. It can be concluded that dimethyl carbonate-blended diesel can effectively reduce particulate emission.

Davidovic, M.; Ferge, T.; Hafner, K.; Maguhn, J.; Muhlberger, F.; Warnecke, R.; Zimmermann, R., 2006, "On-line analysis of gas-phase composition in the combustion chamber and particle emission characteristics during combustion of wood and waste in a small batch reactor," *Environmental Science and Technology*, **39**(6):1393-1402

The emission of particulate matter and gaseous compounds during combustion of wood and refuse-derived fuel in a small batch reactor is investigated by laser mass-spectrometric on-line measurement techniques for gas-phase analysis and simultaneous registration of physical aerosol properties (number size distribution). The gas-phase composition is addressed by a laser-based mass spectrometric method, namely, vacuum-UV single-photon ionization time-of-flight mass spectrometry (VUV-SPI-TOFMS). Particle-size distributions are measured with a Scanning Mobility Particle Sizer spectrometer. Furthermore, a photoelectric aerosol sensor is applied for detection of particle-bound polycyclic aromatic hydrocarbons. The different phases of wood combustion are distinguishable by both the chemical profiles

of gas-phase components (e.g., polycyclic aromatic hydrocarbons, PAH) and the particle-size distribution. Furthermore, short disturbances of the combustion process due to air supply shortages are investigated regarding their effect on particle-size distribution and gas-phase composition, respectively. It is shown that the combustion conditions strongly influence the particle-size distribution as well as on the emission of particle-bound polycyclic aromatic hydrocarbons.

Hung, Hsueh-Fen; Wang, Chiu-Sen, 2006, "Formation of secondary organic aerosols and reactive oxygen species from diluted motorcycle exhaust," *Journal of the Chinese Institute of Chemical Engineers*, **37**(5):491-499

Oxidation of reactive organic gases (ROG) in ambient air produces low-volatility compounds that may condense to form secondary organic aerosols (SOA). Reactive oxygen species (ROS) are also produced in the photochemical reactions involving ROG. In the urban atmosphere, the precursors of SOA and ROS primarily come from vehicular emissions. This study aims at investigating the SOA and ROS formation potentials of diluted motorcycle exhaust under UV irradiation. Exhaust samples were obtained from a 124-cc motorcycle engine under idling conditions. Emissions from the tailpipe were directly collected in a Tedlar bag and diluted with filtered air by a ratio of 5. A filter cassette placed upstream of the bag removed particles in the exhaust while the sample was being collected. The sample in the bag was then irradiated by UV light and the number size distribution of aerosol particles was measured at various time intervals using a Scanning Mobility Particle Sizer spectrometer. The concentrations of gaseous and particulate reactive oxygen species were also determined at intervals. Under continuous UV irradiation, the particle number concentration in the bag peaked at 2-3 h, while the gaseous ROS concentration peaked at 5-7 h. The peak concentration of gaseous ROS was about one order of magnitude higher than the initial value in the diluted motorcycle exhaust. From the difference between gaseous and total ROS concentrations, it was estimated that about 9.6% (by mole number) of the total ROS was in the particulate phase. The results suggest that diluted motorcycle exhaust has a high potential to form ROS and SOA under UV irradiation.

Mamakos A.; Ntziachristos L.; Samaras Z., 2006, "Evaluation of the Dekati mass monitor for the measurement of exhaust particle mass emissions," *Environmental Science and Technology*, **40**(15): 4739-4745

The Dekati mass monitor (DMM) is an instrument which measures the mass concentration of airborne particles in real time by combining aerodynamic and mobility size particle classification. In this study, we evaluate the performance of the DMM by sampling exhaust from five engines and vehicles of different technologies in both steady-state and transient tests. DMM results are found higher than the filter-based particulate matter (PM) by 39 (plus or minus) 24% (range stands for (plus or minus) one standard deviation) for 62 diesel tests conducted in total and 3% and 14% higher, respectively, in two gasoline tests. To explore whether the difference occurs because of the different measurement principles of DMM and filter-based PM, the DMM operation is replicated over steady-state tests by combining an electrical low-pressure impactor (ELPI) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. The correlation of ELPI and SMPS derived mass and filter-based PM is satisfactory ( $R^2 = 0.95$ ) with a mean deviation of 5 (plus or minus) 15%. For the same tests, the correlation of DMM with PM was also high ( $R^2 = 0.95$ ), but DMM exceeded PM by 44 (plus or minus) 23% on average. The comparison of ELPI and SMPS and DMM results reveals that the latter overestimates both the geometric mean diameter and especially the width of the particle mass-weighted size distribution. These findings demonstrate that the statistically significant difference between the DMM and the filter-based PM cannot just originate from the different measurement principles but also from the actual implementation of the combined aerodynamic-mobility measurement in the DMM. Optimizing the DMM will require changes in its design and/or the calculation algorithm to improve the resolution and width of the aerodynamic size distribution recorded.

McMillian, Michael H.; Lawson, Seth A., 2006, "Experimental and modeling study of hydrogen/syngas production and particulate emissions from a natural gas-fueled partial oxidation engine," *International Journal of Hydrogen Energy*, **31**(7): 847-860

In this study, a combustion model was first applied to conditions representing varying compression ratios and equivalence ratios to investigate engine exhaust composition from partial oxidation (POX) of natural gas in reciprocating engines. The model was experimentally validated over a range of equivalence ratios from 1.3 to 1.6 with a spark-ignited single cylinder engine fueled by natural gas. The modeling results matched well with engine gaseous emission data over the experimental range. The model was also extended to higher equivalence ratios to predict H<sub>2</sub> and CO production at engine conditions and stoichiometries representative of homogeneous charge compression ignition (HCCI) engine operation. Secondly, over the same experimental range of equivalence ratios, particulate samples were taken to determine both total particulate mass production (g/hph) via gravimetric measurement as well as particle size distribution and loading via a Scanning Mobility Particle Sizer (SMPS) spectrometer. While experiments indicate hydrogen yields up to 11% using spark ignition (SI), modeling results indicate that greater than 20% H<sub>2</sub> yield may be possible in HCCI operation. Over the experimental range, rich-burn

particulate matter (PM) production is no greater than that from typical lean-burn operation. Finally, an energy balance was performed over the range of engine experimental operation.

## 2005

Aelander, T.; Antikainen, E.; Elonen, E.; Raunemaa, T.; Rautiola, A.; Torkkell, K., 2005, "Particle emissions from a small two-stroke engine: Effects of fuel, lubricating oil, and exhaust aftertreatment on particle characteristics," *Aerosol Science and Technology*, **39**(2):151–161

The effects of fuel and lubricating oil formulation and exhaust catalytic aftertreatment on physical and chemical characteristics of two-stroke engine exhaust particles were studied. The exhaust particles were produced with a professional chainsaw engine. The employed fuels were a 98-octane oxygenated, low-sulfur, low-aromatic reformulated gasoline, which served as a reference, and a 95-octane nonoxygenated alkylate gasoline that had no aromatics and olefins. The applied lubricating oils were a semisynthetic mineral-based oil and a biodegradable ester-based oil. In total eight fuel–lubricating oil–catalyst combinations were studied. The test runs were conducted on a test bench and exhaust was diluted in a full-flow dilution tunnel. The size and number emissions of the exhaust particles were measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer. The organic carbon (OC) and elemental carbon (EC) composition of the particles were analyzed with a thermal–optical transmission analyzer (TOT). In addition, the inorganic ion and metal composition of the particles were quantified, and the gaseous total hydrocarbon (THC), carbon monoxide (CO), and nitrogen oxide (NO<sub>x</sub>) emissions were measured. The volatility characteristics of the exhaust particles were studied with a thermal desorption unit combined with the SMPS. The particle mass (PM) emissions ranged without catalyst from 2.9 to 3.4 g/kWh and with catalyst from 1.7 to 2.4 g/kWh, the catalytic converter thus reducing PM emissions by 19–50%. Without catalyst the alkylate fuel–biodegradable oil combination gave the highest particle mass emissions, but with catalyst with the same fuel–oil mixture the emissions were the lowest. The count median diameter (CMD) of the particles ranged from 57 to 123 nm. Without catalyst, the alkylate fuel–biodegradable oil combination gave the lowest number emissions, but with catalyst with the same fuel–oil combination the emissions were the highest. The catalytic converter reduced the particle size by 22–56 nm, but it also increased the number emissions by a factor of 1.3–2.6. In thermal analysis 88–98% of the exhaust particle volume proved to be volatile, the solid fraction consisting of elemental carbon and metal residues. With the mineral-based lubricating oil, the metal residues appeared in two modes at the evaporation temperatures of 350°C and higher, while in the particles produced with the biodegradable oil the residues were unimodally distributed.

Bertola, Andrea; Boulouchos, Konstantinos; Kaegi, Ralf; Mathis, Urs; Mohr, Martin, 2005, "Influence of diesel engine combustion parameters on primary soot particle diameter," *Environmental Science and Technology*, **39**(6): 1887–1892

Effects of engine operating parameters and fuel composition on both primary soot particle diameter and particle number size distribution in the exhaust of a direct-injected heavy-duty diesel engine were studied in detail. An electrostatic sampler was developed to deposit particles directly on transmission electron microscopy (TEM) grids. Using TEM, the projected area equivalent diameter of primary soot particles was determined. A Scanning Mobility Particle Sizer (SMPS) spectrometer was used for the measurement of the particle number size distribution. Variations in the main engine operating parameters (fuel injection system, air management, and fuel properties) were made to investigate soot formation and oxidation processes. Primary soot particle diameters determined by TEM measurements ranged from 17.5 to 32.5 nm for the diesel fuel and from 24.1 to 27.2 nm for the water-diesel emulsion fuel depending on the engine settings. For constant fuel energy flow rate, the primary particle size from the water-diesel emulsion fuel was slightly larger than that from the diesel fuel. A reduction in primary soot particle diameter was registered when increasing the fuel injection pressure (IP) or advancing the start of injection (SOI). Larger primary soot particle diameters were measured while the engine was operating with exhaust gas recirculation (EGR). Heat release rate analysis of the combustion process revealed that the primary soot particle diameter decreased when the maximum flame temperature increased for the diesel fuel.

Blevins, L.G.; Cauley, T.H., 2005, "Fine particulate formation during switchgrass/coal cofiring," *Journal of Engineering for Gas Turbines and Power*, **127**(3):457–463

Experiments to examine the effects of biomass/coal cofiring on fine particle formation were performed in the Sandia Multi-Fuel Combustor using fuels of pure coal, three combinations of switchgrass and coal, and pure switchgrass. For this work, fine particles with aerodynamic diameter between 10 nm and 1 μm were examined. A constant solid-fuel thermal input of 8 kW was maintained. The combustion products were cooled from 1200 to 420°C during passage through the 4.2 m long reactor to simulate the temperatures experienced in the convection pass of a boiler. Fine particle number densities, mass concentrations, and total integrated number and mass concentrations at the reactor exit were determined using a Scanning Mobility Particle Sizer spectrometer. The fine particle number concentrations for cofiring were much

higher than those achieved with dedicated coal combustion. However the total integrated mass concentration of particles remained essentially constant for all levels of cofiring from 0% coal to 100% coal. The constant mass concentration is significant because pending environmental regulations are likely to be based on particle mass rather than particle size.

Burtscher, H., 2005, "Physical characterization of particulate emissions from diesel engines: a review," *Journal of Aerosol Science*, **36**(7):896–932

Properties of particles emitted from diesel engines and the consequences of these properties for sampling and measuring the particles are reviewed. The influence of aftertreatment devices such as particle traps and catalytic converters on particle properties is demonstrated. Based on the particle properties and results from health effect studies, requirements to metrics, and measurement systems, for example, for type approval testing, are discussed. This discussion is limited to physical properties. Special attention is given to the volatile fraction. We show that care has to be taken when designing the sampling and dilution system, because this step decisively influences what happens with the volatile material, which may remain in the gas phase, condense on solid particles, or form new particles by nucleation. If nucleation occurs, particles formed in the sampling lines may dominate the particle number concentration. A selection of systems for dilution, conditioning and measuring is shown. Systems to determine number, mass, and surface concentrations, size distributions, and carbon concentration are discussed. The discussion is focused on systems developed or adapted recently for the physical characterization of diesel particles.

Ciatti, Stephen A.; Miers, Scott A.; Ng, Henry K., 2005 "Influence of EGR on soot production in a light-duty diesel engine," *2005 Fall Technical Conference of the ASME Internal Combustion Engine Division*, 415–423

A study to explore the effect of EGR upon combustion in a light-duty automotive style diesel engine was performed. The engine used in this study was a Mercedes 1.7L 4 cylinder, direct injected turbodiesel with a common rail injection system. The engine was operated at 2500 RPM, 50% load, with constant rail pressure and injection duration. An endoscope imaging system built by AVL, called the VisioScope trademark was used to acquire in-cylinder optical images of combustion events. These images were processed to extract soot radiation temperatures and soot volume fraction for each pixel. The results were compared to global engine measurements using piezo-electric pressure transducers, an emissions bench, and a Scanning Mobility Particle Sizer (SMPS) to characterize particulates. It was discovered that the optical data correlated well with the global measurements, allowing for in-depth analysis of the mechanisms of emissions formation at three different EGR levels (0%, 10%, 19%). Several conclusions were reached, including the correlation of soot radiation temperature with NO<sub>x</sub> production and the correlation of soot luminosity with engine-out PM. Each of these factors was determined as a function of EGR level.

Ferge, T.; Maguhn, J.; Hafner, K.; Muehlberger, F.; Davidovic, M.; Warnecke, R.; Zimmermann, R., 2005, "On-Line Analysis of Gas-Phase Composition in the Combustion Chamber and Particle Emission Characteristics during Combustion of Wood and Waste in a Small Batch Reactor," *Environmental Science & Technology*, **39**(6):1393–1402

The emission of particulate matter and gaseous compounds during combustion of wood and refuse-derived fuel in a small batch reactor is investigated by laser mass-spectrometric on-line measurement techniques for gas-phase analysis and simultaneous registration of physical aerosol properties (number size distribution). The gas-phase composition is addressed by a laser-based mass spectrometric method, namely, vacuum-UV single-photon ionization time-of-flight mass spectrometry (VUV-SPI-TOFMS). Particle-size distributions are measured with a Scanning Mobility Particle Sizer spectrometer. Furthermore, a photoelectric aerosol sensor is applied for detection of particle-bound polycyclic aromatic hydrocarbons. The different phases of wood combustion are distinguishable by both the chemical profiles of gas-phase components (e.g., polycyclic aromatic hydrocarbons, PAH) and the particle-size distribution. Furthermore, short disturbances of the combustion process due to air supply shortages are investigated regarding their effect on particle-size distribution and gas-phase composition, respectively. It is shown that the combustion conditions strongly influence the particle-size distribution as well as on the emission of particle-bound polycyclic aromatic hydrocarbons.

Giechaskiel, B.; Ntziachristos, L.; Samaras, Z.; Scheer, V.; Casati, R.; Vogt, R., 2005, "Formation potential of vehicle exhaust nucleation mode particles on-road and in the laboratory," *Atmospheric Environment & Atmos. Environ*, **39**(18): 3191–3198

A mobile laboratory equipped with gas analyzers, a particle number counter and a Scanning Mobility Particle Sizer was employed to measure the exhaust particle size distributions of a diesel Euro III passenger car, chasing its exhaust plume on a high-speed track at 50, 100 and 120 km/h. Emissions from the same vehicle were also measured in the laboratory under the same driving conditions, using a partial flow sampling system with constant sampling conditions. The vehicle was equipped with an oxidation

catalyst and was operated on diesel fuel with 280 ppm wt. sulphur content. Similar results for the exhaust aerosol behaviour were found in both sampling environments, despite the different dilution ratio, sampling temperature and residence time of the aerosol in dilute conditions. A relatively constant soot particle mode was formed in all cases and, in addition, a nucleation mode started to form at 100 km/h and became more stable at 120 km/h. No nucleation mode was observed at 50 km/h road load. The similar behaviour of nucleation mode particles both in the chasing and the laboratory tests indicated that such small volatile particles are a true vehicle emission component and not a dilution artefact. Additional measurements in the laboratory with varying engine load revealed that the nucleation mode formation is sensitive to exhaust gas temperature and its occurrence in increased temperature is repeatable and stable for long sampling times. The findings of this study indicate that nucleation mode particles are an actual emission component of diesel passenger cars and they need to be considered in relevant exhaust aerosol characterization studies.

Johnson, J.P.; Kittelson, D.B.; Savstrom, J.C.; Watts, W.F., 2005, "Influence of a catalytic stripper on the response of real time aerosol instruments to diesel exhaust aerosol," *Journal of Aerosol Science*, **36**(9):1089-1107

The objective of this study was to evaluate the response of a suite of portable, real-time aerosol instruments to Diesel exhaust aerosol with and without a catalytic stripper (CS) to determine the change in response as a function of particle size and volatility. The response of the photoemission aerosol sensor (PAS) was strongly influenced by the physical and chemical nature of Diesel aerosol. The presence of a large, predominantly volatile nuclei mode, and/or the presence of volatile material on the surface of the solid carbonaceous agglomerates in the accumulation mode suppressed the PAS response. Removal of the volatile material by passage of the aerosol through the CS enhanced the response, and improved correlations between the PAS, the diffusion charger (DC) and the Scanning Mobility Particle Sizer (SMPS) spectrometer. Data on aerosol size distributions, number, volume, and surface area concentrations with and without the CS in the sample stream are reported.

Johnson, Jason P.; Kittelson, David B.; Watts, Winthrop F., 2005, "Source apportionment of diesel and spark ignition exhaust aerosol using on-road data from the Minneapolis metropolitan area," *Atmospheric Environment*, **39**(11): 2111–2121

Air quality measurements were made on interstate highways in the Minneapolis metropolitan area. Gas and aerosol concentrations were measured on weekdays and weekends. By exploiting the difference in the relative volumes of heavy duty (HD) diesel and light duty (LD) spark ignition (SI) vehicles on weekdays and weekends, we were able to estimate apportioned fuel specific emissions. The on-road, apportioned, fuel specific particle number emissions factors, estimated from condensation particle counter (CPC) measurements were  $1.34 \pm 0.2 \cdot 10^{16}$ /particles  $\text{kg}^{-1}$  for diesels and  $7.1 \pm 1.6 \cdot 10^{15}$  particles  $\text{kg}^{-1}$  for spark ignition vehicles. Estimates from the Scanning Mobility Particle Sizer (SMPS) spectrometer measurements were  $2.1 \pm 0.3 \cdot 10^{15}$  particles  $\text{kg}^{-1}$  for diesels and  $3.9 \pm 0.6 \cdot 10^{14}$  particles  $\text{kg}^{-1}$  for SI vehicles. The difference between CPC and SMPS measurements is mainly due to different lower size detection limits of the instruments, similar to 3 and similar to 10 nm, respectively. On a weekly weighted basis and on weekdays, the majority of particle number was attributed to HD diesel traffic. Weekend production of particles can be primarily attributed to light duty SI automobiles. On a per vehicle basis, HD vehicles produced substantially greater numbers of particles. On a fuel specific basis, HD vehicles produce slightly higher concentrations of particles than light duty vehicles. The relative contribution of LD vehicles to particle number emissions increased as particle size decreased. The HD apportioned size distributions were similar to size distributions measured during other on-road and laboratory studies. The LD apportioned size distribution was bounded by laboratory and on-road size distributions. Our work is representative of summer, highway cruise conditions. It is likely that under cold start and high load operating conditions LD emissions will increase relative to HD emissions.

Johnston, Murray V.; Li, Zhigang; Richter, Henning; Wang, Hai; Yang, Zhiwei; Zhao, Bin, 2005, "Particle size distribution function of incipient soot in laminar premixed ethylene flames: Effect of flame temperature," *Proceedings of the Combustion Institute*, **30**(1): 1441–1448

Particle size distribution functions (PSDFs) of incipient soot formed in laminar premixed 24.2% ethylene-37.9% oxygen-diluent (nitrogen and/or argon) flames with an equivalence ratio of 1.92 were studied by online sampling and Scanning Mobility Particle Sizer spectrometer. Two series of flames were studied to quantify the effect of flame temperature on the characteristics of PSDFs. In the first series, the variation of the flame temperature was accomplished by varying the cold gas velocity. Temperature in the second series of flames was manipulated by the diluent composition from argon to nitrogen. The results show that for flames with the maximum temperature ( $T_{\text{submax}}$ ) around 1800 K the soot PSDFs were distinctively bimodal. As the flame temperature was increased to similar to 1850 K, bimodality faded away. The distribution was unimodal for  $T_{\text{submax}} \leq 1900$  K. The variation of the characteristics of the PSDF as a function of the flame temperature is consistent with the theoretical explanation that



bimodality is the result of competition between persistent particle nucleation and particle-particle coagulation in low-temperature flames.

Lee, Byung Uk; Bae, Gwi-Nam; Kim, Jin Kuk; Lee, Jin Ha; Yeo, Gwon-Koo, 2005, "The Behavior of Combustion Aerosols in an Exhaust Pipe," *The 13th International Pacific Conference on Automotive Engineering*, August 22–24, 2005, Seoul, Korea

The diesel particulate filters have been developed to remove diesel particles known to have adverse health effect from the automotive emission. Understanding of the phenomena of diesel exhaust in an exhaust pipe is needed in designing a DPF. In this study, the phenomena of diesel exhaust in an exhaust pipe were simulated by using combustion aerosols. The test aerosols were generated from a combustion aerosol generator consisting of a burner and a dilution unit. Temperature in an exhaust pipe was constantly controlled by using a heating tape. Particle size distributions were measured by a Scanning Mobility Particle Sizer spectrometer. The particle formation and growth will be discussed.

Lipsky, Eric M.; Robinson, Allen L., 2005, "Design and evaluation of a portable dilution sampling system for measuring fine particle emissions from combustion systems," *Aerosol Science and Technology*, **39**(6):542–553

The size and complexity of current dilution samplers is a major barrier to more wide-spread application of these systems for source characterization. A new, more portable dilution sampler has been designed to provide measurements consistent with the widely cited Caltech dilution sampler. Intercomparison experiments were performed using a diesel engine and wood stove to evaluate the comparability of the new design with a sampler based on the Caltech design. These experiments involved simultaneous operation of multiple dilution samplers from the same source. Filter based measurements included PM<sub>2.5</sub> mass, organic carbon, and elemental carbon emissions. Particle size distributions in the range from 10–480 nm were measured using a Scanning Mobility Particle Sizer spectrometer. The filter-based and integrated-total volume measurements made with the two designs are in good agreement. For example, the average relative bias between the two samplers of PM<sub>2.5</sub> mass emission rate measured with Teflon filters is 1%. Nucleation was intermittently observed in the sampler based on the Caltech design, but rarely observed in the new design. Significant discrepancies in total number emissions between the two samplers occurred during periods of nucleation. Experiments were also conducted to examine the effects of residence time on the diluted emissions. No changes in the filter-based or integrated volume measurements were observed with an additional 40-s residence time, indicating that phase equilibrium is established in the 2.5 s of residence time provided by the dilution tunnel. This conclusion is consistent with theoretical analysis. These results provide new insight into the effects of dilution sampling on measurements of fine particle emissions, providing important data for the ongoing effort of the EPA and ASTM to define a standardized dilution sampling methodology for characterizing emissions from stationary combustion sources.

Mathis, U.; Mohr, M.; Kaegi, R.; Bertola, A.; Boulouchos, K., 2005, "Influence of Diesel Engine Combustion Parameters on Primary Soot Particle Diameter," *Environmental Science & Technology*, **39**(6): 1887–1892

Effects of engine operating parameters and fuel composition on both primary soot particle diameter and particle number size distribution in the exhaust of a direct-injected heavy-duty diesel engine were studied in detail. An electrostatic sampler was developed to deposit particles directly on transmission electron microscopy (TEM) grids. Using TEM, the projected area equivalent diameter of primary soot particles was determined. A Scanning Mobility Particle Sizer (SMPS) spectrometer was used for the measurement of the particle number size distribution. Variations in the main engine operating parameters (fuel injection system, air management, and fuel properties) were made to investigate soot formation and oxidation processes. Primary soot particle diameters determined by TEM measurements ranged from 17.5 to 32.5 nm for the diesel fuel and from 24.1 to 27.2 nm for the water-diesel emulsion fuel depending on the engine settings. For constant fuel energy flow rate, the primary particle size from the water-diesel emulsion fuel was slightly larger than that from the diesel fuel. A reduction in primary soot particle diameter was registered when increasing the fuel injection pressure (IP) or advancing the start of injection (SOI). Larger primary soot particle diameters were measured while the engine was operating with exhaust gas recirculation (EGR). Heat release rate analysis of the combustion process revealed that the primary soot particle diameter decreased when the maximum flame temperature increased for the diesel fuel.

Montajir, Rahman M.; Kawai, Terunao; Goto, Yuichi; Odaka, Matsuo, 2005, "Thermal Conditioning of Exhaust Gas: Potential for Stabilizing Diesel Nano-Particles," *SAE World Congress*, April 11–14, 2005, Detroit, Michigan, USA

Conditioning of diluted exhaust gas by Thermo-Conditioner prior to measurement has been proposed by the GRPE/PMP Research Council of the United Nation in order to achieve stability in nano-particle measurement. In this study the effect of thermo-conditioner on the thermo-physical behavior of

nanoparticle under different conditions have been clarified. Stability in measurement was also attempted depending on the characteristics of nano-particles. Quality of the raw exhaust gas, the dilution ratio and temperature, and the thermal-conditioning temperature were considered as the main parameters. Exhaust gas from a medium-duty DI diesel engine was used for analysis. Scanning Mobility Particle Sizer spectrometer was used for measuring the concentration of nano-particles. It was concluded that the concentration of nuclei-mode particles within the size range of 15-30 nm are significantly influenced by the thermal conditioning temperature. However the concentration of accumulation mode particles having the diameter of about 100 nm experiences no influence. Thermal conditioning of exhaust gas at a temperature of over 300°C is assumed to be sufficient for stabilizing the nano-particles. Hot dilution showed better stability in measurement than cold dilution.

Ntziachristos, L.; Pistikopoulos, P.; Samaras, Z., 2005, "Particle characterization from two-stroke powered two-wheelers," *International Journal of Engine Research*, 6(3):263-275

This paper presents the physical characterization of particle emissions from small two-stroke gasoline two-wheelers. Particle samples were drawn with a dedicated partial dilution system. They were analyzed with various aerosol measurement instruments, including a Scanning Mobility Particle Sizer (SMPS) spectrometer, an electrical low-pressure impactor (ELPI), and a diffusion charger. Solid particles were discriminated by using a thermodenuder. Results suggest that small two-wheeled vehicles are significant emitters of both solid and volatile particles with the volatile fraction being the dominant one. The particle size distribution appears bimodal at high load and log-normal at lower load. The high concentration of condensable species in the exhaust also leads to the formation of volatile particles in the accumulation mode, which is typically not observed in diesel exhaust. Further comparison with a typical diesel passenger car revealed that some older technology two-wheelers emit even higher mass and solid particle counts than the car. However, the paper concludes that technology improvements such as oxidation catalysts and direct injection systems are expected significantly to reduce particulate matter emission rates.

Pagels, Joakum; Wierzbicka, Aneta; Bohgard, Mats; Strand, Michael; Lillieblad, Lena; Sanati, Mehri; Swietlicki, Erik, 2005, "In-Situ Characteristics of Particle Emissions from Biomass Combustion," *Conference: NOSA 2004 Aerosol Symposium: Combustion Aerosols*, Stockholm (Sweden), 11-12 Nov. 2004

In this work we used a Scanning Mobility Particle Sizer spectrometer and an Electrical Low-pressure Impactor to: a) Derive information of the particle morphology through air-borne analysis and b) Identify time and size variations of particle phase components from incomplete combustion and ash-components. The results presented here cover measurements in two moving grate boilers (12 MW operating on moist forest residue and 1.5 MW operating on wood pellets). We have previously shown that PM<sub>1</sub> estimated from Electrical Low-Pressure Impactor (ELPI)-measurements consisted of a rather constant background with peaks correlating with CO and OGC peaks. In the 1.5 MW boiler EC contributed to 34% of PM<sub>1</sub>, while in the 12 MW boiler EC was below 0.5%. Figure 2 shows time variations in the 1.5 MW boiler as the current in three stages of the ELPI-impactor. Note that time-variations increase strongly with particle size. The fraction of the gravimetric mass detected as water-soluble ions (IC) decreased from ~70% for  $d_{ae}$  = 78 and 133 nm to ~25% for 322 and 510 nm particles and increased to around 50% for particles larger than 1  $\mu$ m. In the 12 MW boiler time variations were as low as for 128 nm particles and IC recovery was high for all studied particle sizes. Based on these data we conclude that PM consisting of ash-components are formed with small time variations mainly in mobility-sizes below 250 nm, while Elemental Carbon is emitted at high concentrations during peaks on the time-scale 10-30 s, mainly in particle sizes larger than 150 nm. However, the detailed mixing status of these two particle types/materials is still not known.

Rubino, L.; Phillips, Paul R.; Twigg, Martyn V., 2005, "Measurements of Ultrafine Particle Number Emissions From a Light-Duty Diesel Engine Using SMPS, DMS, ELPI and EEPS," *7th International Conference on Engines for Automobile*, September 11-16, 2005, Naples, Italy

Tightening of automotive particulate matter (PM) emission regulations, driven by health concerns over ultrafine (< 100 nm) and nano-sized (< 50 nm) particles, has focused attention on measurement of particle size and number at the tailpipes of diesel engines. This study presents an investigation of PM emissions by number from a Light-Duty Diesel Engine running on low sulfur fuel using different aftertreatment systems. PM measurements by number and size were conducted over both transient and steady-state engine conditions using a standard CVS dilution tunnel. A Scanning Mobility Particle Sizer (SMPS) spectrometer, Electrical Low Pressure Impactor (ELPI), Differential Mobility Spectrometer (DMS) and an Engine Exhaust Particle Sizer™ (EEPS™) spectrometer were used for PM measurements. The performance of each particle size measurement instrument was assessed, and a comparison provided at similar experimental conditions. A standard diesel oxidation catalyst and particulate filters were used as aftertreatment systems. Particle number emissions for each are compared.

Stetter, John C.; Foster, David E.; Schauer, James J., 2005, "Modern Diesel Particulate Matter Measurements and the Application of Lessons Learned to 2007 Levels and Beyond," *SAE World Congress*, April 11–14, 2005, Detroit, Michigan, USA

Condensation particle counters (CPCs) have been used extensively during the past decade to measure the particle number concentration and, as part of a Scanning Mobility Particle Sizer (SMPS™) spectrometer, the size distribution of ultrafine and fine particle emissions from a wide range of spark ignition and diesel engines. This paper illustrates a method of calibrating the CPC particle counting efficiency, determination of the smallest particle size detection limit, and particle concentration linearity, by generating ultrafine particles with a novel, commercially available electrospray aerosol generator and using a sensitive aerosol electrometer as the traceable reference. The counting efficiencies of several TSI model 3010 CPCs were calibrated against the electrometer using monodisperse particles in the size range of 4.5 to 95 nm. Results from the tests show that the counting efficiencies of the CPCs are in close agreement with each other. The concentration linearity response of the CPCs was tested with monodisperse 50 nm particles and compared against the aerosol electrometer at six particle concentration levels roughly equally spaced across the CPCs' measurement range. The test results demonstrate a linear measurement response of each CPC with correlation coefficients ( $R_{\text{sup } 2}$ ) greater than 0.99. This paper also demonstrates that the lower detection limit of the CPC can be desensitized by adjusting the internal operating parameters of the CPC.

Taishi, Tsuyoshi; Koyama, Tetsuji; Kwon, Soon-Bark; Seto, Takafumi; Sakurai, Hiromu, 2005, "New Measurement System of Nanoparticles in the Automobile Exhaust Gas," *JSAE Technical Paper No. 20055680*, *JSAE Autumn Conference*, Sept. 28–30, 2005, Toyoko, Japan

The aerosol measurement techniques such as Scanning Mobility Particle Sizer (SMPS) spectrometer are one of major methods to evaluate the size distribution of diesel nanoparticles. The charge distribution is important to reduce data from mobility distribution to size distribution. The radioactive sources as aerosol charge neutralizer has been widely used, however, the limit of ion generation rate and the difficulty in handling especially at outdoor measurement remained as problem. In this presentation, we developed a new measurement system for diesel nanoparticles using the microplasma aerosol charger. Characteristics and performance of the system for diesel nanoparticle measurement were reported.

Westerdahl, D.; Fruin, S.; Sax, T.; Fine, P. M.; Sioutas, C., 2005, "Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles," *Atmospheric Environment & Atmos. Environ*, **39**(20): 3597–3610

Recent health studies have reported that ultrafine particles (UFP) (<0.1  $\mu\text{m}$  in diameter) may be responsible for some of the adverse health effects broadly attributed to particulate matter. In urban areas, UFP are produced by combustion sources, such as vehicle exhaust, and by secondary formation in the atmosphere. While UFP can be monitored, few studies have explored the impact of local primary sources in urban areas (including mobile sources on freeways) on the temporal and spatial distribution of UFP. This paper describes the integration of multiple monitoring technologies on a mobile platform designed to characterize UFP and associated pollutants, and the application of this platform in a study of UFP number concentrations and size distributions in Los Angeles. Monitoring technologies included two condensation particle counters (TSI Model 3007 and TSI 3022A) and Scanning Mobility Particle Sizer spectrometers for UFP. Real-time measurements made of  $\text{NO}_x$  (by chemiluminescence), black carbon (BC) (by light absorption), particulate matter-phase PAH (by UV ionization), and particle length (by diffusional charging) showed high correlations with UFP numbers. Average concentrations of UFP and related pollutants varied strongly by location, road type, and truck traffic volumes, suggesting a relationship between these concentrations and truck traffic density.

Zeuthen, Jacob; Livbjerg, Hans, 2005, "Laboratory Investigation of Aerosol Formation in Combustion of Biomass," *NOSA 2004 Aerosol Symposium: Combustion Aerosols*, Stockholm (Sweden), 11–12 Nov. 2004

In this project the formation of aerosol particles and deposits in power plants during combustion of  $\text{CO}_2$ -neutral fuels are investigated. For the experimental work a 173 cm long tubular furnace (diam=25 mm) with laminar flow is used. It is possible to control the temperature up to  $\sim 1200$  deg C in nine separate axial sections along the flue gas flow direction. In the first part of the reactor an inner tube is placed. In this inner tube a flow of inert nitrogen passes pellets of inert alumina impregnated with the salt to be volatilized (e.g. NaCl or KCl). The nitrogen gets saturated and by changing the temperature of the pellets it is possible to adjust the salt concentration in the gas. Other reactive gases ( $\text{SO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{NO}$  and  $\text{O}_2/\text{air}$ ) enter the reactor on the outside of the salt-containing alumina pipe. The temperature is kept constant in the first part of the reactor and is then decreased in the flow direction after a given length. The results obtained so far have shown that the homogeneous nucleation rate of pure salts depends on cooling rate, salt concentration and on the vapor pressure of the salt. Examples of results are shown at figure 1a. Here, two identical experiments are performed with two different salts. Since the vapor pressure of KCl is higher than for NaCl at the same temperature, a higher mass concentration of particles is obtained for this

salt. Due to a lower salt concentration the number concentration of NaCl particles is higher, but the particles are smaller. The particles are analyzed with a number of instruments, including Scanning Mobility Particle Sizer, low pressure cascade impactor and transition electron microscopy. Experiments with introduction of nucleation seeds in the inlet gas have been performed, and it has been found that a suppression of homogeneous nucleation can be observed at rather low number concentrations of seeds. Homogeneous nucleation is favored by rapid cooling and the critical seed concentration for suppression of nucleation is highly dependent on the rate of cooling. Experiments with sulfation of potassium chloride have been performed. By high-temperature filtering it has been found that potassium sulfate is the nucleating agent in aerosol formation during biomass combustion. In the future, the more advanced alkali-sulfur-chloride chemistry will be studied and the mechanisms leading to aerosol formation under biomass combustion conditions in power plants will be studied. The results will be analyzed by model studies including Computational Fluid Dynamics.

## 2004

Aalto, P.; Hameri, K.; Hillamo, R. E.; Hussein, T.; Keskinen, J.; Makela, T.; Pakkanen, T. A.; Parviainen, H.; Pirjola, L.; Valli, A.; Virtanen, A., 2004, "Sniffer' - A novel tool for chasing vehicles and measuring traffic pollutants," *Atmospheric Environment*, **38**(22):3625-3635

To measure traffic pollutants with high temporal and spatial resolution under real conditions a mobile laboratory was designed and built in Helsinki Polytechnic in close co-operation with the University of Helsinki. The equipment of the van provides gas phase measurements of CO and NO<sub>x</sub>, number size distribution measurements of fine and ultrafine particles by an electrical low pressure impactor, an ultrafine condensation particle counter and a Scanning Mobility Particle Sizer spectrometer. Two inlet systems, one above the windshield and the other above the bumper, enable chasing of different type of vehicles. Also, meteorological and geographical parameters are recorded. This paper introduces the construction and technical details of the van, and presents data from the measurements performed during an LIPIKA campaign on the highway in Helsinki. Approximately 90% of the total particle number concentration was due to particles smaller than 50nm on the highway in Helsinki. The peak concentrations exceeded often 200,000 particles/cm<sup>3</sup> and reached sometimes a value of 10<sup>6</sup> particles/cm<sup>3</sup>. Typical size distribution of fine particles possessed bimodal structure with the modal mean diameters of 15-20nm and similar to 150nm. Atmospheric dispersion of traffic pollutions were measured by moving away from the highway along the wind direction. At a distance of 120-140m from the source the concentrations were diluted to one-tenth from the values at 9 m from the source.

Abu-Allaban, M.; Gertler, A. W.; Rogers, C. F., 2004, "A Quantitative Description of Vehicle Exhaust Particle Size Distributions in a Highway Tunnel," *Journal of the Air and Waste Management Association*, **54**(3):360-366

During the period May 18-May 22, 1999, a comprehensive study was conducted in the Tuscarora Mountain Tunnel on the Pennsylvania Turnpike to measure real-world motor-vehicle emissions. As part of this study, size distributions of particle emissions were determined using a Scanning Mobility Particle Sizer spectrometer. Each measured size distribution consisted of two modes: a nucleation mode with midpoint diameter less than 20 nm and an accumulation mode with midpoint diameter less than 100 nm. The nucleation and accumulation components in some distributions also exhibited second maxima, which implies that such particle size distributions are superpositions of two particle size distributions. This hypothesis was utilized in fitting the particle size distributions that exhibited second maxima with four lognormal distributions, two for the nucleation mode and two for the accumulation mode. The fitting assumed that the observed particle size distribution was a combination of two bimodal log-normal distributions, one attributed to the heavy-duty diesel (HDD) vehicles and another attributed either to a different class of HDD vehicles or to the light-duty spark ignition vehicles. Based on this method, estimated particle production rates were  $1.8 \times 10^{13}$  and  $2.8 \times 10^{14}$  particles/vehicle-km for light-duty spark ignition and HDD vehicles, respectively, which agreed with independently obtained estimates.

Aelander, T. J. A.; Leskinen, A. P.; Rantanen, L.; Raunemaa, T. M., 2004, "Characterization of diesel particles: effects of fuel reformulation, exhaust aftertreatment, and engine operation on particle carbon composition and volatility," *Environmental Science and Technology*, **38**(9):2707-2714

Diesel exhaust particles are the major constituent of urban carbonaceous aerosol being linked to a large range of adverse environmental and health effects. In this work, the effects of fuel reformulation, oxidation catalyst, engine type, and engine operation parameters on diesel particle emission characteristics were investigated. Particle emissions from an indirect injection (IDI) and a direct injection (DI) engine car operating under steady-state conditions with a reformulated low-sulfur, low-aromatic fuel and a standard-grade fuel were analyzed. Organic (OC) and elemental (EC) carbon fractions of the particles were quantified by a thermal-optical transmission analysis method and particle size distributions measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer. The particle volatility characteristics were studied with a configuration that consisted of a thermal desorption unit

and an SMPS. In addition, the volatility of size-selected particles was determined with a tandem differential mobility analyzer technique. The reformulated fuel was found to produce 10-40% less particulate carbon mass compared to the standard fuel. On the basis of the carbon analysis, the organic carbon contributed 27-61% to the carbon mass of the IDI engine particle emissions, depending on the fuel and engine operation parameters. The fuel reformulation reduced the particulate organic carbon emissions by 10-55%. In the particles of the DI engine, the organic carbon contributed 14-26% to the total carbon emissions, the advanced engine technology, and the oxidation catalyst, thus reducing the OC/EC ratio of particles considerably. A relatively good consistency between the particulate organic fraction quantified with the thermal optical method and the volatile fraction measured with the thermal desorption unit and SMPS was found.

Andersson, Jon; Preston, Hugh; Warrens, Chris; Brett, Peter, 2004, "Fuel and lubricant effects on nucleation mode particle emissions from a Euro III light-duty diesel vehicle," *Spring Fuels and Lubricants Conference and Exhibition*, June 8–10, 2004, Toulouse, France

The impact of lubricant sulfur and phosphorus levels on the formation of nucleation mode particles was explored in a light-duty diesel vehicle operating over the New European Drive Cycle (NEDC). All measurements were undertaken using a Scanning Mobility Particle Sizer (SMPS) spectrometer, sampling from a conventional Constant Volume Sampler (CVS) system. Rigorous sampling system and vehicle conditioning procedures were applied to eliminate oil carry-over and nanoparticle artifact formation.

An initial vehicle selection process was undertaken on vehicles representing three fuel injection strategies, namely; distributor pump, common rail and unit injector. The vehicles met Euro III specifications and were all equipped with oxidation catalysts. Idle and low load stability were key requirements, since these conditions are the most significant in terms of their propensity to generate nucleation mode particles.

The influence of fuel sulfur on nucleation particle generation is well documented and three fuels were tested at idle to confirm the chosen vehicle's response to fuel sulfur. Traditional European fuels exhibited strong nucleation modes, with the 50 ppm S fuel showing 12% greater nucleation mode levels than the 10 ppm S fuel. Swedish Class 1 Diesel (SWCL1) fuel gave a very stable nucleation particle mode, but at a lower level. This fuel was selected for the lubricant effect study since it was believed that its reduced final boiling point would not overlap with or interfere with the oil discrimination phase of the work.

A total of 9 oils were tested with sulfur and phosphorus levels ranging from 0 to 0.9 and 0 to 0.1%wt respectively. All the oils were fully formulated having SAE "X" W-30 viscosities and a targeted ACEA A5/B5 performance level. Test order was carried out under strict statistical control.

Oil-derived sulfur and phosphorus were both found to increase the production of nucleation mode particles. Little effect of lubricant formulation on accumulation mode particles was observed. Swedish Class 1 (SWCL1) diesel fuel provided substantial reductions in nucleation mode particles.

Bermudez, V.; Desantes, J.M.; Fuentes, E.; Pastor, J.V., 2004, "Methodology for measuring exhaust aerosol size distributions from heavy duty diesel engines by means of a Scanning Mobility Particle Sizer," *Measurement Science and Technology*, **15**(10):2083–2098

A study of the sources of variability in particle size measurements using a dilution minitunnel and a Scanning Mobility Particle Sizer (SMPS) spectrometer has been conducted in order to obtain a comprehensive and repeatable methodology that can be used for measuring the particle size distribution of the exhaust aerosol emitted by a heavy duty diesel engine. The paper includes three experimental phases: an experimental analysis of the SMPS operating parameters' influence; an evaluation of the effect of dilution conditions, such as the dilution ratio and the dilution residence time; and a study of the influence of sampling factors, such as measurement stabilization and the effect of exhaust system pre-conditioning. An examination of the type and degree of influence of each studied factor is presented, recommendations for reducing variability are given and critical parameter values are identified to develop a measurement methodology of low uncertainty that could be applied to a further study concerning the effect of engine operating parameters on the exhaust particle size distribution.

Chang, M.-C. O.; Chow, J. C.; England, G. C.; Hopke, P. K.; Watson, J. G.; Yi, S.-M., 2004, "Measurement of ultrafine particle size distributions from coal-, oil-, and gas-fired stationary combustion sources," *Journal of the Air and Waste Management Association*, **54**(12):1494–1505

A dilution chamber was deployed to sample exhaust from a pilot-scale furnace burning various fuels at a nominal heat input rate of 160 kW/h and 3% excess oxygen. The formation mechanisms of particles smaller than 420 nm in electrical mobility diameter were experimentally investigated by measurement with a Scanning Mobility Particle Sizer (SMPS) spectrometer as a function of aging times, dilution air ratios, combustion exhaust temperatures, and fuel types. The measured particle size distributions in

number concentrations measured show peaks of particle number concentrations for medium sulfur bituminous coal, No. 6 fuel oil, and natural gas at 40-50 nm, 70-100 nm, and 15-25 nm, respectively. For No. 6 fuel oil and coal, the particle number concentration is constant in the range of a dilution air ratio of 50, but the number decreases as the dilution air ratio decreases to 10. However, for natural gas, the particle number concentration is higher at a dilution air ratio of 10 and decreases at dilution air ratios of 20-50. Samples taken at different combustion exhaust temperatures for these fuel types show higher particle number concentrations at 645 K than at 450 K. As the aging time of particles increases, the particles increase in size and the number concentrations decrease. The lifetimes of the ultrafine particles are relatively short, with a scale on the order of a few seconds. Results from this study suggest that an aging time of 10 sec and a dilution air ratio of 20 are sufficient to obtain representative primary particle emission samples from stationary combustion sources.

Corporan, E.; DeWitt, M.; Wagner, M., 2004, "Evaluation of soot particulate mitigation additives in a T63 engine," *Fuel Processing Technology*, **85**(6-7):727-742

The performance of fuel additive candidates to mitigate soot particulate emissions in turbine engines was assessed in a T63 helicopter engine. Seventeen additives, including commercial compounds to reduce emissions in internal combustion engines, diesel cetane improvers, and experimental/proprietary additives, were evaluated. The additives were individually injected into the JP-8 fuel feed to the engine, and evaluated at a minimum of three concentration levels. The engine was operated at two conditions, idle and cruise, to investigate additive effects at different power settings or equivalence ratios. Particulate samples were collected from the engine exhaust using an oil-cooled probe, and analyzed using a suite of particulates instrumentation, which included a condensation nuclei counter (CNC), Scanning Mobility Particle Sizer (SMPS) spectrometer, laser particle counter (LPC) and a tapered element oscillating microbalance (TEOM). Results indicate that the diesel cetane improvers and commercial smoke abatement additives tested had minimal impact on particulate emissions in the T63 turboshaft engine. One proprietary additive was shown to reduce particle number density (PND) by up to 67% at the relatively high concentration of 3000 mg/l. These benefits were observed only at cruise condition, which may provide some insight into the mechanisms by which the additive suppresses the formation or enhances the oxidation of soot particles. Test results with blends of JP-8 and Norpar-13 (normal paraffins) show significant reductions in particulate emissions for both idle and cruise conditions demonstrating the potential environmental benefits of using blends of clean (low aromatic and low sulfur) fuels with JP-8. Comparisons of mass determination with different instruments and preliminary results of chemical characterization of particulate emissions with and without additives are also presented.

Cromas, J.; Ghandhi, J. B., 2004, "Lubricating Oil Contribution to Direct-Injection, Two-Stroke Engine Particulate Emissions," *Small Engine Technology Conference*, September 27-30, 2004, Graz, Austria

Particulate emission measurements were performed on a direct-injection, two-stroke engine that employed a lost-oil lubricating system. The particulate emissions were sampled using a partial-flow dilution system. Particulate mass emission rates were measured using a tapered element microbalance (TEOM), and the results were found to compare favorably with gravimetric tests performed simultaneously. The size distribution was measured using a Scanning Mobility Particle Sizer (SMPS) spectrometer, and the cumulative mass from the measured size distribution was found to agree well with the values measured by the TEOM. The particulate mass emissions were found to be dominated by particulate matter derived from the engine oil. The particulate emissions were found to decrease substantially as the oil flow to the engine was reduced from the baseline case of 1:100 (oil-to-fuel mass ratio). Additionally, the reduction of the oil flow from the baseline case coincided with the removal of a secondary peak centered at 70 nm in the particle size distribution. It was determined that the oil flow rate had to be decreased to 1:450 in order for trends in the combustion-derived particulate matter to be observable. This level is comparable to levels seen in automotive four-stroke engines.

Hasegawa, S.; Hirabayashi, M.; Kobayashi, S.; Kondo, Y.; Moriguchi, Y.; Tanabe, K.; Wakamatsu, S., 2004, "Size distribution and characterization of ultrafine particles in roadside atmosphere," *Journal of Environmental Science and Health - Part A Toxic/Hazardous Substances and Environmental Engineering*, **39**(10):2671-2690

The number concentration and number size distributions of ultrafine particles were measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer at a roadside in early autumn and winter, and the results are discussed with regard to the contribution of traffic activity and meteorological conditions. The number concentration of the <50 nm fraction increased in the morning under calm wind conditions, and this increase corresponded with the increase in total traffic volume and nitric oxide. The increase in ultrafine particles was influenced not only by the increase in total traffic but also by the high contribution of diesel engine vehicles. The number concentration decreased around noon as the wind speed increased, although the total traffic and the number of diesel engine vehicles were at the same level as in the morning. The number size distribution in the morning was bimodal, with a first peak diameter of around

30 nm and a second of around 90 nm in both periods. The volatility of ultrafine particles was investigated using a thermal denuder operating at 250 degrees C. The first peak consisted mainly of volatile components, whereas the second one consisted of solid materials plus some volatile components. These results were consistent with the mass size distribution of elemental and organic carbon. The number size distribution with a peak diameter of around 30 nm was also observed in the afternoon at a suburban site; however, it was produced not by vehicle emissions directly but by photochemical reactions. Although a relatively high number concentration was also observed in the morning at the suburban site due to vehicle emission, the peak diameter ranged from 40 to 90 nm, which was larger than at the roadside.

Haupt, Dan; Nord, Kent; Tingvall, Bror; Ahlvik, Peter; Egeback, Karl-Erik; Andersson, Soren; Blomquist, Micael, 2004, "Investigating the potential to obtain low emissions from a diesel engine running on ethanol and equipped with EGR, catalyst and DPF," *Spring Fuels and Lubricants Conference and Exhibition*, June 8–10, Toulouse, France

Experiments were performed to investigate the potential to achieve low emissions from a diesel engine fueled by ethanol and equipped with a commercially available exhaust after-treatment device, DNO<sub>x</sub><sup>™</sup> from STT Emtec. The DNOsub<sub>x</sub><sup>™</sup> system includes exhaust gas recirculation (EGR) catalysts and a continuously regenerating diesel particulate filter (DPF). Two Euro III classified 9-liter turbocharged, after-cooled diesel engines from Scania were used for the task. One engine was fueled by ethanol and the other by Swedish diesel fuel, EC1. Engine operating conditions of a 22-mode test cycle, including the 13 modes of the European Stationary Cycle (ESC cycle), were used for the tests. The emissions of NO<sub>x</sub> and HC were small for the ethanol-fueled engine, 3.48 and 0.53 g/kWh, respectively, while the emission of CO was higher, 2.07 g/kWh. Estimations of emitted particle mass were calculated by using the software supplied in the Scanning Mobility Particle Sizer (SMPS) spectrometer. The estimations showed that the ethanol engine emitted only ~1/10 of the particle mass emitted by the diesel-fueled engine. A powerful reduction of the regulated emissions was obtained when equipping the ethanol engine with EGR, catalyst and DPF. The emissions of HC, CO and NO<sub>x</sub> decreased down to 0.15, 0.04 and 2.54 g/kWh, respectively, while the estimated particle mass was reduced by 67%. Actually, by using the aftertreatment system, the engine became a Euro IV engine regarding the emissions of HC, CO and NO<sub>x</sub>. The system worked even better with the diesel-fueled engine. The NO<sub>x</sub> emission was reduced by approximately 33% and the estimated particle mass by more than 99%. Calculations showed that the EGR ratio was higher for the diesel engine than for the ethanol engine. Consequently, by applying a higher EGR ratio for the ethanol engine an additional reduction of the NO<sub>x</sub> emissions should be obtained. The results indicate that very low NO<sub>x</sub> and particle emissions could be obtained for an ethanol-fueled diesel engine by using the right aftertreatment equipment. Future studies should investigate the possibility to increase the EGR ratio further. The investigations also underline the need for development of a special particulate filter for ethanol engines.

Hosoya, Mitsuru; Shundo, Seiji; Shimoda, Masatoshi, 2004, "The study of particle number reduction using after-treatment systems for a heavy-duty diesel engine," *SAE 2004 World Congress and Exhibition*, March 8–11, 2004, Detroit, Michigan, USA

To reduce ultra-fine particle number concentration from a heavy-duty diesel engine, the effects of diesel fuel property and after-treatment systems were studied. The reduction of ultra-fine particle number concentration over steady state mode using an 8-liter turbocharged and after-cooled diesel engine was evaluated. PM size distribution was measured by a Scanning Mobility Particle Sizer (SMPS) spectrometer. The evaluation used a commercially available current diesel fuel (Sulfur Content: 0.0036 wt%), high-sulfur diesel fuel (Sulfur Content: 0.046 wt%) and low-sulfur diesel fuel (Sulfur Content: 0.007 wt%). The after-treatment systems were an oxidation catalyst, a wire-mesh-type DPF (Diesel Particle Filter) and a wall-flow type catalyzed DPF. The results show that fine particle number concentration is reduced with a low-sulfur fuel, an oxidation catalyst, a wire-mesh-type DPF (Diesel Particulate Filter) and wall-flow-type catalyzed DPF, respectively. The wall-flow-type catalyzed DPF was the most effective for the particle reduction. The particle number reduction efficiency of the wall-flow-type DPF was 97% in the range of 50 to 70 nm mobility diameter over steady state mode.

Johnson, Tim; Caldow, Robert; Poecher, Arndt; Mirme, A.; Kittelson, David, 2004, "A new electrical mobility particle sizer spectrometer for engine exhaust particle measurements," *SAE 2004 World Congress and Exhibition*, March 8–11, 2004, Detroit, Michigan, USA

Electrical mobility has a long history as a tool for measuring the particle size of engine exhaust emissions. This paper gives a review of these methods as well as more current methods for making exhaust particle measurements. Each of the methods discussed has a limitation especially for making fast (sub-second) measurements. A new instrument is discussed that has been developed by TSI based on a technique developed over the last two decades by the University of Tartu - Estonia. A description of the instrument, the Engine Exhaust Particle Sizer<sup>™</sup> (EEPS<sup>™</sup>) spectrometer, is given as well as engine dynamometer data showing a comparison between the current standards for engine exhaust measurements, the Scanning Mobility Particle Sizing (SMPS<sup>™</sup>) system and the Condensation Particle Counter (CPC). The EEPS

spectrometer compares favorably with the SMPS spectrometer and CPC while providing sub-second response.

Kaegi, R.; Mathis, U.; Mohr, M.; Zenobi, R., 2004, "TEM analysis of volatile nanoparticles from particle trap equipped diesel and direct-injection spark-ignition vehicles," *Atmospheric Environment*, **38**(26):4347-4355

We investigated volatile nanoparticles emitted from two light-duty vehicles using a transmission electron microscope (TEM). Elemental analysis was performed with an energy dispersive system (EDS) attached to the microscope. Differences in stability of volatile nanoparticles during TEM analysis indicated two different chemical compounds. The results are consistent with the thesis that volatile nanoparticles are composed of a more volatile hydrophilic and a less volatile hydrophobic part. Sulfur and potassium were detected in volatile nanoparticles. These elements are likely to play an important role in the formation of volatile nanoparticles. Particle number size distributions derived from a Scanning Mobility Particle Sizer (SMPS) spectrometer and from TEM image analysis were in good agreement.

Kawano, Daisuke; Kawai, Terunao; Naito, Hiroyoshi; Goto, Yuichi; Odaka, Matsuo; Bachalo, William D., 2004, "Comparative measurement of nano-particulates in diesel engine exhaust gas by laser-induced incandescence (LII) and Scanning Mobility Particle Sizer (SMPS) spectrometer," *Spring Fuels and Lubricants Conference and Exhibition*, June 8-10, Toulouse, France

Particulate Matter (PM) from diesel engines is thought to be seriously hazardous for human health. Generally, it is said that the hazard depends on the total number and surface area of particles rather than total mass of PM. In the conventional gravimetric method, only the total mass of PM is measured. Therefore, it is very important to measure not only the mass of PM but also size and number density of particulates. Laser-Induced Incandescence (LII) is a useful diagnostic for transient measurement of soot particulate volume fraction and primary particle size. On the other hand, Scanning Mobility Particle Sizer (SMPS) spectrometer is also used to measure the size distribution of soot aggregate particulates at a steady-state condition. However, the measurement processes and the phenomena used to acquire the information on soot particulate are quite different between the LII and SMPS methods. Therefore, it is necessary to understand the detailed characteristics of both LII and SMPS. In the present study, the size distributions of PM from DI diesel engine are measured by both LII and SMPS simultaneously. In addition, PM mass emission is measured gravimetrically through a dilution tunnel and is separated into SOF and ISOF. The effects of EGR rate and engine load on the results of these particulate measurements are investigated. The different trends in the characteristics of PM emission are shown in each measurement methods for PM. The difference of detailed characteristics between LII and SMPS are illustrated by comparing the measurement results for the particulates. Finally, the problems associated with the measurements using each method are considered and some recommendations have been given for accurate measurement of nanoparticles.

Khatchikian, Peter; Harndorf, Horst; Knopf, Martin, 2004, "Dependence of the Particle Size Distribution on the Injection Pressure of a Modern Diesel Engine With a 1600 Bar Common-Rail Injection System," *2004 FISITA World Automotive Congress*, May 23-27, 2004, Barcelona, Spain

The influence of the injection pressure on the particle size distribution was measured with a Scanning Mobility Particle Sizer spectrometer. Measurements were carried out on a stationary test bench with a 3-l, 6-cylinder, common-rail direct injection engine from a passenger car, by varying the pressure in the rail. The sulfur content of the diesel fuel was < 10 ppm, and the sampling point was the tailpipe or the outlet of a transfer line. First of all, the size distribution was measured with the standard engine calibration. In a second step, only the torque was set to the value of the original operating point (the one with the original rail pressure, etc.) after changing the pressure. This was done by adapting the end of injection. In a third step, the engine was set to the emissions level of the original calibration by varying the EGR rate to set the soot emission (measured in terms of the Bosch Smoke Number, SN), and the start of injection for the NO<sub>x</sub> level. Finally, the size distribution was measured in this configuration but with the sampling point at the outlet of a transfer line. If the original injection pressure of 550 bar is varied in a range of 250-1600 bar and the engine torque is kept at 82 Nm, a slight decrease in the count median diameter (CMD) with increasing rail pressure can be found; on the other hand, the total number concentration (N) is reduced quite significantly. In the step three configuration, the same emissions as the original calibration, both particle size and concentration are almost constant with changing injection pressure. If the residence time of the aerosol prior to dilution is increased by employing a transfer line, a smaller number of slightly larger particles can be measured. With the test procedure and measurement conditions used in this investigation, no negative effect of high injection pressure on the particle size distribution could be found.



King, C. J.; Linak, W. P.; Miller, C.A.; Santoianni, D. A.; Seo, Y.-C.; Shinagawa, T.; Wendt, J. O. L.; Wood, J. P.; Yoo, J.-I., 2004, "High temperature interactions between residual oil ash and dispersed kaolinite powders," *Aerosol Science and Technology*, **38**(9):900–913

The potential use of sorbents to manage ultrafine ash aerosol emissions from residual oil combustion was investigated using a downfired 82 kW laboratory-scale refractory-lined combustor. The major constituents were vanadium (V), nickel (Ni), iron (Fe), and zinc (Zn). The overall ash content of residual oil is very low, resulting in total ash vaporization at 1725 K with appreciable vaporization occurring at temperatures as low as 1400 K. Therefore, the possibility of interactions between ash vapor and sorbent substrates exists. Kaolinite powder was injected at various locations in the combustor. Ash scavenging was determined from particle size distributions (PSDs) measured by a Scanning Mobility Particle Sizer spectrometer. Impactor samples and X-ray fluorescence (XRF) analyses supported these data. Injection of kaolinite sorbent was able to capture up to 60% of all the ash in the residual fuel oil. However, captures of ~30% were more common when sorbent injection occurred downstream of the combustion zone, rather than with the combustion air into the main flame. Without sorbent addition, baseline measurements of the fly ash PSD and chemical composition indicate that under the practical combustion conditions examined here, essentially all of the metals contained in the residual oil form ultrafine particles (<0.1 μm diameter). Theoretical calculations showed that coagulation between the oil ash nuclei and the kaolinite sorbent could account for, at most, 17% of the metal capture which was always less than that measured. The data suggest that kaolinite powders reactively capture a portion of the vapor phase metals. Mechanisms and rates still remain to be quantified.

Liu, MA; Cheung, C S; Pan, KY, 2004, "Effect of Oxygenated Additive on Particle Emission from Diesel Engine," *Journal of Air Force Engineering University (Natural Science Edition) (China)*, **5**(3):71–74

The effect of oxygenated additive on particle emission is investigated. Particle size distribution in exhausts from a diesel engine fueled by the blended fuel with different proportion of oxygenated additive are measured by a Scanning Mobility Particle Sizer spectrometer and an Aerodynamic Particle Sizer spectrometer. Coupled with this, emissions of carbon monoxide, hydrocarbon and nitrogen oxide are also measured with an infrared analyzer, a flame ionization detector and a chemiluminescent analyzer respectively. Results show that there is an effect of oxygenated additive on the particle size distribution from diesel engine. Oxygenated additive can significantly reduce particle emission level and slightly modify emission concentrations of carbon monoxide, hydrocarbon and nitrogen oxide.

Lyyräinen, Jussi; Jokiniemi, Jorma; Kauppinen, Esko; Backman, U.; Vesala, Hannu, 2004, "Comparison of Different Dilution Methods for Measuring Diesel Particle Emissions," *Aerosol Science & Technology*, **38**(91):12–23

Particle emissions from a turbo-charged diesel off-road engine were characterized with DMA + CNC and electron microscopy for comparison of different sampling and dilution systems. Four different sampling methods were used: (1) two ejector diluters, (2) partial flow and ejector diluter, (3) porous tube and ejector diluter, and (4) porous tube diluter. Number size distributions for partial flow and ejector dilution had modes at 25–30 nm and at 45–50 nm independent of the dilution ratio. The mode at 25–30 nm indicated nucleation during dilution in these experiments and was clearly most significant for the partial flow and ejector diluter setup. This was attributed to the temperature difference between exhaust gas, sample line, and partial flow diluter and cold dilution air. For other dilution systems the main mode was at 45 nm and indications of a mode at 15–20 nm were observed depending on the dilution ratio. Especially for the porous tube diluter, the main mechanism for particle growth was condensation on the surfaces of the existing particles. According to this study the best dilution system for obtaining a number size distribution without any significant nucleation effects was the porous tube dilution setup.

Nord, Kent; Haupt, Dan; Ahlvik, Peter; Egeback, Karl-Erik, 2004, "Particulate emissions from an ethanol-fueled, heavy-duty diesel engine equipped with EGR, catalyst and DPF," *Spring Fuels and Lubricants Conference and Exhibition*, June 8–10, 2004, Toulouse, France

Ethanol-fueled engines are considered to be low particulate-emitting engines. This study was performed to investigate the potential to achieve even lower particulate emission if a 9-liter Scania diesel engine, running on ethanol fuel is equipped with emission control. State-of-the-art technology in emission control was applied, e.g., exhaust gas recirculation, EGR, catalysts and a continuous regenerating particle filter, DPF. Particulate emissions were compared with emissions from a 9-liter Scania diesel engine from the same engine family, running on Swedish environmental class 1 diesel fuel. Tailpipe measurements of particle size and distribution were performed with a Scanning Mobility Particle Sizer (SMPS) instrument together with filter sampling. An evaluation of SMPS spectrometer measurements was performed for test conditions specified according to a 22-mode test cycle, which included the test modes in the European Stationary Cycle, ESC. Calculated weighted particle mass from SMPS spectrometer data, and in accordance with ESC, showed that the ethanol engine without emission control emitted approx. 1/12 of particle mass compared to the diesel engine. Weighted particulate emissions were reduced by approx. 96%, when the

engine was fitted with EGR and DPF. The reduction of weighted particulate emissions was even higher when the diesel engine was fitted with EGR and DPF, as high as 99%. Particle size and distribution measurements revealed that particles emitted from the ethanol engine mainly consisted of ultrafine particles (<100 nm), usually had a mean diameter of about 30 nm, while particles emitted from the diesel engine usually had mean diameters of about 60-70 nm and sizes going up to approx. 300 nm. Filter samples analyzed by Scanning electron microscopy and energy dispersive x-ray analysis SEM/EDX showed that the particles, both from the ethanol-fueled engine and the diesel-fueled engine mainly consisted of carbon and that they agglomerated, dependent upon running conditions, chainlike or clot-wise. Raman spectroscopy confirmed that the same elemental carbon was present in particles emitted from diesel and ethanol-fueled engines. The investigations showed that the system used, with EGR and DPF combined, is highly effective in reducing particulate emissions from ethanol- and diesel-fueled diesel engines. A general conclusion is also that the ethanol-fueled engine, equipped with emission control system or not, emitted lower particle mass, smaller particle sizes and approx. the same or a greater number of particles in the emissions than the diesel-fueled engine.

Van Gulijk, C., Marijnissen, J.C.M., Makkee, M., Moulijn, J.A., Schmidt-Ott, A., 2004, "Measuring diesel soot with a Scanning Mobility Particle Sizer and an electrical low-pressure impactor: performance assessment with a model for fractal-like agglomerates," *Journal of Aerosol Science*, **35**:633-655

The Scanning Mobility Particle Sizer (SMPS) spectrometer and the electrical low-pressure impactor (ELPI) are frequently used to measure particle size distributions of combustion aerosols. The instruments are especially popular for diesel exhaust measurements since the emission of "particulate matter" is restricted by legislation. A problem is that the interpretation of the results that these instruments give is not straightforward: fractal-like diesel soot agglomerates in the exhaust have a complex interaction with their carrier gas. This paper focuses on this complex interaction in order to assess its effect on the instruments' output. A theoretical model for the aerodynamic behavior of fractal-like agglomerates is used as a tool to assess the performance of the ELPI and the SMPS spectrometer for the measurement of diesel soot particles. The model couples the aerodynamic diameter and the mobility diameter for fractal-like soot agglomerates. Visual analysis with scanning electron microscopy (SEM) aids model development and instrument performance assessment. It is concluded that the performance of both instruments is affected by the fractal-like structure of diesel soot. The ELPI, if it is set to measure the aerodynamic diameter, gives an underestimation of the apparent size of particles due to their fractal-like structure. As a result, the number of particles is overestimated. The model presented in this paper helps interpretation of the ELPI's response. The SMPS is affected by multiple charging of soot agglomerates, larger than 1000 nm, which are abundantly present in diesel exhaust gas and are not removed by the impaction stage at the input due to their small effective density. The diesel engine in this paper emits many of such particles. Multiple charging leads to an underestimation of the size of the agglomerates and hampers size classification in the instrument.

Zervas, Efthimios; Dorlhene, Pascal; Daviau, Richard; Dionnet, Bernard, 2004, "Repeatability of fine particle measurement of diesel and gasoline vehicles exhaust gas." *Spring Fuels and Lubricants Conference and Exhibition*, June 8-10, 2004, Toulouse, France

Four diesel vehicles and two gasoline ones are used to determine the repeatability of the particle number and size measurements. Two analytical techniques are used: Scanning Mobility Particle Sizer (SMPS) spectrometer and Electrical Low Pressure Impactor (ELPI). The influence of technology (Euro2 and Euro3, diesel and gasoline vehicles, Diesel Particulate Filter (DPF), Gasoline Direct Injection (GDI)) and speed on the particle number and size is presented in the case of steady speeds and the European Driving Cycle (EDC). The repeatability of these measurements is determined at the entire particle distribution. The global 1.96\*Standard Deviation (SD) of the median diameter, determined by SMPS, is 8 nm. The median diameter is difficult to be determined in several cases due to the flat profiles of the emitted particles. The global 1.96\*Relative Standard Deviation (RSD) of the particle number presents a U-like curve, with a minimum value (55-57%) at about 100 nm. These values can reach the 180% (ELPI) and 280% (SMPS) spectrometer at the extremes of this curve.

Zhao, Bin; Yang, Zhiwei; Li, Zhigang; Johnston, M. V.; Wang, Hai. 2004, "Particle size distribution function of incipient soot in laminar premixed ethylene flames: effect of flame temperature," *Proceedings of the Combustion Institute*, **30**: 1441-1448

## 2003

Aigner, M.; Wahl, C., 2003, "Aircraft gas turbine soot emission tests under technical relevant conditions in an altitude test facility and validation of soot measurement technique," *American Society of Mechanical Engineers, International Gas Turbine Institute, Turbo Expo (Publication) IGTI*, **1**:189-200

An aircraft gas turbine was tested in an altitude test facility under technical relevant flight conditions. Test points are the ICAO power settings IDLE, TAKE OFF, CLIMB and DESCENT, as well as CRUISE

conditions. The used measurement techniques are Scanning Mobility Particle Sizer (SMPS) spectrometer 1 and Scanning Electron Microscopy (SEM). It is shown, that the relative Soot Emission Index (compared to take off conditions) is increasing with power setting. The mean diameter of the particles is also increasing with power setting. The particles show a lognormal size distribution for most of the test points. Under TAKE OFF conditions (100% power setting) we found a bimodal size distribution. The SEM pictures show the fractal character of the soot particles with 10 to 15 nm primary particles as well as the agglomerates. To validate the measurement techniques a variable soot generator 2 for nanoparticles was developed. This soot generator was used to check sample line losses. In an additional experiment we used this soot generator to test the trapping efficiency of the recommended filters for the so called ICAO Smoke Number which is the same as SAE- Smoke Number 3 or EPA - Smoke Number 4 .

Ayala, Alberto; Olson, Bernard; Cantrell, Bruce; Drayton, Marcus; Barsic, Nicholas, 2003, "Estimation of diffusion losses when sampling diesel aerosol: A quality assurance measure," *JSAE Technical Paper No. 20030138, 2003 JSAE/SAE International Spring Fuels and Lubricants Meeting*, May 19–22, 2003, Yokohama, Japan

Under the sponsorship of the Coordinating Research Council (CRC), the University of Minnesota (UMN) formed an international research team to investigate the physical and chemical nature of diesel emissions from heavy duty vehicles while operating on highways (CRC Project E-43). These ambient measurements of vehicle emissions following their release into and dilution by the atmosphere guided the development of dilution and sampling procedures for laboratory test cells to simulate on-highway conditions. The importance, visibility, and potential implications of the project prompted the adoption of a quality assurance (QA) plan by an independent implementation team. Because exhaust aerosol characterization for mobile sources lacks prescribed sampling methodologies, standard operating procedures were developed as part of the QA effort to ensure the consistency and validity of the data collected. To verify the daily protocols used, the QA team made surveillance visits to observe UMN team performance on project tasks. Evaluation of instrument performance using aerosols of known size was also done as part of QA system audits conducted to assess the accuracy of particle size measurements. QA for particle concentration measurement was hampered by the lack of a concentration standard, which is a problem common to aerosol science investigations today. Thus, the standard practice in aerosol work is to verify particle concentration by comparing results from two identical condensation particle counters (CPC) and rely on instrument manufacturer calibrations. A fundamental component QA for assessment of instruments and sampling system performance was investigation of particle losses. Along an aerosol sample pathway from source to collection media or measuring instrument, some particles are lost to surfaces. The magnitude of these losses as a function of particle size was determined experimentally by challenging the sampling trains with monodisperse particles in the sub-50 nm aerodynamic diameter size range (nm=10sup -sup 9 meters). Since the most probable loss mechanism was diffusion for sub-50 nm sized particles, theoretical calculations of diffusion loss for 100 nm particles and smaller were also made. Results indicated average sampling train total losses of approximately 50% and 20% for 10 nm and 17 nm size particles, respectively. Measured Scanning Mobility Particle Sizer (SMPS) instrument internal losses were approximately 70% for a 10 nm size mono-disperse aerosol. Measured sample line losses were better predicted by theory if the flow in the sampling lines is considered turbulent. However, the flow in the lines was laminar. This noted discrepancy may be the result of local turbulence created by valves and bends in the sample lines and is an area recommended for additional investigation. Finally, application of a particle loss correction to particle size distributions from the study increased SMPS number concentrations at 10 nm by approximately a factor of 5 and at 20 nm by a factor of 2.

Brundish, K. D.; Hilton, M.; Jefferies, M.; Johnson, M. P.; Miller, M. N.; Wilson, C. W., 2003, "Measurement of smoke particle size and distribution within a gas turbine combustor," *American Society of Mechanical Engineers, International Gas Turbine Institute, Turbo Expo (Publication) IGTI*, 2:455–462

The objective of the work described in this paper was to identify a method of making measurements of the smoke particle size distribution within the sector of a gas turbine combustor, using a Scanning Mobility Particle Sizing (SMPS) analyser. As well as gaining a better understanding of the combustion process, the principal reasons for gathering these data was so that they could be used as validation for Computational Fluid Dynamic (CFD) and chemical kinetic models. Smoke mass and gaseous emission measurements were also made simultaneously. A "water cooled," gas sampling probe was utilized to perform the measurements at realistic operating conditions within a generic gas turbine combustor sector. Such measurements had not been previously performed and consequently initial work was undertaken to gain confidence in the experimental configuration. During this investigation, a limited amount of data were acquired from three axial planes within the combustor. The total number of test points measured were 45. Plots of the data are presented in 2 dimensional contour format at specific axial locations in addition to axial plots to show trends from the primary zone to the exit of the combustor. Contour plots of smoke particle size show that regions of high smoke number concentration once formed in zones close to the fuel injector persist in a similar spatial location further downstream. Axial trends indicate that the average smoke particle size and number concentration diminishes as a function of

distance from the fuel injector. From a technical perspective, the analytical techniques used proved to be robust. As expected, making measurements close to the fuel injector proved to be difficult. This was because the quantity of smoke in the region was greater than 1000 mg/m<sup>3</sup>. It was found necessary to dilute the sample prior to the determination of the particle number concentration using SMPS. The issues associated with SMPS dilution are discussed.

Clark, Nigel N.; Gautam, Mridul; Boyce, James; Xie, Wenwei; Mehta, Sandeep; Jarett, Ron; Rapp, Byron, 2003, "Heavy-duty vehicle exhaust plume study in the NASA/Langley wind tunnel," *JSAE Technical Paper No. 20030290, 2003 JSAE/SAE International Spring Fuels and Lubricants Meeting*, May 19–22, 2003, Yokohama, Japan

Concern over health effects associated with diesel exhaust and debate over the influence of high number counts of particles in diesel exhaust prompted research to develop a methodology for diesel particulate matter (PM) characterization. As part of this program, a tractor truck with an electronically managed diesel engine and a dynamometer were installed in the Old Dominion University (ODU) Langley full-scale wind tunnel. This arrangement permitted repeat measurements of diesel exhaust under realistic and reproducible conditions and permitted examination of the steady exhaust plume at multiple points. Background particle size distribution was characterized using a Scanning Mobility Particle Sizer (SMPS) spectrometer. In addition, a remote sampling system consisting of a SMPS, PM filter arrangement, and carbon dioxide (CO<sub>2</sub>) analyzer, was attached to a roving gantry allowing for exhaust plume sampling in a three-dimensional grid. Raw exhaust CO<sub>2</sub> levels and truck performance data were also measured. The plume centerline was mapped and dilution ratios were determined and mapped. At 55 mph truck wheel speed and steady wind speed operation, at 200 inches behind the stack at the plume centerline, the dilution ratio was about 75, and at 337 inches behind the stack the dilution ratio was about 125. The truck was also operated through repeated transient tests, from 40 to 55 mph, with a wind speed of 47 mph. CO<sub>2</sub> variations in the plume were clearly detected during the transients. Steady-state PM size measurements were made at scan rates of 90 seconds. Ambient samplers located in the tunnel determined PM loading by mass. This paper presents the methodology used, discusses the plume location and dilution, and discusses results from ambient samplers. Particle sizing results are given in a companion paper.

Gautam, Mridul; Clark, Nigel N.; Mehta, Sandeep; Boyce, James A.; Rogers, Fred; Gertler, Alan, 2003, "Concentrations and size distributions of particulate matter emissions from a Class-8 heavy-duty diesel truck tested in a wind tunnel," *JSAE Technical Paper No. 20030291, 2003 JSAE/SAE International Spring Fuels and Lubricants Meeting*, May 19–22, 2003, Yokohama, Japan

In an effort to develop engine/vehicle test methods that will reflect real-world emission characteristics, West Virginia University (WVU) designed and conducted a study on a Class-8 tractor with an electronically controlled diesel engine that was mounted on a chassis dynamometer in the Old Dominion University Langley full-scale wind tunnel. With wind speeds set at 88 km/hr in the tunnel, and the tractor operating at 88 km/hr on the chassis dynamometer, a Scanning Mobility Particle Sizer (SMPS) spectrometer was employed for measuring PM size distributions and concentrations. The SMPS spectrometer was housed in a container that was attached to a three-axis gantry in the wind tunnel. Background PM size-distributions were measured with another SMPS unit that was located upstream of the truck plume. Ambient temperatures were recorded at each of the sampling locations. The truck was also operated through transient tests with vehicle speeds varying from 65 to 88 km/hr, with a wind speed of 76 km/hr. Sampling of the plume, with the truck operating at 88 km/hr revealed unimodal distributions with geometric mean diameter (GMD) values ranging from 55 to 80 nm. When size distributions from five locations in the plume were corrected for concentration with respect to dilution, they were found to be similar. Size distributions and concentrations of PM emissions from the tractor operating at 88 km/hr were found to agree with those from few other heavy-duty diesel vehicles that this team of researchers had previously tested in the field using the WVU Transportable Heavy-duty Vehicle Emissions Testing Laboratory. Under idle operation a distinct nuclei mode was detected with GMD varying from 14 to 24 nm.

Hirabayashi, Motohiro, 2003, "Size distribution of Ultra-Fine Particles in the Roadside Atmospheres," *JSAE Technical Paper No. 20035679, JSAE Autumn Conference*, September 17–19, 2003, Tokyo, Japan

Size distribution of ultra-fine particle was measured by using the Scanning Mobility Particle Sizer in the roadside of three places with different traffic conditions, in order to grasp the influence of exhaust ultra-fine particles to the atmospheric environment. The high-concentration, ultra-fine particles under the 50 nm diameter that is thought it originates in the vehicle exhaust, was observed in each roadside atmospheres. It was found that the heavy-duty vehicles have contributed largely for the particles under 50 nm diameter, and the contribution of exhaust soot is large for the particles above 50 nm diameter.

Hope A. M., 2003, "Development of detection techniques and diagnostics for airborne carbon nanoparticles," *Technical Report*, Sandia National Laboratories, Albuquerque, NM; Livermore, CA, US, Report No. 2003-8666

We have recorded time-resolved LII signals from a laminar ethylene diffusion flame over a wide range of laser fluences at 532 nm. We have performed these experiments using an injection-seeded Nd:YAG laser with a pulse duration of 7 ns. The beam was spatially filtered and imaged into the flame to provide a homogeneous spatial profile. These data were used to aid in the development of a model, which will be used to test the validity of the LII technique under varying environmental conditions. The new model describes the heating of soot particles during the laser pulse and the subsequent cooling of the particles by radiative emission, sublimation, and conduction. The model additionally includes particle heating by oxidation, accounts for the likelihood of particle annealing, and incorporates a mechanism for nonthermal photodesorption, which is required for good agreement with our experimental results. In order to investigate the fast photodesorption mechanism in more detail, we have recorded LII temporal profiles using a regeneratively amplified Nd:YAG laser with a pulse duration of 70 ps to heat the particles and a streak camera with a temporal resolution of approximately 65 ps to collect the signal. Preliminary results confirm earlier indications of a fast mechanism leading to signal decay rates of much less than a nanosecond. Parameters to which the model is sensitive include the initial soot temperature, the temperature of the ambient gas, and the partial pressure of oxygen. In order to narrow the model uncertainties, we have developed a source of soot that allows us to determine and control these parameters. Soot produced by a burner is extracted, diluted, and cooled in a flow tube, which is equipped with a Scanning Mobility Particle Sizer (SMPS) spectrometer for characterization of the aggregates.

Johnson, M. P.; Hilton, M.; Waterman, D. R.; Black, J. D., 2003, "Development of techniques to characterize particulates emitted from gas turbine exhausts," *Measurement Science and Technology*, **14**

Particles emitted from aircraft play a role in the formation of contrails and it is essential to characterize them to understand the physical and chemical processes that are happening. Current methods for measuring aircraft particulate emissions study the reflectance of samples collected in filter papers. A series of experiments to more fully characterize particulates has been performed on a small-scale gas turbine engine. An intrusive sampling system conforming to current ICAO regulations for aircraft emissions was used with a Scanning Mobility Particle Sizer (SMPS) spectrometer. Non-intrusive measurements were made using laser induced incandescence (LII) and samples were taken from the exhaust to analyze using a transmission electron microscope. Results obtained from different techniques showed good agreement with each other. As engine power conditions increased, both the SMPS and LII indicated that the mass of soot had decreased. Differences were observed between measurements of diluted and undiluted samples. The mean particle size decreased with dilution but the size distribution became bi-modal. The study has shown how significant the sampling environment is for measuring particulates and careful techniques need to be used to ensure that accurate, consistent results can be obtained.

Johnson, T; Caldow, R; Pucher, A; Mirme, A; Kittelson, D, 2003, "An Engine Exhaust Particle Sizer™ spectrometer for transient emission particle measurements," *9th Diesel Engine Emissions Reduction (DEER) Workshop 2003*, Newport, RI (US), 08/24/2003-08/28/2003

There has been increased interest in obtaining size distribution data during transient engine operation where both particle size and total number concentrations can change dramatically. Traditionally, the measurement of particle emissions from vehicles has been a compromise based on choosing between the conflicting needs of high time resolution or high particle size resolution for a particular measurement. Currently the most common technique for measuring submicrometer particle sizes is the Scanning Mobility Particle Sizer™ (SMPS™) system. The SMPS system gives high size resolution but requires an aerosol to be stable over a long time period to make a particle size distribution measurement. A Condensation Particle Counter (CPC) is commonly used for fast time response measurements but is limited to measuring total concentration only. This paper describes a new instrument, the Engine Exhaust Particle Sizer™ (EEPS™) spectrometer, which has high time resolution and a reasonable size resolution. The EEPS spectrometer was designed specifically for measuring engine exhaust and, like the SMPS system, uses a measurement based on electrical mobility. Particles entering the instrument are charged to a predictable level, then passed through an annular space where they are repelled outward by the voltage from a central column. When the particles reach electrodes on the outer cylindrical (a column of rings), they create a current that is measured by an electrometer on one or more of the rings. The electrometer currents are measured multiple times per second to give high time resolution. A sophisticated real-time inversion algorithm converts the currents to particle size and concentration for immediate display.

Khalek, Imad A.; Fritz, Steven G.; Paas, Norbert, 2003, "Particle size distribution and mass emissions from a mining diesel engine equipped with a dry system technologies emission control system," *JSAE Technical Paper No. 20030244, 2003 JSAE/SAE International Spring Fuels and Lubricants Meeting*, May 19–22, 2003, Yokohama, Japan

Particle size distribution, number, and mass emissions from the exhaust of a 92 kW 1999 Isuzu 6BG1 non-road naturally aspirated diesel engine were measured. The engine exhaust was equipped with a Dry System Technologies(R) (DST) auxiliary emission control device that included an oxidation catalyst, a heat exchanger, and a disposable paper particulate filter. Particle measurement was taken during the ISO 8178 8-mode test for engine out and engine with the DST using a Scanning Mobility Particle Sizer (SMPS) spectrometer in parallel to the standard filter method (SFM), specified in 40 CFR, Part 89. The DST efficiency of removing particles was about 99.9 percent based on particle number, 99.99 percent based on particle mass derived from number and size. However, the efficiency based on mass derived from the SFM was much lower on the order of 90 to 93 percent. This discrepancy in particle mass efficiency between the two methods was mainly due to poor correlation between the mass derived from number and size and the mass measured using the SFM. This poor correlation was obtained for the DST out particulate matter (PM) emission and not for engine out. For DST out PM, the SFM tends to exaggerate the mass emissions of particles due to the condensation/adsorption of gas phase non-aerosol volatile compounds on the Palflex T60A20 filters used for particle mass measurement, thus resulting in a lower efficiency of particle removal. If the health effect of particles is due to the mass and number of their physical characteristics, particle measurement using the SFM may need rethinking, particularly if the nature of the measured product is dominated by volatile rather than soot.

Khalek, Imad A.; Spears, Matt; Charmley, William, 2003, "Particle size distribution from a heavy-duty diesel engine: Steady-state and transient emission measurement using two dilution systems and two fuels," *SAE 2003 World Congress*, March 3–6, 2003, Detroit, Michigan, USA

Particle size distribution and number concentration were measured in the dilute exhaust of a heavy-duty diesel engine for steady-state and transient engine operation using two different dilution systems that included a full flow CVS that was coupled to an ejector pump (CVS-EP), and a double-ejector, micro-dilution tunnel (DEMDT) that was connected to engine exhaust close to turbocharger outlet. Measurements were performed using a Scanning Mobility Particle Sizer (SMPS) spectrometer, an electrical low-pressure impactor (ELPI), and a parallel flow diffusion battery (PFDB). Fuels with sulfur content of about 385 ppm and 1 ppm were used for this work. The PFDB performed well in measuring nanoparticles in the size range below 56 nm when compared with the SMPS spectrometer. This was especially valid when a distinct log-normal size distribution in the size range below 56 nm in diameter, the upper size limit of the PFDB, was present. The dilution method and the sulfur content in the fuel influenced the characteristics of the size distributions significantly under both steady-state and the FTP hot-start transient cycle. The CVS-EP resulted in more particle growth and higher number of nanoparticles compared to the DEMDT. Harmonizing the measurement method of total particle number and size emissions from engines by providing a reference condition to variables such as dilution temperature, dilution rate, dilution ratio, relative humidity, dilution and transfer line residence time, will be a key next step in developing a protocol and a standard operating procedure for such measurement. Until then, different researchers will continue to produce different results in measuring total particle size and number, depending on the sampling and dilution system selected.

Kweon, Chol-Bum; Okada, Shusuke; Stetter, John C.; Christenson, Charles G.; Shafer, Martin M.; Schauer, James J.; Foster, David E., 2003, "Effect of fuel composition on combustion and detailed chemical/physical characteristics of diesel exhaust," *JSAE Technical Paper No. 20030098, 2003 JSAE/SAE International Spring Fuels and Lubricants Meeting*, May 19–22, 2003, Yokohama, Japan

An experimental study was performed to investigate the effect of fuel composition on combustion, gaseous emissions, and detailed chemical composition and size distributions of diesel particulate matter (PM) in a modern heavy-duty diesel engine with the use of the enhanced full dilution tunnel system of the Engine Research Center (ERC) of the UW Madison. Detailed description of this system can be found in our previous reports. The experiments were carried out on a single-cylinder, 2.3-liter D.I. diesel engine equipped with an electronically controlled unit injection system. The operating conditions of the engine followed the California Air Resources Board (CARB) 8-mode test cycle. The fuels used in the current study include baseline No. 2 diesel (Fuel A: sulfur content = 352 ppm), ultra-low-sulfur diesel (Fuel B: sulfur content = 14 ppm), and Fisher Tropsch (F-T) diesel (sulfur content = 0 ppm). Samples were collected on a series of Teflon and baked quartz fiber filters to evaluate mass loading, elemental and organic carbon (EC/OC), sulfates (SO<sub>4</sub><sup>2-</sup>), and trace metals for the three different fuel compositions. A Scanning Mobility Particle Sizer (SMPS) spectrometer was used to measure particle number concentrations and size distributions. Results show that the fuel composition significantly affected combustion, mass loading, chemical composition, and number concentrations and size distributions of PM. Mass loading, EC, and sulfates were significantly lowered with the advanced diesel fuels (Fuel B and

F-T fuel) at higher loads. The advanced diesel fuels also produced more nucleimode particles and less accumulation-mode particles than Fuel A at high load conditions. In contrast, the former produced less nuclei-mode particles and more accumulation mode particles than the latter at the medium load conditions.

Kweon, Chol-Bum; Okada, Susuke; Stetter, John C.; Christenson, Charles G.; Shafer, Martin M.; Schauer, James J.; Foster, David E., 2003, "Effect of injection timing on detailed chemical composition and particulate size distributions of diesel exhaust," *JSAE Technical Paper No. 20030099, 2003 JSAE/SAE International Spring Fuels and Lubricants Meeting*, May 19–22, 2003, Yokohama, Japan

An experimental study was carried out to investigate the effects of fuel injection timing on detailed chemical composition and size distributions of diesel particulate matter (PM) and regulated gaseous emissions in a modern heavy-duty D.I. diesel engine. These measurements were made for two different diesel fuels: No. 2 diesel (Fuel A) and ultralow-sulfur diesel (Fuel B). A single-cylinder, 2.3-liter D.I. diesel engine equipped with an electronically controlled unit injection system was used in the experiments. PM measurements were made with an enhanced full-dilution tunnel system at the Engine Research Center (ERC) of the University of Wisconsin-Madison (UW Madison). The engine was run under 2 selected modes (25% and 75% loads at 1200 rpm) of the California Air Resources Board (CARB) 8-mode test cycle. Samples were collected on a series of Teflon and baked quartz fiber filters to evaluate mass loading, elemental and organic carbon (EC/OC), and sulfates ( $\text{SO}_{4\text{sup } 2\text{sup } -}$ ) for several injection timings with the two different fuel compositions. A Scanning Mobility Particle Sizer (SMPS) spectrometer was used to measure particle number concentrations and size distributions in the residence time chamber (RTC). Results show that the advanced injection timing significantly affected detailed chemical composition and size distributions of the PM. The advanced injection timing significantly decreased EC and modestly decreased OC, particularly at the higher load (mode 6). The impact on OC with changing fuel composition and the fuel composition's effect on changes in OC with injection timing were negligible and statistically insignificant. Sulfates did not change with the injection timing and/or engine load for Fuel B, however, Fuel A showed significant variations in the sulfates with the injection timing and/or engine load.

Lapuerta, M; Armas, O; Gomez, A, 2003, "Diesel particle size distribution estimation from digital image analysis," *Aerosol Science and Technology*, **37**:369–381

One of the most serious problems associated with Diesel engines is pollutant emissions, especially nitrogen oxides and particulate matter. However, although current emissions standards in Europe and America with regard to light vehicles and heavy duty engines refer to the particulate limit in mass units, there has been increasing concern of late to know the size and number of particles emitted by engines. This interest has been promoted by the latest studies about the harmful effects of particles on health and is enhanced by recent changes in internal combustion engine technology. This study is focused on the implementation of a method to determine the particle size distribution that could be appropriate for the current methodology of vehicle certification in Europe. This method uses an automated Digital Image Analysis Algorithm (DIAA) to determine particle size trends from Scanning Electron Microscope (SEM) images of filters charged in a partial dilution system used for measuring specific particulate emissions. The experimental work was performed on a stationary electric generation direct injection Diesel engine with 0.5 MW (671 hp) rated power, which is considered as a typical engine in middle power industries. Particulate size distributions obtained using DIAA were compared with distributions obtained using an Optical Particle Counter (OC) and a Scanning Mobility Particle Sizer (SMPS) spectrometer, the latter currently considered as the most reliable technique. Although the number concentration detected by this method does not represent the real flowing particle concentration, the algorithm gives a fair reproduction of the trends observed with on-line techniques (SMPS and OC) when the engine load is varied.

Lehmann, U.; Mohr, M.; Schweizer, T.; Ruetter, J., 2003, "Number size distribution of particulate emissions of heavy-duty engines in real world test," *Atmospheric Environment*, **37**(37):5247–5259

Five in-service engines in heavy-duty trucks complying with Euro II emission standards were measured on a dynamic engine test bench at EMPA. The particulate matter (PM) emissions of these engines were investigated by number and mass measurements. The mass of the total PM was evaluated using the standard gravimetric measurement method, the total number concentration and the number size distribution were measured by a Condensation Particle Counter (lower particle size cut-off: 7 nm) and an Electrical Low Pressure Impactor (lower particle size: 32 nm), respectively. The transient test cycles used represent either driving behaviour on the road (real-world test cycles) or a type approval procedure. They are characterized by the cycle power, the average cycle power and by a parameter for the cycle dynamics. In addition, the particle number size distribution was determined at two steady-state operating modes of the engine using a Scanning Mobility Particle Sizer spectrometer. For quality control, each measurement was repeated at least three times under controlled conditions. It was found that the number size distributions as well as the total number concentration of emitted particles could be measured with a good repeatability. Total number concentration was between  $9 \times 10^{13}$  and  $1 \times 10^{13}$  particles/s ( $3 \times 10^{13}$  -  $7 \times 10^{14}$  p/kWh) and mass concentration was between 0.09 and 0.48 g/kWh. For all

transient cycles, the number mean diameter of the distributions lay typically at about 120 nm for aerodynamic particle diameter and did not vary significantly. In general, the various particle measurement devices used reveal the same trends in particle emissions. We looked at the correlation between specific gravimetric mass emission (PM) and total particle number concentration. The correlation tends to be influenced more by the different engines than by the test cycles.

Maguhn, J.; Karg, E.; Kettrup, A.; Zimmermann, R., 2003, "On-line Analysis of the Size Distribution of Fine and Ultrafine Aerosol Particles in Flue and Stack Gas of a Municipal Waste Incineration Plant: Effects of Dynamic Process Control Measures and Emission Reduction Devices," *Environmental Science & Technology*, **37**(20):4761-4770

The size distribution of particles in the waste gas of a municipal waste incineration plant (23 MW) was measured on-line at two sampling points in the flue-gas duct (700 and 300 degree C) as well as in the stack gas (80 degree C). The measurements were performed during both stable combustion conditions and transient operating conditions. The particle measurements were carried out by a mobile system consisting of a home-designed sampling system with dilution device and a Scanning Mobility Particle Sizer (SMPS) spectrometer for the particle size range 17-600 nm as well as an Aerodynamic Particle Sizer (APS) spectrometer for the size range 500 nm-30  $\mu$  m. The APS and SMPS data were combined using a special method and a home written software tool. The maximum of the particle-size distribution in the flue gas of the incinerator shifts from about 90 nm at the 700 degree C sampling point to about 140 nm at the 300 degree C point, showing the particle growth by coagulation processes and condensation of inorganic and organic gaseous species with decreasing temperature. This finding is consistent with the measured concentration profiles of gaseous organic chemical species in the flue gas. While at flue-gas temperatures of 600-800 degree C a rich pattern of polycyclic aromatic hydrocarbon species (PAH) is observable, the PAH concentrations are considerably reduced further downstream of the flue-gas channel, where the temperature drops below 500 degree C. Condensation and reactive bonding of gaseous chemicals onto particulate matter is, among other reasons, responsible for the depletion of gas-phase species. Process control measures, such as firing the backup burners or cleaning of the grate with pressurized air, can cause dynamic changes of the particle-size distribution. Furthermore the flue-gas cleaning measures have great impact onto both the particle concentration and the size distribution. For this reason the impact of one particular emission reduction device, the wet electrostatic dust precipitator (wet-ESP), is evaluated. The wet-ESP reduces considerably the particle concentration over the whole size range. Behind the flue-gas processing units a broad maximum in the particle-size distribution occurs at about 70 nm, but no pronounced particle-size distribution could be observed. The particle concentration level at this maximum is about 3 magnitudes lower than in the raw flue gas. However, intermittent periods lasting for several minutes of high emissions of ultrafine particles with  $d < 40$  nm were observed. These particles are most likely formed by nucleation processes behind the wet-ESP from gas-phase constituents of the stack gas.

McMurry, P. H.; Cao, F.; Kittelson, D. B.; Park, K., 2003, "Relationship between particle mass and mobility for diesel exhaust particles," *Environmental Science & Technology*, **37**(3):577-583

We used the aerosol particle mass analyzer (APM) to measure the mass of mobility-classified diesel exhaust particles. This information enabled us to determine the effective density and fractal dimension of diesel particles as a function of engine load. We found that the effective density decreases as particle size increases. TEM images showed that this occurs because particles become more highly agglomerated as size increases. Effective density and fractal dimension increased somewhat as engine load decreased. TEM images suggest that this occurs because these particles contain more condensed fuel and/ or lubricating oil. Also, we observed higher effective densities when high-sulfur EPA fuel (approx. 360 ppm S) was used than for Fischer-Tropsch fuel (approx. 0 ppm S). In addition, the effective density provides the relationship between mobility and aerodynamic equivalent diameters. The relationship between these diameters enables us to intercompare, in terms of a common measure of size, mass distributions measured with the Scanning Mobility Particle Sizer (SMPS) spectrometer and a MOUDI impactor without making any assumptions about particle shape or density. We show that mass distributions of diesel particles measured with the SMPS-APM are in good agreement with distributions measured with a MOUDI and a nano-MOUDI for particles larger than approx 60 nm. However, significantly more mass and greater variation were observed by the nano-MOUDI for particles smaller than 40 nm than by the SMPS-APM.

Okada, Shusuke; Kweon, Chol-Bum; Stetter, John C.; Foster, David E.; Shafer, Martin M.; Christensen, Charles G.; Schauer, James J.; Schmidt, Alexandra M.; Silverberg, Amy M.; Gross, Deborah S., 2003, "Measurement of trace metal composition in diesel engine particulate and its potential for determining oil consumption: ICPMS (inductively coupled plasma mass spectrometer) and ATOFMS (aerosol time of flight mass spectrometer) measurements," *SAE 2003 World Congress*, March 3-6, 2003, Detroit, Michigan, USA

Current regulations stipulate acceptable levels of particulate emissions based on the mass collected on filters obtained by sampling in diluted exhaust. Although precise, this gives us only aggregated



information. In addition to the mass-based measurements, detailed chemical analysis of the particulate matter (PM) is performed, additional subtle information about the combustion process can be revealed. This paper reports the results of detailed chemical analysis of trace metal in the PM emitted from a single-cylinder, heavy-duty diesel engine. The trace metal concentrations are used as an indicator of oil consumption. Two techniques were used to make the trace metal concentration measurements. PM was captured on filters and trace metals were quantified with an Inductively Coupled Plasma Mass Spectrometer (ICPMS), and also an Aerosol Time-of-Flight Mass Spectrometer (ATOFMS) was used to perform particle size and composition measurements in real time. Particle size distributions were also measured using a Scanning Mobility Particle Sizer (SMPS) spectrometer. Aerosol Time-of-Flight Mass Spectrometers have been widely used to study atmospheric aerosols, but only limited ATOFMS measurements have been collected for internal combustion engine exhaust emissions. The instrument yields data of a single particle size and composition for a particle size range from 0.2 to 3 micrometers. Mass spectral composition analyses are completed at a rate of up to  $t \sim 200$  particles per minute. In this work we compare trace metals, elemental carbon (EC) and organic carbon (OC) concentrations from the ATOFMS with more traditional filter-based and particle size distribution data to assess the utility of using ATOFMS for real-time fundamental engine exhaust studies. The data presented covers the operating modes of California Air Resources Board (CARB 8 mode) emissions test. Dramatic changes in trace metals, EC and OC, and PM size with changes in the engine operating modes are evident from both filters and ATOFMS, which illustrates that the chemical and physical characteristics of the PM, especially trace metals, EC and OC concentrations, are highly dependant on the engine operating conditions. The results show chemical compositions of each particle and the contribution of the lubricating oil to the PM.

Wong, C. P.; Chan, T. L.; Leung, C. W., 2003, "Characterisation of diesel exhaust particle number and size distributions using mini-dilution tunnel and ejector-diluter measurement techniques," *Atmospheric Environment*, **37**(31):4435–4446

This paper presents the characteristics of diesel exhaust particle number and size distributions. These were measured for different engine load conditions from 10% to 100% of full engine load at a maximum torque of constant speed, using mini-dilution tunnel and sampling (MDTS) and ejector-diluter and sampling (EDS) systems. The exhaust particles sampled were analyzed using a Scanning Mobility Particle Sizer (SMPS) spectrometer. In general, the particle number concentration increased with increasing engine load using both measurement systems. Comparing the particle number and volume concentrations, the MDTS system measures a lower level in the nanoparticle range,  $D_p < 50$  nm, but a higher level in the accumulation mode size range,  $50 < D_p < 670$  nm, than the EDS system. The measurements also showed that the MDTS system shifted the particle count median diameter (CMD) to larger particle diameter and the particle number size and volume distribution for all engine load conditions. It is mainly because the mini-dilution tunnel leads to the particle transformations of nucleation and condensation taking place simultaneously when the exhaust particle emissions are cooled and diluted. However, the effect of coagulation on the total number particle concentration was shown to be negligible. On the other hand, the EDS measurement system can minimize the particle transformations taking place on the exhaust particle number and size distributions during the heated dilution process. Hence, the EDS measurement system can provide more reliable diesel exhaust particle number and size distributions than MDTS measurement system.

Zhao, B.; Yang, Z.; Johnston, M. V.; Wang, H.; Wexler, A. S.; Balthasar, M.; Kraft, M., 2003, "Measurement and numerical simulation of soot particle size distribution functions in a laminar premixed ethyleneoxygen-argon flame," *Combustion and Flame*, **133**(1–2):173–188

Spatially resolved measurement of the soot particle size distribution function (PSDF) was made in a laminar premixed ethylene-argon-oxygen flame ( $\phi = 2.07$ ) using a Scanning Mobility Particle Sizer spectrometer. The emphasis of the study was to follow the evolution of the PSDF from the onset of particle inception to particle mass growth. At the onset of soot inception, the PSDF was found to follow a power-law dependence on particle diameter. The PSDF becomes bimodal at larger height above the burner surface, and remains bimodal throughout the flame. Numerical simulation using a kinetic model proposed previously and a stochastic approach to solve aerosol dynamics equations again showed a bimodal PSDF. Further analysis revealed that bimodality is intrinsic to an aerosol process involving particle-particle coagulation and particle nucleation dominated by monomer dimerization.

Zhao, B.; Yang, Z.; Wang, J.; Johnston, M. V.; Wang, H., 2003, "Analysis of Soot Nanoparticles in a Laminar Premixed Ethylene Flame by Scanning Mobility Particle Sizer," *Aerosol Science and Technology*, **37**(8):611–620

Mobility size distributions of soot particles produced from a fuel-rich, laminar premixed ethylene flat flame were obtained by in situ probe sampling and online analysis using a nano Scanning Mobility Particle Sizer spectrometer. The emphasis of the work was the development of an in situ sampling technique to follow the evolution of nanoparticles formed in flames. Particle size distribution functions were obtained along the centerline of the flame in a spatially resolved manner. Considerable efforts were

made to eliminate particle losses in the sample probe. To this end, the effect of dilution on particle losses in the sample probe was systematically studied. It is demonstrated that particle losses due to coagulation and diffusive wall deposition were negligible using a dilution ratio greater than  $\sim 10^4$ . The sampling technique is shown to be capable of closely following the evolution of particle size distribution from the nucleation mode to mass growth mode dominated by particle coagulation and gas-surface reactions. Beyond the particle nucleation region of the flame, the size spectra were found to be distinctively bimodal, indicating sustained particle nucleation throughout the flame studied.

## 2002

Blevins, L. G.; Cauley III, T. H., 2002, "Fine particulate formation during biomass/coal cofiring," *ASME Heat Transfer Div Publ HTD*, **372**(3):295–299

Experiments to examine the effects of biomass/coal cofiring on fine particle formation were performed in the Sandia Multi-Fuel Combustor using fuels of pure coal, 3 combinations of switchgrass and coal, and pure switchgrass. A constant thermal input was maintained. The combustion products were cooled during passage through the 4.2 m long reactor to simulate the temperatures experienced in the convection pass of a boiler. Fine particle number densities, mass concentrations, and total number concentrations for particles between 10 nm and 1  $\mu$ m at the reactor exit were determined using a Scanning Mobility Particle Sizer spectrometer. The results indicate that the fine particle loading for cofiring is higher than that achieved with dedicated coal combustion but lower than that achieved with dedicated switchgrass combustion. 9 Refs.

Fogh, C.; Jensen, J. N.; Livbjerg, H.; Lund, C.; Nielsen, M. T.; Poulsen, K.; Sander, B.; Simonsen, P., 2002, "Formation and emission of fine particles from two coal-fired power plants," *Combustion Science and Technology*, **174**(2):79–113

A field study of the formation and emission of fine particles from two Danish full-scale, coal-fired power plants at Avedriv similar to Qrket and Nordjyllandsv similar to Qrket was performed using a so-called multi-platform approach. The measurements were made simultaneously at three positions in the plants: platform 1 at the inlet to the electrostatic precipitator, platform 2 at the outlet of the electrostatic precipitator, and platform 3 in the stack. The combined data from the three measuring platforms provide information about all key particle processes in the plant, such as the generation of particles in the boiler, the ESP filtering efficiency, the influence of the flue gas desulfurization plant, and the emissions from the stack. 23 Refs.

Franke, H. U., 2002, "Exhaust gas concept for EURO 4 passenger car diesel engine. Subproject: Various measurements and analyses of particle emissions, carried out on the 1.9 l PDE engine equipped with diesel particle filter," Report, Magdeburg Univ.

A complex research, leading to accomplishment of EURO 4 exhaust limits, should be examined in the case of a Diesel engine, equipped with a direct injection PDE system. Tests have been done on the stationary test bench, as well as on two passenger cars driven on the chassis dynamometer. Diesel fuel with a sulphur content of 45 ppm has been used. To decrease the soot ignition temperature, different additives have been employed. An oxidation catalyst with DeNO<sub>x</sub> effect found a place in front of the Diesel particle filter (DPF). The data of particle mass, particle size distribution particle number as well as morphological structure of particles and agglomerates have been examined by the use of a Berner low pressure impactor, a Scanning Mobility Particle Sizer (SMPS) spectrometer and a morphological measurement system. Generally, the particle emission reduction has been observed at all engine loads with every further modification of an engine control unit, when DPF designed by HJS company was applied. The efficiency of HJS DPF no. 1 was 98,1% by 25% load, 97,9% by 50% load and 87,9% by 75% engine load. A similar conclusion can be also done for the HJS DPF no. 2. The morphological research results are more or less similar for both HJS DPF versions. The mean agglomerate diameter is about 600 nm in front of DPF. However, there are less single particles but many drops with and without solid nuclei behind DPF. Single particles then usually have size of 8 nm, drops about 50 nm. The highest particle filter efficiency of DPF designed by OM company (version no. 1) has been found in the case of the test car VP 91, particularly by 25% engine load. The efficiency rate was 74,6%. Due to a very high soot concentration, caused by an ECU modification, it was not possible to carry out the same measurements on the stationary engine test bench. In the case of OM DPF no. 2, tests on the stationary test bench have shown that the particle filter efficiency is approximately 45%. Besides, the results show that DPF regeneration requires an improvement of exhaust gas temperature, especially within an idle operation mode. Unfortunately, available hardware and software equipment was not enough to reach this higher temperature level at every operation mode of tested PDE Diesel engine.

Jeuland, N.; Dementhon, J. B.; Plassat, G.; Coroller, P.; Momique, J. C.; Belot, G.; Bruchet, D., 2002, "Performances and durability of DPF (diesel particulate filter) tested on a fleet of Peugeot 607 Taxis -

First and second test phases results," *Powertrain and Fluid Systems Conference and Exhibition*, October 21–24, 2002, San Diego, California, USA

The use of diesel engines has strongly increased during the last years and now represents 30% of the sales in Europe and up to 50% of the number of cars in circulation for some countries. This success is linked not only to the economical aspect of the use of such vehicles, but also to the recent technological improvements of these engines. The new technical solutions (high pressure direct injection, turbocharging ...) have indeed allowed the increase of these engine performances while decreasing their fuel consumption, pollutant emissions and noise level. From an environmental point of view, diesel engines are nevertheless penalized by their particulate and NO<sub>x</sub> emissions. The study and the treatment of the particulate, highly criticized for their potential impact on health, are the subject of numerous works of characterization and developments. PSA Peugeot-Citroen has recently launched its particulate filter technology on several types of vehicles. In order to evaluate the durability of this technology over a long period of time, a study program has been set-up by ADEME (French environmental agency), IFP Powertrain, PSA Peugeot-Citroen and G7 (a Parisian taxi company). The objective is to study the evolution of several taxis and their after-treatment system performances over 80,000 km mileage in hard urban driving conditions, which corresponds to the recommended mileage before the first DPF maintenance. More specifically, the following evaluations are being performed at regular intervals (around 20,000 km):

- Regulated gaseous pollutant emissions on NEDC cycle (New European Driving Cycle);
- Particulate emissions, by mass measurement on NEDC but also by particle number and size measurement with SMPS (Scanning Mobility Particle Sizer) technique on NEDC and on unconventional steady-state running points;
- Unregulated pollutant emissions (hydrocarbons speciation C<sub>1</sub>-C<sub>13</sub>, oxygenated compounds). The results obtained until now have not shown any degradation of the particulate filter efficiency. This paper presents the methodology setup and the explanation of the first results obtained. Indeed, a more specific study has shown that most of the aerosols measured with SMPS are composed of liquid fractions, mainly sulfates due to sulfur coming from the fuel, but also from the lubricant. The impact of sulfates stored on the catalyst surface during low-temperature running phases and removed during high-temperature running phases has also been outlined.

Kweon, Chol-Bum; Foster, David E.; Schauer, James J.; Okada, Shusuke, 2002, "Detailed chemical composition and particle size assessment of diesel engine exhaust," *Powertrain and Fluid Systems Conference and Exhibition*, October 21–24, 2002, San Diego, California, USA

A dilution source sampling system has been incorporated into the exhaust measurement system of a research single-cylinder diesel engine. To allow more detailed assessment of the individual chemical components of the diesel particulate matter (PM) the exhaust dilution system includes a residence time chamber (RTC) to allow for residence times of 30 to 60 seconds in the second stage of dilution before sampling. Samples are collected on a range of different filters where mass loading, elemental and organic carbon (ECOC), trace metals, sulfate ions (SO<sub>4</sub>), particle-phase organic compounds, and semi-volatile organic compounds are evaluated. In addition, particle size distributions have been determined using a Scanning Mobility Particle Sizer (SMPS) spectrometer. Results show that the chemical composition of the particulate matter is highly dependent on the engine operating conditions. There is a dramatic shift in the ratio of elemental carbon to organic compounds and in the sulfate ions (SO<sub>4</sub>) and trace metals when the engine is traversed across a load and speed range. Similarly there is a shift in the particle size range for which there is virtually no impact on the mass loading.

Sasaki, Sousuke; Nakajima, Toru, 2002, "Study on the measuring method of vehicular PM size distribution to simulate the atmospheric dilution process," *Powertrain and Fluid Systems Conference and Exhibition*, October 21–24, 2002, San Diego, California, USA

The measuring method of vehicular particulate matter (PM) size distribution to simulate the atmospheric dilution process was studied. PM size distribution was measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer. To simulate the atmospheric dilution process with a chassis dynamometer test, a chasing experiment was done in order to obtain reference data. A light-duty diesel truck was selected as a basic test vehicle. Three sizes of prototype partial flow diluters (PPFD) were made to reproduce the PM size in the atmosphere. The PM sizes of the chasing experiment and the PPFDF experiment was roughly agreed. Differences in the data obtained from a full flow dilution tunnel and the chasing experiments were investigated. The length of the transfer tube greatly affected the smaller side of the PM number concentration

Zellbeck, H.; Friedrich, J.; Starosta, H.; Schulze, T.; Blei, S.; Liebsch, S.; Reuss, H.C.; Kaemmer, A.; Deutschmann, R., "Methods and tools for system optimization of combustion engines at Dresden University of Technology" *Emission control 2002*. Proceedings, Dresden (Germany), June 10–11, 2002

Reducing fuel consumption and exhaust gas emission of internal combustion engines at Dresden University of Technology computer simulation, experimental tests and hardware in the loop simulations (HiL) are developed and used. Based on MATLAB/SIMULINK mean value models were made for

optimizing system performance of super charged combustion engines as well as injection systems. A HiL-Simulation is shown, to influence Sauter mean diameter by a directly actuated piezo Diesel injector in which the fuel is injected through 2 rows of different sized nozzle holes. Also tests of electronic engine control systems are done by HiL-simulation. Optimizing combustion development process a measurement system was developed supported by the FVV to detect liquid Diesel fuel wetting the piston bowl. With the presented Scanning Mobility Particle Sizer the particle size distribution of row emission and after filter was measured. The filter efficiency was over 99.9%. On a high dynamic engine test bench the real vehicle driving on the road is simulated (HiL) measuring exhaust gas emission by a fast exhaust gas analyzer. During an acceleration mode peaks of HC and NO-emission as well as smoke emission were detected.

## 2001

Abu-Qudais, M.; Kittelson, D.; Matson, A., 2001, "Combination of methods for characterization diesel engine exhaust particulate emissions," *JSME International Journal, Series B: Fluids and Thermal Engineering*, **44**(1):166–170

This paper describes a study of the exhaust aerosols produced by a diesel engine. A combination of techniques for collecting and measure particulate matter in a diluted exhaust gases are presented. Three techniques have been used: a Micro Orifice Uniform Deposit Impactor (MOUDI), a Low Pressure Impactor (LPI) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. A direct injection naturally aspirated diesel engine was used in the study at three different equivalence ratios: 0.3, 0.45, and 0.6 at an engine speed of 1 400 rpm which is rated torque speed. Mass concentration measurements made with the MOUDI were in qualitative, but not quantitative, agreement with those calculated from the aerosol volume concentrations measured by the SMPS. The particulate matter obtained from the LPI was analyzed using transmission electron microscope and was found to be comprised of individual spherical particles ranging from 10 nm to 50,nm with a mean size of approximately 25 nm. Some conclusions about the size distribution measurement possibilities can be drawn. 4 Refs.

Armas, O.; Ballesteros, R.; Gomez, A., 2001, "Morphological Analysis of Particulate Matter Emitted by a Diesel Engine using Digital Image Analysis Algorithms and Scanning Mobility Particle Sizer," *SAE SP, Diesel emission control systems*, **1641**:31–38

This paper shows the morphological analysis results of particulate matter emissions from an indirect injection diesel engine working in two operating modes using Digital Image Analysis Algorithms (DIAA) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. Two typical engine operating modes were selected among the collection of steady stages, which reproduce the sequence of operating conditions that the vehicles equipped with this type of engines must follow during the transient cycle established in the European Emission Directive 70/220. The DIAA results were obtained from images of particulate matter collected in filters. The filters were charged in a dilution mini-tunnel, connected up-stream and down-stream of a cyclone-based particulate filtration system, which was coupled to the engine exhaust line. The images were obtained by a Scanning Electron Microscope (SEM) from samples taken directly from the charged filters. All the obtained results were compared with the SMPS measurements at the same operating condition and the same sampling points.

Jamriska, M.; Morawska, L., 2001, "A model for determination of motor vehicle emission factors from on-road measurements with a focus on submicrometer particles," *Science of the Total Environment*, **264**(3):241–255

The multiplicity of parameters that influence traffic-related emissions and that are often very difficult to measure or predict, makes the assessment of traffic emissions a very complicated process, strongly dependent on local conditions and usually associated with a high degree of error. The aim of this paper was to develop, calibrate and test a simple model for 'on-road' measurements of traffic emission factors as part of a major program focused on the assessment of traffic contribution to fine and ultrafine emissions to the whole air shed and to local areas in south-east Queensland, Australia. A mathematical model developed was based on the mass balance concept for on-road assessment of traffic-related emission rates. The model requires fewer experimental data points as input and is more applicable to the common on-road testing situation, when no more than two monitors of a specific pollutant can be used. The model was tested and calibrated using experimental data on particle number concentration collected at a road-monitoring site using the Scanning Mobility Particle Sizer, and was applied to assess the emission factors of submicrometer particles emitted by traffic. The average emission factor obtained using the box model and the experimental data from road measurements was  $1.75 \times 10^{14}$  particles  $\text{km}^{-1}$  vehicle $^{-1}$ , with a standard error of 67.6%. While the emission factor obtained was comparable with some results obtained from dynamometer studies and applied to the vehicle mix at the sampling site, they were significantly higher than the factors reported by other studies.

Kim, W.-S.; Kim, S. H.; Lee, D. W.; Lee, S.; Lim, C. S.; Ryu, J. H., 2001, "Size Analysis of Automobile Soot Particles Using Field-Flow Fractionation," *Environmental Science & Technology*, **35**(6):1005–1012

Soot particles emitted from various automobile engines are analyzed for size distributions using field-flow fractionation (FFF). Soot samples are prepared for FFF analysis using a three-step procedure, where a layer of soot particles is focused between the layers of n-hexane and water, followed by dispersing of particles in water containing 0.05% Triton X-100. The mean diameters determined by FFF show similar trends with those obtained from dynamic light scattering (DLS) and scanning electron microscopy (SEM). Data from FFF are also compared with those from an on-line Scanning Mobility Particle Sizer (SMPS) spectrometer. SMPS size distributions extend further to larger size than those of FFF distributions, which indicates the three-step sample preparation procedure effectively disaggregates the agglomerated particles. Although the amount of particulate matter (PM) emitted from a heavy-duty diesel engine is much higher than that from a light-duty diesel engine, the size distributions of soot particles show no significant difference between heavy- and light-duty diesel engines. The engine-operating mode (engine speed and load rate) does not seem to affect significantly the size distribution of soot particles. It was found that the PM from a turbo-charged diesel engine contains a higher percentage of particles smaller than 100 nm than an engine with a naturally aspirated (NA) air-inhalation system. As for gasoline engines, the PM collected after the catalytic converter has a narrower size distribution than those collected before and has a higher percentage of particles smaller than 100 nm.

Lee, Dai Woon; Kim, Sun Hui; Kim, Won-Suk; Lee, Seungho; Lim, Cheol Soo; Ryu, Jung Ho, 2001, "Size analysis of automobile soot particles using field-flow fractionation," *Environmental Science & Technology*, **35**(6):1005-1012

Soot particles emitted from various automobile engines are analyzed for size distributions using field-flow fractionation (FFF). Soot samples are prepared for FFF analysis using a three-step procedure, where a layer of soot particles is focused between the layers of n-hexane and water, followed by dispersing of particles in water containing 0.05% Triton X-100. The mean diameters determined by FFF show similar trends with those obtained from dynamic light scattering (DLS) and scanning electron microscopy (SEM). Data from FFF are also compared with those from an on-line Scanning Mobility Particle Sizer (SMPS) spectrometer. SMPS size distributions extend further to larger size than those of FFF distributions, which indicates the three-step sample preparation procedure effectively disaggregates the agglomerated particles. Although the amount of particulate matter (PM) emitted from a heavy-duty diesel engine is much higher than that from a light-duty diesel engine, the size distributions of soot particles show no significant difference between heavy- and light-duty diesel engines. The engine-operating mode (engine speed and load rate) does not seem to affect significantly the size distribution of soot particles. It was found that the PM from a turbo-charged diesel engine contains a higher percentage of particles smaller than 100 nm than an engine with a naturally aspirated (NA) air-inhalation system. As for gasoline engines, the PM collected after the catalytic converter has a narrower size distribution than those collected before and has a higher percentage of particles smaller than 100 nm.

Lehmann, U.; Mohr, M., 2001, "Influence of different configurations of a catalyst and a trap on particulate emission of a diesel passenger car," *International Journal of Vehicle Design*, **27**(1–4):228–241

New particulate emission measurements performed on a diesel passenger car to see the influence of different configurations of aftertreatment systems are reported. Five combinations of a particle trap and an oxidation catalyst are investigated. These configurations are discussed in view of particulate emission, measured by number and mass. All measurements were carried out at a chassis dynamometer of the EMPA. A diesel passenger car with an IDI engine was operated at four steady state conditions. Exhaust gas was diluted in a standard constant volume sampler (CVS) device (full flow dilution tunnel). Particulate size distributions were measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer and gravimetric measurements were performed according to regulations. Furthermore measurements without CVS tunnel were done by using external dilution units to see the influence of the sampling method. We used a thermo desorber to distinguish volatile and non-volatile aerosol fractions and we analyzed filter samples for determination of organic soluble fraction, water-soluble fraction and sulphur content. Huge differences depending on the configuration of the aftertreatment system and load were observed. In general it was found that a large reduction of particulate emission could be obtained by using a particle trap. A catalyst converter has minor effect on particulate emission. Nucleation of new particles was observed under certain conditions depending on configuration and sampling method. 8 Refs.

Wieser, U.; Gaegauf, Ch.; Macquat, Y., 2001, "Particle emissions from wood-fired boilers - Examination of emission rates in practical use," *Technical Report No. ENET-210221*, Oekozentrum, Langenbruck (Switzerland), in German

This report made for the Swiss Federal Office of Energy (SFOE), the Swiss Agency for the Environment, Forests and Landscape (SAEFL) as well as for the air pollution inspection offices of 5 Swiss Cantons,

presents the results of measurements made on 14 different types of wood-fired heating appliances and boilers with respect to their emissions of particulate matter, including units fired with wood logs, wood chippings and pellets. For comparison, the results for fossil-fuel-fired units are also quoted. The laboratory equipment and the measurement methods used are described which were used to determine the total amount of particles emitted and, in particular, the emissions of particles in the sub micrometer range. The results of the measurements made are presented and discussed in detail. A second phase of the project is described which allowed the measurement of particulate matter emissions in the field. The report is concluded with a discussion on the collection of data on particle emissions using SMPS (Scanning Mobility Particle Sizer) analysis methods, particle, size distribution and particle emission factors.

## 2000

Burtscher, H., 2000, "Comparison of particle emissions from different combustion systems," *Journal of Aerosol Science*, **31**(S1):S620–S621

Some examples of the properties of particles from combustion of petrol and diesel are given. The Scanning Mobility Particle Sizer (SMPS) spectrometer measures size distributions. To obtain information on particle mass, a low-pressure impactor (LPI) is used in series with DMA. The DMA classifies particles according to their mobility, the LPI according to mb. 2 Refs.

Chang, Y. C.; Tseng, H. H.; Hong, G. I.; Wang, K. T., 2000, "Measurement of smoke properties of incenses made in three countries," *Journal of Aerosol Science*, **31**(S1):S488–S489

Smoke properties of incenses made in different countries are investigated. The system used consists of a flow control/conditioning console, a smoke chamber (0.035 m<sup>3</sup>), an electrical resistance type of ignition control unit, and particle measuring instrumentation. The results showed that the smoke properties such as CMD and MMAD<sub>50</sub> were very similar for all three incense samples. The maximum total mass and the total number concentrations differ from each other. 3 Refs.

Harris, J. Stephen; Maricq, M. Matti, 2000, "Signature size distributions for diesel and gasoline engine exhaust particulate matter," *Journal of Aerosol Science*, **32**:749–764

The size distributions of diesel exhaust particulate matter measured from a large number of vehicles and test engines, using a variety of diesel fuels, collapse onto a single characteristic lognormal distribution, when normalized by total particle number and plotted against a scaled diameter. Distinctly different characteristic distributions are observed for direct injection and for port injection gasoline vehicles. These signature distributions are compared to numerically calculated self-preserving size distributions which incorporate fractal dimensions <3 to describe the dendritic nature of soot. However, the coagulation models alone are incapable of reproducing the observed characteristic distributions; compared to the diesel engine data they are conspicuously asymmetric and too steep on the large particle diameter side of the distribution. We discuss the possibility that soot oxidation also plays a role in determining the shape of the characteristic distributions, and the possibility that these signatures could be utilized to distinguish soot emissions from other aerosols.

Johnson, M. P.; Hilton, M.; Burrows, R.; Madden, P., 2000, "Characterization of particulates emitted from gas turbine exhausts," *Journal of Aerosol Science*, **31**(S1):S618–S619

The structure and morphology of soot particles obtained from civil gas turbines were analyzed using a variety of techniques. A Scanning Mobility Particle Sizer system (SMPS) spectrometer and transmission electron microscope (TEM) were used to characterize the particle emitted from aircraft gas turbine exhausts. Both techniques determined the total soot content, including particle size distribution and qualitative and quantitative measurements. 1 Refs.

Mohr, M.; Schmatloch, V.; Zaugg, H. P., 2000, "Investigation of fine particle emissions of small oil fired burners," *Proceedings of second European conference on small burner and heating technology, ECSBT 2*, **1**:179-186, Surface, porous and catalytic burners. Burner technology. Thermo acoustics. Emissions. Sensors and burner control, Stuttgart, Germany

An experimental study on particle emissions of various small oil fired burners installed in a test boiler has been carried out using a Scanning Mobility Particle Sizer (SMPS) spectrometer. By means of this instrument particles were characterized by number and size in the size range between 6 nm to 200 nm. The experiments were mostly carried out under steady state conditions. A standard fuel and a low sulfur fuel were tested in the project. Number size distributions were measured as a function of air fuel ratio, fuel quality and gap between nozzle and stabilization disk. The results revealed particle concentrations at standard conditions of about  $1 \times 10^6$  particles/cm<sup>3</sup> and did not show large variation between different technologies. The mode of the distributions was between 10 to 15 nm. Only a weak influence of the

burner set-up on the particle concentration was observed as long as the CO concentration was within a reasonable range. On the other hand a significant effect of the fuel was found on the particle emissions.

Ristovski, Z. D.; Morawska, L.; Hitchins, J.; Thomas, S.; Greenaway, C.; Gilbert, D., 2000, "Particle emissions from compressed natural gas engines," *Journal of Aerosol Science*, **31**(4):403–413

This paper presents the results of measurements conducted to determine particle and gas emissions from two large compressed natural gas (CNG) spark ignition (SI) engines. Particle size distributions in the range from 0.01-30  $\mu\text{m}$ , and gas composition were measured for five power settings of the engines: 35, 50, 65, 80 and 100% of full power. Particle emissions in the size range between 0.5 and 30  $\mu\text{m}$ , measured by the Aerodynamic Particle Sizer (APS) spectrometer, were very low at a level below two particles  $\text{cm}^{-3}$ . These concentrations were comparable with average ambient concentration, and were not considered in the succeeding analysis. Both engines produce significant amounts of particles in the size range between 0.015 and 0.7  $\mu\text{m}$ , measured by the Scanning Mobility Particle Sizer (SMPS) spectrometer. Maximum number of concentrations of about  $1 \times 10^7$  particles  $\text{cm}^{-3}$  were very similar for both engines. The CMDs were in the range between 0.020 and 0.060  $\mu\text{m}$ . The observed levels of particulate emission are in terms of number of the same order as emissions from heavy duty diesel engines. On the other hand, emissions of CO and NO<sub>2</sub> of 5.53 and 3.33 g  $\text{kW h}^{-1}$ , respectively, for one of the tested engines, were considerably lower than set by the standards. According to the specifications for the gas emissions, provided by the US EPA, this engine can be considered as a "low-emission" engine, although emissions of submicrometer particles are of the same order as heavy-duty diesel vehicles.

Ristovski, Z. D.; Tass, I.; Morawska, L.; Saxby, W., 2000, "Investigation into the emission of fine particles, formaldehyde, oxides of nitrogen and carbon monoxide from natural gas heaters," *Journal of Aerosol Science*, **31**(S1):S490–S491

The range of concentration levels of submicrometer particles, formaldehyde (HCHO), oxides of nitrogen (NO<sub>x</sub>), and carbon monoxide (CO) that arises from the standard operation of four typical flueless natural gas heaters are investigated. The results showed that both fine particle mass and number emission rates are low and the mass ones are close to 0.046 ng/J. This showed that the emission of particles during the combustion of natural gas in domestic heaters do not contribute to the indoor particle concentration. Natural gas heaters are a larger contributor of organic compounds such as formaldehyde and NO<sub>2</sub>. 2 Refs.

Zimmermann, R.; Maguhn, J.; Kettrup, A., 2000, "On-line analysis of combustion aerosols in the state of formation (900-300 degree C) at industrial incinerators," *Journal of Aerosol Science*, **31**(S1):S622–S623

On-line analytical methods were used to analyze combustion aerosols from industrial plants. Particle size distribution of the combustion aerosol were measured with a mobile aerosol measurement system, which includes an Aerodynamic Particle Sizer (APS) spectrometer and a Scanning Mobility Particle Sizer (SMPS) spectrometer. Results show that the measurements of the particle size distribution of the combustion aerosol probed at a 23 MW municipal waste incinerator (MWI) reveals a shift of the maximum of the particle size distribution to larger diameters upon the travel through the boiler section. The growth in particle diameter is due to reactive attachment of gas phase species and agglomeration process.

## 1999

Harrison, Roy M.; Brear, Fred; Shi, Ji Ping, 1999, "Particle size distribution from a modern heavy duty diesel engine," *Science of the Total Environment*, **235**(1–3):305–317

The particle size distribution emitted from a modern heavy duty diesel engine was investigated. It was found that the operation of the engine test bed system, primary and secondary dilution system and instruments were stable and that particle size distributions could be measured repeatedly under specific conditions. The particle size distribution was determined at different engine speed and load. The mode in the number distribution was smaller than 25 nm at engine speed 1600 rev./min and between 40-60 nm at 2600 rev./min. Good agreement of size distributions measured by SMPS and ELPI instruments was found in the region of overlap, and with measurements made with an electron microscope and image analyser. Total particle volume concentrations calculated from SMPS and ELPI data agreed well with those estimated from filter mass data. The measured particle size distribution and total number concentration changes with the dilution conditions and humidity of dilution air. The mechanism is being investigated and preliminary results suggest that sulphuric acid is involved in homogeneous nucleation of new particles during the dilution process.

Kayes, D.; Hochgreb, S., 1999, "Mechanisms of particulate matter formation in spark-ignition engines. 3. Model of PM," *Environmental Science and Technology*, **33**(22):3978–3992

Recent health concerns over airborne particulate matter (PM) have prompted examination of the mechanisms by which PM is formed in spark ignition (SI) internal combustion engines. A study was undertaken in order to understand the effects of dilution on measured PM, to examine and model the

effect of steady state engine operating conditions on engine-out PM, and to characterize the effect of transient engine conditions on particle growth and dynamics. Particle dynamics in diluted SI and compression ignition (CI) engine exhaust are examined and discussed in the context of SI exhaust dilution. Temperature measurements in the exhaust pipe and dilution tunnel reveal the degree of mixing between exhaust and dilution air, the effect of flow rate on heat transfer from undiluted and diluted exhaust to the environment, and the minimum permissible dilution ratio for a maximum sample temperature of 52 C. Measurements of PM concentrations as a function of dilution ratio, using a Scanning Mobility Particle Sizer (SMPS) spectrometer, show the competing effects of temperature and particle/vapor concentrations on particle growth dynamics, which result in a range of dilution ratios—from 13 to 18—where the effect of dilution ratio, independent of flow rate, is kept to a minimum and is therefore optimal in order to achieve repeatable PM concentration measurements. Particle dynamics in transit through the dilution tunnel are measured and compared to previous research. PM emissions are strongly affected by steady state engine parameters that affect global and local air/fuel ratios, the concentration of liquid fuel in the cylinder, and the availability of soot precursors. PM emissions vary by up to six orders of magnitude between the fuels tested, when at the same fuel/air equivalence ratio. Minimum PM concentrations are emitted at a global fuel/air ratio within 10% of stoichiometric, with the exact value depending on the particular fuel, and concentrations can increase by more than three orders of magnitude when the fuel/air ratio is either increased or decreased 30% from stoichiometric. Burning liquid fuel is a significant source of PM, as evidenced by the fact that open valve fuel injection increases PM emissions by up to three orders of magnitude relative to closed valve injection. Coolant and oil temperatures, spark timing, and Exhaust Gas Recirculation (EGR) affect PM through their effect on intake port and cylinder temperatures, as well as through the effect on the availability of liquid fuel in the cylinder. Particles derived from oil consumption were found to be between zero and 40% of the total PM concentration for the oils used in the present experiments. Differences in PM emissions with and without the catalytic converter are not statistically significant. Particulate number and mass concentrations plus particle sizes are addressed in the present paper, as is the correlation between PM and emissions of gaseous pollutants—hydrocarbons (HCs), oxides of nitrogen (NO<sub>x</sub>), oxides of carbon (CO and CO sub 2 )—as well as oxygen and characteristic temperatures and pressures during the engine cycle. A model of PM formation via homogeneous- and heterogeneous-phase reactions, growth via condensation and adsorption/absorption of vapors, and diminution via oxidation explains the observed behavior of PM emissions with respect to each of the engine, fuel, and dilution parameters above. PM emissions during transient engine operation are generally a first-order time response with characteristic times similar to those involved in the fuel evaporation process, suggesting that PM emissions respond to instantaneous engine conditions and may be modeled using a quasi-steady state application of the model.

Kerminen, V.-M.; Makela, T.; Hillamo, R.; Rantanen, L., 1999, "Relation between particle number and mass size distribution in the diesel car exhaust," *Journal of Aerosol Science*, **30**(S1):S777-S778

Presented are results demonstrating that detailed investigation of diesel exhaust particles requires measuring both the particle number of mass size distribution. It is shown that valuable information on diesel particle mass size distribution up to sizes of about 0.2–0.3 μm can be obtained by converting the corresponding number size distribution appropriately.

Maricq, M. M.; Podsiadlik, D. H.; Chase, R. E., 1999, "Examination of the size-resolved and transient nature of motor vehicle particle emissions," *Environmental Science & Technology*, **33**(10):1618-1626

This paper reports mass measurements, size distributions, and the transient response of tailpipe particulate emissions from 21 recent model gasoline vehicles. Transient measurements are made for the FTP drive cycle (and limited ECE tests) using a Scanning Mobility Particle Sizer and an electrical low-pressure impactor. The particles emitted in vehicle exhaust have diameters in the 10-300 nm diameter range, with a mean diameter of about 60 nm. Particle emissions during the drive cycles occur as narrow peaks that correlate with vehicle acceleration. Cold start emissions generally outweigh those from a hot start by more than a factor of 3. Particulate mass deduced from the transient distributions agrees semiquantitatively with gravimetric measurements. Tailpipe particulate emissions from the recent model gasoline vehicles tested are very low, with mass emission rates ranging downward from 7 mg/mi for a light-duty truck during the cold start phase of the FTP drive cycle to less than or equal to 0.1 mg/mi during phase 2 for nearly half of the test vehicles. Three high-mileage (>100 K mi) test vehicles exhibited similarly low particulate emission rates. The FTP-weighted 3-bag average is under 2 mg/mi for all the conventional gasoline vehicles tested.

Maricq, M. M.; Podsiadlik, D. H.; Chase, R. E., 1999, "Gasoline Vehicle Particle Size Distributions: Comparison of Steady State, FTP, and US06 Measurements," *Environmental Science & Technology*, **33**(12):2007-2015

Factors influencing the number and size of tailpipe particles from port injection, spark ignition vehicles are examined by comparing emissions recorded during steady-state operation and those obtained from FTP and US06 drive cycles. Size distributions are measured using the Scanning Mobility Particle Sizer



(SMPS) spectrometer and the electrical low-pressure impactor (ELPI). Steady-state particulate emissions are examined as a function of vehicle speed and air to fuel ratio. The emission rates increase moderately with increasing speed but climb steeply with decreasing A/F. This is consistent with the observations from transient drive cycle measurements where particulate emissions occur predominantly during periods of heavy acceleration. As expected from the more aggressive speed and acceleration of the US06 cycle, the per mile particulate emission rates are higher than for phases 2 and 3 of the FTP. For the eight vehicles tested, the US06 mass emissions range from 1.2 to 9.6 mg/mi. Use of a US06-compliant calibration leads to a factor of 2 reduction of particulate emissions, in both number and mass, over the drive cycle.

## 1998

Abdul-Khalek, I. S.; Kittelson, D. B.; Graskow, B. R.; Wei, Q.; Brear, F., 1998, "Diesel exhaust particle size: measurement issues and trends," *Proceedings of the 1998 SAE International Congress & Exposition, In-Cylinder Diesel Particulate and NOx Control SAE Special Publications*, SAE, Warrendale, PA, USA, **1326**:133-145

Measurements of exhaust particle size distribution and number concentration produced by a modern, low emission diesel engine were made. The influences of operating conditions, an exhaust catalyst, and dilution conditions were determined. The engine was tested under the operating conditions of the ISO 8 and 11 Mode tests. Measurements were performed on the exhaust of a medium-duty, turbocharged, aftercooled, direct injection Diesel engine, using a mini-dilution system, Scanning Mobility Particle Sizer (SMPS) spectrometer, and condensation particle counter (CPC). In the case of the engine catalyst studies, an electrical aerosol analyzer (EAA) and a condensation nucleus counter (CNC) were used. Results show that number size distribution are bi-modal and log-normal in form. In most cases, more than half of the particle number are present in the nuclei mode. 27 Refs.

Ahmed, S. H.; Hayat, S. O., 1998, "Effect of ashless additive technologies on the number and size distribution of fine particles emitted from diesel engines," *Diesel engines - particulate control. Proceedings*, pp. 1-13, 105 page(s), Book analytic

The Scanning Mobility Particle Sizer was used in a comparative study of two diesel fuels, (a) low sulphur EN 590 diesel and (b) ultra low sulphur 'city diesel' and several ashless additive technologies to determine their effect on fine particle emissions from a heavy duty single cylinder and a light duty multi-cylinder engine. The two fuels provided significantly different particle number emissions but similar size distributions. An additive formulation containing a non metallic ashless combustion enhancer provided significant reductions of between 45% and 85% in particle number emissions, in both the test fuels with no shift in the particle size distribution. The multifunctional additive package containing a dispersant and a lubricity agent did not significantly alter the number of particles emitted or the particle size distribution.

Bach, C.; Heeb, N.; Mattrel, P.; Mohr, M., 1998, "Effect-based assessment of exhaust emissions from automobiles," Report 58 page(s), Swiss Federal Laboratory for Materials Testing and Research (EMPA), Report No. EMPA—160928/2

As a result of current exhaust emission specifications for gasoline and diesel vehicles, alternative or reformulated energy sources cannot be simply compared with each other on the basis of the regulated test and measuring methods. For this reason a measuring procedure for an 'effect-based assessment of exhaust emissions' has been developed together with a rating matrix. Subsequently, this was used as an example on one vehicle type with a gasoline, natural gas and diesel engine. The aim of the project was to compare the exhaust emissions with respect to the ozone formation potential, greenhouse gases, acid formation, carcinogenic gases, and particles as well as energy consumption. The integral measurements were performed by using the conventional standard bag method, gas and liquid chromatography, and a Scanning Mobility Particle Sizer (SMPS) spectrometer. In order to be able to investigate the exhaust emissions during critical conditions of the cycle such as cold start or acceleration procedures, measurements were carried out continuously as well. For these tests, the conventional analyser for the regulated air pollutants were used as well as an on-line mass spectrometer for the measurement of methane, ethane, benzene, xylene and toluene. The report summarizes the results.

Graskow, B. R.; Kittelson, D. B.; Abdul-Khalek, I. S.; Ahmadi, M. R.; Morris, J. E., 1998, "Characterization of exhaust particulate emissions from a spark ignition engine," *Proceedings of the 1998 SAE International Congress & Exposition, In-Cylinder Diesel Particulate and NOx Control SAE Special Publications*, SAE, Warrendale, PA, USA, **1326**:155-165

Several experiments were undertaken to measure and characterize exhaust particulate emissions from a spark ignition engine. Experiments were performed using three different fuels with measurements made both upstream and downstream of the catalytic converter. Emissions were characterized in terms of number-weighted break, specific particle emissions and particulate size distribution. 26 Refs.

Kittelson, D. B., 1998, "Engines and Nanoparticles: a Review," *Journal of Aerosol Science*, **29**(5): 575–588

Most of the particle number emitted by engines is in the nanoparticle range, D (50 nm, while most of the mass is in the accumulation mode, 50 nm(D (1000 nm, range. Nanoparticles are typically hydrocarbons or sulfate and form by nucleation during dilution and cooling of the exhaust, while accumulation mode particles are mainly carbonaceous soot agglomerates formed directly by combustion. Emission standards on diesel engines have led to dramatic reductions in particle mass emitted. However, a new HEI study shows that some low-emission diesel engines emit much higher concentrations of nanoparticles than older designs and other low-emission designs. Many recent studies suggest that at similar mass concentrations; nanometer size particles are more dangerous than micron size particles. This has raised questions about whether nanoparticle (number based) emission standards should be imposed. Unlike mass, number is not conserved. It may change dramatically by nucleation and coagulation during dilution and sampling, making it very difficult to design a standard. Furthermore, if nanoparticles are a problem, spark ignition engines may also have to be controlled.

Ristovski, Z. D.; Morawska, L.; Bofinger, N. D.; Hitchins, J., 1998, "Submicrometer and Supermicrometer Particulate Emission from Spark Ignition Vehicles," *Environmental Science and Technology*, **32**(24):3845-3852

Particulate emissions from 11 gasoline-powered and 2 liquefied petroleum (LPG)-powered passenger vehicles were characterized during the Accelerated Simulation Mode driving cycles on a chassis dynamometer. The test fleet consisted of 10 catalyst-equipped vehicles operated with unleaded gasoline (5 Ford Falcons and 5 Holden Commodores), 2 LPG-powered vehicles (both Ford Falcons), and 1 older type noncatalyst vehicle operated with leaded gasoline. Particulate characterization included determination of total particulate number concentration and size distribution using the Scanning Mobility Particle Sizer (SMPS) spectrometer and the Aerodynamic Particle Sizer (APS) spectrometer. The average particle number concentrations in the SMPS spectrometer range for all modes was lower for Ford Falcons and somewhat higher for Commodores, with values of  $1.5 \times 10^4$  and  $4.1 \times 10^4 \text{ cm}^{-3}$ , respectively. This difference is significant and was observed for all modes. The number concentration levels were higher for the LPG-fueled cars ( $8.4 \times 10^4 \text{ cm}^{-3}$ ) and for the leaded gasoline-powered vehicle ( $7.9 \times 10^4 \text{ cm}^{-3}$ ). There was not a significant variation in particle count median diameter in the SMPS and the APS ranges, either for different operating conditions of the vehicles investigated or between different vehicle groups. The observed size distributions were bimodal with average values of CMD ranging from 39.1 to 60.2 nm in the SMPS range and from 0.9 to 1.4  $\mu\text{m}$  in the APS range. The results obtained from this study can be used as a first order estimation toward emission inventories for vehicle groups included in the investigations.

## 1997

Matter, U.; Siegmann, K., 1997, "The influence of particle filter and fuel additives on turbo diesel engine exhaust," *Journal of Aerosol Science*, **28**(S1):S51–S52

Particulate emission from modern turbo diesel engines on test benches is investigated. Photoemission sensors, an aethalometer and a Scanning Mobility Particle Sizer (SMPS) spectrometer are employed to characterize the particles, together with filter analyses of sampled soot. Commercially available fuel additives and particle filters are investigated in their ability of reducing particulate emission. Promising techniques of sensing particulate diesel exhaust are evaluated.

## 1996

Christensen, K. A.; Livbjerg, H., 1996, "Field study of submicron particles from the combustion of straw," *Aerosol Science & Technology*, **25**(2):185–199

The evolution of small aerosol particles accompanying the combustion of straw for energy production is investigated. A sampling equipment specially designed for field measurements is described and characterized. The aerosol is studied by low-pressure cascade impactor and Scanning Mobility Particle Sizer, the particle morphology by transmission electron microscopy, and the chemical composition by energy dispersive x-ray analysis. The combustion gas contains 3-500  $\text{mg}/\text{Nm}^3$  of submicron particles with a mean diameter of approximately 0.3  $\mu\text{m}$ . The particles consist of almost pure potassium chloride and sulphate. The formation mechanism is analyzed by a theoretical simulation of the chemical reactions and the aerosol change during cooling of the flue gas. It is concluded that some sulphation of KCl occurs in the gas phase although the sulphate concentration is much lower than predicted by an equilibrium assumption. The theoretical simulation proves that the fine mode particles can be formed by homogeneous nucleation of either KCl or  $\text{K}_2 \text{SO}_4$  as the first step and further growth occurs by coagulation and diffusive condensation of both KCl and  $\text{K}_4 \text{SO}_4$  on existing particles. 25 refs., 13 figs., 2 tabs.

Owens, T. M.; Biswas, P., 1996, "Vapor phase sorbent precursors for toxic metal emissions control from combustors," *Industrial and Engineering Chemistry Research*, **35**(3):792-798

A bench scale system incorporating a high temperature flow reactor has been utilized to study the reaction of metallic species and sorbent compounds. The reaction and interaction between a lead precursor and a vapor phase silica precursor is studied. The effect of chlorine and the effect of sorbent to metal feed ratio on lead speciation and size distribution is determined. The size distributions of the reacted compounds are measured by a Scanning Mobility Particle Sizer (SMPS) spectrometer and an optical particle counter (OPC). The product powders are also collected and their compositions established by X-ray diffraction (XRD) or, if an amorphous powder is collected, by infrared and raman spectroscopy. Experiments have been performed at 500, 750, 1,000, and 1,250 C and for reactor residence times of 0.5-1.0 s. For example, at 750 C, for a lead feed in conjunction with a vapor phase silica precursor compound, the mean particle size exiting the reactor is significantly larger than for a lead only feed or a silicon only feed. The composition of the resulting aerosol is determined, via infrared and Raman spectroscopy, to be lead silicate,  $PbSiO_3$ . Higher temperature results are similar. At lower temperatures, silica particles are not formed in the reactor. Higher silica precursor feed rates result in particles large enough for capture by conventional pollution control equipment.

Wu, C.-Y.; Arar, E.; Biswas, P., 1996, "Mercury capture by aerosol transformation in combustion environments," Book Monograph, Published at Pittsburgh, PA, USA, the 1996 Annual Meeting of Air and Waste Management Association 89, Nashville, TN, USA

Mercury released from coal combustors and waste incinerators poses a potential environmental concern. Unlike most other heavy metals that are emitted in particulate form, mercury has been reported to be released mainly in the elemental vapor phase that is not effectively captured in typical particulate control devices. In this work, we examine the oxidation of mercury in air at high temperature environments. The objective is to effectively transform mercury into mercury oxide which has a higher possibility to form particles because of its lower vapor pressure. The study was conducted in a new reactor with real-time measurement of particle size distribution (by Scanning Mobility Particle Sizer) and composition analysis by x-ray diffraction for particles on filters and by Cold Vapor Atomic Absorption Spectroscopy for gaseous species collected in impingers). Temperature was varied from 320 degree C to 1200 degree C and residence time was varied from 1.5 s to 45 s. Although oxidation was observed when the residence time was increased, the experimental results showed that the oxidation rate was too slow to be effective for mercury capture in practical combustion systems. Studies are needed for alternative approaches to capture mercury vapor such as the use of novel sorbent materials.

## 1995

Biswas, P.; Owens, T. M.; Wu, C. Y., 1995, "Control of toxic metal emissions from combustors using vapor phase sorbent materials," *Journal of Aerosol Science*, **26**:S217-S218

In this paper, results of experiments on the evolution of the lead aerosol size distribution in different environments are reported to develop better insights into particle formation and growth mechanisms. The use of vapor phase sorbents for metals capture in high temperature gas streams are also reported. The experimental system used in this work consists of a flow reactor with real time measurement of particle size distributions (Scanning Mobility Particle Sizer) at the exit of the reactor. Lead is introduced by atomizing a solution of lead acetate. Depending on the experiment, chlorine is also introduced into the system. The sorbent is inlet to the reactor in the vapor form, carried in an argon stream. The particles are collected at the exit of the reactor for chemical characterization by x-ray diffraction. Experiments were conducted for a range of temperatures (500 to 1200 degree C), flow rates and inlet concentrations.

## 1994

Huglin, Ch.; Gaegauf, Ch.; Burtscher, H., 1994, "Size distribution and photoelectric activity of particles in wood combustion exhaust," *Journal of Aerosol Science*, **25**(S1):113-114

In order to study particulate emissions from wood combustion, size distributions of two different systems has been measured. A wood stove in different phases of the combustion and a wood chip burner have been used for these measurements. Size distribution of emissions were found to differ significantly in different phases of wood combustion.

## 1992

Hautanen, Jukka; Kilpelainen, Markku; Kauppinen, Esko I.; Jokiniemi, Jorma K.; Lehtinen, Kari E. J., 1992, "Electrical Agglomeration of Fly Ash Particles," *Aerosol Science*, **23**(S1):S783-S786

Electrical agglomeration of charged aerosol particles in alternating electrical field has been studied. Experimental system and preliminary results are presented.

Joutsensaari, Jorma; Kauppinen, Esko I.; Ahonen, Petri; Lind, Terttaliisa M.; Ylatao, Sampo I.; Jokiniemi, Jorma K.; Hautanen, Jukka; Kilpelainen, Markku, 1992, "Aerosol Formation in Real Scale Pulverized Coal Combustion," *Aerosol Science*, **23**(S1):S241–S244

Aerosol formation in pulverized coal combustion have been studied experimentally at the real scale power plant. Combustion aerosol mass and number size distributions have been determined, when burning bituminous coal from Poland. Mass size distributions have been measured by low pressure impactor and number distributions by differential electrical mobility (DMA) method.

Makela, Jyrki M.; Aalto, Pasi; Gorbunov, Boris Z.; Korhonen, Pekka, 1992, "Size Distributions from Aerosol Spark Generator," *Aerosol Science*, **23**(S1):S233–S236

An aerosol spark generator for producing ultrafine particles from metals is described. The aerosol particle size distributions from the spark generator have been measured by both Diffusion Battery-CNC and DMA-Electrometer systems. The average diameter of the generated particles can be varied to be between 2 and 15 nm. The measurements on the particle size distributions are described.

Patschull, J.; Roth, P., 1992, "Charge and Size Distribution of Particles Emitted From a Di-Diesel Engine," *Aerosol Science*, **23**(S1):S229–S232

In diesel engines solid particle phases are formed as a result of incomplete combustion in fuel rich zones of the diffusion-controlled spray combustion. The whole process is composed of particle inception and surface growth associated with agglomeration of the monomeres to irregularly shaped particle aggregates. Furthermore, diesel particles are electrically charged by electrons and ions produced during the involved chemical reactions. As the flame ionization causes a bipolar diffusion charging very early in the engine cycle, the contact electrifications by surface interactions and the thermoelectric charging might lead to an unbalanced charge distribution. The present paper reports on some measurements of particle charge and size distribution performed on the diluted diesel exhaust obtained under different engine operation conditions. The size distributions of the particles as well as the fraction and the balance of naturally charged particles were determined from electrical mobility measurements. Furthermore, the number of elementary charges per particle was investigated by classifying and counting particles of a discrete mobility class. The evaluation of these measurements resulted in charge distributions of the naturally charged particles.

## 1983

Neville, M.; McCarthy, J.F.; Sarofim, A.F., 1983, "Size Fractionation of Submicrometer Coal Combustion Aerosol for Chemical Analysis," *Atmospheric Environment*, **17**(12):2599–2604

The application of an electrostatic classifier for discrete size classification of sub-um fly-ash particles was examined for the purpose of determining the size dependence of elemental concentration. From transmission electron microscopy studies, the size fractionated output for these particles from the electrostatic classifier was found to have larger effective mean diameter and a broader distribution than would be expected from the theory for singly charged particles. Four discrete size cuts of the sub-um particles, produced from the combustion of Montana lignite in a laboratory scale combustion system, with mean diameters of 10, 20, 30 and 40 nm were analyzed by INAA. The concentration of the volatile and trace species which condense well after the birth of the sub-um particles (Neville and Sarofim, 1983) as the combustion gases are being cooled were found to be systematically enriched with decreasing particle size. The observed  $d^{-1}$  relationship is consistent with a hypotheses that the volatile and trace elements condense on the surface of the sub-um particles with the rate of deposition being controlled by mass transfer.

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## Filtration

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### 2013

Buha, J., Fissan, H., and Wang, J., 2013, "Filtration behavior of silver nanoparticle agglomerates and effects of the agglomerate model in data analysis," *Journal of Nanoparticle Research*, **15**(7): 1709

In many data evaluation procedures for particle measuring devices and in filtration models, spherical particles are assumed. However, significant fractions of aerosol particles are agglomerates of small primary spheres. The morphology of particles in filtration processes may not be known a priori and if the filtration data are processed with wrong assumption, errors can be induced. In this work, we have quantified such errors for the case of open-structured agglomerates. Filtration efficiency tests with polydisperse silver nanoparticle agglomerates and their sintered spheres were performed. After the sintering process, particles with a compact structure with the shape close to a sphere are obtained, which are referred to as sintered spheres in the present study. The testing method involved generation of particulate forms, passing the particles through the testing section, and measurement of the particle number concentrations and size distributions before and after the filter. Measurements of the aerosols upstream and downstream of the filter were conducted using Scanning Mobility Particle Sizer (SMPS) spectrometers (TSI Inc.), which covered the range from 10 to 480 nm. Particles were additionally characterized from the electron microscopic images and the average primary particle size was determined to be 16.8 nm. The number-size distribution curves were obtained and used for penetration calculation. The penetration was dependent on the particle size and morphology. Silver- sintered spheres were captured with a lower efficiency than agglomerates with the same mobility diameter because of the stronger interception effect for agglomerates. Data analysis of the number-size distribution for agglomerates was processed based on sphere assumption and using the model for open-structured agglomerates developed by Lall and Friedlander. The efficiencies based on total concentrations of number, surface and volume were affected when the agglomerate model was used. The effect was weakest for the total number efficiency and strongest for the total volume efficiency. Filtration efficiency curves for agglomerates were additionally plotted as a function of the volume equivalent diameter or number of primary particles per agglomerate, because the latter parameters provide more information for the agglomerate structure.

Chen, S.-C., Wang, J., Fissan, H., and Pui, D. Y. H., 2013, "Exposure assessment of nanosized engineered agglomerates and aggregates using Nuclepore filter," *Journal of Nanoparticle Research*, **15**(10): 1955

Nuclepore filter collection with subsequent electron microscopy analysis for nanosized agglomerates (20-500 nm in mobility diameter) was carried out to examine the feasibility of the method to assess the personal engineered nanoparticle exposure. The number distribution of the nanoparticles collected on the filter surface was obtained by visual counting and converted to the distribution in the air using validated capillary tube models. The model was validated by studying the overall penetrations of nanoparticles (Ag and soot) with different agglomeration degrees through 1  $\mu\text{m}$  pore diameter Nuclepore filters at different face velocities (2–15 cm/s). In the model, the effects of the maximum length of agglomerates on interception deposition and the dynamic shape factor on impaction deposition were taken into account. Results showed that the data of the overall penetration were in very good agreement with the properly applied models. A good agreement of filter surface collection between the validated model and the SEM analysis of this study was obtained, indicating a correct particle number distribution in the air can be converted from the Nuclepore filter surface collection and this method can be applied for quantitative engineered nanoparticle exposure assessment.

Hasolli, N., Park, Y. O., and Rhee, Y. W., 2013, "Experimental Study on Filtration Performance of Flat Sheet Multiple-Layer Depth Filter Media for Intake Air Filtration," *Aerosol Science and Technology*, **47**(March): 1334–1341

Depth filter media are usually composed of multiple layers to attain optimal values of main filtration parameters such as pressure drop and particle collection efficiency (PCE). Understanding the performance of the single layers that make up the filter media can contribute to attaining these optimum values. For the purpose of this study, we have developed two samples of depth filter media, 2LM and 3LM, composed of two and three layers, respectively. Samples of the media and single layers have been prepared in flat sheet form. Filtration performance of these samples has been evaluated using a lab-scale flat sheet filtermedia test unit with KCl as the test aerosol. Results from these tests have been compared with those from an automated filter tester (AFT) with NaCl as the test aerosol. These media have been

characterized based on filter media properties and data from the filtration performance tests and the effect of layers on filtration performance has been observed. Pressure drop data from the tests with a standard test unit and our laboratory test unit are similar, and the difference in data is attributed to high inlet concentration of KCl particles. The overall filtration performance is strongly dependent on the final layer regarding pressure drop and particle of two- and three-layer filter media. Other constituent layers can be seen as contributors to the dust-holding capacity of the filter media and can reduce the dust load for the final layer.

Nelson, A. J., Page, M. a., Ginsberg, M. D., and Rood, M. J., 2013, "Bench-Scale Aerosol Filtration Test System and Evaluation of an Acoustic Bioaerosol Removal Device for Indoor Air Streams," *Aerosol Science and Technology*, **47**: 1285–1292

It is important to have well-defined, reproducible methods to evaluate and compare newly developed air filtration equipment. To facilitate accurate assessment of air purification devices at the bench scale, an experimental system was designed, built, and documented to evaluate particulate removal efficiency (PRE) of air filtration devices based on principles used in ASHRAE standards. The system was then carefully characterized and used to evaluate PRE and total energy consumption of a novel acoustically enhanced impaction (AEI) air purification device. The AEI device demonstrated 99.998% PRE of 0.5–1.5  $\mu\text{m}$  diameter KCl particles while causing a 120 Pa pressure drop and requiring a total of 3.0 W/l of air treated at indoor ambient conditions. A single element of the AEI device operated in a biological safety level 2 facility was then used to evaluate PRE of bioaerosol consisting of *Bacillus cereus* (BC) spores. PRE of BC was  $99.86 \pm 0.05\%$  at indoor ambient temperature and pressure. This research describes the use of the Bench-scale Air Purification Testing and Evaluation Chamber (BAP-TEC) to experimentally evaluate and compare PRE and total energy requirements of novel air purification devices at the bench scale (280–1400 alpm). Further, an AEI device containing a fibrous filter media and high intensity sound field in the same control volume is evaluated using the BAP-TEC. Temporally resolved PRE of a bioaerosol by the AEI is also presented.

Zhuang, Z., Bergman, M. S., Eimer, B. C., and Shaffer, R. E., 2013, "Laboratory face seal leakage evaluation of n95 filtering facepiece respirators against nanoparticles and "all size" particles.," *Journal of occupational and environmental hygiene*, **10**(9): 496–504

National Institute for Occupational Safety and Health (NIOSH)-certified N95 filtering facepiece respirators (FFRs) are used for respiratory protection in some workplaces handling engineered nanomaterials. Previous NIOSH research has focused on filtration performance against nanoparticles. This article is the first NIOSH study using human test subjects to compare N95 FFR face seal leakage (FSL) performance against nanoparticles and "all size" particles. In this study, estimates of FSL were obtained from fit factor (FF) measurements from nine test subjects who participated in previous fit-test studies. These data were analyzed to compare values obtained by: 1) using the PortaCount Plus Respirator Fit Tester (8020A, TSI Inc., MN, USA) alone (measurable particle size range 20 nm to > 1,000 nm, hereby referred to as the "all size particles test"), and 2) using the PortaCount Plus Respirator Fit Tester with N95-Companion™ accessory (8095, TSI Inc., Minn.) accessory (negatively charged particles, size range ~40 to 60 nm, hereby referred to as the "nanoparticles test"). Log-transformed FF values were compared for the "all size particles test" and "nanoparticles test" using one-way analysis of variance tests (significant at  $P < 0.05$ ). For individual FFR models, geometric mean (GM) FF using the "nanoparticles test" was the same or higher than the GM FFs using "all size particles test." For all three FFR models combined, GM FF using the "nanoparticles test" was significantly higher than the GM FF using "all size particles test" ( $P < 0.05$ ). These data suggest that FSL for negatively charged ~40-60 nm nanoparticles is not greater than the FSL for the larger distribution of charged and uncharged 20 to > 1,000 nm particles.

## 2012

Li, L. I. N., Zuo, Z., Japuntich, D. A., and Pui, D. Y. H., 2012, "Evaluation of Filter Media for Particle Number, Surface Area and Mass Penetrations," *The Annals of Occupational Hygiene*, **56**(5): 581–594

The National Institute for Occupational Safety and Health (NIOSH) developed a standard for respirator certification under 42 CFR Part 84, using a TSI 8130 automated filter tester with photometers. A recent study showed that photometric detection methods may not be sensitive for measuring engineered nanoparticles. Present NIOSH standards for penetration measurement are mass-based; however, the threshold limit value/missible exposure limit for an engineered nanoparticle worker exposure is not yet clear. There is lack of standardized filter test development for engineered nanoparticles, and development of a simple nanoparticle filter test is indicated. To better understand the filter performance against engineered nanoparticles and correlations among different tests, initial penetration levels of one fiberglass and two electret filter media were measured using a series of polydisperse and monodisperse aerosol test methods at two different laboratories (University of Minnesota Particle Technology Laboratory and 3M Company). Monodisperse aerosol penetrations were measured by a TSI 8160 using NaCl particles from 20 to 300 nm. Particle penetration curves and overall penetrations were measured by

Scanning Mobility Particle Sizer (SMPS) spectrometer, condensation particle counter (CPC), nanoparticle surface area monitor (NSAM), and TSI 8130 at two face velocities and three layer thicknesses. Results showed that reproducible, comparable filtration data were achieved between two laboratories, with proper control of test conditions and calibration procedures. For particle penetration curves, the experimental results of monodisperse testing agreed well with polydisperse SMPS measurements. The most penetrating particle sizes (MPPSs) of electret and fiberglass filter media were ~50 and 160 nm, respectively. For overall penetrations, the CPC and NSAM results of polydisperse aerosols were close to the penetration at the corresponding median particle sizes. For each filter type, power-law correlations between the penetrations measured by different instruments show that the NIOSH TSI 8130 test may be used to predict penetrations at the MPPS as well as the CPC and NSAM results with polydisperse aerosols. It is recommended to use dry air (<20% RH) as makeup air in the test system to prevent sodium chloride particle deliquescing and minimizing the challenge particle dielectric constant and to use an adequate neutralizer to fully neutralize the polydisperse challenge aerosol. For a simple nanoparticle penetration test, it is recommended to use a polydisperse aerosol challenge with a geometric mean of ~50 nm with the CPC or the NSAM as detectors.

Shi, B., and Gustén, J., 2012, "Influence of Filter Fiber Material on Removal of Ultrafine and Submicron Particles Using Carbon Fiber Ionizer- Assisted Intermediate Air Filters," *ASHRAE Transactions*, 602–612

Ultrafine and submicron particles are possibly highly related with respiratory and cardiopulmonary diseases and syndromes. Many studies show that an ionizer operated upstream of a ventilation air filter could enhance the particle collection efficiency of the filter, without affecting the pressure drop. However, most of these studies are based on one single fiber material and does not develop any comparison among different ventilation filters. This study investigates the influence of the fiber material and filter class on the enhanced efficiency. The influence of ion concentration is also considered. The experiments were conducted in a full-scale filter test rig and a chamber with 100% air re-circulation. Seven ventilation filters of class F5-F9 (MERV11-15), made of three types of fiber materials were tested in the filter test rig. A Scanning Mobility Particle Sizer (SMPS) spectrometer was utilized to count ultrafine and submicron particles in the upstream and downstream air flow. In the chamber test, a charged synthetic filter of class G4 (MERV 8) was mounted in the air re-circulation system. A P-TRAK Ultrafine Particle Counter, a CI-500 optical particle counter and a Dust-TRAK Aerosol Monitor were used to measure ultrafine and submicron particles. The results show, under the given experimental conditions, that ionization could substantially improve the filtration efficiency. For example, assisted by the ionizer, the coarse G4 (MERV 8) filter showed a filtration efficiency that increased by about 30 to 40 %-units for submicron particles. For the same ion concentration, the enhanced efficiency of charged synthetic filters is higher than that of glass fiber filters, especially in the ultrafine particle size fraction. Similarly, with the same ion concentration, the charged synthetic filters of higher class showed higher enhanced efficiencies than the charged synthetic filters of lower class. Measurements showed a negligible generation of ozone, both in the filter test rig and in the test chamber experiments

## 2011

Cho, H.-W., Yoon, C.-S., Lee, J.-H., Lee, S.-J., Viner, A., and Johnson, E. W., 2011, "Comparison of pressure drop and filtration efficiency of particulate respirators using welding fumes and sodium chloride.," *The Annals of occupational hygiene*, 55(6): 666–80

Respirators are used to help reduce exposure to a variety of contaminants in workplaces. Test aerosols used for certification of particulate respirators (PRs) include sodium chloride (NaCl), dioctyl phthalate, and paraffin oil. These aerosols are generally assumed to be worst case surrogates for aerosols found in the workplace. No data have been published to date on the performance of PRs with welding fumes, a hazardous aerosol that exists in real workplace settings. The aim of this study was to compare the performance of respirators and filters against a NaCl aerosol and a welding fume aerosol and determine whether or not a correlation between the two could be made. Fifteen commercial PRs and filters (seven filtering facepiece, two replaceable single-type filters, and six replaceable dual-type filters) were chosen for investigation. Four of the filtering facepiece respirators, one of the single-type filters, and all of the dual-type filters contained carbon to help reduce exposure to ozone and other vapors generated during the welding process. For the NaCl test, a modified National Institute for Occupational Safety and Health protocol was adopted for use with the TSI Model 8130 automated filter tester. For the welding fume test, welding fumes from mild steel flux-cored arcs were generated and measured with a SIBATA filter tester (AP-634A, Japan) and a manometer in the upstream and downstream sections of the test chamber. Size distributions of the two aerosols were measured using a Scanning Mobility Particle Sizer spectrometer. Penetration and pressure drop were measured over a period of aerosol loading onto the respirator or filter. Photos and scanning electron microscope images of clean and exposed respirators were taken. The count median diameter (CMD) and mass median diameter (MMD) for the NaCl aerosol were smaller than the welding fumes (CMD: 74 versus 216 nm; MMD: 198 versus 528 nm, respectively). Initial penetration and peak penetration were higher with the NaCl aerosol. However, pressure drop increased much more

rapidly in the welding fume test than the NaCl aerosol test. The data and images clearly show differences in performance trends between respirator models. Therefore, general correlations between NaCl and weld fume data could not be made. These findings suggest that respirators certified with a surrogate test aerosol such as NaCl are appropriate for filtering welding fume (based on penetration). However, some respirators may have a more rapid increase in pressure drop from the welding fume accumulating on the filter. Therefore, welders will need to choose which models are easier to breathe through for the duration of their use and replace respirators or filters according to the user instructions and local regulations.

## 2010

Seto, T., Furukawa, T., Otani, Y., Uchida, K., and Endo, S., 2010, "Filtration of Multi-Walled Carbon Nanotube Aerosol by Fibrous Filters," *Aerosol Science and Technology*, **44**(9): 734–740

Filtration efficiency of multi-walled carbon nanotube (MWCNT) aerosol by fibrous filter was evaluated experimentally. Mono-mobility test aerosols with electrical mobility diameter of 100, 200, and 300 nm were generated by the atomization of MWCNT aqueous suspension followed by mobility classification with a differential mobility analyzer (DMA). By analyzing the shape of classified aerosol particles under a scanning electron microscope, it was found that the DMA-classified 300 nm particles were fibrous in shape and had uniform diameter of about 60 nm and length of 2.1 micrometer. On the other hand, 100 nm and 200 nm particles contained a fairly large amount of multiply charged fibrous particles with a larger diameter. These test aerosols were challenged to a medium performance fibrous filter at various filtration velocities. As a result, fibrous particles were captured by fibrous filter at a higher collection efficiency than the spherical particles with the same mobility. By analyzing the single fiber capturing efficiency, interception incorporating the rotation of fibrous particles is found to be the dominant capturing mechanism for the fibrous particles in the studied size range.

Zhang, Q., Welch, J., Park, H., Wu, C.-Y., Sigmund, W., and Marijnissen, J. C. M., 2010, "Improvement in nanofiber filtration by multiple thin layers of nanofiber mats," *Journal of Aerosol Science*, **41**(2): 230–236

Nanofiber filtration is drawing great interest nowadays because of its large surface collection area as well as low air resistance. In this study, electrospun nanofiber mats of different thicknesses were evaluated for their filter quality factors. Shorter-term electrospun fiber mats exhibited a better quality factor than those longer-term electrospun ones. Multiple thin layers of nanofiber mats to improve the filter quality of the nanofiber filters were then evaluated. Filtration test results showed that the filter made up of multiple thin layers of nanofiber mats had a filter quality factor much higher than the single thick layer nanofiber mat. Better thickness uniformity in the multi-layer structure due to stacking compensation and smaller fiber diameters in nanofibers of short-term deposition time are two possible reasons for the improvement of the filter quality.

## 2009

Li, H.-W., Wu, C.-Y., Tepper, F., Lee, J.-H., and Lee, C. N., 2009, "Removal and retention of viral aerosols by a novel alumina nanofiber filter," *Journal of Aerosol Science*, **40**(1): 65–71

Nanomaterial, due to its unique physical, chemical and biological properties compared to its bulk counterparts, has the potential to provide a product superior to its bulk predecessor. In this study, a novel alumina nanofiber filter was assessed for its removal and retention capability for MS2 aerosol. Its physical removal efficiency in the 10–400nm range was 94.35%, while its viable removal efficiency was 98.87%, which was slightly lower than three conventional HEPA filters tested. However, its pressure drop was much lower than HEPA filters, yielding a higher filter quality than HEPA filters. The average extracted fraction from the nanofiber filter was  $8.64 \times 10^{-2} \pm 7.00 \times 10^{-2}$ , which is three orders lower than other HEPA filters, demonstrating that the viruses were effectively retained in the nanofiber filter. Furthermore, the performance of the nanofiber filter showed no dependence on relative humidity. In conclusion, this novel alumina nanofiber filter presents advantageous potential for removal and retention of viral aerosol agents.

## 2008

Boskovic, L., Agranovski, I. E., Altman, I. S., and Braddock, R. D., 2008, "Filter efficiency as a function of nanoparticle velocity and shape," *Journal of Aerosol Science*, **39**(7): 635–644

The filtration efficiency of a conventional fibrous filter was investigated with particular emphasis on the removal of particles with different shapes. A previous study has shown that particles of spherical shape are removed from the gas carrier with efficiencies which are higher when compared to cubic particles of the same aerodynamic size. In this project, to challenge our previously made explanation, spherical PSL and cubic MgO particles were tested along with particles of sodium chloride (NaCl) of intermediate shape (cubic particles with rounded edges) at a range of filtration velocities from 5 to 20 cm/s. It was found that particles of NaCl are removed with efficiencies lower than those for PSL particles but higher than the



efficiencies for cubic particles of MgO, at the lowest filtration velocity when inertial effects are negligible. The rounded NaCl particles, depending on the geometry of the contact, could either land on the rounded corner and hence roll, land on a sharp edge and hence tumble, or slide. This range of options alters the probability of detachment of the particle. The difference between the filter efficiencies for cubic MgO particles and intermediate shaped NaCl particles is decreasing with the increase in velocity. With increasing velocity, the filtration efficiency of the cubic MgO particles, exceeds the filtration efficiency for the intermediate shaped NaCl particles, due to the dominating inertial effects of the denser, and hence heavier, MgO particles. This paper shows the results of these experiments and, we hope, will ignite the interest of the aerosol community towards further theoretical analysis of the phenomenon.

## 2007

Japuntich, Daniel; Franklin, Luke; Pui, David; Kuehn, Thomas; Kim, Seong; Viner, Andrew, 2007, "A comparison of two nano-sized particle air filtration tests in the diameter range of 10 to 400 nanometers," *Journal of Nanoparticle Research*, **9**(1):93-107

Two different air filter test methodologies are discussed and compared for challenges in the nano-sized particle range of 10-400 nm. Included in the discussion are test procedure development, factors affecting variability and comparisons between results from the tests. One test system which gives a discrete penetration for a given particle size is the TSI 8160 Automated Filter tester (updated and commercially available now as the TSI 3160) manufactured by the TSI, Inc., Shoreview, MN. Another filter test system was developed utilizing a Scanning Mobility Particle Sizer™ (SMPS™) to sample the particle size distributions downstream and upstream of an air filter to obtain a continuous percent filter penetration versus particle size curve. Filtration test results are shown for fiberglass filter paper of intermediate filtration efficiency. Test variables affecting the results of the TSI 8160 for NaCl and dioctyl phthalate (DOP) particles are discussed, including condensation particle counter stability and the sizing of the selected particle challenges. Filter testing using a TSI 3936 SMPS sampling upstream and downstream of a filter is also shown with a discussion of test variables and the need for proper SMPS volume purging and filter penetration correction procedure. For both tests, the penetration versus particle size curves for the filter media studied follow the theoretical Brownian capture model of decreasing penetration with decreasing particle diameter down to 10 nm with no deviation. From these findings, the authors can say with reasonable confidence that there is no evidence of particle thermal rebound in the size range.

## 2006

Kim, Seong; Wang, Huaping; Imagawa, Masayuki; Chen, Da-Ren; Pui, David, 2006, "Experimental and Modeling Studies of the Stream-Wise Filter Vibration Effect on the Filtration Efficiency," *Aerosol Science and Technology*, **40**(6):389-395

The stream-wise vibration effect of a fibrous filter is studied experimentally and numerically for the purpose of evaluating filtration efficiency. The particle sizes range from 0.02 to 10  $\mu\text{m}$  and the face velocity ranges from 3 to 10 cm/s. The vibrational peak velocity also varied from 0 to 50 cm/s. The filtration efficiency for this wide size range is obtained by combining the individual test results for fine particles (0.02 to 0.5  $\mu\text{m}$ ) and large particles (0.5 to 10.0  $\mu\text{m}$ ). For the fine particle experiment, Arizona Road Dust (ARD) test particles are generated by an atomizer after an ultrasonic process and measured by a Scanning Mobility Particle Sizer (SMPS) spectrometer. For the large particle experiment, the test particles are generated by a fluidized bed and measured by an Aerodynamic Particle Sizer (APS). When the particles are generated by the atomizer after ultrasonication, the majority of the particles are in nano scale without the agglomerates on the large particle surface, while particles generated by the fluidized bed are mostly in micro-scale because many nanoparticles are agglomerated on large particle surface. The filtration efficiency increases with the vibrational peak velocity in the impaction-dominant region ( $D_p > 0.1 \mu\text{m}$ ) and diffusion-dominant region ( $D_p$ )

## 2003

Chen, C.-C.; Huang, S.-H., 2003, "Loading characteristics of a miniature wire-plate electrostatic precipitator," *Aerosol Science & Technology*, **37**(2):109-121

In this work, in order to investigate the particle loading effects on the performance of an electrostatic precipitator (ESP), simultaneous measurements of the dust cake thickness accumulated on the collection plates, ESP's collection efficiency, corona discharge characteristics, and ozone concentration were conducted experimentally. A laboratory scale single stage wire-plate ESP was used for the aerosol loading test. Two kinds of particulate matter, cement and aluminum oxide ( $\text{Al}_2\text{O}_3$ ), were generated by using a Palas Powder Disperser. A displacement meter was used to monitor the dust cake thickness accumulated on the collection plates. A Scanning Mobility Particle Sizer was used to measure the particle size distribution and number concentration upstream and downstream of the ESP. Ozone generated by the ESP was sampled 20 cm downstream of the ESP exit and measured with an ozone analyzer. The Dioctyl Phthalate (DOP) was also used as a liquid challenge agent in order to investigate the loading effects of liquid particles on the ESP performance.

The results showed that when challenged with cement particles, the ion current decreased with increasing dust cake thickness under a constant electrical field strength. Moreover, the collection efficiency and ozone generated by corona discharge decreased as the loading test progressed. For example, when the dust layer was about 5 mm in thickness, the output current and the ozone concentration decreased about 33 and 44%, respectively, and the collection efficiency (300 nm particle) decreased about 4% at a fixed electrical field strength of -4.2 kV/cm. However, the ion current increased as aluminum oxide particles deposited on the collection plates. The increase in ozone concentration and aerosol penetration was mainly due to the occurrence of back corona, evidenced by the existence of the caves on the surface of the dust layers. In the case of testing with cement particles, the ion current rises after about 20 min of loading test and then decreases with time, while ozone concentration increases synchronously.

## 2001

Chen C.-C.; Chen W.-Y.; Huang S.-H.; Lin W.-Y.; Kuo Y.-M.; Jeng F.-T., 2001, "Experimental study on the loading characteristics of needlefelt filters with micrometer-sized monodisperse aerosols," *Aerosol Science and Technology*, **34**(3):262-273

In this work, three types of needlefelt filters, made of Polyester (PE), Ryton Sulfar (RS), and Polyaramid (PA), were tested to investigate the aerosol loading characteristics of fabric filters when challenged with micrometer-sized monodisperse potassium sodium tartrate (PST) particles. A fibrous filter with packing density of 9%, thickness of 0.38 mm, and fiber diameter of 5.1  $\mu\text{m}$  was included for comparison. A vibrating orifice monodisperse aerosol generator was used to produce three different sizes (5, 10, and 20  $\mu\text{m}$ ) of PST particles for aerosol loading experiment. An ultrasonic atomizing nozzle and a TSI constant output nebulizer were used to generate polydisperse PST particles for the aerosol penetration test. The aerosol penetration of submicrometer-sized particles through the filters was measured by using a Scanning Mobility Particle Sizer spectrometer. An Aerodynamic Particle Sizer was used to measure the penetration fraction of aerosol particles larger than 0.8  $\mu\text{m}$ . The pressure drop across the filter was monitored by using pressure transducers, which were calibrated against an inclined manometer. Airflows of 5, 10, 20, and 30 cm/s were used to study the flow dependency. The aerosol penetration results showed that the particles larger than 3  $\mu\text{m}$  did not penetrate the clean fabric filters tested in the present study. The loading curves (plots of pressure drop against sampling time) displayed three regions: an initial region of fast increase, a transition region, and a final linear region after the dust formation point. After the formation point of the dust cake, both fabric and fibrous filters shared the same slope (of the loading curves). The slope of different regions of the loading curves was determined by many factors, such as size of challenge aerosol, face velocity, surface treatment, and the compressibility of the dust cake forming on the filter. The method of final surface treatment was found to be critical to the performance of the fabric filters. In order to avoid the unnecessary rise in air resistance, the melting clumps formed during final surface treatment should be as thin and narrow as possible, just enough to support the filter bag cleaning. From the standpoint of filter quality and energy consumption, the low filtration velocity has to be adopted whenever possible, because high filtration velocity not only led to lower filter quality (in particular for submicrometer-sized particles) but also created dust cake of lower porosity, which caused an extra rise in pressure drop across the dust cake.

Huang, Sheng-Hsiu, Chen, Chih-Chieh, 2001, "Ultrafine Aerosol Penetration through Electrostatic Precipitators," *Environmental Science and Technology*, **36**(21):4625-4632

This work measures the penetration of ultrafine particles through a single-stage and a two-stage ESP as a function of particle size. Also studied herein are how parameters including particle size, rate of airflow through the ESP, and voltage of the discharging electrode affect aerosol penetration through the ESP. Monodisperse particles with sizes between 10 and 60 nm were generated as the challenge aerosols to investigate the particle charges given by an ESP. A comparison of experimental and theoretical results confirms that a partial charging regime exists when the particle diameter is several tens of nanometers. Experimental results indicated that aerosol penetration through the single- and two-stage ESPs increased significantly for particles below 20 and 50 nm, respectively. However, the exact regime depends on the parameters including airflow rate, applied voltage, and configuration of the ESP. Phenomena such as ionic flow, particle space charge, and flow turbulence may significantly affect the collection efficiency of an ESP for ultrafine particles. To achieve the same collection efficiency, it is more economical to use single-stage ESPs to collect particles less than 16 nm from the standpoint of energy consumption. However, it is more economical to use two-stage ESPs to collect particles larger than 16 nm.

## 1998

Chen, C.-C.; Hang, S.-H., "The effects of particle charge on the performance of a filtering facepiece," *American Industrial Hygiene Association Journal*, **59**(4):227-233

This study quantitatively determined the effect of electrostatic charge on the performance of an electret filtering facepiece. Monodisperse challenge corn oil aerosols with uniform charges were generated using a modified vibrating orifice monodisperse aerosol generator. The aerosol size distributions and concentrations

upstream and downstream of an electret filter were measured using an Aerodynamic Particle Sizer spectrometer, an Aerosizer® Particle Size Analyzer, and a Scanning Mobility Particle Sizer spectrometer. The aerosol charge was measured by using an aerosol electrometer. The tested electret filter had a packing density of about 0.08, fiber size of 3 µm, and thickness of 0.75 mm. As expected, the primary filtration mechanisms for the micrometer-sized particles are interception and impaction, especially at high face velocities, while electrostatic attraction and diffusion are the filtration mechanisms for submicrometer-sized aerosol particles. The fiber charge density was estimated to be 1.35 times 10<sup>-5</sup> coulomb per square meter. After treatment with isopropanol, most of fiber charges were removed, causing the 0.3-µm aerosol penetration to increase from 36 to 68%. The air resistance of the filter increased slightly after immersion in the isopropanol, probably due to the coating of impurities in isopropanol. The aerosol penetration decreased with increasing aerosol charge. The most penetrating aerosol size became larger as the aerosol charge increased, e.g., from 0.32 to 1.3 µm m when the aerosol charge increased from 0 to 500 elementary charges.

## 1994

Siag, A. M.; Tennal, K. B.; Mazumder, M. K., 1994, "Determination of fiber charge density of electret filters," *Particulate Science and Technology*, 351-355

Particle collection in electret filters is enhanced by electrostatic forces resulting from the embedded charge on the fibers of the filters. Currently, there is no simple method available for determining the charge density on the electret fibers. Since charge on the fibers can degrade with loading or during storage, a simple evaluation technique is desirable. Aerosol consisting of organic oil mist generally used for testing HEPA filters should not be used for evaluating electret filters since organic aerosol droplets rapidly degrade the electrostatic charge effect. In this study experimental measurements of penetration were made on electret filters challenged with monodisperse, polystyrene latex spheres, PLS, of known size and charge. The experimental data on penetration were used to separate the contributions of the individual collection parameters and to determine the effective density of charge on the fibers. Collection efficiency due to mechanical filtration mechanisms was measured using filters in which the electret charge had been effectively neutralized by loading the filters with an oil aerosol. Collection due to dielectrophoretic force was obtained using aerosol with zero charge. The effective charge density on the fibers was determined by comparing measurements of penetration for particles having known size and charge with the predictions of available theoretical equations. Details of this method of determining the density of charge on the fibers are described here. Experimental determinations of charge density showed reasonable agreement with the charge densities given by the manufacturers of the tested filters.

## 1992

Wake, D.; Brown, R.C.; Trottier, R.A.; Liu, Y., 1992, "Comparison of the Efficiency of Respirator Filters and Filtering Facepieces Against Radon Daughter Aerosols and Laboratory Generated Aerosols," *Aerosol Science*, 23(S1):S757-S760

Respirator filters and filtering facepieces have been tested for filtration efficiency against radon daughters in a flourspar mine. The test method involved the use of sampling filters exposed to natural radon daughters in air filtered by the test respirators. Respirators with a filtration efficiency high enough for them to be considered suitable for use against toxic dusts generally reduced radon daughter levels by 90% or more, though nuisance dust masks were ineffective. The measured penetration of radon daughters through the former types of filter correlated reasonably well with the penetration of 0.1µm neutralized monodisperse aerosol and with that of the BS4400 sodium chloride aerosol, measured in the laboratory. Test filters were exposed to radon daughters in the mine for 8 hours as a simulation of their behavior during a working shift, but their performance was not altered by this and they did not show measurable radioactivity as a result of it.

Ylatalo, Sampo I.; Kauppinen, Esko I.; Hautanen, Jukka; Joutsensaari, Jorma; Ahonen, Petri; Lind, Terttaliisa M.; Jokiniemi, Jorma K.; Kilpelainen, Markku, 1992, "On the Determination of Electrostatic Precipitator Efficiency by Differential Mobility Analyzer," *Aerosol Science*, 23(S1):S795-S798

In order to determine penetration curve of the electrostatic precipitator (ESP) as a function of aerosol particle diameter in the range of 10-1000 nm measurement series were carried out in real scale power plant conditions. Differential mobility particle sizing (DMPS) system was used to measure the particle mobility distributions before and after ESP. MICRON-algorithm (constrained regularization) was used to invert mobility distributions to the corresponding number distributions. Penetration curve was calculated from the measured number distributions.

Zhang, Zhiqun; Liu, Benjamin Y. H., 1992, "Experimental Study of Aerosol Filtration in the Transition Flow Regime," *Aerosol Science and Technology*, 16(44):227-235

An experimental study was carried out to investigate the performance of fibrous filters in the transition flow regime, where the fiber radius is of the same order of magnitude as the mean free path of gas

molecules. Filter media with the mean fiber diameter of 0.21-0.72  $\mu\text{m}$  and the packing density of 0.053-0.08 were used in this study. The transition flow conditions were achieved by reducing the gas pressure in the filter test apparatus. Experiments were performed in the pressure range of 0.1-1.0 atm using monodisperse particles of 0.04-0.45  $\mu\text{m}$  diameter range. The particles were generated by the electrostatic classification technique. The particulate penetration through the filter and the corresponding pressure drop were measured at face velocities ranging from 6 to 19 cm/s. Experimental results showed that the penetration decreased by three to four orders of magnitude as the pressure was reduced from 1.0 to 0.1 atm. It was found that the most penetrating particle size increased slightly as the gas pressure was reduced. The pressure drop across the filter was also found to decrease significantly as the pressure was reduced.

## 1990

Yu, Aoyuan, 1990, "A Simple and Reliable Testing Method for Particle-Collection Efficiency of Ultra Low Penetration Air (ULPA) Filters," *Swiss Contamination Control*, **3**:259-261

As specified in American IES Recommended Practice IES-RP-cc-001-86, Ultra Low Penetration Air (ULPA) filter is the filter having a minimum particle-collection efficiency of 99.999% for particulate diameters  $>0.12 \mu\text{m}$  in size. And yet, the Recommended Practice does not give a detailed description of so-called <<Dual Laser Tests>> procedure. Here, the author presents a simple and reliable testing method using cold DOP as a challenge aerosol source and employing TSI Model 3071 Electrostatic Classifier and Model 3020 CNC (Condensation Nucleus Counter) to measure the efficiency of ULPA filter on 0.10  $\mu\text{m}$  (or 0.12  $\mu\text{m}$ ) in size. The author doesn't think the specified  $> 0.12 \mu\text{m}$  of particle size is a necessary criterion for ULPA filter. Testing <<on 0.12  $\mu\text{m}$ >> particulate diameter for ULPA is a better solution, due to its good comparability and labor- and time-savings. Again, CNC does not need dilution air, and the conventional error from dilution will be avoided. The paper also makes some comments on current ULPA filter test procedures, and suggest using on <<on 0.10  $\mu\text{m}$ >> as a reference particle size for ULPA filters.

## 1987

Holton, Patricia M.; Tackett, Denise Lynne; Willeke, Klaus, 1987, "Particle Size-Dependent Leakage and Losses of Aerosols in Respirators," *American Industrial Hygiene*, **48**(10):848-854

Measuring particle size-dependent leakage into and losses inside a respirator reveals the deposition mechanisms occurring at the leak site and the flow dynamics inside the respirator. This study investigated particle size-dependent leakage and deposition within the mask by examining the leakage into the mask for different hole locations, probe locations, hole shapes, hole lengths and hole sizes. The shape of the leak has an effect on particle size-dependent leakage. Probe and leak location tests indicated that not only does the total measured leakage change but also the size-dependence of the leakage changes depending on the leak and probe locations. When the leak site is in the chin area, the clean air entering through the filters at the chin helps to carry the inward leakage into the breathing zone. Particle size-dependent leakage does occur and is due to both inertial entry losses at the leak site and within the mask, and diffusional losses within the mask and leak site. Particle size-dependent curves change shape as the hole size changes with relatively more larger particles entering through the small hole size.

Holton, Patricia M.; Willeke, Klaus, 1987, "The Effect of Aerosol Size Distribution and Measurement Method on Respirator Fit," *American Industrial Hygiene*, **48**(10):855-860

The particle size-dependent leakage into a respirator was examined by measuring the leakage of particle sizes between 0.07 to 4.4  $\mu\text{m}$  through three hole sizes in a negative-pressure half-mask respirator worn by a human subject. This investigation showed that the size distribution of an aerosol test agent and the measurement method have an effect on the leakage measured in a quantitative filter test. For instance, the ratio of percent leakage measured by light scattering between test aerosols with count median diameters of 2.2 and 0.28  $\mu\text{m}$  can be as large as 5:1. Likewise, the ratio of the percent leakage measured by a particle count method vs. a mass method of detection of the same polydisperse aerosol with a count medium diameter equal to 2.2  $\mu\text{m}$  can be as high as 4:1. The mass leakage into a mask with a leak is also greater for an exposure aerosol with a count median diameter between 0.15 to 0.30  $\mu\text{m}$  compared to exposure aerosols with larger count median diameters for aerosols with the same mass concentration.

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## Fundamental Research and Chemistry

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### 2013

Abramson, E., Imre, D., Beránek, J., Wilson, J., and Zelenyuk, A., 2013, "Experimental determination of chemical diffusion within secondary organic aerosol particles.," *Physical Chemistry Chemical Physics : PCCP*, **15**(8): 2983–91

Formation, properties, transformations, and temporal evolution of secondary organic aerosol (SOA) particles depend strongly on SOA phase. Recent experimental evidence from both our group and several others indicates that, in contrast to common models' assumptions, SOA constituents do not form a low-viscosity, well-mixed solution, yielding instead a semisolid phase with high, but undetermined, viscosity. We find that when SOA particles are made in the presence of vapors of semi-volatile hydrophobic compounds, such molecules become trapped in the particles' interiors and their subsequent evaporation rates and thus their rates of diffusion through the SOA can be directly obtained. Using pyrene as the tracer molecule and SOA derived from  $\alpha$ -pinene ozonolysis, we find that it takes  $\sim 24$  hours for half the pyrene to evaporate. Based on the observed pyrene evaporation kinetics we estimate a diffusivity of  $2.5 \times 10^{-21} \text{ m}^2 \text{ s}^{-1}$  for pyrene in SOA. Similar measurements on SOA doped with fluoranthene and phenanthrene yield diffusivities comparable to that of pyrene. Assuming a Stokes-Einstein relation, an approximate viscosity of  $10^8 \text{ Pa s}$  can be calculated for this SOA. Such a high viscosity is characteristic of tars and is consistent with published measurements of SOA particle bounce, evaporation kinetics, and the stability of two reverse-layered morphologies. We show that a viscosity of  $10^8 \text{ Pa s}$  implies coalescence times of minutes, consistent with the findings that SOA particles formed by coagulation are spherical on the relevant experimental timescales. Measurements on aged SOA particles doped with pyrene yield an estimated diffusivity  $\sim 3$  times smaller, indicating that hardening occurs with time, which is consistent with the increase in SOA oligomer content, decrease in water uptake, and decrease in evaporation rates previously observed with aging.

Antonsson, E., Bresch, H., Lewinski, R., Wassermann, B., Leisner, T., Graf, C., Langer, B., and Rühl, E., 2013, "Free nanoparticles studied by soft X-rays," *Chemical Physics Letters*, **559**: 1–11

The use of nanoparticle beams for the preparation and characterization of isolated nanoscopic matter is reviewed. It is a general preparation scheme which is shown to be of broad use, whenever the intrinsic properties of nanoscopic matter without any interactions with other particles or substrates are of interest. The size regime spans a few nanometers up to the micron sizes, wherein a large variety of substances and materials can be studied. Similarly broad are the detection schemes and properties, which can be investigated by this approach. This review covers various properties of isolated nanoparticles, including their size, surface properties, optical constants, as well as their local electronic structure.

Liu, C., Chu, B., Liu, Y., Ma, Q., Ma, J., He, H., Li, J., and Hao, J., 2013, "Effect of mineral dust on secondary organic aerosol yield and aerosol size in  $\alpha$ -pinene/NO<sub>x</sub> photo-oxidation," *Atmospheric Environment*, **77**: 781–789

Although it is a significant contributor to atmospheric particles, the role of mineral dust in secondary organic aerosol (SOA) formation has not been fully recognized. In this study, alumina was chosen as the surrogate to investigate the effect of mineral dust on  $\alpha$ -pinene/NO<sub>x</sub> photo-oxidation in a 2 m<sup>3</sup> smog chamber at 30 C and 50% relative humidity (RH). Results showed that alumina seeds could influence both the SOA yield and the aerosol size in the photo-oxidation process. Compared to the seed-free system, the presence of alumina seeds resulted in a slight reduction of SOA yield, and also influenced the final concentration of O<sub>3</sub> in the chamber. As an important oxidant of  $\alpha$ -pinene, the decrease in O<sub>3</sub> concentration could reduce the formation of semi-volatile compounds (SVOCs) and consequently inhibited SOA formation. In addition, the size of aerosol was closely related with the mass loading of alumina seeds. At low alumina concentration, SVOCs condensed onto the pre-existing seed surface and led to aerosol size growth. When alumina concentration exceeded about 5  $\mu\text{g m}^{-3}$ , SVOC species that condensed to each seed particle were dispersed by alumina seeds, resulting in the decrease in aerosol size.

Nah, T., Kessler, S. H., Daumit, K. E., Kroll, J. H., Leone, S. R., and Wilson, K. R., 2013, "OH-initiated oxidation of sub-micron unsaturated fatty acid particles," *Physical Chemistry Chemical Physics: PCCP*, **15**(42): 18649–63

The heterogeneous reaction of OH radicals with sub-micron unsaturated fatty acid particles in the presence of H<sub>2</sub>O<sub>2</sub> and O<sub>2</sub> is studied to explore how surface OH addition reactions initiate chain reactions that rapidly transform the chemical composition of an organic particle. In the presence of 20.7 ppm [H<sub>2</sub>O<sub>2</sub>] in a 10% mixture of O<sub>2</sub> in N<sub>2</sub>, the effective uptake coefficients of oleic acid, linoleic acid and linolenic acid are found to be  $1.72 \pm 0.08$ ,  $3.75 \pm 0.18$  and  $5.73 \pm 0.14$ , respectively. These effective uptake coefficients are larger than unity, providing clear evidence for particle-phase secondary chain chemistry. The effective uptake coefficient increases linearly with the number of C=C double bonds in the unsaturated fatty acid molecule. Elemental composition analysis reveals that there is an addition of, on average,  $0.57 \pm 0.02$ ,  $0.61 \pm 0.01$  and  $0.73 \pm 0.04$  O atoms per reactive loss of oleic acid, linoleic acid and linolenic acid, respectively, which suggests that OH addition to the C=C double bond is not the sole reaction pathway that consumes the molecular species. These results suggest the potential presence of secondary reactions that consume the unsaturated fatty acid molecular species without increasing the particulate oxygen content. As the unsaturated fatty acid particles become more oxygenated, volatilization also becomes significant. The magnitudes of the effective uptake coefficients are found to be dependent on the concentrations of OH, O<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> in the flow reactor. A plausible reaction mechanism is presented to show how surface OH addition reactions initiate chain reactions that rapidly transform an unsaturated organic particle's physicochemical properties.

Nelson, A. J., Page, M. a., Ginsberg, M. D., and Rood, M. J., 2013, "Bench-Scale Aerosol Filtration Test System and Evaluation of an Acoustic Bioaerosol Removal Device for Indoor Air Streams," *Aerosol Science and Technology*, **47**: 1285–1292

It is important to have well-defined, reproducible methods to evaluate and compare newly developed air filtration equipment. To facilitate accurate assessment of air purification devices at the bench scale, an experimental system was designed, built, and documented to evaluate particulate removal efficiency (PRE) of air filtration devices based on principles used in ASHRAE standards. The system was then carefully characterized and used to evaluate PRE and total energy consumption of a novel acoustically enhanced impaction (AEI) air purification device. The AEI device demonstrated 99.998% PRE of 0.5–1.5 μm diameter KCl particles while causing a 120 Pa pressure drop and requiring a total of 3.0 W/l of air treated at indoor ambient conditions. A single element of the AEI device operated in a biological safety level 2 facility was then used to evaluate PRE of bioaerosol consisting of *Bacillus cereus* (BC) spores. PRE of BC was  $99.86 \pm 0.05\%$  at indoor ambient temperature and pressure. This research describes the use of the Bench-scale Air Purification Testing and Evaluation Chamber (BAP-TEC) to experimentally evaluate and compare PRE and total energy requirements of novel air purification devices at the bench scale (280–1400 alpm). Further, an AEI device containing a fibrous filter media and high intensity sound field in the same control volume is evaluated using the BAP-TEC. Temporally resolved PRE of a bioaerosol by the AEI is also presented.

Pawar, A. A., and Venkataraman, C., 2013, "Pulse-Heat Aerosol Reactor (PHAR): Processing Thermolabile Biomaterials and Biomolecules into Nanoparticles with Controlled Properties," *Aerosol Science and Technology*, **47**(4): 383–394

This work addresses the challenge of processing thermolabile biomaterials and biomolecules into nanoparticles without compromising structural integrity and activity. Control of size, structure, and crystallinity of nanoparticles is desirable for drug targeting and controlled release applications. An innovation using pulse-heat aerosol processing is demonstrated through a design of a pulse-heat aerosol reactor. Nanoparticle aerosol lipid matrices (NALM), of stearic acid, were synthesized under different processing conditions, obtained through pulse heating (fixed duration, three heat-pulse levels) and solvent selection. Operation with continuous heating resulted in ill-conditioned, multimodal size distributions. Pulse heating resulted in the synthesis of NALM with mean mobility diameter in the range of 56–183 nm and narrow unimodal size distributions (geometric size distribution, GSD=1.5–1.7). Under higher pulse levels, particles with larger mean mobility diameter were formed, which had shell-type structures, compared with smaller, solid particles under operation with low level or no heating. NALM with different degree of crystallinity were produced under different processing conditions, at which different drop temperature is expected to result. The activity of glucose oxidase enzyme was preserved, when subjected to high pulse-heat (gas temperature of 110°C) aerosol processing. These results establish pulse-heat aerosol processing as a single-step, continuous method to process heat-sensitive biomaterials and biomolecules into nanoparticles with controlled properties, while avoiding thermal damage.

Carlsson, P. T. M., Keunecke, C., Krüger, B. C., Maaß, M.-C., and Zeuch, T., 2012, "Sulfur dioxide oxidation induced mechanistic branching and particle formation during the ozonolysis of  $\beta$ -pinene and 2-butene.," *Physical Chemistry Chemical Physics: PCCP*, **14**(45): 15637–40

Recent studies have suggested that the reaction of stabilised Criegee Intermediates (CIs) with sulfur dioxide ( $\text{SO}_2$ ), leading to the formation of a carbonyl compound and sulfur trioxide, is a relevant atmospheric source of sulfuric acid. Here, the significance of this pathway has been examined by studying the formation of gas phase products and aerosol during the ozonolysis of  $\beta$ -pinene and 2-butene in the presence of  $\text{SO}_2$  in the pressure range of 10 to 1000 mbar. For  $\beta$ -pinene at atmospheric pressure, the addition of  $\text{SO}_2$  suppresses the formation of the secondary ozonide and leads to highly increased nopinone yields. A complete consumption of  $\text{SO}_2$  is observed at initial  $\text{SO}_2$  concentrations below the yield of stabilised CIs. In experiments using 2-butene a significant consumption of  $\text{SO}_2$  and additional formation of acetaldehyde are observed at 1 bar. A consistent kinetic simulation of the experimental findings is possible when a fast CI +  $\text{SO}_2$  reaction rate in the range of recent direct measurements [Welz et al., *Science*, 2012, 335, 204] is used. For 2-butene the addition of  $\text{SO}_2$  drastically increases the observed aerosol yields at higher pressures. Below 60 mbar the  $\text{SO}_2$  oxidation induced particle formation becomes inefficient pointing to the critical role of collisional stabilisation for sulfuric acid controlled nucleation at low pressures.

Dey, L., and Venkataraman, C., 2012, "A Wet Electrostatic Precipitator (WESP) for Soft Nanoparticle Collection," *Aerosol Science and Technology*, **46**(7): 750–759

Many nanoparticle collection devices have limitations related to retention of particle integrity from bounce, shattering, or aggregation. Suspensions of soft nanoparticles (e.g., proteins, lipids) are required for drug delivery and therapy. To enable direct collection of soft nanoparticles into liquid media, a wet electrostatic precipitator (WESP) was designed and evaluated in this work. Different sections were used for ion generation and particle charging, for minimal contact between the corona wire and particles, which were charged using positive nitrogen ions. WESP dimensions and operating parameters were optimized using charge distribution modeling. The prototype WESP was designed for operation with a continuous flow of liquid over the collection plate, to allow continuous particle collection from the exit stream of an aerosol reactor. The collection efficiency of the WESP, in dry and wet modes, was measured using aerosols of monodisperse polystyrene latex (PSL), polydisperse sucrose, and stearic acid (soft lipid) particles, through SMPS measurements, corrected for diffusional losses, at the entry and exit of the device. Measured collection efficiency was 70%–90% for particles of sizes 80–600 nm diameter in reasonable agreement with theoretical estimates. However, for small particles (20–80 nm diameter) measured collection efficiency ranged 40%–70%, significantly lower than theoretical estimates, possibly from incomplete neutralization of negative charges attained during air-jet atomization. Transmission electron microscopy (TEM) images and dynamic light scattering (DLS) measurements confirm that wet collection produces a suspension of free, unaggregated nanoparticles with sizes similar to their measured mean mobility diameter.

Ku, B. K., and Evans, D. E., 2012, "Investigation of Aerosol Surface Area Estimation from Number and Mass Concentration Measurements: Particle Density Effect," *Aerosol Science and Technology*, **46**(4): 473–484

For nanoparticles with nonspherical morphologies, e.g., open agglomerates or fibrous particles, it is expected that the actual density of agglomerates may be significantly different from the bulk material density. It is further expected that using the material density may upset the relationship between surface area and mass when a method for estimating aerosol surface area from number and mass concentrations (referred to as "Maynard's estimation method") is used. Therefore, it is necessary to quantitatively investigate how much the Maynard's estimation method depends on particle morphology and density. In this study, aerosol surface area estimated from number and mass concentration measurements was evaluated and compared with values from two reference methods: a method proposed by Lall and Friedlander for agglomerates and a mobility based method for compact nonspherical particles using well-defined polydisperse aerosols with known particle densities. Polydisperse silver aerosol particles were generated by an aerosol generation facility. Generated aerosols had a range of morphologies, count median diameters (CMD) between 25 and 50 nm, and geometric standard deviations (GSD) between 1.5 and 1.8. The surface area estimates from number and mass concentration measurements correlated well with the two reference values when gravimetric mass was used. The aerosol surface area estimates from the Maynard's estimation method were comparable to the reference method for all particle morphologies within the surface area ratios of 3.31 and 0.19 for assumed GSDs 1.5 and 1.8, respectively, when the bulk material density of silver was used. The difference between the Maynard's estimation method and surface area measured by the reference method for fractal-like agglomerates decreased from 79% to 23% when the measured effective particle density was used, while the difference for nearly spherical particles decreased from 30% to 24%. The results indicate that the use of particle density of agglomerates improves the accuracy of the Maynard's estimation method and that an effective density should be taken

into account, when known, when estimating aerosol surface area of nonspherical aerosol such as open agglomerates and fibrous particles.

Pawar, A. A., Chen, D.-R., and Venkataraman, C., 2012, "Influence of precursor solvent properties on matrix crystallinity and drug release rates from nanoparticle aerosol lipid matrices.," *International journal of pharmaceutics*, **430**(1-2): 228–37

The crystallinity of drug-loaded lipid nanoparticles is believed to affect drug release rates; however, effective control over lipid crystallinity has not been achieved by current lipid nanoparticle preparation methods. The present study investigates control over the crystallinity of drug-loaded nanoparticle aerosol lipid matrices (NALM) through differences in evaporation rates of precursor solution drops and the subsequent control over drug release rates from these matrices. Gefitinib-loaded NALM were synthesized in an aerosol reactor using precursor solutions of gefitinib and stearic acid at a ratio of 1:4 w/w in organic solvents with high (dichloromethane) and low (ethyl acetate and chloroform) vapor pressures. Mean mobility diameter measured using a Scanning Mobility Particle Sizer was in the range of 123–132 nm with a unimodal distribution and a geometric standard deviation of 1.6–1.9. A layered particle structure was observed using transmission electron microscopy, which suggests partial drug enrichment in the surface layer. Higher drug loading (20% w/w) and uniform entrapment efficiencies (~100%) were achieved. The initial drug to lipid ratio (1:4 w/w) of the precursor solution was preserved in the synthesized lipid matrices. The crystallinity of the gefitinib-loaded lipid matrix was measured using X-ray diffraction and differential scanning calorimetry. In vitro drug release from gefitinib-loaded NALM in phosphate buffered saline (pH 7.2) over 10 days showed an initial fast release period followed by a prolonged sustained release period with varying release rates. Gefitinib-loaded NALM synthesized at higher evaporation rates exhibited lower degrees of crystallinity and faster drug releases. These results suggest the determinant role of lipid crystallinity manipulated by differing evaporation rates during aerosol synthesis on drug releases from nanometer-sized lipid matrices.

Rutter, a. P., Shakya, K. M., Lehr, R., Schauer, J. J., and Griffin, R. J., 2012, "Oxidation of gaseous elemental mercury in the presence of secondary organic aerosols," *Atmospheric Environment*, **59**: 86–92

Gaseous elemental mercury (GEM;  $\text{Hg}^0$  (g)) was oxidized by ozone and secondary hydroxyl radicals generated by the chemistry associated with the formation of secondary organic aerosols. The reaction was investigated in a 9-m<sup>3</sup> Teflon batch reactor. The losses of GEM in ozone-only experiments compared well with numerical model predictions based on published reaction rates, and a second order rate analysis gave a reaction rate of  $(7.4 \pm 0.5) \times 10^{-19} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ , which was statistically indistinct from recent publications. Furthermore, the net oxidation of GEM observed in the SOA reaction system agreed well with a numerical model based on the GEM-ozone reaction rate determined in this study and a published GEM-OH oxidation rate. Recent modeling studies of mercury atmospheric cycling have found that use of laboratory-based GEM-ozone reaction rate coefficients caused overestimation of GEM oxidation, while theoretical studies cast doubt over the viability of the GEM-ozone oxidation reaction in the real atmosphere. The results presented here suggest that the reaction is viable in the atmosphere and that recent published reaction rates for GEM and ozone are pertinent for use in atmospheric models. An average of GEM-ozone rates determined during this and recent studies was  $6.9 \pm 0.9 \times 10^{-19} \text{ cm}^3 \text{ molecules}^{-1} \text{ s}^{-1}$ . This value is recommended for use in future modeling studies.

## 2011

Abbas, Z., Holmberg, J. P., Hellström, A. K., Hagström, M., Bergenholtz, J., Hassellöv, M., and Ahlberg, E., 2011, "Synthesis, characterization and particle size distribution of TiO<sub>2</sub> colloidal nanoparticles," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, **384**(1-3): 254–261

Nanoparticles of controlled size, well defined shape, pure phase and of clean surfaces are ideal model systems to investigate surface/interfacial reactions. In this study we have explored the possibility of synthesizing TiO<sub>2</sub> nanoparticles in the size range of 7–20 nm under well controlled experimental conditions. A simple method based on the hydrolysis of TiCl<sub>4</sub> was used to obtain particles having surfaces free from organics. Stable dispersions of TiO<sub>2</sub> nanoparticles of various sizes were obtained by optimizing the reaction/dialysis time and temperature. The synthesized TiO<sub>2</sub> particles were found to be predominantly of anatase phase and narrow particle size distributions were obtained. The TiO<sub>2</sub> particles were characterized with respect to their phase, size and shape by X-ray diffraction (XRD) and transmission electron microscopy (TEM), respectively. Particle size distribution in a colloidal dispersion was obtained by the electrospray Scanning Mobility Particle Sizer (ES-SMPS) method and compared with an average particle size determined from dynamic light scattering (DLS). The average particle sizes obtained by the DLS and ES-SMPS methods were in good agreement, while a primary particle size of ~4 nm was found in X-ray diffraction irrespective of the particle size in solution. Early stages of the nucleation process were monitored by the ES-SMPS method. These results show that small particles of 4–5 nm are initially formed and it is highly likely that large particles are formed due to aggregation of primary particles.



Fang, W., Lei, G., Shan, X., Liu, F., Wang, Z., and Sheng, L., 2011, "A VUV photoionization organic aerosol mass spectrometric study with synchrotron radiation," *Journal of Electron Spectroscopy and Related Phenomena*, **184**(3-6): 129–133

A photoionization aerosol time-of-flight mass spectrometer (ATOFMS) has been developed for on-line analysis of organic compounds in aerosol particles using tunable vacuum ultraviolet (VUV) synchrotron radiation. Aerosol particles can be sampled directly from atmospheric pressure and are focused through an aerodynamic lens assembly into the mass spectrometer. The particles are vaporized when they impact on a heater, and then the nascent vapor is softly photoionized by synchrotron radiation. The degree of fragmentation of molecule can be controlled either by the heater temperature or by the photon energy. Thus, fragment-free tunable VUV mass spectra are obtained by tuning the photon energy close to the ionization energies (IEs) of the sample molecules. The direct determination of the IEs of benzophenone (9.07 eV), salicylic acid (8.72 eV), and urea (9.85 eV) are measured from the photoionization efficiency spectra with uncertainties of  $\pm 50$  meV. Ab initio calculations have been employed to predict the theoretical ionization energy.

Pawar, A. a., and Venkataraman, C., 2011, "Droplet-Phase Synthesis of Nanoparticle Aerosol Lipid Matrices with Controlled Properties," *Aerosol Science and Technology*, **45**(7): 811–820

Nanoparticle lipid matrices are being recently explored for controlled drug delivery applications. The degree of crystallinity of the lipid matrix governs drug loading capacity and release rates. Current methods of preparing nanoparticle lipid matrices, like high pressure homogenization and solvent-emulsification evaporation, offer limited or poor control over lipid crystallinity. The present work explores a droplet-phase aerosol synthesis method to prepare nanoparticle aerosol lipid matrices (NALM). Control of evaporation rate was used to achieve control over crystallinity of NALM. NALM were synthesized in an aerosol reactor using precursor solutions of stearic acid in organic solvents with widely different vapour pressures, resulting in different rates of evaporation. Mean mobility diameter, measured using a Scanning Mobility Particle Sizer (SMPS) spectrometer, ranged from 94–127 nm, with a unimodal distribution and geometric standard deviation of 1.7–1.9. Aerodynamic diameters measured using gravimetric analysis of samples collected in a micro-orifice cascade impactor, ranged from 423–608 nm. Relatively higher peak intensities and peak areas in X-ray diffractograms, and higher melting enthalpies in the differential scanning calorimetry (DSC) thermograms were obtained for NALM synthesized under lower evaporation rates, using lower vapour pressure solvents, similar to that of bulk stearic acid. In contrast, NALM synthesized at higher evaporation rates, using higher vapour pressure solvents consistently showed a lower degree of crystallinity, evidenced by blunt X-ray diffraction peaks and lower melting enthalpies in DSC analysis. These results suggest the formation of non-equilibrium crystal structures within NALM, under the influence of faster evaporation rates, leading to a lower degree of crystallinity. The work demonstrates proof of concept of droplet-phase aerosol synthesis for preparing nanoparticle lipid matrices and for achieving control over their crystallinity.

Rattanavaraha, W., Rosen, E., Zhang, H., Li, Q., Pantong, K., and Kamens, R. M., 2011, "The reactive oxidant potential of different types of aged atmospheric particles: An outdoor chamber study," *Atmospheric Environment*, **45**(23): 3848–3855

The reactive oxygen species (ROS) potential of aged diesel exhaust particulate matter (PM) and other aged aerosol systems in the presence and absence of an urban hydrocarbon environment was assessed. Experiments were performed in a 274m<sup>3</sup> dual outdoor Teflon film chamber. Filter samples were taken to assess the oxidant generation associated with PM by an optimized dithiothreitol (DTT) method. Diesel exhaust PM had a higher ROS response when it was in the presence of an urban hydrocarbon mixture and was associated with significant O<sub>3</sub> production. For all the aged dilute diesel systems, ROS expression increased by a factor of 2–4 over fresh diesel particles. Other particle systems were also investigated. A low ROS was observed in most of the nighttime experiments, including the nighttime aerosols from SO<sub>2</sub> with O<sub>3</sub> and SO<sub>2</sub> aged by itself. However, when all the systems were compared, aged diesel exhaust tended to express very high ROS potentials, with secondary organic aerosols from an  $\alpha$ -pinene + toluene + an urban HC mixture giving the highest ROS response.

Smith, M. L., Kuwata, M., and Martin, S. T., 2011, "Secondary Organic Material Produced by the Dark Ozonolysis of  $\alpha$ -Pinene Minimally Affects the Deliquescence and Efflorescence of Ammonium Sulfate," *Aerosol Science and Technology*, **45**(2): 244–261

The hygroscopic phase transitions and growth factors of mixed particles having as components ammonium sulfate and secondary organic material (SOM) were measured. The SOM was generated by the dark ozonolysis of  $\alpha$ -pinene, and organic particle mass concentrations of 1.63 and 12.2  $\mu\text{g m}^{-3}$  were studied. The hygroscopic properties were investigated using a 1 $\times$ 3 tandem differential mobility analyzer (1 $\times$ 3-TDMA). The 1 $\times$ 3-TDMA takes advantage of the hysteresis between solid-to-aqueous and aqueous-to-solid phase transitions to determine the efflorescence and deliquescence relative humidities (ERH and DRH, respectively) of materials. Overall, the influence of the SOM produced by the dark ozonolysis of  $\alpha$ -

pinene on the ERH and DRH of ammonium sulfate was small, shifting for example the DRH from 80% for pure ammonium sulfate to 77% for organic volume fractions of 0.96. The ERH likewise shifted by only a small amount across this composition range, specifically from 31 to 29%. The SOM produced at the lower organic particle mass concentrations shifted ERH and DRH even less, indicating an influence of SOM chemical composition on phase transitions. The hygroscopic growth factors of the mixed particles were adequately modeled across the range of studied RH (50 to 83%) using volume-averaged growth factors of the pure materials. The results for ERH, DRH, and the growth factors were all consistent with a model of phase separation between the inorganic and organic phases in individual particles, at least for the studied RH values (<83%) and for SOM prepared by  $\alpha$ -pinene ozonolysis.

## 2010

Chacon-Madrid, H. J., Presto, A. a, and Donahue, N. M., 2010, "Functionalization vs. fragmentation: n-aldehyde oxidation mechanisms and secondary organic aerosol formation.," *Physical chemistry chemical physics : PCCP*, **12**(42): 13975–82

Because of their relatively well-understood chemistry and atmospheric relevance, aldehydes represent a good model system for carbon-carbon fragmentation reactions in organic-aerosol aging mechanisms. Small aldehydes such as ethanal and propanal react with OH radicals under high NO<sub>x</sub> conditions to form formaldehyde and ethanal, respectively, with nearly unit yield. CO<sub>2</sub> is formed as a coproduct. This path implies the formation of the C<sub>n-1</sub> aldehyde, or an aldehyde with one fewer methylene group than the parent. However, as the carbon number of the n-aldehyde increases, reaction with the carbon backbone becomes more likely and the C<sub>n-1</sub> formation path becomes less important. In this work we oxidized n-pentanal, n-octanal, n-undecanal and n-tridecanal with OH radicals at high NO<sub>x</sub>. The C<sub>n-1</sub> aldehyde molar yields after the peroxy radical + NO reaction were 69 ± 15, 36 ± 10, 16 ± 5 and 4 ± 1%, respectively. Complementary structure-activity relationship calculations of important rate constants enable estimates of branching ratios between several intermediates of the C<sub>n</sub>-aldehyde reaction with OH: C<sub>n</sub> peroxyacyl nitrate versus C<sub>n</sub> alkoxyacyl radical formation, C<sub>n-1</sub> alkyl nitrate versus C<sub>n-1</sub> alkoxy radical, and C<sub>n-1</sub> aldehyde formation versus isomerization products. We also measured SOA mass yields, which we compare with analogous n-alkanes to understand the effect of fragmentation on organic-aerosol formation.

Han, B., Kim, H. J., and Kim, Y. J., 2010, "Fine particle collection of an electrostatic precipitator in CO<sub>2</sub>-rich gas conditions for oxy-fuel combustion.," *The Science of the total environment*, **408**(21): 5158–64

The collection of particles in CO<sub>2</sub>-enriched environments has long been important for the capture of CO<sub>2</sub> in order to clean gases via oxy-fuel combustion. We here report on the collection characteristics of fine and ultrafine particles using an electrostatic precipitator (ESP) in a CO<sub>2</sub>-enriched atmosphere. In order to understand the characteristics of particle collection in CO<sub>2</sub>-rich gas mixtures, the ionic properties of a CO<sub>2</sub>-enriched atmosphere was also investigated. The electrical mobility of the ions in a CO<sub>2</sub>-enriched atmosphere was found to be about 0.56 times that found in a conventional air atmosphere, due to the higher mass of CO<sub>2</sub> gas compared to that of air. The low electrical mobility of ions resulted in a low corona current under CO<sub>2</sub>-enriched conditions. The collection efficiency of particles in a CO<sub>2</sub>-rich atmosphere for a given power consumption was thus somewhat lower than that found in air, due to the low quantity of particle charging in CO<sub>2</sub>-enriched air. At the same time, higher temperatures led to the higher electrical mobility of ions, which resulted in a greater collection efficiency for a given power. The presence of a negative corona also led to a greater collection efficiency of particles in an ESP than that achieved for a positive corona.

Liu, C., Liu, Y., Ma, Q., and He, H., 2010, "Mesoporous transition alumina with uniform pore structure synthesized by alumisol spray pyrolysis," *Chemical Engineering Journal*, **163**(1-2): 133–142

Mesoporous transition alumina was prepared by spray pyrolysis of an alumisol precursor, without the use of structure-directing reagents. The prepared alumina exhibited a well-defined mesoporous structure with narrow pore size distribution (3.0–4.7 nm). Preparation temperature played a major role in the phase formation and the porosity of prepared alumina. The phase transformation of boehmite to gamma-Al<sub>2</sub>O<sub>3</sub> and theta-Al<sub>2</sub>O<sub>3</sub> was observed at 800 and 1200°C, respectively. An increase in preparation temperature resulted in a decrease in surface area and pore volume and an increase in pore size. Longer residence time promoted the crystallinity of prepared alumina effectively, but it also led to the formation of particles with smaller surface area, lower pore volume, larger pore size, and broader pore size distribution. Additionally, while alumisol concentration had little effect on the surface area, its reduction led to a decrease in pore volume and pore size and caused the formation of bimodal pore size distribution. The as-prepared alumina also exhibited excellent thermal stability, showing great application potential for industry.

Zelenyuk, A., Ezell, M. J., Perraud, V., Johnson, S. N., Bruns, E. a., Yu, Y., Imre, D., Alexander, M. L., and Finlayson-Pitts, B. J., 2010, "Characterization of organic coatings on hygroscopic salt particles and their atmospheric impacts," *Atmospheric Environment*, **44**(9): 1209–1218

The photooxidation of  $\alpha$ -pinene in the presence of NO<sub>2</sub>, with and without added NaNO<sub>3</sub> seed particles, has been studied in a large-diameter flow tube. Particles formed by homogeneous nucleation and by condensation on the pre-existing seeds were sampled at various stages of the reaction, dried using four diffusion dryers, size selected at different mobility diameters (dm) using a differential mobility analyzer (DMA), and characterized with a single particle mass spectrometer (SPLAT II). It was found that homogeneously nucleated particles are spherical, have a density ( $\rho$ ) of  $1.25 \pm 0.02 \text{ g cm}^{-3}$  ( $\pm 2\sigma$ ) and contain a significant amount of organic nitrates. The mass spectra of the low volatility products condensed on the NaNO<sub>3</sub> seed particles were found to be virtually the same as in the case of homogeneous nucleation. The data show that the presence of even a submonolayer of organics on the NaNO<sub>3</sub> particles causes water retention that leads to a decrease in particle density and that the amount of water retained increases with organic coating thickness. Thicker coatings appear to inhibit water evaporation from the particle seeds altogether. This suggests that in the atmosphere, where low volatility organics are plentiful, some hygroscopic salts will retain water and have different densities and refractive indices than expected in the absence of the organic coating. This water retention combined with the organic shell on the particles can potentially impact light scattering by these particles and activity as cloud condensation nuclei (CCN), as well as heterogeneous chemistry and photochemistry on the particles.

## 2009

Kulkarni, P., Deye, G. J., and Baron, P. a., 2009, "Bipolar diffusion charging characteristics of single-wall carbon nanotube aerosol particles," *Journal of Aerosol Science*, **40**(2): 164–179

Bipolar diffusion charging characteristics of airborne single-wall carbon nanotube (SWCNT) agglomerates were investigated in the mobility diameter range of 100–1000nm. Neutral fractions of three types of SWCNT aerosols following bipolar charge equilibrium in a radioactive source were experimentally measured to infer their electrical charging characteristics. Significant deviation from Boltzmann and Fuchs stationary charge equilibrium was observed, with neutral fractions of SWCNT particles lower by 30–53% compared to that of spherical particles of the same mobility. Particles with mobility diameter larger than 400nm showed high electrical charging efficiencies compared to that of mobility-equivalent spherical particles. Higher charging efficiencies of SWCNT particles were attributed to their higher electrical capacitance resulting from complex nonspherical morphologies. Numerical calculations using idealized fiber geometries confirmed the qualitative trend in the experimental data. The electrical capacitance of nanotubes particles deduced from experimentally measured neutral fractions were also found to be higher by a factor ranging from 1.6 to 4.6 compared to that of mobility-equivalent spherical particles, indicating high charge carrying capacity. The charging-equivalent diameters of nanotube particles were computed and were found to be higher than their mobility diameter by a factor of 2.85–4.34.

Signorell, R., and Bertram, A., 2009, "Physical chemistry of aerosols.," *Physical chemistry chemical physics : PCCP*, **11**(36): 7759

Here we report a new method for measuring the heterogeneous chemistry of sub-micron organic aerosol particles using a continuous flow stirred tank reactor. This approach is designed to quantify the real time heterogeneous kinetics, using a relative rate method, under conditions of low oxidant concentration and long reaction times that more closely mimic the real atmosphere than the conditions used in a typical flow tube reactor. A general analytical expression, which couples the aerosol chemistry with the flow dynamics in the chamber is developed and applied to the heterogeneous oxidation of squalane particles by hydroxyl radicals (OH) in the presence of O<sub>2</sub>. The particle phase reaction is monitored via photoionization aerosol mass spectrometry and yields a reactive uptake coefficient of  $0.51 \pm 0.10$ , using OH concentrations of  $1\text{--}7 \times 10^8 \text{ molecule cm}^{-3}$  and reaction times of 1.5–3 h. In general, this approach provides a new way to connect the chemical aging of organic particles measured at short reaction times and high oxidant concentrations in flow tubes with the long reaction times and low oxidant conditions in smog chambers and the real atmosphere.

Winterhalter, R., Herrmann, F., Kanawati, B., Nguyen, T. L., Peeters, J., Vereecken, L., and Moortgat, G. K., 2009, "The gas-phase ozonolysis of beta-caryophyllene (C(15)H(24)). Part I: an experimental study.," *Physical chemistry chemical physics : PCCP*, **11**(21): 4152–72

The gas phase reaction of ozone with beta-caryophyllene was investigated in a static glass reactor at 750 Torr and 296 K under various experimental conditions. The reactants and gas phase products were monitored by FTIR-spectroscopy and proton-transfer-reaction mass spectrometry (PTR-MS). Aerosol formation was monitored with a Scanning Mobility Particle Sizer (SMPS) spectrometer and particulate

products analysed by liquid chromatography/mass spectrometry (HPLC-MS). The different reactivity of the two double bonds in beta-caryophyllene was probed by experiments with different ratios of reactants. An average rate coefficient at 295 K for the first-generation products was determined as  $1.1 \times 10^{-16} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ . Using cyclohexane as scavenger, an OH-radical yield of  $(10.4 \pm 2.3)\%$  was determined for the ozonolysis of the more reactive internal double bond, whereas the average OH-radical yield for the ozonolysis of the first-generation products was found to be  $(16.4 \pm 3.6)\%$ . Measured gas phase products are CO, CO<sub>2</sub> and HCHO with average yields of  $(2.0 \pm 1.8)\%$ ,  $(3.8 \pm 2.8)\%$  and  $(7.7 \pm 4.0)\%$ , respectively for the more reactive internal double bond and  $(5.5 \pm 4.8)\%$ ,  $(8.2 \pm 2.8)\%$  and  $(60 \pm 6)\%$ , respectively from ozonolysis of the less reactive double bond of the first-generation products. The residual FTIR spectra indicate the formation of an internal secondary ozonide of beta-caryophyllene. From experiments using HCOOH as a Criegee intermediate (CI) scavenger, it was concluded that at least 60% of the formed CI are collisionally stabilized. The aerosol yield in the ozonolysis of beta-caryophyllene was estimated from the measured particle size distributions. In the absence of a CI scavenger the yield ranged between 5 and 24%, depending on the aerosol mass. The yield increases with addition of water vapour or with higher concentrations of formic acid. In the presence of HCHO, lower aerosol yields were observed. This suggests that HCOOH adds to a Criegee intermediate to form a low-volatility compound responsible for aerosol formation. The underlying reaction mechanisms are discussed and compared with the results from the accompanying theoretical paper.

## 2008

Fukuhara, N., Suzuki, K., Takeda, K., and Nihei, Y., 2008, "Characterization of environmental nanoparticles," *Applied Surface Science*, **255**(4): 1538–1540

In this study, we attempted to characterize environmental nanoparticles and particle diameter distributions in the atmospheric environment neighboring a traffic route by using a Scanning Mobility Particle Sizer (SMPS) spectrometer. The composition of the environmental nanoparticles was analyzed using time-of-flight secondary ion mass spectrometry (TOF-SIMS). It was observed that the environmental nanoparticles showed peaks at 20 nm and 100 nm. The secondary ions C<sup>+</sup>, O<sup>+</sup>, Si<sup>+</sup>, and SiH<sub>3</sub><sup>+</sup> were strongly detected in the environmental nanoparticles with a peak at 20 nm. On the other hand, the secondary ions NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, K<sup>+</sup>, and Ca<sup>+</sup> were detected in the environmental nanoparticles with a peak at 100 nm. Moreover, it was found that the secondary ion spectral patterns of the organic compounds were different for each particle diameter. Hence, we concluded that the combination of the SMPS with TOF-SIMS is a powerful technique to characterize environmental nanoparticles.

Han, B., Hudda, N., Ning, Z., and Sioutas, C., 2008, "Enhanced unipolar charging of concentration-enriched particles using water-based condensational growth," *Journal of Aerosol Science*, **39**(9): 770–784

A novel methodology with high efficiency for charging fine and ultrafine particles was developed and evaluated. The technique has been realized by combining the versatile aerosol concentration enrichment system (VASES) developed by our group and a newly developed unipolar charger equipped with carbon fiber ionizers. Particles are grown to super-micron droplets via condensation of ultrapure deionized water and then concentrated by virtual impaction in the VACES. The grown droplets are charged with negligible ozone generation in the carbon fiber unipolar charger, and subsequently dried to original particle size distribution using a diffusion dryer, while preserving the acquired charges. This new methodology was investigated for different particle sizes, chemical compositions and concentrations. The number of charges on particles was highly dependent on particle concentration as well as particle size; larger particles and smaller particle concentrations led to a greater number of charges per particle. The average electrical mobility of the charged particles exceeded  $1.4 \times 10^{-1} \text{ cm}^2/(\text{Vs})$ , corresponding, on average, to more than 250 elementary charges per particle for concentrations in the range of  $1.4\text{--}1.9 \times 10^5 \text{ particles/cm}^3$  for ammonium nitrate, ammonium sulfate and glutaric acid. A lower number of charges per particle was observed for PSL particles, probably due to their hydrophobic nature, and thus, number of charges per particle averaged to about 98, 191 and 349 for polystyrene latex (PSL) particles of 99, 130 and 170 nm, respectively, at particle concentrations of  $1.1\text{--}1.7 \times 10^5 \text{ particles/cm}^3$ . Even in the case of PSL, these charges far-exceed those obtained by traditional corona chargers.

Lu, Z., Hao, J., Hu, L., and Takekawa, H., 2008, "The compaction of soot particles generated by spark discharge in the propene ozonolysis system," *Journal of Aerosol Science*, **39**(10): 897–903

The compaction of soot aggregates generated by spark discharge is reported in the propene ozonolysis experiment. Since propene is not an organic aerosol precursor, the compaction cannot be explained by an earlier hypothesis attributing the compaction to the capillary forces of condensed organic matter in the small angle cavities of the aggregates [Saathoff, Naumann et al. (2003). Coating of soot and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> particles by ozonolysis products of alpha-pinene. *Journal of Aerosol Science*, **34**, 1297–1321]. Several species among the gas-phase products from propene ozonolysis are proposed to contribute to the structural change of soot particles. As the compaction would directly change themorphology and the size

distribution of soot particles, it is expected to induce important changes to the soot particles' physical and chemical properties, which in turn would have significant atmospheric implications.

Paganini, E., Mariotti, G., Gasperetti, S., Vallebona, C., Predolin, L., Muré, E., Palleschi, V., Salvetti, A., and Tognoni, E., 2008, "Multi-diagnostic approach to characterize the onset of formation of nanoparticles in a premixed laminar ethylene/air flame," *Spectrochimica Acta Part B: Atomic Spectroscopy*, **63**(2): 191–201

The onset of formation of nanoparticles in a premixed laminar ethylene/air flame was studied using optical and non-optical diagnostic techniques. The optical techniques (Visible Imaging, Spectroscopic analysis of optical emission, Laser Induced Incandescence and Optical Extinction) were checked against the direct measurement of particles size distribution done with a commercial sampler. The diagnostic techniques were set up on a standard burner (McKenna). The combustion conditions were varied by tuning two experimental parameters: the equivalent ratio  $\Phi$  and the cold gases velocity  $v$ . Values of  $\Phi$  were investigated in the range 1.3 to 2.2, in combination with cold unburned gas velocities of 5, 7 and 10 cm/s. By varying the combustion conditions, a transition in the signal detected by the different techniques was observed. The transition was put in correspondence with the sensitivity of the given technique to the onset of the nanoparticles formation. A comparison of the sensitivity of the different techniques was performed. The different sensitivities of the diagnostics and the different information that they provide confirmed the need for a multi-diagnostics setup for the characterization of nanoparticle production during the combustion process.

## 2006

Destailats H.; Lunden M.M.; Singer B.C.; Coleman B.K.; Hodgson A.T.; Weschler C.J.; Nazaroff W.W., 2006, "Indoor secondary pollutants from household product emissions in the presence of ozone: A bench-scale chamber study," *Environmental Science and Technology*, **40**(14): 4421–4428

Ozone-driven chemistry is a source of indoor secondary pollutants of potential health concern. This study investigates secondary air pollutants formed from reactions between constituents of household products and ozone. Gas-phase product emissions were introduced along with ozone at constant rates into a 198-L Teflon-lined reaction chamber. Gas-phase concentrations of reactive terpenoids and oxidation products were measured. Formaldehyde was a predominant oxidation byproduct for the three studied products, with yields for most conditions of 20-30% with respect to ozone consumed. Acetaldehyde, acetone, glycolaldehyde, formic acid, and acetic acid were each also detected for two or three of the products. Immediately upon mixing of reactants, a Scanning Mobility Particle Sizer detected particle nucleation events that were followed by a significant degree of secondary particle growth. The production of secondary gaseous pollutants and particles depended primarily on the ozone level and was influenced by other parameters such as the air-exchange rate. Hydroxyl radical concentrations in the range  $0.04\text{--}200 \times 10^5$  molecules/cm<sup>3</sup> were determined by an indirect method. OH concentrations were observed to vary strongly with residual ozone level in the chamber, which was in the range 1-25 ppb, as is consistent with expectations from a simplified kinetic model. In a separate chamber study, we exposed the dry residue of two products to ozone and observed the formation of gas-phase and particle-phase secondary oxidation products.

Dodds, Scott M.; Dreyfus, Matthew A.; Dykins, John; Johnston, Murray V.; Tolocka, Michael P., 2005, "Cholesterol ozonolysis: Kinetics, mechanism and oligomer products," *Journal of Physical Chemistry A*, **109**(28): 6242–6248

Fine particles of cholesterol were reacted with ozone under pseudo-first-order conditions in an aerosol bag reactor. Gas-phase ozone was monitored using an ozone meter. Particle size distribution functions were determined using a Scanning Mobility Particle Sizer, which selected particle sizes for introduction into a photoionization aerosol mass spectrometer (PIAMS). PIAMS was used to determine the concentration of cholesterol in the aerosol as a function of reaction time. Dilution corrected rate coefficients were used to calculate the reactive uptake coefficient for ozone onto cholesterol particles as  $(2.8 \pm 0.4) \times 10^6$ . Uptake was found to be independent of particle diameter for the sizes studied (100 and 200 nm), suggesting that the uptake is surface mediated. The reaction products were also collected on filters and analyzed by electrospray ionization (ESI) mass spectrometry with both direct infusion and liquid chromatography sample introduction. The main primary reaction products contained one, two, or three oxygens added to the cholesterol moiety. Secondary oligomeric products were also observed, consisting of covalently bound dimers and trimers. Tandem mass spectrometry was used to confirm the expected structures of these compounds. The dimers appear to be acyl hydroperoxides, consistent with a previously reported mechanism for the reaction in a nonparticipating solvent. Finally, the magnitude of the uptake coefficient confirms that cholesterol is suitable as a local source tracer for source apportionment of ambient organic aerosol. 46 Refs.

Dreyfus, Matthew A.; Heaton, Katherine J.; Johnston, Murray V.; Saul, Thomas D.; Tolocka, Michael P.; Wang, Shenyi; Zordan, Christopher A., 2006, "Chemistry of particle inception and growth during alpha-pinene ozonolysis," *Environmental Science and Technology*, **40**(6): 1843–1848

A flow-tube reactor was used to study the formation of particles from alpha-pinene ozonation. Particle phase products formed within the first 3–22 s of reaction were analyzed online using a Scanning Mobility Particle Sizer and two particle mass spectrometers. The first, a photoionization aerosol mass spectrometer (PIAMS), was used to determine the molecular composition of nascent particles between 30 and 50 nm in diameter. The second, a nano-aerosol mass spectrometer (NAMS), was used to determine the elemental composition of individual particles from 50 nm to below 10 nm in diameter. Molecular composition measurements with PIAMS confirm that both the stabilized Criegee intermediate and hydroperoxide channels of alpha-pinene ozonolysis are operative. However, these channels alone cannot explain the high oxygen content of the particles measured with NAMS. The carbon-to-oxygen mole ratios of suspected nucleating agents are in the range of 2.25–4.0, while the measured ratios are from 1.9 for 9 nm particles to 2.5 and 2.7 for 30 and 50 nm particles, respectively. The large oxygen content may arise by co-condensation of small oxygenated molecules such as water or multistep reactions with ozone, water, or other species that produce highly oxygenated macromolecules. In either case, the increasing ratio with increasing particle size suggests that the aerosol becomes less polar with time.

Epstein, Hila; Afergan, Eyal; Moise, Tamar; Richter, Yoram; Rudich, Yinon; Golomb, Gershon, 2006, "Number-concentration of nanoparticles in liposomal and polymeric multiparticulate preparations: Empirical and calculation methods," *Biomaterials*, **27**(4):651–659

The actual number of particles in formulations of nanoparticles (NP) is of importance for quality assurance, comprehensive physicochemical characterization, and pharmacodynamics. Some calculation methods that have been previously employed are limited because they rely on several assumptions and are not applicable for certain preparations. Currently there are no validated experimental methods for determining the particle number-concentration ( $N_c$ ) of liposomal and polymeric nanoparticulate preparations (<500 nm). This study examines a new empirical method for counting the number of particles in nanoparticulate formulations including drug-containing liposomes and polymeric NP. In the new method, suspended NP are nebulized to form aerosol droplets which are dried and counted using a Scanning Mobility Particle Sizer (SMPS) spectrometer. Experiments were conducted with three different preparations, empty liposomes (200 and 400 nm), drug-loaded liposomes (200 nm), and polymeric NP (150 nm). It was verified that no detrimental morphological or structural changes of the formulations have been induced by the SMPS technique, and that the obtained  $N_c$  values represent the original particles. It is concluded that nano-formulations with concentrations of up to  $10^7$  particles per  $1\text{ cm}^3$  air, corresponding to approximately  $10^{12}$  particles per 1 ml solution, can be directly counted within the size range of 30–900 nm. The measured values are compared to newly developed theoretical calculations to assess the viability of these calculations.

Abbey, Erin D.; Crofton, Mark W.; Kalitan, Danielle M.; Petersen, Eric L.; Rickard, Matthew J.A.; Traum, Matthew J., 2005, "A facility for gas- and condensed-phase measurements behind shock waves," *Measurement Science and Technology*, **16**(9) 1716–1729

A shock-tube facility consisting of two, single-pulse shock tubes for the study of fundamental processes related to gas-phase chemical kinetics and the formation and reaction of solid and liquid aerosols at elevated temperatures is described. Recent upgrades and additions include a new high-vacuum system, a new gas-handling system, a new control system and electronics, an optimized velocity-detection scheme, a computer-based data acquisition system, several optical diagnostics, and new techniques and procedures for handling experiments involving gas/powder mixtures. Test times on the order of 3 ms are possible with reflected-shock pressures up to 100 atm and temperatures greater than 4000 K. Applications for the shock-tube facility include the study of ignition delay times of fuel/oxidizer mixtures, the measurement of chemical kinetic reaction rates, the study of fundamental particle formation from the gas phase, and solid-particle vaporization, among others. The diagnostic techniques include standard differential laser absorption, FM laser absorption spectroscopy, laser extinction for particle volume fraction and size, temporally and spectrally resolved emission from gas-phase species, and a Scanning Mobility Particle Sizer for particle size distributions. Details on the set-up and operation of the shock tube and diagnostics are given, the results of a detailed uncertainty analysis on the accuracy of the test temperature inferred from the incident-shock velocity are provided, and some recent results are presented. 37 Refs.

Choi, Jong Hyun; Koshland, Catherine P.; Lucas, Donald; Sawyer, Robert F., 2005, "Photochemical interaction of polystyrene nanospheres with 193 nm pulsed laser light," *Journal of Physical Chemistry B*, **109**(50):23905-23910

The photochemical interaction of 193 nm light with polystyrene nanospheres is used to produce particles with a controlled size and morphology. Laser fluences from 0 to 0.14 J/cm<sup>2</sup> at 10 and 50 Hz photofragment nearly monodisperse 110 nm spherical polystyrene particles. The size distributions before and after irradiation are measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer, and the morphology of the irradiated particles is examined with a transmission electron microscope (TEM). The results show that the irradiated particles have a smaller mean diameter (similar to 25 nm) and a number concentration more than an order of magnitude higher than nonirradiated particles. The particles are formed by nucleation of gas-phase species produced by photolytic decomposition of nanospheres. A nondimensional parameter, the photon-to-atom ratio (PAR), is used to interpret the laser-particle interaction energetics. 39 Refs.

Choi, Jong Hyun; Koshland, Catherine P.; Lucas, Donald; Sawyer, Robert F.; Stipe, Christopher B., 2005, "NaCl particle interaction with 193-nm light: Ultraviolet photofragmentation and nanoparticle production," *Journal of Applied Physics*, **97**(12):1–9

The interaction of nanoscale NaCl particles with 193-nm photons is studied to better understand particle disintegration and production by ultraviolet photofragmentation. The particles are irradiated in a constrained air stream with laser fluences from 0.08 to 0.23 J cm<sup>2</sup> with single and multiple pulses striking the particles. The resulting particle size distributions are measured with a Scanning Mobility Particle Sizer and the morphology is analyzed qualitatively by scanning electron microscopy (SEM). Photofragmentation of NaCl particles at 193 nm produces gas-phase species as well as small solid-phase fragments without significantly heating the particles or creating a plasma. The irradiated particles have a mean diameter from 20 to 55 nm (depending on the photon energy) and a number concentration an order of magnitude higher than the 118 nm mean diameter nonirradiated particles. The SEM images before and after 193-nm irradiation reveal that the irradiated particles are less fractal and more spherical. 52 Refs.

Czoschke, N. M.; Jang, M.; Northcross, A. L., 2005, "Semi empirical model for organic aerosol growth by acid-catalyzed heterogeneous of organic carbonyls," *Environmental Science and Technology*, **39**(1):164–174

Aerosol growth by heterogeneous reactions of diverse carbonyls in the presence and absence of acidified seed aerosols was studied in a 4 m long flow reactor (2.5 cm i.d.) and a 2-m<sup>3</sup> indoor Teflon film chamber under darkness. The acid catalytic effects on heterogeneous aerosol production were observed for diverse carbonyls in various ranges of humidities and compositions of seed inorganic aerosols. Particle population data measured by a Scanning Mobility Particle Sizer were used to calculate organic aerosol growth. To account for the aerosol growth contributed by heterogeneous reactions, the increase in organic aerosol mass was normalized by the organic mass predicted by partitioning or the square of predicted organic mass. The carbonyl heterogeneous reactions were accelerated in the presence of acid catalysts (H<sub>2</sub>SO<sub>4</sub>), leading to higher aerosol yields than in their absence. The experimental data from aerosol yields in the flow reactor were semiempirically fitted to the model parameters to predict the

organic aerosol growth. The model parameters consist of environmental characteristics and molecular structure information of organic carbonyls. Basicity constants of carbonyls were used to describe the proton affinity of carbonyls for the acid catalysts. Particle environmental factors, such as humidity, temperature, and inorganic seed composition, were expressed by excess acidity and the parameters obtained from an inorganic thermodynamic model. A stepwise regression analysis of the aerosol growth model for the experimental data revealed that either the chemical structure information of carbonyls or characteristic environmental parameters are statistically significant in the prediction of organic aerosol growth. It was concluded that this model approach is applicable to predict secondary organic aerosol formation by heterogeneous reaction.

Stewart, K. D.; Andino, J. M., 2005, "Studies of the uptake of gaseous ethyl-3-ethoxy propionate onto ammonium sulfate and ammonium nitrate aerosol particles," *Thirteenth International Conference on Modeling, Monitoring and Management of Air Pollution*, Cordoba (Spain), 16–18 May 2005, 519–528

Understanding the heterogeneous (gas-particle) interactions that occur in the troposphere is of seminal importance for modelers that assess the atmospheric fates and transport of gases. One means of understanding these interactions is to calculate the mass uptake coefficient of a gaseous species onto common anthropogenic aerosols via an experimental means. The effective uptake behavior of gaseous Ethyl-3-Ethoxy Propionate (E3EP) onto ammonium sulfate and ammonium nitrate aerosols of varying total surface areas was studied using a custom manufactured flow system. E3EP is an organic species that is commonly found in paint and paint products. Ammonium sulfate and ammonium nitrate aerosols were studied because of their abundance in the troposphere. Gas phase E3EP concentrations were obtained using a solid phase microextraction technique (SPME) coupled to a gas chromatograph with a flame ionization detector (GC-FID). Analyses of the ammonium sulfate and ammonium nitrate aerosols were carried out using a Scanning Mobility Particle Sizing system (SMPS) spectrometer. As expected, losses of gaseous E3EP onto the aerosol surfaces decreased with decreasing initial surface area. The calculated initial effective uptake coefficients were shown to vary, depending on aerosol composition. Due to the lack of published literature data regarding the heterogeneous interactions of organic species in the troposphere, these studies provide a significant enhancement to the overall knowledge base.

## 2004

Chan, W. R.; Khlystov, A. Y.; Mandiro, M.; Pandis, S. N.; Stanier, C. O., 2004, "A method for the in situ measurement of fine aerosol water content of ambient aerosols: The dry-ambient aerosol size spectrometer (DAASS)," *Aerosol Science & Technology*, **38**(S1):215–228

Hygroscopic growth of atmospheric particles affects a number of environmentally important aerosol properties. Due to the hysteresis exhibited by the aerosol hygroscopic growth, the physical state of particles and the amount of aerosol water are uncertain within a wide range of relative humidities (RHs) found in the troposphere, leading to uncertainties in optical and chemical properties of the aerosol. Here we report the design and tests of an automated system that was built to assess the amount of aerosol water at atmospheric conditions. The system consists of two Scanning Mobility Particle Sizers™ (SMPS™) and an aerodynamic particle (APS) that measure the aerosol size distribution between 3 nm and 10 μm in diameter. The inlets of the instruments and their sheath air lines are equipped with computer-controlled valves that direct air through Nafion dryers or bypass them. The Nafion dryers dehydrate the air streams to below 30% RH at which point ambient particles are expected to lose most or all water. The switch between the dried and the ambient conditions occurs every 7 min and is synchronized with the scan times of the aerosol spectrometers. In this way the system measures alternatively dried (below 30% RH) and ambient aerosol size distributions. A comparison of the ambient RH and the dried RH size distributions and the corresponding integrated volume concentrations provides a measure of the physical state of particles and the amount of aerosol water. The aerosol water content can be treated as a growth factor or as an absolute quantity and can be calculated as a time series or as a function of RH (humidigram). When combined with aerosol composition measurements, the DAASS can be used to compare hygroscopic growth models and measurements. 48 Refs.

Hermesdorf, D.; Bonnamy, A.; Suhm, M. A.; Signorell, R., 2004, "Infrared spectra of phenanthrene particles generated by pulsed rapid expansion of CO<sub>2</sub> solutions," *Physical Chemistry Chemical Physics*, **6**(19): 4652–4657

A new apparatus for the pulsed rapid expansion of supercritical solutions (RESS) is presented. Phenanthrene particles with diameters of 240 nm were generated by expanding dilute solutions of phenanthrene in CO<sub>2</sub>. The number size distribution and the shape of the particles were analyzed off-line with scanning electron microscopy and on-line with a Scanning Mobility Particle Sizer and extinction measurements. From these investigations and from infrared spectra of the aerosol particles, refractive index data for phenanthrene were determined in the mid-infrared region of the spectrum.



Heszler, P.; Landstrom, L.; Marton, Z., 2004, "Early stage of the material removal during ArF laser ablation of graphite," *Applied Physics A: Materials Science and Processing*, **79**(3):579–585

Material removal during ArF excimer laser ablation of graphite at atmospheric pressure was investigated by two independent methods; 1) by observation of the propagating properties of the shock wave generated by the carbonaceous ejecta and 2) by in situ measurement of the size distribution of carbon nanoparticles condensing in the ablation plume. This latter was carried out by a Scanning Mobility Particle Sizer system based on a differential mobility analyzer. The performed measurements indicate that the material removal during ArF laser ablation consists of two steps at fluences above the threshold fluence. First, a thin layer of carbon (of the order of 1 nm) is removed by a quick desorption process, leading to shockwave formation. This process takes place in a ns time scale, and desorption rate estimations reveal that this cannot be explained by thermal surface evaporation. Since to our knowledge there is no thermal process that could account for the estimated desorption rate, it is argued that this is a fast photochemical (i.e., non-thermal) process. The size distribution of the condensed nanoparticles related to this step shows a rising edge at diameters below 10 nm. At fluences above the ablation threshold, the majority of the material is ejected in the second phase, resulting in condensation of carbon nanoparticles, peaking at similar to 50 nm diameters in the size spectrum. Both shockwave formation and material removal are also detected well below the ablation threshold fluence, which is attributed to the photochemical process. 39 Refs.

Ludwig, Chr.; Mohr, M., 2004, "On-Line Characterization of Aerosols Formed in a Jet Flow Condenser for Analytical Applications," *Paul Scherrer Institut Scientific Report 2003, Volume V*

With a jet flow condenser (JFC) aerosol particles can be generated from gases containing high boiling substances. This can be used for transferring these substances from the hot carrier gas into an analytical device. In this study we have investigated the particle number size distribution (NSD) produced in two JFCs with different geometry. Experiments were performed with elemental zinc as a model substance. Zinc has been volatilized in a tubular furnace and the particles generated in the JFC were characterized on-line by a Scanning Mobility Particle Sizer (SMPS) spectrometer.

Virtanen A.; Ristimäki J.; Keskinen J., 2004, "Method for Measuring Effective Density and Fractal Dimension of Aerosol Agglomerates," *Aerosol Science and Technology*, **38**(5):437–446

A method to find particle effective density and the fractal dimension, based on simultaneous size distribution measurements with SMPS and ELPI, is introduced. A fitting procedure is used to find the particle density as a function of particle size and the fractal dimension. The method was tested by simulation and by experimental measurements of particles with varying morphology. For fractal dimension values between 2.2 and 3.0, fractal dimension was measured with an accuracy of 0.1, and effective density was measured with 15% relative accuracy.

## 2003

Carroll, B.; Chandramouli, B.; Jang, M.; Kamens, R. M., 2003, "Particle growth by acid-catalyzed heterogeneous reactions of organic carbonyls on preexisting aerosols," *Environmental Science and Technology*, **37**(17):3828-3837

Aerosol growth by the heterogeneous reactions of different aliphatic and alpha, beta-unsaturated carbonyls in the presence/absence of acidified seed aerosols was studied in a 2 m long flow reactor (2.5 cm i.d.) and a 0.5-m<sup>3</sup> Teflon film bag under darkness. For the flow reactor experiments, 2,4-hexadienal, 5-methyl-3-hexen-2-one, 2-cyclohexenone, 3-methyl-2-cyclopentenone, 3-methyl-2-cyclohexenone, and octanal were studied. The carbonyls were selected based on their reactivity for acid-catalyzed reactions, their proton affinity, and their similarity to the ring-opening products from the atmospheric oxidation of aromatics. To facilitate acid-catalyzed heterogeneous hemiacetal/acetal formation, glycerol was injected along with inorganic seed aerosols into the flow reactor system. Carbonyl heterogeneous reactions were accelerated in the presence of acid catalysts (H<sub>2</sub>SO<sub>4</sub>), leading to higher aerosol yields than in their absence. Aldehydes were more reactive than ketones for acid-catalyzed reactions. The conjugated functionality also resulted in higher organic aerosol yields than saturated aliphatic carbonyls because conjugation with the olefinic bond increases the basicity of the carbonyl leading to increased stability of the protonated carbonyl. Aerosol population was measured from a series of sampling ports along the length of the flow reactor using a Scanning Mobility Particle Sizer spectrometer. Fourier transform infrared spectrometry of either an impacted liquid aerosol layer or direct reaction of carbonyls as a thin liquid layer on a zinc selenide FTIR disk was employed to demonstrate the direct transformation of chemical functional groups via the acid-catalyzed reactions. These results strongly indicate that atmospheric multifunctional organic carbonyls, which are created by atmospheric photooxidation reactions, can contribute significantly to secondary organic aerosol formation through acid-catalyzed heterogeneous reactions. Exploratory studies in 25- and 190-m<sup>3</sup> outdoor chambers were also implemented to demonstrate the formation of high molecular weight organic structures. The reaction of

ozone with alpha-pinene to generate secondary organic aerosols (SOAs) was performed in the presence of background aerosol consisting of a mixture of wood soot and diesel soot. Results strongly suggest that indigenous sulfuric acid associated with the combustion of fossil fuels (e.g., diesel soot) can initiate acid-catalyzed heterogeneous reactions of SOAs on the particle phase.

Docherty, K. S.; Ziemann, P. J., 2003, "Effects of Stabilized Criegee Intermediate and OH Radical Scavengers on Aerosol Formation from Reactions of  $\beta$ -Pinene with O<sub>3</sub>," *Aerosol Science & Technology*, **37**(11):877–891

The formation of secondary organic aerosol (SOA) from reactions of O<sub>3</sub> with  $\beta$ -pinene, an exocyclic monoterpene prominent in the ambient atmosphere, was studied in an environmental chamber using a thermal desorption particle beam mass spectrometer for chemical analysis and a Scanning Mobility Particle Sizer for aerosol yield measurements. Potential reaction pathways for SOA formation were investigated in a series of experiments conducted using various scavengers for stabilized Criegee intermediates (SCI) and OH radicals, both of which are formed in the reaction. The major particulate products were compounds less volatile than pinic acid, a low-volatility dicarboxylic acid that was identified but was a minor component of the aerosol. The aerosol mass spectrum and yield were relatively insensitive to the identity of the SCI scavenger, indicating that association reactions of scavengers with SCI were not important in SOA formation. The mass spectrum of the aerosol also did not depend on the identity of the OH radical scavenger. SOA yields, on the other hand, were significantly larger when cyclohexane was used as an OH radical scavenger, compared to those measured for reactions conducted using alcohols or aldehydes. This dependence indicates that radical pathways play a major role in SOA formation in this reaction. Furthermore, the results show that reaction of OH radicals with scavengers used in laboratory studies can perturb the radical chemistry in such a way as to significantly impact SOA yields. We propose that this effect is due to increases in the ratio [hydroperoxy radicals]/[organic peroxy radicals] when alcohols or aldehydes are used as OH radical scavengers. This apparently enhances the rate of reaction of hydroperoxy radicals with key radical intermediates in SOA formation, effectively short-circuiting the reaction system into pathways leading to more volatile products.

Hope A. M., 2003, "Development of detection techniques and diagnostics for airborne carbon nanoparticles," *Technical Report*, Sandia National Laboratories, Albuquerque, NM; Livermore, CA, US, Report No. 2003–8666

We have recorded time-resolved LII signals from a laminar ethylene diffusion flame over a wide range of laser fluences at 532 nm. We have performed these experiments using an injection-seeded Nd:YAG laser with a pulse duration of 7 ns. The beam was spatially filtered and imaged into the flame to provide a homogeneous spatial profile. These data were used to aid in the development of a model, which will be used to test the validity of the LII technique under varying environmental conditions. The new model describes the heating of soot particles during the laser pulse and the subsequent cooling of the particles by radiative emission, sublimation, and conduction. The model additionally includes particle heating by oxidation, accounts for the likelihood of particle annealing, and incorporates a mechanism for nonthermal photodesorption, which is required for good agreement with our experimental results. In order to investigate the fast photodesorption mechanism in more detail, we have recorded LII temporal profiles using a regeneratively amplified Nd:YAG laser with a pulse duration of 70 ps to heat the particles and a streak camera with a temporal resolution of approximately 65 ps to collect the signal. Preliminary results confirm earlier indications of a fast mechanism leading to signal decay rates of much less than a nanosecond. Parameters to which the model is sensitive include the initial soot temperature, the temperature of the ambient gas, and the partial pressure of oxygen. In order to narrow the model uncertainties, we have developed a source of soot that allows us to determine and control these parameters. Soot produced by a burner is extracted, diluted, and cooled in a flow tube, which is equipped with a Scanning Mobility Particle Sizer<sup>TM</sup> (SMPS<sup>TM</sup>) for characterization of the aggregates.

Hopke, P. K.; Lee, D.-W.; Mavliev, R.; Rasmussen, D. H.; Wang, H.-C., 2003, "Comparison of experimental and theoretical heterogeneous nucleation on ultrafine carbon particles," *Journal of Physical Chemistry B*, **107**(50):13813–13822

Using a modified turbulent mixing CNC, the heterogeneous nucleation of different compounds (working fluids) on nanometer sized carbon particles was examined. The working fluids were dibutyl phthalate, octadecane, octadecanol, and octadecanoic acid. Based on the particle size distributions measured with a Scanning Mobility Particle Sizing system, nucleation and consequent growth were examined with respect to different temperature and vapor pressure for each working fluid. Nucleation rates for all conditions were calculated from the fitted size distribution data by subtracting the residual nonactivated particle concentration for each condition. Experimental nucleation rates were compared to the calculated ones based on Fletcher's heterogeneous nucleation theory. This theory matches well with the experiments with octadecanol and octadecanoic acid, and at high supersaturation ratios for dibutyl phthalate.

However, the theory shows discrepancies with the observed phenomena at low supersaturation for dibutyl phthalate, and especially for octadecane. Several possible hypotheses for the discrepancies and observed particle growth are discussed. 33 Refs.

Jang, M.; Kamens, R. M.; Lee, S., 2003, "Organic aerosol growth by acid-catalyzed heterogeneous reactions of octanal in a flow reactor," *Atmospheric Environment*, **37**(15):2125–2138

Octanal was chosen as a model carbonyl compound to study aerosol growth by the heterogeneous acid-catalyzed reactions. The heterogeneous reactions of octanal in the presence/absence of acidified seed aerosols were conducted in a 2 m flow reactor (2.5 cm ID) under darkness in the presence of background seed aerosols. To facilitate hemiacetal/acetal formation via the acid-catalyzed heterogeneous reaction of octanal, 1-nonanol was also co-injected with inorganic seed aerosols into the flow reactor system. The aerosol population was measured from a series of sampling ports down the flow reactor as a function of distance using a Scanning Mobility Particle Sizer (SMPS) spectrometer. These results were used to predict the apparent rate constants for heterogeneous reactions of octanal, optimizing experimentally observed aerosol growth from condensation and heterogeneous reactions. The aldehyde heterogeneous reactions were accelerated in the presence of an acid catalyst ( $\text{H}_2\text{SO}_4$ ), and led to higher aerosol yields, than when  $\text{H}_2\text{SO}_4$  was not present in the seed aerosol. Our results showed that the log of the organic aerosol yield is inversely related to humidity in the presence of an acid-catalyst ( $R^2=0.98$ ). When aerosols were not catalyzed with an acid, aerosol growth %yields were neither sensitive nor linear with %RH ( $R^2=0.18$ ). These results may be explained by the fact that acid-catalyzed heterogeneous reactions of aldehydes are significantly accelerated as the acidity increases. The SMPS was also operated in two different aerosol-sampling flow rates to characterize particle off-gassing while particles were inside the SMPS. The off-gassing of organic compounds from the particle phase was smaller with an acid catalyst than without an acid catalyst. Fourier transform infrared spectrometry (FTIR) was employed to demonstrate the direct transformation of chemical functional groups by acid-catalyzing the reactions of octanal with 1-nonanol as a thin liquid layer on a zinc selenide (ZnSe) FTIR disk. It was concluded that carbonyls, which are produced by atmospheric photochemical oxidation reactions, can significantly contribute to secondary organic aerosol formation through acid-catalyzed heterogeneous reactions.

Jaoui M.; Leungsakul S.; Kamens R.M., "Gas and Particle Products Distribution from the Reaction of  $\beta$ -Caryophyllene with Ozone," *Journal of Atmospheric Chemistry*, **45**(3):261–287

Gas and particulate reaction products from the ozonolysis of  $\beta$ -caryophyllene (**I**) in the presence of atmospheric air were investigated using a combination of gas chromatography-mass spectrometry (GC-MS) and high performance liquid chromatography (HPLC). A Scanning Mobility Particle Sizer system (3936, TSI) and a Condensation Particle Counter (3025A, TSI) were used to study secondary organic aerosol formation. The nighttime oxidation was carried out in a large outdoor smog chamber (190 m<sup>3</sup>). A wide range of ring retaining and ring opening products in the gas and particle phase are reported over the course of the reaction. On average, measured gas and particle phase products accounted for ~64% of the reacted  $\beta$ -caryophyllene (**I**) carbon. Measurements show that a number of reaction products with low vapor pressure (e.g.,  $\beta$ -caryophyllone aldehyde (**IV**),  $\beta$ -norcaryophyllone aldehyde (**V**),  $\beta$ -caryophyllonic acid (**VIII**),  $\beta$ -14-hydroxycaryophyllonic acid (**XIV**)) were found in the sample taken during the first 20 min of the reaction and may play an important role in the early formation of secondary organic aerosol. A detailed mechanism is proposed to account for most products observed in this investigation.

Kim, D. H.; Kim, D. S.; Lee, K. W.; Park, S. H.; Song, Y. M., 2003, "Brownian coagulation of polydisperse aerosols in the transition regime," *Journal of Aerosol Science*, **34**(7):859–868

Brownian coagulation of aerosols in the transition regime was studied at Knudsen numbers ranging from 0.8 to 5.5. Coagulation rates were obtained from the change in total number concentration of polydisperse oleic acid and the sodium chloride aerosols with geometric mean diameters between 25 and 150 nm in a closed chamber at atmospheric pressure. Particle size distributions and concentrations were measured by Scanning Mobility Particle Sizer™ (TSI) and Condensation Nuclei Counter (TSI), respectively. The experimental coagulation rates took deposition rates (diffusional wall loss rates) into account, because coagulation and diffusion deposition occur simultaneously in a closed chamber. In addition, to obtain the accurate coagulation rates of the experiments, two approaches were used and compared with each other. The experimental results were observed to follow the theoretical results of Dahneke and Fuchs, which showed that coagulation coefficients increased just a little in the transition regime. 15 Refs.

Knight, M.; Petrucci, G. A., 2003, "Study of residual particle concentrations generated by the ultrasonic nebulization of deionized water stored in different container types," *Analytical Chemistry*, **75**(17):4486–4492

A Scanning Mobility Particle Sizer has been used to quantify residual particle number and mass concentrations generated by ultrasonic nebulization of deionized (DI) water stored in a variety of bottles.

High variability of residual particles was found not only between different bottle types but also between different bottles of the same type. Degradation of the water quality, quantified as increased residual mass and number concentrations as a function of time, occurred to varying degrees for water stored in different bottle types. Overall, glass bottles showed the highest residual particle concentrations and exhibited the poorest stability over time. After a storage period of 3 weeks, DI water stored in Pyrex bottles showed average increases in particle mass and number densities in the aerosol of over 250% and 60%, respectively. Total dissolved impurity levels in the water increased from 110 to 290 ng/ml over the 3-week period. It is hypothesized that leaching from the bottle walls increases impurity levels in the water over time. Leaching was observed for both glass and polymer bottles. Contrary to this trend, residual particle concentrations from deionized water stored in Teflon bottles showed a net decrease during the measurement period. With respect to absolute residual particle concentrations and storage stability, a Teflon bottle yielded the best performance. Total residual particle mass and number densities for Teflon were less than a factor of 15% and 1%, respectively, as compared to residual particle levels observed for the Pyrex bottle. Absolute dissolved impurity levels in the water for the Teflon bottle decreased from 7.8 to 3.7 ng/ml over the 4-week period.

Leung, J. K. C.; Tso, M. Y. W.; Lam, J. H. C.; Zhou, Q. F., 2003, "Direct measurement of attachment of  $^{220}\text{Rn}$  progeny on aerosols by atomic force microscopy," *Nuclear Instruments and Methods in Physics Research, Section A, Accelerators, Spectrometers, Detectors and Associated Equipment*, **508**(3)

Atomic force microscopy (AFM) is becoming a powerful tool for the study of nuclear tracks in materials such as CR-39. Coupled with its capability of observing near nm aerosol particles, we have utilized the AFM to observe the radon progeny-loaded aerosol particles deposited on surfaces of CR-39 and to observe the corresponding etch pits produced by the  $\alpha$ -particles emitted from the radon progenies. A special platform was built so that after the aerosol particles on the CR-39 have been scanned and recorded, the CR-39 can be etched and then scanned for the etch pits at the same location. Both  $^{222}\text{Rn}$  and  $^{220}\text{Rn}$  progenies were used in the study. The progenies were generated by the appropriate radon sources and mixed with aerosol particles generated by aerosol generators. The aerosol size distributions were analyzed by a Scanning Mobility Particle Sizer spectrometer. Some of the limitations and difficulties of the technique will be described. The results enable us to examine the attachment process including multiple attachments of radon progenies on aerosols.

## 2002

Bonn, B.; Moortgat, G. K.; Schuster, G., 2002, "Influence of water vapor on the process of new particle formation during monoterpene ozonolysis," *Journal of Physical Chemistry A*, **106**(12):2869–2881

This study was focused on the investigation of the influence of water vapor on the size distribution of the newly formed aerosol particles during the reaction of monoterpenes and ozone measured by a Scanning Mobility Particle Sizer (TSI 3936). Measurements made during reactions of selected exocyclic monoterpenes  $\beta$ -pinene and sabinene, 1 ppmv) with ozone (0.5 ppmv) showed a decrease of the particle number concentration and total aerosol volume with increasing water vapor. On the contrary, number concentration and total aerosol volume were not affected or less affected by the presence of water vapor during similar experiments with the endocyclic monoterpenes  $\alpha$ -pinene and  $\Delta^3$ -carene. However, when the reactant concentrations of  $\alpha$ -pinene and ozone were lowered to 50 and 110 ppbv, a similar decreasing effect of water vapor on the nucleation was observed as found in the exocyclic monoterpene reactions, whereas an increase of the produced aerosol volume was measured. These observations contradict the assumption that the dicarboxylic acids, such as pinic acid in the case of  $\alpha$ - and  $\beta$ -pinene, produced by the unimolecular decomposition of the excited Criegee intermediate, are responsible for the observed nucleation. The dicarboxylic acids should therefore be independent of the concentration of water vapor. On the contrary, we bring evidence that new particle formation proceeds via the stabilized Criegee intermediate. Furthermore, the intermolecularly (e.g.,  $\beta$ -pinene) or intramolecularly (e.g.,  $\alpha$ -pinene) formation of secondary ozonides acting as nucleation precursors explains the observed effect of water vapor. The results of similar experiments with added formic acid and carbonyl compounds are supportive of this assumption.

## 2001

Ball, J. C.; Karlsson, R. S.; Maricq, M. M.; Szente, J. J., 2001, "Homogeneous aerosol formation by the chlorine atom initiated oxidation of toluene," *Journal of Physical Chemistry A*, **105**(1):82–96

The photolysis of  $\text{Cl}_2$  molecules in the presence of toluene and oxygen, at levels of  $\sim 10^{14}$  radicals/ $\text{cm}^{-3}$ , initiates a sequence of chemical reactions that rapidly produce an aerosol. Size distributions of the aerosol particles are examined, using a Scanning Mobility Particle Sizer, as a function of time, photolysis energy, the initial concentrations of toluene and chlorine, and of added NO and  $\text{HO}_2$ . The number of particles and the volume of aerosol both exhibit a steep nonlinear increase as the initial chlorine atom level is raised. Surprisingly, the number of particles displays a strong inverse dependence on the initial

toluene concentration, whereas the aerosol volume remains nearly unaffected by toluene level. Kinetic measurements of particle formation made using a flow reactor reveal an incubation period after initiation of the  $\text{Cl} + \text{C}_6\text{H}_5\text{CH}_3$  reaction, followed by steep increases in particle number and volume. The particle number rapidly reaches a plateau, whereas the aerosol volume continues to increase with time. The earliest observed particles are unexpectedly large, with mean diameters as high as 100 nm; a continuous growth from <10 nm is generally not observed. Both  $\text{NO}$  and  $\text{HO}_2$  suppress aerosol formation. These observations prompt us to postulate a mechanism whereby a minor reaction channel between chlorine atoms and benzylperoxy radicals to produce a Criegee intermediate controls the number of critical nuclei. This rate-limiting step is followed by rapid condensation of semivolatile compounds onto the nuclei. Because the aerosol volume can represent 10%, or more, of the toluene consumed, this necessarily includes products from the major oxidation pathways. As part of this work, we report 295 K rate constants of  $k_4 = (8 \pm 2) \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$  for the benzylperoxy self-reaction, and  $k_6 = (2.7 \pm 0.5) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$  for its reaction with  $\text{NO}$ .

Chen, Ying-Yuan; Lee, Whei-May Grace, 2001, "The effect of surfactants on the deliquescence of sodium chloride," *Aerosol Science & Technology*, **36**(2):229-242

This study investigated the deliquescence of sodium chloride aerosol with surfactants at a retention time of 4.24 sec. Two surfactants used in this study, glutaric acid and pyruvic acid, are found in atmospheric aerosol and have a high hydrophile-lipophile balance (HLB) value. The experimental system consisted of a relative humidity conditioner, a tandem differential mobility analyzer (TDMA) and a Scanning Mobility Particle Sizing (SMPS) system. Results obtained from the observation of TDMA presented the deliquescence point of sodium chloride aerosol at 75% RH. In addition, the growth size of sodium chloride aerosol was observed to be 79.47 nm and the growth ratio was 77.94%, when the initial size of aerosol was 101.82 nm. Surfactants were internally mixed with sodium chloride at six different weight fractions, i.e. 2.5, 5, 10, 20, 40, and 60% by weight of surfactants in dry aerosol. Both surfactants apparently decreased the deliquescence point of sodium chloride, in which the lowest deliquescence point appeared at about 71% RH when the weight fraction of surfactants is 60% by weight of surfactants in dry aerosol. Moreover, the smallest size of sodium chloride aerosol with surfactants was 142.7 nm at 60% by weight of pyruvic acid in dry aerosol. Our results further demonstrate that the deliquescence point and size of sodium chloride aerosol with surfactants are related to the weight fraction of surfactants.

Hurley, M. D.; Sokolov, O.; Wallington, T. J.; Takekawa, H.; Karasawa, M.; Klotz, B.; Barnes, I.; Becker, K. H., 2001, "Organic Aerosol Formation during the Atmospheric Degradation of Toluene," *Environmental Science & Technology*, **35**(7):1358-1366

Organic aerosol formation during the atmospheric oxidation of toluene was investigated using smog chamber systems. Toluene oxidation was initiated by the UV irradiation of either toluene/air/ $\text{NO}_x$  or toluene/air/ $\text{CH}_3\text{ONO}/\text{NO}$  mixtures. Aerosol formation was monitored using Scanning Mobility Particle Sizer and toluene loss was monitored by in-situ FTIR spectroscopy or GC-FID techniques. The experimental results show that the reaction of OH radicals,  $\text{NO}_3$  radicals and/or ozone with the first generation products of toluene oxidation are sources of organic aerosol during the atmospheric oxidation of toluene. The aerosol results fall into two groups, aerosol formed in the absence and presence of ozone. An analytical expression for aerosol formation is developed and values are obtained for the yield of the aerosol species. In the absence of ozone the aerosol yield, defined as aerosol formed per unit toluene consumed once a threshold for aerosol formation has been exceeded, is 0.075 plus or minus 0.004. In the presence of ozone the aerosol yield is 0.108 plus or minus 0.004. This work provides experimental evidence and a simple theory confirming the formation of aerosol from secondary reactions.

Jang, M.; Kamens, R. M., 2001, "Atmospheric secondary aerosol formation by heterogeneous reactions of aldehydes in the presence of a sulfuric acid aerosol catalyst," *Environmental Science & Technology*, **35**(24):4758-4766

Particle growth by the heterogeneous reaction of aldehydes was evaluated in 0.5 m<sup>3</sup> Teflon film bags under darkness in the presence of background seed aerosols. The aldehydes used were as follows: glyoxal, butanal, hexanal, octanal, and decanal. To study acid catalyst effects on aldehyde heterogeneous reactions, one of the Teflon bags was initially filled with seed aerosols composed of ammonium sulfate-aerosol acidified with sulfuric acid. These results were compared to particle growth reactions that contained only ammonium sulfate as a background seed aerosol. The gas-phase aldehydes were then added to the Teflon bags. In selected experiments, 1-decanol was also added to the Teflon bags with aldehydes to clarify particle growth via a heterogeneous hemiacetal/acetal formation in the presence/absence of an acid catalyst. The particle size distribution and growth were measured using a Scanning Mobility Particle Sizer<sup>TM</sup> (TSI-SMPS<sup>TM</sup>), and the results were applied to predicting aerosol growth and size distribution changes by condensation and heterogeneous reactions. Aerosols created from the heterogeneous reactions of aldehydes were collected directly on an ungreased zinc selenide (ZnSe) FTIR disk (25 mm in diameter) by impaction. The ZnSe disks were directly analyzed for product

functional groups in the aerosol phase using a Fourier transform infrared (FTIR) spectrometer with a deuterated triglycine sulfate (DTGS) detector. Aerosol growth by heterogeneous aldehyde reactions proceeds via a hydration, polymerization process, and hemiacetal/acetal formation from the reaction of aldehydes with alcohols. These aldehyde heterogeneous reactions were accelerated in the presence of an acid catalyst,  $H_2SO_4$ , and led to higher aerosol yields than when  $H_2SO_4$  was not present in the seed aerosol. The FTIR spectra obtained from the growing aerosol, also illustrated aldehyde group transformation in the particle phase as a function of the heterogeneous reaction. It was concluded that aldehydes, which can be produced by atmospheric photochemical reactions, can significantly contribute on secondary aerosol formation through heterogeneous reactions in the presence of an acid catalyst.

Jaoui, M.; Kamens, R. M., 2001, "Mass balance of gaseous and particulate products analysis from alpha-pinene/ $NO(x)$ /air in the presence of natural sunlight," *Journal of Geophysical Research*, **106**(D12):12,541-12,558

The daytime oxidation of alpha-pinene in the presence of natural sunlight and oxides of nitrogen,  $NO(x)$ , was studied in a large outdoor smog chamber using gas chromatograph mass spectrometry as the method of identification and quantification. A Scanning Mobility Particle Sizer system (3936, TSA) and a Condensation Particle Counter (3025A, TSA) were used to study the secondary organic aerosol formation, and a filter pack/denuder sampling system was used for simultaneously collecting gas phase and particle phase products for analysis. A gas chromatograph coupled to an electron impact mass spectrometry (GCEIMS) method is described for the identification and quantification of gas and aerosol products. The sensitivity of the method was sufficient to produce good quality mass spectra over the range of concentrations used in this study, and compares very well with classical methods based on derivatization and liquid chromatography-ion trap mass spectroscopy methods. Mass balances for gaseous and aerosol reaction products are reported over the course of the reaction. More than 16 products were identified and quantified. On average, measured gas and particle phase products accounted for approximately 54 to approximately 71 percent of the carbon reacted alpha-pinene. Measurements show that 10-hydroxypinonic acid, 10-hydroxypinonaldehyde, 4-oxopinonic acid, and 10-oxopinonic acid are observed in the early stage in the aerosol phase and may play an important role in the early formation of secondary aerosols.

Yamada, Y.; Koizumi, A.; Miyamoto, K.; Ichitsubo, H.; Fukutsu, K.; Shimo, M.; Tokonami, S., 2001, "Studies on characterization of radon and its progeny," In Technical Report 'exposure assessment and reduction of environmental radiation' 39 pages, National Inst. of Radiological Sciences, Chiba (Japan), Report No. NIRS-R-45

A basic study for characterization of radon and its progeny has been carried out. The main work is to establish a measurement technique of size distribution of radon progeny aerosols. The aerosols are ranged from sub-nanometer to micrometer, so that the impaction method and the diffusion method were introduced. From size measurements using the ELPI (Electrical Low Pressure Impactor), it was suggested that grease or oil coating does not interfere with the electrical size measurements but prevent particle rebound and/or redispersion. As the impactor measures two size distributions on number and activity, we can get a very important information on attachment phenomenon. In the comparison of size distribution with the Scanning Mobility Particle Sizer (SMPS) spectrometer in EML (Environmental Measurements Laboratory, New York), there was a relatively good agreement with our SDB (Screen-type Diffusion Battery) or the MOUDI (Micro-Orifice Uniform Deposit Impactor). And our original GSA (Graded Screen Array) based on the SDB showed a very sharp peak of unattached fraction below 1 nm. We try to apply the measurement technique for a field study and to get radon progeny aerosol data which affects the DCF (Dose Conversion Factor).

Combined wet electrostatic aerosol removal and energy recovery, 2001, UVE TECHNICAL, REPORT No. NEI-DK-3925, dk-TEKNIK ENERGI OG MILJOe, Soeborg, Denmark, in Danish

## 2000

Ehara, K.; Mulholland, G. W.; Hagwood, R. C., 2000, "Determination of Arbitrary Moments of Aerosol Size Distributions from Measurements with a Differential Mobility Analyzer," *Aerosol Science & Technology*, **32**(5):434-452

A method to determine arbitrary moments of aerosol size distributions from differential mobility analyzer measurements has been proposed. The proposed method is based on a modification of the algorithm developed by Knutson and Whitby to calculate the moments of electrical mobility distributions. For this modification, the electrical mobility and the charge distribution have been approximately expressed by power functions of the particle diameter. To evaluate the validity of the approximation, we have carried out numerical simulations for typical size distributions. We have found that for typical narrowly distributed aerosols such as polystyrene latex particles and particles that arise in the tandem differential mobility analyzer configuration, the distribution parameters can be accurately determined by

this method. For a log-normally distributed aerosol, the accuracy of the distribution parameters determined by this method has been evaluated as a function of the geometric standard deviation. We have also compared the accuracy of the proposed method with other existing methods in the case of the asymmetric Gaussian distribution.

Martin, S. T.; Han, J.-H., 2000, "An aerosol chemical reactor for coating metal oxide particles with  $(\text{NH}_4)_2\text{SO}_4\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ . Manipulation of the sulfate coating," *Journal of Crystal Growth*, **219**(3):290-299

One important chemical and morphological class of atmospheric particles consists of mineral dusts contained as inclusions in crystalline and aqueous sulfate particles. The mineral dust surface plays an important role in regulating the sulfate phase by providing a surface for the heterogeneous nucleation of the crystal from supersaturated aqueous sulfate phases. In principle, the efficacy of mineral dusts in promoting phase transitions can be investigated through laboratory process studies. To do so, however, a particle generator is necessary. The present work describes the generation of metal oxide particles (viz.  $\alpha\text{-Fe}_2\text{O}_3$ ,  $\alpha\text{-Al}_2\text{O}_3$ ,  $\text{Al}_6\text{Si}_2\text{O}_{13}$ , and amorphous  $\text{SiO}_2$ ) coated by  $(\text{NH}_4)_2\text{SO}_4$ . The diameter of the oxide core and the thickness of the sulfate coating are regulated from 30 to 400 nm and 13 to 81 nm, respectively. Sizing is based upon aerosol particles collected by electrostatic precipitation and subsequently imaged by transmission electron microscopy. Depending on reactor conditions, the fraction of particles containing a metal oxide core varies from 0.01 to 0.86, as inferred from number size distributions measured with a Scanning Mobility Particle Sizer spectrometer. The single most important reactor variable affecting fraction of particles containing a metal oxide core is the total sulfate loading. No dependencies on the effects of different chemical surface properties of the metal oxide particles on the coating process have been observed in our laboratory to date. Rather, the number size distribution of metal oxide core particles appears important without regard to chemical surface properties.

## 1999

Dassios, K. G.; Pandis, S. N., 1999, "Mass accommodation coefficient of ammonium nitrate aerosol," *Atmospheric Environment*, **33**(18):2993-3003

The mass transfer rate of pure ammonium nitrate between the aerosol and gas phases was quantified experimentally by the use of the tandem differential mobility analyzer/Scanning Mobility Particle Sizer (TDMA/SMPS) technique. Ammonium nitrate particles 80-220 nm in diameter evaporated in purified air in a laminar flow reactor under temperatures of 20-27 degree C and relative humidities in the vicinity of 10%. The evaporation rates were calculated by comparing the initial and final size distributions. A theoretical expression of the evaporation rate incorporating the Kelvin effect and the effect of relative humidity on the equilibrium constant is developed. The measurements were consistent with the theoretical predictions but there was evidence of a small kinetic resistance to the mass transfer rate. The discrepancy can be explained by a mass accommodation coefficient ranging from 0.8 to 0.5 as temperature increases from 20-27 degree C. The corresponding timescale of evaporation for submicron NH<sub>4</sub>NOSUB3 particles in the atmosphere is of the order of a few seconds to 20 min. (Author abstract) 31 Refs.

Holub, R. F.; Reimer, G. M.; Hopke, P. K.; Hovorka, J.; Krcmar, B.; Smrz, P. K., 1999, "'Geoaerosols': their origin, transport and paradoxical behavior: a challenge to aerosol science," *Journal of Aerosol Science*, **30**(S1):S111-S112

Aerosols in the range 3 to 200 nm in a cave and in an experimental mine were measured using Diffusion Particle Sizer (DPS) and Scanning Mobility Particle Sizer (SMPS) spectrometer. The areas were also sampled for volatile organics by means of TENEX adsorbers, and for the filterable inorganics and organics by means of porous membrane filters. Some of the filters were measured by means of electron microscope and gas chromatography to determine the concentration and composition of the inert aerosols. The concentration and composition of the radioactive aerosols was measured by means of standard nuclear counting equipment. Simultaneously, the outdoor inert and radioactive aerosols were also sampled. The results for both DPS (cave) and SMPS (mine) were basically the same. 5 Refs.

Martin, S.; Han, J., 1999, "Generation, characterization, and phase transitions of  $(\text{NH}_4)_2\text{SO}_4\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$  aerosols internally mixed with metal oxide particles," *Journal of Aerosol Science*, **30**(S1):S827-S828

Aerosols composed of a water soluble  $(\text{NH}_4)_2\text{SO}_4\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$  components and a water insoluble  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ , or  $\text{ZrO}_2$  core are reported. An internally mixed aerosol is shown to be generated in several steps. The effects of changing the controllable parameters on particle size and number density are reported. Unity saturation was obtained at 80% relative humidity at 298 K. In the absence of heterogeneous nuclei, efflorescence did not occur until the relative humidity is 35%. Heterogeneous nuclei had a significant impact on the crystallization relative humidity. 1 Refs.

Noziere, B.; Barnes, I.; Becker, K.-H., 1999, "Product study and mechanisms of the reactions of alpha - pinene and of pinonaldehyde with OH radicals," *Journal of Geophysical Research*, **104**(D19):23645–23656

The reactions of alpha -pinene and of its main oxidation product, pinonaldehyde (3-acetyl-2,2-dimethyl-cyclobutyl-ethanal), with OH radicals have been studied in the laboratory using Fourier transform infrared spectroscopy for real-time monitoring of the gas-phase chemical species and a Scanning Mobility Particle Sizer system (3071 A, TSI) for the study of secondary aerosol formation. All gas-phase molar yields were quantified using calibrated reference of the pure compound, except for the nitrate products. The results were: for the alpha -pinene experiments in the presence of NO<sub>x</sub>, pinonaldehyde, (87±20)%; total nitrates (18±9)%; formaldehyde, (23±9)%; acetone (9±6)%; for the alpha -pinene experiments in the absence of NO<sub>x</sub>: pinonaldehyde, (37±7)%; formaldehyde, (8±1)%; acetone, (7±2)%; for the pinonaldehyde experiments in the presence of NO, formaldehyde (152±56)% and acetone (15±7)%. The aerosol measurements showed that the condensed products accounted for the missing carbon in the gas-phase balance. The partitioning of the products into the condensed phase was found to be potentially significant under experimental conditions but less than 10% for initial alpha -pinene concentrations lower than 10<sup>13</sup> molecule cm<sup>-3</sup> and hence negligible under atmospheric conditions in the absence of aerosol seeds. On the basis of these results a comprehensive mechanism for the gas-phase reaction of alpha -pinene with OH in the presence of NO<sub>x</sub> has been proposed, including quantitative values for all the involved branching ratios.

## 1997

Lee, W.-M. G.; Shih, P.-M.; Wang, C.-S., 1997, "The influence of relative humidity on the size of atmospheric aerosol," *J. Environ. Sci. Health, Part A: Environ. Sci. Eng. Toxic Hazard. Subst. Control*, **A32**(4):1085–1097

The relative humidity of ambient air is very often over 75% throughout the year in Taiwan. Thus, the influence of relative humidity on the growth of hygroscopic aerosols in the atmosphere is particularly important. In this study, a TDMA system (Tandem Differential Mobility Analyzer) equipped with newly developed a SMPS (Scanning Mobility Particle Sizer) was used to observe the sizes of submicron ammonium sulfate aerosols, from 63.8nm to 224.7nm, at five controlled relative humidities, 50%, 75%, 81%, 85% and 89%. It is the first time that the hygroscopic growths and deliquescence points of submicron aerosols of ammonium sulfate have ever been closely observed by use of the SMPS. The experimental results showed that the abrupt size changes of aerosols at their deliquescence points can be more precisely observed than in previous studies. It was also shown that there was no apparent growth in an ammonium sulfate aerosol until relative humidities was over 75%. At relative humidities between 81% and 89%, the growth ratio of size was about a factor of 1.3 to 1.6 and the water content of ammonium sulfate aerosols by mass was about 47% to 66%.

Mouradian, S.; Skogen, J. W.; Dorman, F. D.; Zarrin, F.; Kaufman, S. L.; Smith, L. M., 1997, "DNA analysis using an electrospray Scanning Mobility Particle Sizer," *Analytical Chemistry*, **69**(5):919–925

Samples of single-stranded DNA (6.1 kDa, 20 nucleotides in length) and double-stranded DNA (300 kDa, 500 base-pairs) in similar or equal 20m M -ammonium acetate of pH 6.8 or 7.8 were sprayed (similar or equal 50 nl/min) through a capillary under reduced pressure into a neutralizing chamber held at minus 2.5 kV. The charge associated with multiply charged species was neutralized with a 210 Po small alpha -particle source to give mainly neutral or singly charged species. The aerosol was transferred to a TSI Model 3071 Differential Mobility Analyzer (DMA), the air flow in which was 20 L/min; a potential of up to 5 kV was applied to the central rod where most airborne particles were collected, and the cylinder wall was earthed. Gas-phase molecules of a specific electrophoretic mobility passed through the DMA and were monitored in a condensation particle detector saturated with butanol. Analytes were separated and detected at the pmol to fmol level. Measured electrophoretic mobility diameters correlated with the mol. wt. of the analytes. Different apparent densities were observed for single- and double-stranded DNA.

## 1996

Lee, Whei-May Grace; Chen, Chou-Yuan; Huang, Shou-Lin; Lee, Chung-Te, 1996 "Deliquescent growth of inorganic-salt aerosols observed by a combined system of TDMA and integrated nephelometer," *Journal of Aerosol Science*, **27**(S1): S313–S314

The deliquescent growth of three inorganic-salt aerosols were studied using the Tandem Differential Mobility Analyzer and Scanning Mobility Particle Sizer (TDMA plus SMPS), and integrated nephelometer. The two systems were connected in parallel and operated simultaneously, and the inorganic-salt considered were NaCl, NH<sub>4</sub>NH<sub>3</sub>, and NH<sub>4</sub>SO<sub>4</sub>. A comparison of the two systems showed that integrated nephelometer provides better observation on deliquescent point. However, the heat from its lamp may change the liquid state of aerosol and would delay the deliquescent point. 3 Refs.



Owens, T. M.; Biswas, P., 1996, "Vapor phase sorbent precursors for toxic metal emissions control from combustors," *Industrial and Engineering Chemistry Research*, **35**(3):792-798

A bench scale system incorporating a high temperature flow reactor has been utilized to study the reaction of metallic species and sorbent compounds. The reaction and interaction between a lead precursor and a vapor phase silica precursor is studied. The effect of chlorine and the effect of sorbent to metal feed ratio on lead speciation and size distribution is determined. The size distributions of the reacted compounds are measured by a Scanning Mobility Particle Sizer (SMPS) spectrometer and an optical particle counter (OPC). The product powders are also collected and their compositions established by X-ray diffraction (XRD) or, if an amorphous powder is collected, by infrared and raman spectroscopy. Experiments have been performed at 500, 750, 1,000, and 1,250 C and for reactor residence times of 0.5-1.0 s. For example, at 750 C, for a lead feed in conjunction with a vapor phase silica precursor compound, the mean particle size exiting the reactor is significantly larger than for a lead only feed or a silicon only feed. The composition of the resulting aerosol is determined, via infrared and Raman spectroscopy, to be lead silicate,  $\text{PbSiO}_3$ . Higher temperature results are similar. At lower temperatures, silica particles are not formed in the reactor. Higher silica precursor feed rates result in particles large enough for capture by conventional pollution control equipment.

## 1993

Rogak, Steven N.; Flagan, Richard C., 1993, "The Mobility and Structure of Aerosol Agglomerates," *Aerosol Science and Technology*, **18**:25-47

The relation between the aerosol agglomerate structure and transition regime mobility can be explained with a simple model incorporating fractal geometry and results from computer simulations. The model is consistent with previous measurements.

## 1992

Daumer, B.; Niessner, R.; Klockow, D., 1992, "Laboratory Studies of the Influence of Thin Organic Films on the Neutralization Reaction of  $\text{H}_2\text{SO}_4$  Aerosol with Ammonia," *Journal of Aerosol Science*, **23**(4):315-325

The neutralization reaction of micro-encapsulated sulfuric acid aerosol with gaseous ammonia has been investigated. Experimental parameters were: influence of different coating substances, coating thickness, relative humidity and reaction time. The reaction of  $\text{H}_2\text{SO}_4$  aerosol with  $\text{NH}_3$  was measured as a function of time by chemical characterization of the particulate and gaseous phase before and after mixing of the reactants in a flow reactor. A remarkable decrease in the reaction rate was observed, depending on molecular structure of the organic coating substance and the thickness of coating. Straight chain molecules like n-hexadecane and n-hexadecanol strongly retard the reaction. Branched chain molecules as coating showed no significant effect, probably because of their inability to form tight films. Naturally occurring organic coating substances affect the neutralization, too. The mass transport resistance of the films investigated are interpreted in terms of the accommodation and reaction coefficient.

Hameri, Kaarle; Rood, Mark; Hansson, Hans-Christen, 1992, "Hygroscopic Properties of a NaCl Aerosol Coated with Organic Compounds," *Aerosol Science*, **23**(S1):S437-S440

Atmospheric aerosol particles consist of a wide range of mixtures of inorganic and organic compounds. The hygroscopic properties of atmospheric aerosol particles (e.g., NaCl) has been studied extensively by numerous experimental and theoretical techniques (Winkler, 1988; Covert et al, 1984; Rood et al 1987 and Hansson et al, 1990). Results from experimental research has indicated that atmospheric aerosol particles with diameters less than  $1 \mu\text{m}$  are hygroscopic and such behavior has been associated with the existence of sulphate, nitrate and chloride salts.

Kutz, S.; Schmidt-Ott, A, 1992, "Characterization of Agglomerates by Condensation-Induced Restructuring," *Aerosol Science*, **23**, (S1):S357-S360

Most solid aerosol particles are agglomerates of smaller primary particles adhering to each other by weak forces. The structure of agglomerates, in particular the density of the primary particle packing determines their behavior in many respects. This is the motivation for finding a method that classifies agglomerates in terms of their packing density. In this paper we want to introduce such a method, which in principle allows to discriminate between particles of different packing densities. It is based on the property of agglomerates to increase their density, reaching a structure of lower internal energy if a vapor condenses on them.

Wake, D.; Brown, R.C.; Trottier, R.A.; Liu, Y., 1992, "Comparison of the Efficiency of Respirator Filters and Filtering Facepieces Against Radon Daughter Aerosols and Laboratory Generated Aerosols," *Aerosol Science*, **23**(SI):S757-S760

Respirator filters and filtering facepieces have been tested for filtration efficiency against radon daughters in a flourspar mine. The test method involved the use of sampling filters exposed to natural radon daughters in air filtered by the test respirators. Respirators with a filtration efficiency high enough for them to be considered suitable for use against toxic dusts generally reduced radon daughter levels by 90% or more, though nuisance dust masks were ineffective. The measured penetration of radon daughters through the former types of filter correlated reasonably well with the penetration of 0.1 $\mu$ m neutralized monodisperse aerosol and with that of the BS4400 sodium chloride aerosol, measured in the laboratory. Test filters were exposed to radon daughters in the mine for 8 hours as a simulation of their behavior during a working shift, but their performance was not altered by this and they did not show measurable radioactivity as a result of it.

## 1991

Matsoukas, Themis; Friedlander, Sheldon K., 1991, "Dynamics of Aerosol Agglomerate Formation," *Journal of Colloid and Interface Science*, **146**(2):495-506

The dynamics of the formation of metal oxide agglomerates was studied by introducing metal containing salts of magnesium and zinc into a flat flame. Distributions of the mobility equivalent diameter of the agglomerates formed downstream from the flat flame were self-preserving and in good agreement with the theory for the agglomeration of power law (fractal-like) agglomerates in the free molecule regime. Theory indicates that after sufficiently long times, the average size of the agglomerates varies inversely with the size of the primary particles composing the agglomerates, for a given volumetric concentration of aerosol material. The effect of primary particle size is significant, and supported by experimental evidence.

Reischl, G.P., 1991, "The Relationship of Input and Output Aerosol Characteristics for an Ideal Differential Mobility Analyzer Particle Standard," *Journal of Aerosol Science*, **22**(3)297-312

A careful review of the components of a generator-charger-classifier (GCC) system for monodisperse particle generation is given and its influence on the properties of the resulting size distribution is discussed. The concentrations, mean diameters and geometric standard deviations of the output aerosol are investigated for different input aerosols with diameters from 5 nm to 250 nm and geometric standard deviations from 1.2 to 2.0, by number and volume size distributions. The general trends are discussed in detail.

## 1986

Blackford, David B.; Simons, Gary R., 1986, "Particle Size Analysis of Carbon Black," *Particle and Particle Systems Characterization*, **4**(1-4):112-117

A new technique is described for measuring the size distribution of submicrometer sized powders, such as carbon black. Powder is first dispersed as an aerosol and the subsequent particle size measured with a differential mobility classifier and an aerosol concentration detector. The technique has been used to provide a size distribution for seven ASTM/D1765 designated grades of carbon black (N110, N234, N299, N330, N650, N683, N762). In addition, four of these samples (N234, N330, N683, N762) are standards designated by ASTM D24 committee on carbon black as Standard Reference Blacks C3, B3, D3, A3, respectively. This paper will discuss the effectiveness of the dispersion technique and present distribution data on the seven ASTM grades of carbon black.

## 1985

Hansson, Hans-Christen; Ahlberg, Mats S., 1985, "Dynamic Shape Factors of Sphere Aggregates in an Electric Field and Their Dependence on the Knudsen Number," *Journal of Aerosol Science*, **16**(1):69-79

Dynamic shape factors in an electric field for sphere aggregates of up to five monodisperse particles and their dependence on the Knudsen number in the range  $Kn=0.2-1.2$ , were investigated using a commercial electrostatic classifier (TSI 3071). The results have been compared with data from the literature obtained with electrical inertial spectrometers as well as with diffusion batteries and conclusions are drawn about the probable orientation of the aggregates during settling. For clusters of more than three primary particles no preferred orientation was observed over the Knudsen range investigated and the measured dynamic shape factors showed good agreement with those obtained with inertial spectrometers (aerosol centrifuges) indicating a random orientation during settling in both types of instruments. Chains of 3 and 4 primary particles were found to be oriented with their polar-axis parallel with the electric field during settling for all particle sizes studied. For doublets a significant increase of the dynamic shape factor with

increasing Knudsen number was found. Extrapolations from a linear regression line gave dynamic shape factors of 1.036 and 1.073 for  $Kn=0$  and 1.28, respectively. The value of 1.036 is in good agreement with that expected for doublets settling with their polar-axis parallel with the electric field and the value of 1.073 is in agreement with recently published experimental data at  $Kn=1.28$  obtained with diffusion batteries in which doublets should move with random orientation. A similar behavior was observed for triplet clusters. At Knudsen numbers below 0.5 these oblate shaped particles seemed to settle with their polar axis perpendicular with the electric field but at higher Knudsen numbers they seemed to move with random orientation. The observed change in orientation with increasing Knudsen number for these triplet particles was however not significant.

## 1982

Hoffmann, J.; Bunz, H.; Schock, W., 1982, "Dynamical Behaviour of Polydisperse Aerosol Particles in a Smog Chamber," *Aerosol Science*, **23**(S1):S861–S864

Aerosol particles containing different salt species (NaCl and Nitrate Salts) were produced and injected into a smog chamber of 3.7 m<sup>3</sup> volume. The chamber is thermally isolated and temperature differences below 0.5 K can be obtained in the entire volume. This allows to maintain stable thermodynamic conditions for small droplets beyond the point of deliquescence (up to relative humidities of about 97%) of the salt particles. Most of the measurements described here were made at relative humidities below the point of deliquescence. Changes in particle concentrations and changes in particle size spectra due to coagulation and other physical processes were measured using an optical particle counter and a differential mobility particle sizer. The measurements were compared to model calculations using the NACHE code. In the code in addition to the mechanical processes the uptake of water is modeled as a function of relative humidity, particle composition and particle size distribution.

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## Instrumentation Development

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### 2013

Wang, D., Kam, W., Cheung, K., Pakbin, P., and Sioutas, C., 2013, "Development of a Two-Stage Virtual Impactor System for High Concentration Enrichment of Ultrafine, PM 2.5 , and Coarse Particulate Matter," *Aerosol Science and Technology*, **47**(3): 231–238

A two-stage particle concentration enrichment system was developed to provide highly concentrated particles at low flow rates, for applications in areas such as toxicity studies of particulate matter (PM) as well as for increasing the signal-to-noise ratio in online particle sampling instruments. The current system is an extension of the Versatile Aerosol Concentration Enrichment System (VACES) developed at University of Southern California and operates by placing a second-stage miniature virtual impactor (VI) downstream of the VACES. Particles are sequentially enriched through each stage. Laboratory evaluations were conducted using various types of polydisperse particles to simulate typical ambient PM components as well as monodisperse polystyrene latex (PSL) particles. The system's configuration was tested by adjusting the intermediate flow rate, which is the intake flow of the second-stage VI (or minor flow of the first-stage VIs), for which 15 L/min was determined to be optimal in terms of maximizing the overall concentration enrichment. Particle size distributions before and after concentration enrichment were compared using a Scanning Mobility Particle Sizer spectrometer. Overall, our results indicate that the sampled particles were relative consistently enriched by factors of 100–120 (i.e., a concentration enrichment efficiency 75–85% of the ideal value) based on both PM mass and number concentrations, and along with similar physical properties of the size distribution (i.e., mode, median). Continuous and time-integrated field tests using urban ambient PM also showed consistent enrichment factors (by roughly 100–120 times) for number and mass concentrations, black carbon, and PM-bound polycyclic aromatic hydrocarbons.

### 2012

Dey, L., and Venkataraman, C., 2012, "A Wet Electrostatic Precipitator (WESP) for Soft Nanoparticle Collection," *Aerosol Science and Technology*, **46**(7): 750–759

Many nanoparticle collection devices have limitations related to retention of particle integrity from bounce, shattering, or aggregation. Suspensions of soft nanoparticles (e.g., proteins, lipids) are required for drug delivery and therapy. To enable direct collection of soft nanoparticles into liquid media, a wet electrostatic precipitator (WESP) was designed and evaluated in this work. Different sections were used for ion generation and particle charging, for minimal contact between the corona wire and particles, which were charged using positive nitrogen ions. WESP dimensions and operating parameters were optimized using charge distribution modeling. The prototype WESP was designed for operation with a continuous flow of liquid over the collection plate, to allow continuous particle collection from the exit stream of an aerosol reactor. The collection efficiency of the WESP, in dry and wet modes, was measured using aerosols of monodisperse polystyrene latex (PSL), polydisperse sucrose, and stearic acid (soft lipid) particles, through SMPS measurements, corrected for diffusional losses, at the entry and exit of the device. Measured collection efficiency was 70%–90% for particles of sizes 80–600 nm diameter in reasonable agreement with theoretical estimates. However, for small particles (20–80 nm diameter) measured collection efficiency ranged 40%–70%, significantly lower than theoretical estimates, possibly from incomplete neutralization of negative charges attained during air-jet atomization. Transmission electron microscopy (TEM) images and dynamic light scattering (DLS) measurements confirm that wet collection produces a suspension of free, unaggregated nanoparticles with sizes similar to their measured mean mobility diameter.

Kang, J. S., Lee, K. S., Lee, K. H., Sung, H. J., and Kim, S. S., 2012, "Characterization of a Microscale Cascade Impactor," *Aerosol Science and Technology*, **46**(9): 966–972

Although a cascade impactor is widely used for bioaerosol sampling and separation, the conventional system has some shortcomings when it is applied for the portable device. In the present study, a microscale cascade impactor system was developed using a soft lithography process to provide good portability and cost efficiency. The system described here included three impaction stages, and a rectangular-shaped jet nozzle was employed with a 0.5 L/min airflow rate. The collection efficiency of each stage was evaluated using polystyrene latex (PSL) (0.2–2.5  $\mu\text{m}$ ) and ammonium sulfate (10–700 nm) particles. The 50% cutpoints of each stage were numerically calculated to be 1.06, 0.55, and 0.26  $\mu\text{m}$ , and the experimental values were found to be 1.19, 0.51, and 0.27  $\mu\text{m}$ , respectively. The bounce and re-

entrainment in the impaction zone were reduced by spin-coating of silicon oil onto the microchannel surface. The overall particle losses were less than 10% for the 0.2–2.5  $\mu\text{m}$  particles and 9%–12% for the 0.05–0.20  $\mu\text{m}$  particles.

## 2011

Fang, W., Lei, G., Shan, X., Liu, F., Wang, Z., and Sheng, L., 2011, "A VUV photoionization organic aerosol mass spectrometric study with synchrotron radiation," *Journal of Electron Spectroscopy and Related Phenomena*, **184**(3-6): 129–133

A photoionization aerosol time-of-flight mass spectrometer (ATOFMS) has been developed for on-line analysis of organic compounds in aerosol particles using tunable vacuum ultraviolet (VUV) synchrotron radiation. Aerosol particles can be sampled directly from atmospheric pressure and are focused through an aerodynamic lens assembly into the mass spectrometer. The particles are vaporized when they impact on a heater, and then the nascent vapor is softly photoionized by synchrotron radiation. The degree of fragmentation of molecule can be controlled either by the heater temperature or by the photon energy. Thus, fragment-free tunable VUV mass spectra are obtained by tuning the photon energy close to the ionization energies (IEs) of the sample molecules. The direct determination of the IEs of benzophenone (9.07 eV), salicylic acid (8.72 eV), and urea (9.85 eV) are measured from the photoionization efficiency spectra with uncertainties of  $\pm 50\text{meV}$ . Ab initio calculations have been employed to predict the theoretical ionization energy.

Hsiao, T.-C., Chen, D., Greenberg, P. S., and Street, K. W., 2011, "Effect of geometric configuration on the collection efficiency of axial flow cyclones," *Journal of Aerosol Science*, **42**(2): 78–86

The particle collection efficiencies of axial flow cyclones with eight different geometric configurations, operated at 50 lpm aerosol flowrate, have been evaluated in this study. The geometric variation of test cyclones includes the optional addition of an upside-down cup, two vortex finder lengths, and two cyclone base shapes. Under various configurations, the cutoff aerodynamic particle size of axial flow cyclones changed from 272 to 448 nm. Our study shows that configuration effects on the collection efficiency of axial flow cyclones are different from those of tangential flow cyclones. The observation of different geometric effects on particle collection by axial and tangential flow cyclones is attributed to the flow pattern difference between cyclones of two types. It is further concluded that the optimal configuration for axial flow cyclones is with an abrupt contraction base, without an upside-down cup and with an increased vortex finder length. A simple model combining the model of Leith and Licht (1972) and the tubing loss in 901 bends at high Reynolds numbers has also been proposed to predict the particle collection efficiency curve of the optimal axial flow cyclone among those tested.

## 2010

Han, B., Kim, H. J., and Kim, Y. J., 2010, "Fine particle collection of an electrostatic precipitator in CO<sub>2</sub>-rich gas conditions for oxy-fuel combustion," *The Science of the total environment*, **408**(21): 5158–64

The collection of particles in CO<sub>2</sub>-enriched environments has long been important for the capture of CO<sub>2</sub> in order to clean gases via oxy-fuel combustion. We here report on the collection characteristics of fine and ultrafine particles using an electrostatic precipitator (ESP) in a CO<sub>2</sub>-enriched atmosphere. In order to understand the characteristics of particle collection in CO<sub>2</sub>-rich gas mixtures, the ionic properties of a CO<sub>2</sub>-enriched atmosphere was also investigated. The electrical mobility of the ions in a CO<sub>2</sub>-enriched atmosphere was found to be about 0.56 times that found in a conventional air atmosphere, due to the higher mass of CO<sub>2</sub> gas compared to that of air. The low electrical mobility of ions resulted in a low corona current under CO<sub>2</sub>-enriched conditions. The collection efficiency of particles in a CO<sub>2</sub>-rich atmosphere for a given power consumption was thus somewhat lower than that found in air, due to the low quantity of particle charging in CO<sub>2</sub>-enriched air. At the same time, higher temperatures led to the higher electrical mobility of ions, which resulted in a greater collection efficiency for a given power. The presence of a negative corona also led to a greater collection efficiency of particles in an ESP than that achieved for a positive corona.

## 2008

Han, B., Hudda, N., Ning, Z., and Sioutas, C., 2008, "Enhanced unipolar charging of concentration-enriched particles using water-based condensational growth," *Journal of Aerosol Science*, **39**(9): 770–784

A novel methodology with high efficiency for charging fine and ultrafine particles was developed and evaluated. The technique has been realized by combining the versatile aerosol concentration enrichment system (VASES) developed by our group and a newly developed unipolar charger equipped with carbon fiber ionizers. Particles are grown to super-micron droplets via condensation of ultrapure deionized water and then concentrated by virtual impaction in the VACES. The grown droplets are charged with negligible ozone generation in the carbon fiber unipolar charger, and subsequently dried to original

particle size distribution using a diffusion dryer, while preserving the acquired charges. This new methodology was investigated for different particle sizes, chemical compositions and concentrations. The number of charges on particles was highly dependent on particle concentration as well as particle size; larger particles and smaller particle concentrations led to a greater number of charges per particle. The average electrical mobility of the charged particles exceeded  $1.4 \times 10^{-1} \text{ cm}^2/(\text{Vs})$ , corresponding, on average, to more than 250 elementary charges per particle for concentrations in the range of  $1.4\text{-}1.9 \times 10^5$  particles/ $\text{cm}^3$  for ammonium nitrate, ammonium sulfate and glutaric acid. A lower number of charges per particle was observed for PSL particles, probably due to their hydrophobic nature, and thus, number of charges per particle averaged to about 98, 191 and 349 for polystyrene latex (PSL) particles of 99, 130 and 170 nm, respectively, at particle concentrations of  $1.1\text{-}1.7105$  particles/ $\text{cm}^3$ . Even in the case of PSL, these charges far-exceed those obtained by traditional corona chargers.

Orsini, D. a., Rhoads, K., McElhoney, K., Schick, E., Koehler, D., and Hogrefe, O., 2008, "A Water Cyclone to Preserve Insoluble Aerosols in Liquid Flow—An Interface to Flow Cytometry to Detect Airborne Nucleic Acid," *Aerosol Science and Technology*, **42**(5): 343–356

A miniature cyclone was designed to gently capture fine aerosols into a continuous liquid flow. The geometry of the cyclone was designed so that the friction of the turning air swirls a 100  $\mu\text{l}$  volume of water at the base of the cone, creating a standing liquid vortex which coats the inside deposition surface. The collection efficiency of the cyclone was characterized as a function of insoluble particle size, both in stand-alone operation and preceded by aerosol growth by water vapor condensation. The aerosol growth lowered the smallest collected particle size and created synonymous sample-into-substrate material conditions at the point of impact. The cyclone collection efficiencies were higher than 88% for the fluorescent polystyrene latex bead diameter sizes 50–3000 nm. The cyclone was further interfaced to a flowcytometer to detect airborne nucleic acid (as a virus test aerosol) in the cyclone sample flow. The flow cytometer, which is commonly used for single cell identification via fluorescence, was modified to accept a continuous sample flow (nominal  $60 \mu\text{l min}^{-1}$ ) from the cyclone for real-time detection. A rod-shaped plant virus (Tobamovirus) and a protein-enveloped insect virus (Baculovirus) were aerosolized, collected by the cyclone, and stained inline using the nucleic acid dyes SYBR Green I, SYTO-9, and SYTO-24 (Molecular Probes, Inc.). In addition, an Environmental Scanning Electron Microscope (ESEM) was used to confirm the collection of single virus particles and qualitatively evaluate the degree to which the aerosolization and collection process affected the integrity of the virus.

## 2006

Zhu, Yifang; Yu, Nu; Kuhn, Thomas; Hinds, William, 2006, "Field Comparison of P-TRAK and Condensation Particle Counters," *Aerosol Science and Technology*, **40**(6):422–430

The P-TRAK ultrafine particle counter is a portable version of a condensation particle counter (CPC). Both instruments detect particle number concentrations in real time but have different detection limits. The P-TRAK has been widely used for indoor air quality evaluation and aerosol research. However, there is very limited information about the reliability and precision of this instrument and its comparability with other similar instruments. The purpose of this study was to compare a P-Trak<sup>®</sup> Ultrafine Particle Counter with a standard CPC and evaluate its applicability to ambient air monitoring. This study was carried out near the Interstate 405 freeway (I-405) in Los Angeles. Measurements were made at increasing distances from the freeway on both sides at night as well as inside and outside of two 2-bedroom apartments located near the freeway. A CPC and a Scanning Mobility Particle Sizer (SMPS) spectrometer were collocated with two P-Trak counters and measurement results compared. In general, higher correlations were observed between P-Trak counter and CPC data for indoor measurements than outdoor. The highest P-Trak counter and CPC correlation ( $r^2 = 0.9385$ ) was detected inside Apartment 2, which is located farther away from the freeway than Apartment 1. The poorest correlation occurred at 30 m downwind from the freeway. In that case, the P-Trak counter reported about 25% of ultrafine particle concentration that CPC did. A sigmoid (S-shape) function was fitted to observed P-Trak counter to CPC ratios and geometric mean diameters of the corresponding ultrafine particle size distributions. Overall, we concluded the P-Trak counter worked reasonably well when sampled indoor air. However, it has significant limitations in detecting freshly emitted ultrafine particles from vehicles. The P-Trak counter underestimated ultrafine particles especially for particles smaller than its activation size which was found to be approximately 250–30 nm. Caution must be given in interpreting data collected by P-Trak monitors near combustion sources.

## 2005

Asbach, C.; Fissan, H.; Kuhlbusch, T.A.J., 2005, "Investigation on the gas particle separation efficiency of the gas particle partitioner," *Atmospheric Environment*, **39**(40)

A gas particle partitioner (GPP, US patent 6,761,752 B2) that allows highly efficient separation of gas and particles with no effect on the thermodynamic conditions and substantially no change of the composition

of the gas has been developed. The GPP is a coaxial arrangement with inner and outer electrode. It utilizes a corona discharge to electrically charge the particles and a strong electric field in a separate unit to take them out of the sample flow. Several measures were taken to minimize an effect of the corona discharge on the gas composition. The GPP is designed such that when switched on, the sample flow is particle free, whereas when switched off, the sample flow contains a representative sample of the aerosol. The GPP as described in this manuscript was designed to meet the requirements for precise artifact correction with particle mass concentration monitors, such as the TEOM. This paper focuses on the gas particle separation efficiency of the instrument. The separation efficiency was determined for both, (ultra-) fine and coarse particles. The (ultra-) fine particles were generated with diameters ranging from 18 to 255 nm of polystyrene latex particles and their size distributions measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer. Coarse particles with diameters between 4.5 and 10.7  $\mu\text{m}$  were generated from a sodium chloride solution and characterized with an Aerodynamic Particle Sizer (APS) and a tapered element oscillating microbalance (TEOM). The investigations showed that the separation efficiency was very near 100% for all particles with diameters larger than 25 nm, whereas it decreased for smaller diameters. Particles of size 18 nm were separated from the gas flow with an efficiency of approximately 97%. Along with near 100% separation efficiency, the additional gas concentrations were 42 ppbV for  $\text{O}_3$  and 15 ppbV for  $\text{NO}_2$ . 13 Refs.

Biswas, Subhasis; Fine, Philip M.; Geller, Michael D.; Hering, Susanne V.; Sioutas, 2005, Constantinos "Performance Evaluation of a Recently Developed Water-Based Condensation Particle Counter," *Aerosol Science & Technology*, **39**(5):419–427

This study provides an intercomparison of the performance of a newly developed water-based condensation particle counter (WCPC) and a more widely used butanol-based CPC (TSI 3022A). Four test aerosols (ammonium nitrate, ammonium sulfate, adipic acid, and glutaric acid) were generated and tested in the laboratory before the instruments were deployed at four field locations (USC/downtown LA, I-710 Freeway, Pacific coast, and Los Angeles International Airport). Both instruments sampled the same incoming aerosol. Selected experiments utilized a differential mobility analyzer to select a particle size upstream of the CPCs. Evaluation of performance was based on the response of the instruments to varying particle composition, concentrations, and size. The results indicated good correlation between the two CPCs, with  $R^2$  values ranging from 0.74–0.99. Good agreement was found between the two instruments for particle concentrations between 0 and 40,000 particles/ $\text{cm}^3$ , with WCPC/TSI 3022A ratios between 0.8 and 1.2. Due to differences in the photometric mode calibration of these instruments, the ratio drops to 0.6–0.8 between 40,000–100,000 particles/ $\text{cm}^3$ . However, the ratio rises again for lab aerosols above 100,000 particles/ $\text{cm}^3$  to 1.0–1.1. Results of this evaluation show that the W-CPC is a reliable particle-counting technology for particle concentrations encountered downstream of a DMA as well as in some ambient environments (< 40,000 particles/ $\text{cm}^3$ ).

Chen, Chih-Chieh; Huang, Sheng-Hsiu; Kuo, Yu-Mei; Lin, Wane-Yun; Shih, Tung-Sheng; Weng, Yi-Mei, 2005, "Development of a size-selective inlet-simulating ICRP lung deposition fraction," *Aerosol Science and Technology*, **39**(5):437–443

A size-selective inlet made of polyurethane filter foam was designed and fabricated to simulate a portion of the ICRP respiratory deposition curve. A downstream aerosol measuring device then could be used to generate aerosol concentration data that represented the fraction reaching the respiratory system. This article introduces useful knowledge about porous foam penetration for particle size ranges below those reported in the previous studies. Different porosities of polyurethane foam filters were tested for aerosol penetration. Among the parameters operated in this work were (1) foam porosity (ppi), (2) filter thickness, (3) face velocity, and (4) packing density of the filter foams. Di-octyl ph-thalate was used as the test agent. A constant output atomizer and an ultrasonic atomizing nozzle were used to generate polydisperse submicrometer- and micrometer-sized particles, respectively. Aerosol concentrations and size distributions upstream and downstream of the filter foams were monitored by using a Scanning Mobility Particle Sizer (for particles with diameters smaller than 0.7  $\mu\text{m}$ ) and an Aerodynamic Particle Sizer spectrometer (for particles larger than 0.7  $\mu\text{m}$ ). The aerosol output was neutralized by a radioactive source. A lognormal-distribution curve with a mode of 0.25  $\mu\text{m}$  and a GSD of 6.2 was set as the primary target curve simulating the light-work ICRP deposition model. The results showed that the most penetrating size (also referred to as collection minimum) of the filter foams decreased upon increasing the foam porosity, packing density, and face velocity. In this work, the highest foam porosity and packing density we could acquire were 100 ppi and 0.2, respectively. By adjusting the face velocity, the most penetrating size was moved to 0.25  $\mu\text{m}$ , which happened to be the most penetrating size for ICRP light-work criterion. The whole aerosol penetration curve could further fit to the modified ICRP curve by adjusting the filter thickness. There are numerous ways to match the ICRP definition. This size-selective inlet becomes even more versatile if the auxiliary detector and vacuum system are operated under different flow rates to simulate light-to-heavy workloads. 19 Refs.

Girshick, S.L.; Hafiz, J.; Heberlein, J.; McMurry, P.H.; Mukherjee, R.; Renault, T.; Wang, X., 2005 "System for in situ characterization of nanoparticles synthesized in a thermal plasma process," *Plasma Chemistry and Plasma Processing*, **25**(5):439–453

We have designed a particle diagnostic system that is able to measure particle size and charge distributions from low stagnation pressure and high temperature (2000–4000 K) environments in near real time. This system utilizes a sampling probe interfaced to an ejector to draw aerosol from the low pressure chamber. Particle size and charge distributions are measured with a Scanning Mobility Particle Sizer spectrometer. A hypersonic impactor is mounted in parallel with the Scanning Mobility Particle Sizer to collect particles for off-line microscopic analysis. This diagnostic system has been used to measure size and charge distributions of nanoparticles (Si, Ti, Si-Ti-N, etc.) synthesized with our thermal plasma reactor. We found that the mean particle size increases with operating pressure and reactant flow rates. We also found that most particles from our reactor are neutral for particles smaller than 20 nm, and that the numbers of positively and negatively charged particles are approximately equal. Inc. 33 Refs.

Lee, D.; Park, K.; Zachariah, M.R., 2005, "Determination of the size distribution of polydisperse nanoparticles with single-particle mass spectrometry: The role of ion kinetic energy," *Aerosol Science and Technology*, **39**(2):162–169

We develop a method to determine size and size distribution (30–150 nm) of polydisperse nanoparticles using a laser ablation/ ionization time-of-flight single-particle mass spectrometer that extends the work first described by Reents and Ge. We found a composition independent "power law" dependence between the total peak area and original particle volume that enables one to determine particle volume directly from a particles mass spectrum. This power-law relationship suggests that some ions ablated and ionized from a particle are selectively lost during transport from the laser ablation/ionization region to the detector. A numerical calculation of ion trajectories shows that ion loss is highly dependent on the initial kinetic energy of ions. We show that the size-dependent energetic ions formed by the laser-particle interaction lead to power-law relationship between the cube root of peak area and particle diameter. The results demonstrate that particle size distributions measured with the mass spectrometer are in good agreement with those measured with a Scanning Mobility Particle Sizer spectrometer.

Lipsky, Eric M.; Robinson, Allen L., 2005, "Design and evaluation of a portable dilution sampling system for measuring fine particle emissions from combustion systems," *Aerosol Science and Technology*, **39**(6):542–553

The size and complexity of current dilution samplers is a major barrier to more wide-spread application of these systems for source characterization. A new, more portable dilution sampler has been designed to provide measurements consistent with the widely cited Caltech dilution sampler. Intercomparison experiments were performed using a diesel engine and wood stove to evaluate the comparability of the new design with a sampler based on the Caltech design. These experiments involved simultaneous operation of multiple dilution samplers from the same source. Filter based measurements included PM<sub>2.5</sub> mass, organic carbon, and elemental carbon emissions. Particle size distributions in the range from 10–480 nm were measured using a Scanning Mobility Particle Sizer spectrometer. The filter-based and integrated-total volume measurements made with the two designs are in good agreement. For example, the average relative bias between the two samplers of PM<sub>2.5</sub> mass emission rate measured with Teflon filters is 1%. Nucleation was intermittently observed in the sampler based on the Caltech design, but rarely observed in the new design. Significant discrepancies in total number emissions between the two samplers occurred during periods of nucleation. Experiments were also conducted to examine the effects of residence time on the diluted emissions. No changes in the filter-based or integrated volume measurements were observed with an additional 40-s residence time, indicating that phase equilibrium is established in the 2.5 s of residence time provided by the dilution tunnel. This conclusion is consistent with theoretical analysis. These results provide new insight into the effects of dilution sampling on measurements of fine particle emissions, providing important data for the ongoing effort of the EPA and ASTM to define a standardized dilution sampling methodology for characterizing emissions from stationary combustion sources. 20 Refs.

Riebel, Ulrich; Stommel, Yves Gorat, 2005, "A corona-discharge-based aerosol neutralizer designed for use with the SMPS-system", *Journal of Electrostatics*, **63**(6–10):917–921

The widely used Scanning Mobility Particle Sizer (SMPS) spectrometer for measuring submicron particles uses radioactivity to charge aerosol particles to a known charge distribution. Because of numerous restrictions and safety issues concerning radioactive sources, corona-discharge-based neutralizers are an attractive alternative. The newly developed electrical neutralizer consists of an aerosol chamber with an AC electrical discharge. Combining small residence times, high ion concentrations, small electric field strengths, discharge in the aerosol chamber itself and an automatic regulation of the ion input results in a very effective neutralizing device. Experiments with an initially uncharged aerosol have shown that charging of submicron particles to the bipolar equilibrium charge distribution was possible for



volumetric flow rates of up to 1.5 L/min and concentrations of at least  $5 \times 10^6$  1/cm<sup>3</sup>. Particle loss (>@3 nm) and particle production (>@3 nm) were not observed. 9 Refs.

Taishi, Tsuyoshi; Koyama, Tetsuji; Kwon, Soon-Bark; Seto, Takafumi; Sakurai, Hiromu, 2005, "New Measurement System of Nanoparticles in the Automobile Exhaust Gas," *JSAE Technical Paper No. 20055680*, *JSAE Autumn Conference*, Sept. 28–30, 2005, Toyoko, Japan

The aerosol measurement techniques such as Scanning Mobility Particle Sizer (SMPS) spectrometer are one of major methods to evaluate the size distribution of diesel nanoparticles. The charge distribution is important to reduce data from mobility distribution to size distribution. The radioactive sources as aerosol charge neutralizer has been widely used, however, the limit of ion generation rate and the difficulty in handling especially at outdoor measurement remained as problem. In this presentation, we developed a new measurement system for diesel nanoparticles using the microplasma aerosol charger. Characteristics and performance of the system for diesel nanoparticle measurement were reported.

Yoon, Young Jun; Cheevers, Sinead; Jennings, S. Gerard; O'Dowd, Colin D., 2005, "Performance of a venturi dilution chamber for sampling 3–20 nm," *Journal of Aerosol Science*, **36**(4):535–540

The transmission efficiency of a venturi mixing and dilution system was investigated with laboratory generated aerosol by comparison of two condensation particle counters (CPCs). The transmission efficiency exceeded 95% for particle sizes between 3 and 20 nm. The use of the diluter system is demonstrated through comparison with total concentrations derived from a nano-Scanning Mobility Particle Sizer (nSMPS) applied to measuring a rapidly changing atmospheric nucleation mode. The study indicates that the diluted-CPC sampler can resolve rapidly changing, and more intense peaks in excess of  $10^6$  cm<sup>-3</sup>, which are otherwise missing or under-sampled by the nSMPS.

## 2004

Chakrabarti, B.; Singh, M.; Sioutas, C., 2004, "Development of a near-continuous monitor for measurement of the Sub-150 nm PM mass concentration," *Aerosol Science & Technology*, **38**(S1):239–252

Population exposures to ambient particulate matter (PM) have recently received considerable attention due to the association between ambient particle concentrations and mortality. Recent toxicological studies suggest that ultrafine PM (diameter <100 nm) may be responsible for the observed health effects. However, even though ultrafine mass concentrations vary drastically over short time scales in the atmosphere, no monitor currently measures ultrafine PM mass continuously. The need for monitors that can perform ultrafine particle concentration measurement in shorter time intervals is of paramount importance to environmental health, as such a monitor can lead to substantial improvements in population exposure assessment to ambient ultrafine PM. In this study, a modified BAM (Beta Attenuation Monitor) is employed to measure near-ultrafine (i.e., <0.15  $\mu\text{m}$  or  $\text{PM}_{0.15}$ ) particulate mass concentration. The BAM is preceded by a 0.15  $\mu\text{m}$  cutpoint impactor, which is designed to have very low pressure drop. The BAM is operated in a 2 h cycle at a downwind receptor site in the Los Angeles Basin in Claremont. Among the other instruments collocated with the BAM are Scanning Mobility Particle Sizer (SMPS) spectrometer, an aerodynamic particle sizer (APS), and a Micro-Orifice Uniform Deposit Impactor (MOUDI). Our results indicate that the  $\text{PM}_{0.15}$  mass concentrations obtained by means of the modified BAM and MOUDI are in excellent agreement. The  $\text{PM}_{0.15}$  SMPS-to-BAM concentration ratio is generally smaller than 1 and follows a rather distinct diurnal profile, with a maximum towards the middle of the day and minima during the early morning and nighttime periods, presumably due to the classification of fractal-structured ultrafine particles in the accumulation mode by the SMPS. The lack of correlation between  $\text{PM}_{2.5}$  and  $\text{PM}_{0.15}$  mass concentrations further corroborates the need for developing monitors such as the modified BAM for the documentation of the short-term variation of ultrafine mass measurements. 24 Refs.

Fine, P. M.; Misra, C.; Singh, M.; Sioutas, C., 2004, "Development and Evaluation of A Compact Facility for Exposing Humans to Concentrated Ambient Ultrafine Particles," *Aerosol Science & Technology*, **38**(1):27–35

This article presents the development and evaluation of a very compact facility for exposing humans to concentrated ambient ultrafine particles ( $d_a < 0.15 \mu\text{m}$ ). The human ultrafine particle concentrator (UFPC) operates at an intake flow rate of 1200 liters per minute (LPM). The concentrator is preceded by an ultrafine impactor which separates the accumulation mode from ultrafine mode particles under a very low pressure drop (1.5 kPa), a feature that is essential in enabling human inhalation studies of ultrafine concentrated ambient particulates (CAP). A key feature of the UFPC is a new cooling system, consisting of a programmable refrigerated circulator, which produces the supersaturation that is necessary to grow ultrafine PM to supermicrometer sizes so that they can be concentrated by means of conventional virtual impaction. The new cooling system allows for entirely automated operation of the UFPC. The UFPC was characterized in field experiments, in which the concentration enrichment of ultrafine particles was

determined based on their number and mass concentration as well as on chemical composition including elemental carbon (EC), inorganic ions (sulfate and nitrate), and polycyclic aromatic hydrocarbons (PAH). Tests were conducted at minor-to-total flow ratios varying from 2.5-5% (hence at minor flow rates between 30-60 LPM). Measurements with the Scanning Mobility Particle Sizer (SMPS) spectrometer showed a near-ideal increase in number concentrations (corresponding to the ratio of total-to-minor flow rate) of ultrafine particles after enrichment. The concentration enrichment was uniform across the entire particle diameter range of 15-660 nm. Similar results were obtained for EC and PAH concentrations (measured by an Aethalometer). Time-integrated filter-based tests, conducted to characterize the system for ultrafine PM mass and inorganic ion concentrations showed that the average enrichment factor was very close to the ideal values, indicating near-perfect collection efficiency with minimal particle losses. 21 Refs.

Johnson, Tim; Caldwell, Robert; Poecher, Arndt; Mirme, A.; Kittelson, David, 2004, "A new electrical mobility particle sizer spectrometer for engine exhaust particle measurements," *SAE 2004 World Congress and Exhibition*, March 8-11, 2004, Detroit, Michigan, USA

Electrical mobility has a long history as a tool for measuring the particle size of engine exhaust emissions. This paper gives a review of these methods as well as more current methods for making exhaust particle measurements. Each of the methods discussed has a limitation especially for making fast (sub-second) measurements. A new instrument is discussed that has been developed by TSI based on a technique developed over the last two decades by the University of Tartu - Estonia. A description of the instrument, the Engine Exhaust Particle Sizer™ (EEPS™) spectrometer, is given as well as engine dynamometer data showing a comparison between the current standards for engine exhaust measurements, the Scanning Mobility Particle Sizing (SMPS™) system and the Condensation Particle Counter (CPC). The EEPS spectrometer compares favorably with the SMPS spectrometer and CPC while providing sub-second response.

Ludwig, Chr.; Mohr, M., 2004, "On-Line Characterization of Aerosols Formed in a Jet Flow Condenser for Analytical Applications," *Paul Scherrer Institut Scientific Report 2003*, Volume V

With a jet flow condenser (JFC) aerosol particles can be generated from gases containing high boiling substances. This can be used for transferring these substances from the hot carrier gas into an analytical device. In this study we have investigated the particle number size distribution (NSD) produced in two JFCs with different geometry. Experiments were performed with elemental zinc as a model substance. Zinc has been volatilised in a tubular furnace and the particles generated in the JFC were characterized on-line by a Scanning Mobility Particle Sizer (SMPS) spectrometer.

## 2003

Chen, C.-C.; Huang, S.-H., 2003, "Loading characteristics of a miniature wire-plate electrostatic precipitator," *Aerosol Science & Technology*, 37(2):109-121

In this work, in order to investigate the particle loading effects on the performance of an electrostatic precipitator (ESP), simultaneous measurements of the dust cake thickness accumulated on the collection plates, ESP's collection efficiency, corona discharge characteristics, and ozone concentration were conducted experimentally. A laboratory scale single stage wire-plate ESP was used for the aerosol loading test. Two kinds of particulate matter, cement and aluminum oxide (Al<sub>2</sub>O<sub>3</sub>), were generated by using a Palas Powder Disperser. A displacement meter was used to monitor the dust cake thickness accumulated on the collection plates. A Scanning Mobility Particle Sizer was used to measure the particle size distribution and number concentration upstream and downstream of the ESP. Ozone generated by the ESP was sampled 20 cm downstream of the ESP exit and measured with an ozone analyzer. The Dioctyl Phthalate (DOP) was also used as a liquid challenge agent in order to investigate the loading effects of liquid particles on the ESP performance. The results showed that when challenged with cement particles, the ion current decreased with increasing dust cake thickness under a constant electrical field strength. Moreover, the collection efficiency and ozone generated by corona discharge decreased as the loading test progressed. For example, when the dust layer was about 5 mm in thickness, the output current and the ozone concentration decreased about 33 and 44%, respectively, and the collection efficiency (300 nm particle) decreased about 4% at a fixed electrical field strength of -4.2 kV/cm. However, the ion current increased as aluminum oxide particles deposited on the collection plates. The increase in ozone concentration and aerosol penetration was mainly due to the occurrence of back corona, evidenced by the existence of the caves on the surface of the dust layers. In the case of testing with cement particles, the ion current rises after about 20 min of loading test and then decreases with time, while ozone concentration increases synchronously.

Johnson, T; Caldow, R; Poecher, A; Mirme, A; Kittelson, D, 2003, "An Engine Exhaust Particle Sizer™ spectrometer for transient emission particle measurements," *9th Diesel Engine Emissions Reduction (DEER) Workshop 2003*, Newport, RI (US), 08/24/2003—08/28/2003

There has been increased interest in obtaining size distribution data during transient engine operation where both particle size and total number concentrations can change dramatically. Traditionally, the measurement of particle emissions from vehicles has been a compromise based on choosing between the conflicting needs of high time resolution or high particle size resolution for a particular measurement. Currently the most common technique for measuring submicrometer particle sizes is the Scanning Mobility Particle Sizer (SMPS™) system. The SMPS system gives high size resolution but requires an aerosol to be stable over a long time period to make a particle size distribution measurement. A Condensation Particle Counter (CPC) is commonly used for fast time response measurements but is limited to measuring total concentration only. This paper describes a new instrument, the Engine Exhaust Particle Sizer™ (EEPS™) spectrometer, which has high time resolution and a reasonable size resolution. The EEPS spectrometer was designed specifically for measuring engine exhaust and, like the SMPS system, uses a measurement based on electrical mobility. Particles entering the instrument are charged to a predictable level, then passed through an annular space where they are repelled outward by the voltage from a central column. When the particles reach electrodes on the outer cylindrical (a column of rings), they create a current that is measured by an electrometer on one or more of the rings. The electrometer currents are measured multiple times per second to give high time resolution. A sophisticated real-time inversion algorithm converts the currents to particle size and concentration for immediate display.

Lapuerta, M; Armas, O; Gomez, A, 2003, "Diesel particle size distribution estimation from digital image analysis," *Aerosol Science and Technology*, **37**:369–381

One of the most serious problems associated with Diesel engines is pollutant emissions, especially nitrogen oxides and particulate matter. However, although current emissions standards in Europe and America with regard to light vehicles and heavy duty engines refer to the particulate limit in mass units, there has been increasing concern of late to know the size and number of particles emitted by engines. This interest has been promoted by the latest studies about the harmful effects of particles on health and is enhanced by recent changes in internal combustion engine technology. This study is focused on the implementation of a method to determine the particle size distribution that could be appropriate for the current methodology of vehicle certification in Europe. This method uses an automated Digital Image Analysis Algorithm (DIAA) to determine particle size trends from Scanning Electron Microscope (SEM) images of filters charged in a partial dilution system used for measuring specific particulate emissions. The experimental work was performed on a stationary electric generation direct injection Diesel engine with 0.5 MW (671 hp) rated power, which is considered as a typical engine in middle power industries. Particulate size distributions obtained using DIAA were compared with distributions obtained using an Optical Particle Counter (OC) and a Scanning Mobility Particle Sizer (SMPS) spectrometer, the latter currently considered as the most reliable technique. Although the number concentration detected by this method does not represent the real flowing particle concentration, the algorithm gives a fair reproduction of the trends observed with on-line techniques (SMPS and OC) when the engine load is varied.

Singh, M.; Misra, C.; Sioutas, C., 2003, "Field evaluation of a personal cascade impactor sampler (PCIS)," *Atmospheric Environment*, **37**(34):4781–4793

This paper presents the field evaluation of a personal cascade impactor sampler (PCIS). PCIS is a miniaturized cascade impactor, consisting of four impaction stages, followed by an after-filter. Particles are separated in the following aerodynamic particle diameter ranges: <0.25, 0.25-0.5, 0.5-1.0, 1.0- 2.5 and 2.5-10 micro m. The PCIS operates at a flow rate of 9 liters per minute (l/min) using a very high efficiency, battery-operated light pump at a pressure drop of 11 in H sub 20 (2.7 kPa). For field evaluation, the PCIS was collocated with other samplers including the micro-orifice uniform deposit impactor (MOUDI), Scanning Mobility Particle Sizer (SMPS) spectrometer and aerodynamic particle sizer (APS) in Los Angeles and Claremont, CA. PCIS and MOUDI agree very well for coarse particulate matter (PM) (PM<sub>2.5-10</sub>) mass concentrations. The fine PM (PM<sub>2.5</sub>) mass as measured by PCIS is in excellent agreement with SMPS-APS measurement (1.02 times) and slightly higher ( 1.1 times) than the MOUDI measurements. Time-integrated (size fractionated) PM<sub>2.5</sub> mass, inorganic ions (nitrate and sulfate), elemental carbon (EC) and organic carbon (OC) concentrations obtained with PCIS and MOUDI were found to be in very good agreement with few differences in the <0.25 micro m size fraction, especially for OC and nitrate measurements. Near-continuous and size fractionated PM<sub>2.5</sub> nitrate and total carbon measurements by PCIS and MOUDI using the ADI and Sunset labs monitors are in close agreement for all size fractions, indicating that any differences between MOUDI and PCIS measurements for time-integrated data might be due to artifacts associated with long-term sampling and not to differences in individual cut-points. The performance of the PCIS was also evaluated in wind tunnel tests at wind speeds up to 8 km/h. These tests showed that particle sampling efficiency and separation characteristics of the PCIS are unaffected by the wind speeds for particles up to 10 micro m in aerodynamic diameter.

## 2002

Chen, C.-C.; Huang, S.-H.; Kuo, Y.-M.; Wu, C.-H., 2002, "Aerosol penetration through silica gel tubes," *Aerosol Science & Technology*, **36**(4):457-468

Silica gel is commonly used by industrial hygienists to collect gases and vapors in the work place, in particular air contaminants with high polarity. The collected air pollutants are then treated and analyzed to identify their type and to determine the concentration using various methods and instrumentations. In addition to collection of gaseous pollutants, the silica gel tubes are also used for acid mist collection according to the listed official analytical methods (e.g., NIOSH method 7903 and OSHA method ID-165SG). However, the filtration characteristics of silica gel tubes have not been thoroughly investigated. A constant output aerosol generator and an ultrasonic atomizing nozzle were used to generate submicrometer-sized and micrometer-sized aerosol particles, respectively. A Scanning Mobility Particle Sizer and an aerodynamic particle sizer were used to measure particles smaller and larger than 0.6  $\mu\text{m}$ , respectively. Potassium sodium tartrate and dioctylphthalate were used as the solid and liquid test agents, respectively. Two types of SKC silica gel tubes (Cat No. 226-10 and 226-10-03) were examined for aerosol penetration, air resistance, and loading characteristics. The results show that the aerosol penetration through the silica gel tubes could be as high as 80% at the penetration maximum (or collection minimum) under the normal sampling flow of 0.5 L/min, well within the inertial impaction dominated region. Two glass wool plugs and one urethane plug between sorbent sections and at the back end of the SKC 226-10 contributed about 22% of the total air resistance, and the remaining 78% of the air resistance was caused by the silica gel. When the filtration efficiency by these separators was deduced, the aerosol penetration at the most penetrating size was as high as 90%. The aerosol penetration increased and the penetration curve shifted to a smaller particle size as the sampling flow increased. However, this increase in aerosol penetration of particles smaller than the penetration maximum reached a maximum and then decreased as the sampling flow was increased beyond 1.5 L/min (equivalent filtration velocity of 93 cm/s), a clear evidence of inertial impaction surpassing the diffusion deposition. As a result, the use of silica gel tubes for acid mist collection may not be appropriate if the behavior of the complete aerosol size distribution is not considered as part of the assessment of these devices. 17 Refs.

## 2001

Abbey, E.; Petersen, E.; Rickard, M.; Traum, M.; Welle, R., 2001, "A new shock-tube facility for studying combustion phenomena in mixtures containing condensed species," *Proceedings of the National Heat Transfer Conference*, **1**:947-955

Two shock tubes at The Aerospace Corporation have been refurbished for the study of fundamental processes related to the formation and reaction of solid and liquid aerosols at elevated temperatures. One shock tube is intended for the study of powdered aerosols and other condensed-phase species, while the second tube is designed for the study of gas-phase reactants. Recent upgrades and additions include a new high-vacuum system, an optimized velocity-detection scheme, a computer-based data acquisition system, and new techniques and procedures for handling experiments involving gas/powder mixtures. Diagnostic techniques include laser extinction for particle volume fraction and size, optical pyrometry for particle temperature, laser-light scattering for particle size and number density, temporally and spectrally resolved emission from gas-phase species, and a Scanning Mobility Particle Sizer spectrometer. Details on the setup and operation of the shock tube and diagnostics for the study of heterogeneous combustion processes are given, and sample results are presented. 24 Refs.

## 2000

Robert M. G.; Peter O. W., 2000, "Laser-induced incandescence and elastic-scattering measurements of particulate-matter volume fraction changes during passage through a dilution tunnel," *Report, Sandia National Labs*. 11 page(s), Report No. SAND2000-8736C

Modern diesel engines produce far less mass of particulate matter than their predecessors, but this advance has been achieved at the expense of a significant increase in the number of sub-micron sized particles. This change in soot morphology has created the need for new instrumentation capable of measuring small volumes and sizes of particulate matter in a reasonable period of time, and preferably in real-time. Laser-induced incandescence and laser elastic scattering are complementary techniques suitable for this task. Optical measurements are presented for a diesel engine exhaust and compared with measurements performed using a Scanning Mobility Particle Sizer spectrometer. This study investigates the effects of exhaust dilution and temperature control of the sampling system. It is also shown that laser-induced vaporization of low temperature volatile material is a potentially valuable technique for measuring the volatile component of exhaust particulate matter.

## 1999

Kaufman, S. L.; Caldow, R.; Dorman, F. D.; Irwin, K. D.; Poecher, A., 1999, "Conversion efficiency of the TSI Model 3480 electrospray aerosol generator using sucrose," *Journal of Aerosol Science*, **30**(S1):S373-S374

In this study, measurements of the overall conversion efficiency from solute to aerosol for the Model 3480 Electrospray Aerosol Generator (EAG) were carried out to learn whether further increases in aerosol yield would be possible. The measurements enabled to determine the size-distribution and concentration of the aerosol. Data was obtained with which to estimate the aerosol concentrations provided by the EAG. 3 Refs.

## 1998

Hilton, M.; Black, J. D., 1998, "Detection of soot particles in gas turbine engine combustion gases using non intrusive FTIR spectroscopy," *Proceedings of Society of Photo-Optical Instrumentation Engineers*, Bellingham, WA, 3493:20-31

Fourier Transform IR (FTIR) spectroscopy for making non-intrusive measurements of gas turbine exhaust gases and laser-induced incandescence (LII) for measuring soot content are being evaluated in EU Brite EuRam project AEROJET. Soot concentrations in modern air-engine exhausts are very low (typically less than 0.2 mg/cu m) with mean particle sizes less than 100 nm. The standard extractive filter paper soot measurement gives results expressed in terms of SAE smoke number, typically less than 10 SAE for modern engines. IR studies of exhaust gases from a modified air engine that produces high levels of particulates showed a broadband baseline shift caused by soot which depended on engine running conditions. Higher levels of particulates were studied in the exhaust of a small liquid kerosene fuelled combustor sector rig using an FTIR spectrometer. The IR broadband background that depends on both soot particle size and number density was compared to combustion efficiency. Preliminary results showed that for smoke numbers less than 5, broadband emission increased as combustion efficiency decreased and unburnt hydrocarbon content increased. Results obtained at the same running conditions using LII, soot measured in an extracted sample using an optical obscuration smoke meter, particle size distributions using a Scanning Mobility Particle Sizer (SMPS) spectrometer, and FTIR spectrometer measurements are compared.

Li, Y.-T.; Koropchak, J. A., 1998, "Investigations of particle characteristics and carrier effects on particle beam LC-MS," *Instrumentation Science & Technology*, **26**(4):389-407

We investigated the characteristics of the particles resulting from a thermospray nebulizer and two different desolvation systems for particle beam liquid chromatography-mass spectrometry (PB-LC-MS). The particle size distributions and shapes/morphologies were characterized using a Scanning Mobility Particle Sizer (SMPS) spectrometer and high resolution transmission electron microscopes (HRTEM), respectively. The carrier effects of ammonium acetate were evaluated with these particle characteristics and the PB-LC-ITMS responses. The addition of ammonium acetate changed the particle size distributions and shapes/morphologies, and also significantly increased the analyte transmission efficiency in both the SMPS and the PB LC-ITMS. However, ammonium acetate did not cause increases in particle sizes and particle size decreases were even observed in some cases. The observed particle shapes and morphologies were different for caffeine and 3,3 prime -dimethoxybenzidine, and also depended on the desolvation methods. These results were interpreted to be due to the combined carrier effects of fragmenting large aerosols, increasing analyte solubility, and neutralizing the charged particles. The fragmentation of large aerosol particles via the addition of the semivolatile carrier was also hypothesized as a major process reducing the gravitational loss of aerosols and increasing the analyte transport efficiency. In the other set of experiments based on UV absorption, the mass balance of caffeine was evaluated. Both the SMPS and the mass balance data suggested that the porous membrane desolvation system could discriminate the small particles which, at least in part, resulted in the poor analyte transmission efficiency and nonlinear calibration behaviors of PB LC-MS. (Author abstract) 31 Refs.

## 1997

Chen, D.-R.; Pui, D. Y. H., 1997, "Experimental investigation of scaling laws for electrospraying: Dielectric constant effect," *Aerosol Science & Technology*, **27**(3):367-380

Experiments were performed to investigate the effect of liquid dielectric constant on existing scaling laws for the electrospraying process. The variations of the droplet size and the emitted current were measured as a function of the dielectric constant for the electrospray operating in the cone-jet mode. Eight different solvents with dielectric constants,  $\kappa$ , ranging from 12.5 to 182 were tested. The residue particle size distributions were measured using a TSI Scanning Mobility Particle Sizer, (SMPS) spectrometer. The produced liquid droplet sizes were then calculated from the known solution concentrations. The results show that: (1) For the produced droplet size,  $D[d]$ , experimental data are in agreement with the scaling

law ( $G(\kappa) = 1.66 \kappa^{-1} [1/6]$ ) proposed by Gañán-Calvo et al. (1994) for solvents with high dielectric constants. The derivation in low dielectric constant cases may be that the assumption of Gañán-Calvo (1994) on the characteristic length may not hold for the present system; (2) for the emitted current,  $I$ , experimental data deviate from those given by Fernández de la Mora and Loscertales (1994) except for the case of benzyl alcohol. The difference may be partially explained by the use of different electrolytes. A larger deviation is found in comparing with the equation,  $f(\kappa) = 6.46\kappa^{1/4}$ , provided by Gañán-Calvo et al. (1994). The reasons may be due to the inapplicable characteristic length assumption and the role of ion drifting current in the total emitted current. Based on the experimental data collected, some problems are pointed out regarding the criteria proposed in the previous studies for predicting the minimum liquid feed rate. A formula is given based on these data to estimate the maximum feed rate and to explain the observations reported in Chen et al. (1995).

## 1992

Ylatalo, Sampo I.; Kauppinen, Esko I.; Hautanen, Jukka; Joutsensaari, Jorma; Ahonen, Petri; Lind, Terttaliisa M.; Jokiniemi, Jorma K.; Kilpelainen, Markku, 1992, "On the Determination of Electrostatic Precipitator Efficiency by Differential Mobility Analyzer," *Aerosol Science*, **23**(S1):S795–S798

In order to determine penetration curve of the electrostatic precipitator (ESP) as a function of aerosol particle diameter in the range of 10-1000 nm measurement series were carried out in real scale power plant conditions. Differential mobility particle sizing (DMPS) system was used to measure the particle mobility distributions before and after ESP. MICRON-algorithm (constrained regularization) was used to invert mobility distributions to the corresponding number distributions. Penetration curve was calculated from the measured number distributions.

## 1991

Adachi, Motoaki; Romay, Francisco J.; Pui, David Y. H., 1991, "High-Efficiency Unipolar Aerosol Charger Using a Radioactive Alpha Source," *Journal of Aerosol Science*, **23**(2):123–127

A new design of a unipolar charger has been developed. The charger consists of a radioactive source placed between two screen electrodes enclosed by a Plexiglass tube. The electric field in the charger is aligned with aerosol flow. The new charger is capable of charging ultrafine aerosols efficiently and with low particle losses. The charger was evaluated thoroughly both theoretically and experimentally. The basic equations for the charging process in this charger were numerically solved for monodisperse, ultrafine aerosols under various operating conditions. From the calculation results, the particle charge level and loss rate within the charger were found to depend on two dimensionless parameters defined in the paper. In the experiment, the particle charge level and penetration rate were measured for particle sizes of 10-30 nm, operating voltages of 3-9 kV, flow rates of carrier gas of 2-5.21 min<sup>-1</sup>, and pressures of 0.46 and 1 atm. The experimental results which were obtained under similar conditions estimated by the dimensionless parameters were found to agree with the theoretical predictions. The charger gave a unit charge to 50% of the 10 nm diameter particles with 20% particle losses. The charger was found to work also under low pressure as a high-efficiency charger.

Kauppinen, Esko I., 1991, "On the Determination of Continuous Submicron Liquid Aerosol Size Distributions with Low Pressure Impactors," *Aerosol Science and Technology*, **16**(3):171

The aspects associated with the determination of continuous submicron aerosol size distributions using multijet low pressure impactors have been studied. Multiple sets of error free and noisy simulated data sets have been inverted and impactors have been compared with the differential mobility particle size analyses (DMA) method using well defined, laboratory generated liquid oleic acid aerosols tagged with ammonium fluorescein. Impactors included in this study were the Berner-type impactor HAUKE 25.015 (BLPI), modified University of Washington Mark 5 impactor (KLPI) and the impactor designed at the University of Florida (LLPI), as described by Hillamo and Kauppinen (1991), Kauppinen and Hillamo (1989) and Vanderpool et al. (1990), respectively. The inversion of simulated error free impactor data (i.e., data with perfect kernel functions) for unimodal submicron aerosols with small (2.5%) stage mass error estimate yields results very close to input distributions, when the method based on constrained regularization (Wolfenbarger and Seinfeld, 1990, 1991) is used in the inversion. When the error estimate is increased, inverted spectra are flattened. However, they remain clearly unimodal. When normally distributed random error is added to the data and the error estimate for each data point equals to the standard deviation of the random error, the fraction of bi- and trimodal inverted spectra increases with increasing the random error level and the asymmetry of the kernel functions. When the random error level and data error estimates are equal to or smaller than 10%, inverted spectra are mainly unimodal close to input distribution for both error free and noisy data. The inversion of impactor data from the detailed laboratory experiments (i.e., the data with real kernel functions) indicate, that only BLPI kernel functions are accurate enough to yield unimodal distributions close to those measured with DMA. When the stage mass error estimate is increased beyond the stage mass determination error, unimodal spectra

also for KLPI and LLPI are found. The decrease of the BLPI stage mass error estimate below the experimental error increase the agreement with DMA results. In most cases the error estimate for BLPI stage masses can be decreased to 2.5%, indicating the validity of both BLPI submicron kernel functions and fluorometric method used to determine stage mass concentrations.

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# Nanotechnology

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## 2013

Antonsson, E., Bresch, H., Lewinski, R., Wassermann, B., Leisner, T., Graf, C., Langer, B., and Rühl, E., 2013, "Free nanoparticles studied by soft X-rays," *Chemical Physics Letters*, **559**: 1–11

The use of nanoparticle beams for the preparation and characterization of isolated nanoscopic matter is reviewed. It is a general preparation scheme which is shown to be of broad use, whenever the intrinsic properties of nanoscopic matter without any interactions with other particles or substrates are of interest. The size regime spans a few nanometers up to the micron sizes, wherein a large variety of substances and materials can be studied. Similarly broad are the detection schemes and properties, which can be investigated by this approach. This review covers various properties of isolated nanoparticles, including their size, surface properties, optical constants, as well as their local electronic structure.

Pawar, A. A., and Venkataraman, C., 2013, "Pulse-Heat Aerosol Reactor (PHAR): Processing Thermolabile Biomaterials and Biomolecules into Nanoparticles with Controlled Properties," *Aerosol Science and Technology*, **47**(4): 383–394

This work addresses the challenge of processing thermolabile biomaterials and biomolecules into nanoparticles without compromising structural integrity and activity. Control of size, structure, and crystallinity of nanoparticles is desirable for drug targeting and controlled release applications. An innovation using pulse-heat aerosol processing is demonstrated through a design of a pulse-heat aerosol reactor. Nanoparticle aerosol lipid matrices (NALM), of stearic acid, were synthesized under different processing conditions, obtained through pulse heating (fixed duration, three heat-pulse levels) and solvent selection. Operation with continuous heating resulted in ill-conditioned, multimodal size distributions. Pulse heating resulted in the synthesis of NALM with mean mobility diameter in the range of 56–183 nm and narrow unimodal size distributions (geometric size distribution, GSD=1.5–1.7). Under higher pulse levels, particles with larger mean mobility diameter were formed, which had shell-type structures, compared with smaller, solid particles under operation with low level or no heating. NALM with different degree of crystallinity were produced under different processing conditions, at which different drop temperature is expected to result. The activity of glucose oxidase enzyme was preserved, when subjected to high pulse-heat (gas temperature of 110°C) aerosol processing. These results establish pulse-heat aerosol processing as a single-step, continuous method to process heat-sensitive biomaterials and biomolecules into nanoparticles with controlled properties, while avoiding thermal damage.

## 2012

Byeon, J. H., and Kim, J.-W., 2012, "Aerosol fabrication of thermosensitive nanogels and in situ hybridization with iron nanoparticles," *Applied Physics Letters*, **101**(2): 023117

Collison atomized n-isopropylacrylamide (NIPAM) droplets were thermally treated with different furnace wall temperatures to form nanogels in an aerosol state. The size of the aerosol nanogels decreased from 40.3nm to 32.8 nm by increasing the temperature from 40C to 90C due to the coil-to-globule transition of the NIPAM. A serial reactor consisting of a spark generator coupled to a collision atomizer was further employed to efficiently (>90% in production yield) fabricate biocompatible (78.8% in cell viability)-magnetic (30.3emug<sup>-1</sup> in saturation magnetization) hybrid nanogels of NIPAM and iron nanoparticles

Byeon, J. H., and Kim, Y.-W., 2012, "Aerosol copper initiated core-shell nanoparticle synthesis and micropatterning," *New Journal of Chemistry*, **36**(11): 2184

We report the development of copper-silver core-shell nano-particles where the aerosol copper nanoparticles served as the seeds for the deposition of silver atoms on their surface. The copper particles were also electrostatically patterned on the substrate to fabricate silver micropatterns.

Li, L. I. N., Zuo, Z., Japuntich, D. A., and Pui, D. Y. H., 2012, "Evaluation of Filter Media for Particle Number, Surface Area and Mass Penetrations," *The Annals of Occupational Hygiene*, **56**(5): 581–594

The National Institute for Occupational Safety and Health (NIOSH) developed a standard for respirator certification under 42 CFR Part 84, using a TSI 8130 automated filter tester with photometers. A recent study showed that photometric detection methods may not be sensitive for measuring engineered nanoparticles. Present NIOSH standards for penetration measurement are mass-based; however, the threshold limit value/permissible exposure limit for an engineered nanoparticle worker exposure is not



yet clear. There is lack of standardized filter test development for engineered nanoparticles, and development of a simple nanoparticle filter test is indicated. To better understand the filter performance against engineered nanoparticles and correlations among different tests, initial penetration levels of one fiberglass and two electret filter media were measured using a series of polydisperse and monodisperse aerosol test methods at two different laboratories (University of Minnesota Particle Technology Laboratory and 3M Company). Monodisperse aerosol penetrations were measured by a TSI 8160 using NaCl particles from 20 to 300 nm. Particle penetration curves and overall penetrations were measured by Scanning Mobility Particle Sizer (SMPS) spectrometer, condensation particle counter (CPC), nanoparticle surface area monitor (NSAM), and TSI 8130 at two face velocities and three layer thicknesses. Results showed that reproducible, comparable filtration data were achieved between two laboratories, with proper control of test conditions and calibration procedures. For particle penetration curves, the experimental results of monodisperse testing agreed well with polydisperse SMPS measurements. The most penetrating particle sizes (MPPSs) of electret and fiberglass filter media were ~50 and 160 nm, respectively. For overall penetrations, the CPC and NSAM results of polydisperse aerosols were close to the penetration at the corresponding median particle sizes. For each filter type, power-law correlations between the penetrations measured by different instruments show that the NIOSH TSI 8130 test may be used to predict penetrations at the MPPS as well as the CPC and NSAM results with polydisperse aerosols. It is recommended to use dry air (<20% RH) as makeup air in the test system to prevent sodium chloride particle deliquescing and minimizing the challenge particle dielectric constant and to use an adequate neutralizer to fully neutralize the polydisperse challenge aerosol. For a simple nanoparticle penetration test, it is recommended to use a polydisperse aerosol challenge with a geometric mean of ~50 nm with the CPC or the NSAM as detectors.

## 2011

Abbas, Z., Holmberg, J. P., Hellström, A. K., Hagström, M., Bergenholtz, J., Hassellöv, M., and Ahlberg, E., 2011, "Synthesis, characterization and particle size distribution of TiO<sub>2</sub> colloidal nanoparticles," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, **384**(1-3): 254–261

Nanoparticles of controlled size, well defined shape, pure phase and of clean surfaces are ideal model systems to investigate surface/interfacial reactions. In this study we have explored the possibility of synthesizing TiO<sub>2</sub> nanoparticles in the size range of 7–20 nm under well controlled experimental conditions. A simple method based on the hydrolysis of TiCl<sub>4</sub> was used to obtain particles having surfaces free from organics. Stable dispersions of TiO<sub>2</sub> nanoparticles of various sizes were obtained by optimizing the reaction/dialysis time and temperature. The synthesized TiO<sub>2</sub> particles were found to be predominantly of anatase phase and narrow particle size distributions were obtained. The TiO<sub>2</sub> particles were characterized with respect to their phase, size and shape by X-ray diffraction (XRD) and transmission electron microscopy (TEM), respectively. Particle size distribution in a colloidal dispersion was obtained by the electrospray Scanning Mobility Particle Sizer (ES-SMPS) method and compared with an average particle size determined from dynamic light scattering (DLS). The average particle sizes obtained by the DLS and ES-SMPS methods were in good agreement, while a primary particle size of ~4 nm was found in X-ray diffraction irrespective of the particle size in solution. Early stages of the nucleation process were monitored by the ES-SMPS method. These results show that small particles of 4–5 nm are initially formed and it is highly likely that large particles are formed due to aggregation of primary particles.

Pawar, A. a., and Venkataraman, C., 2011, "Droplet-Phase Synthesis of Nanoparticle Aerosol Lipid Matrices with Controlled Properties," *Aerosol Science and Technology*, **45**(7): 811–820

Nanoparticle lipid matrices are being recently explored for controlled drug delivery applications. The degree of crystallinity of the lipid matrix governs drug loading capacity and release rates. Current methods of preparing nanoparticle lipid matrices, like high pressure homogenization and solvent-emulsification evaporation, offer limited or poor control over lipid crystallinity. The present work explores a droplet-phase aerosol synthesis method to prepare nanoparticle aerosol lipid matrices (NALM). Control of evaporation rate was used to achieve control over crystallinity of NALM. NALM were synthesized in an aerosol reactor using precursor solutions of stearic acid in organic solvents with widely different vapour pressures, resulting in different rates of evaporation. Mean mobility diameter, measured using a Scanning Mobility Particle Sizer (SMPS) spectrometer, ranged from 94–127 nm, with a unimodal distribution and geometric standard deviation of 1.7–1.9. Aerodynamic diameters measured using gravimetric analysis of samples collected in a micro-orifice cascade impactor, ranged from 423–608 nm. Relatively higher peak intensities and peak areas in X-ray diffractograms, and higher melting enthalpies in the differential scanning calorimetry (DSC) thermograms were obtained for NALM synthesized under lower evaporation rates, using lower vapour pressure solvents, similar to that of bulk stearic acid. In contrast, NALM synthesized at higher evaporation rates, using higher vapour pressure solvents consistently showed a lower degree of crystallinity, evidenced by blunt X-ray diffraction peaks and lower melting enthalpies in DSC analysis. These results suggest the formation of non-equilibrium crystal

structures within NALM, under the influence of faster evaporation rates, leading to a lower degree of crystallinity. The work demonstrates proof of concept of droplet-phase aerosol synthesis for preparing nanoparticle lipid matrices and for achieving control over their crystallinity.

Voigt, T., Ripperger, S., and Sachweh, B., 2011, "Controlled Generation of Nanoscale Organic Pigments by Adiabatic Expansion in Laval Nozzles and in Expanding Free Jets," *Aerosol Science and Technology*, **45**(12): 1456–1467

As organic pigments are produced mainly in wet chemical processes, a novel gas to particle process for generating organic nanoscale pigments is introduced. This process is based on the expansion of a nearly saturated gas flow from an overpressure regime to ambient pressure conditions. The expansion is achieved in a Laval nozzle and subsequently continues as an expanding free jet in an expansion chamber. This expansion chamber is equipped with an adjustable cooling system. Copper phthalocyanine and Paliogen Red, two organic pigments, were used as test materials. By controlling the temperature in the expansion chamber, particularly in the free jet, nanoscale pigments with high (needle shape) to low (grainy shape) aspect ratios are generated. Particle size distribution measurements as well as scanning electron microscopy and transmission electron microscopy analyses show that newly generated particles differ significantly in size and shape from the cubic-shaped bulk material. Particles with low aspect ratio are below 100 nm in all dimensions. Hence, specific control of temperatures in the expanding free jet offers a tool to produce nanoscale organic pigments with defined particle sizes.

## 2010

Byeon, J. H., and Kim, J.-W., 2010, "Production of carbonaceous nanostructures from a silver-carbon ambient spark," *Applied Physics Letters*, **96**(15): 153102

Using silver-carbon ambient sparks, hollow carbon nanospheres or multiwall carbon nanotubes were produced separately from carbon encapsulated silver nanoparticles ( $-1,400 \text{ K s}^{-1}$ ) during relatively slow ( $-800 \text{ K s}^{-1}$ ) or fast ( $-2,900 \text{ K s}^{-1}$ ) cooling process. Different cooling processes (i.e., different exposures within high temperature) caused the formation of different carbon precipitates in the process of silver mediated graphitization: for  $-2,900 \text{ K s}^{-1}$  and  $-1,400 \text{ K s}^{-1}$ , respectively, obtained tubelike and sphere (encapsulated and hollow)-like carbonaceous nanostructures

Hung, D. V., Tong, S., Nakano, Y., Tanaka, F., Hamanaka, D., and Uchino, T., 2010, "Measurements of particle size distributions produced by humidifiers operating in high humidity storage environments," *Biosystems Engineering*, **107**(1): 54–60

Particle size distributions of nano- and ultrasonic mists, were measured under storage environments with high humidity. Mists generated using nanomist humidifiers have average particle diameters  $< 100 \text{ nm}$ . whilst produced by ultrasonic humidifiers have particle diameters in the range  $200 \text{ nm}$ - $1250 \text{ nm}$ . Nanomist humidifiers are thought to be able to provide improved capability for generating ultrafine mists for high humidification. Experiments were performed inside the container equipped with humidifiers, a refrigerator and a system controller. A Scanning Mobility Particle Sizer (SMPS) spectrometer and a light scattering spectrometer were employed to measure particle sizes. Size distributions were measured under ambient air and controlled temperature and humidity ( $5 \text{ C}$  and  $80\%$ ,  $90\% \text{ RH}$ ), at time intervals of 2, 4 and 18 h. A modified nanomist generator operated at frequencies of 40, 50, 60 Hz. Results showed that the number mode of the particle size distribution produced by the nanomist humidifier was  $64 \text{ nm}$ , about 3 times smaller than that of ultrasonic mist. The particle concentration of the nanomist humidifier was about 4 times finer than that of ultrasonic mist. The number concentration of the nanomist humidifier depended greatly on the generator frequency and humidity. The Nukiyama-Tanasawa equation estimated the size distribution better than the Rosin-Rammler equation. It appears that the nanomist can provide high humidification with reduced wetting of the stored produce, or the corrugated cardboard boxes used to pack the produce, thereby assisting in the control of fresh produce deterioration and microorganism growth.

Lee, S.-B., Jung, J. H., Bae, G.-N., and Lee, D.-J., 2010, "In-Situ Characterization of Metal Nanopowders Manufactured by the Wire Electrical Explosion Process," *Aerosol Science and Technology*, **44**(12): 1131–1139

In order to understand the size distributions of metal nanopowders inside manufacturing equipment operated at elevated pressures, a Scanning Mobility Particle Sizer is used to carry out in-situ measurements of metal nanopowders manufactured by the wire electrical explosion process. A pressure reducer and rotating disk diluter are used for conditioning metal nanopowder samples appropriate for real-time aerosol instruments operated at atmospheric pressure. Based on measurement data collected downstream of the evaporation chamber, the production of metal nanopowders shows good stability and uniformity for a total number concentration of approximately  $5 \times 10^7 \text{ particles/cm}^3$ , and a unimodal size distribution with a mean diameter of approximately  $170 \text{ nm}$ . Using an aerosol electrometer and two sets of electrostatic classifiers, positively charged particles slightly outnumber negatively charged particles.

The performance of the rotating disk diluter is confirmed by comparing the size distributions of metal nanopowders diluted with five different dilution factors, ranging from 235 to 2500. SEM and TEM image analysis indicates that most metal nanopowders manufactured by this process consist of aggregated particles, and their size distributions obtained from SEM images are similar to those measured by the SMPS. The changes in particle size distribution at each stage of the manufacturing process, including the evaporation chamber, trap buffer, cyclone, and mesh filter, are also monitored using the above in-situ monitoring system. The resulting in-situ measurement data can be used for design modifications of equipment, as well as for investigating the sources of nanopowder release to the workplace environment.

Seto, T., Furukawa, T., Otani, Y., Uchida, K., and Endo, S., 2010, "Filtration of Multi-Walled Carbon Nanotube Aerosol by Fibrous Filters," *Aerosol Science and Technology*, **44**(9): 734–740

Filtration efficiency of multi-walled carbon nanotube (MWCNT) aerosol by fibrous filter was evaluated experimentally. Mono-mobility test aerosols with electrical mobility diameter of 100, 200, and 300 nm were generated by the atomization of MWCNT aqueous suspension followed by mobility classification with a differential mobility analyzer (DMA). By analyzing the shape of classified aerosol particles under a scanning electron microscope, it was found that the DMA-classified 300 nm particles were fibrous in shape and had uniform diameter of about 60 nm and length of 2.1 micrometer. On the other hand, 100 nm and 200 nm particles contained a fairly large amount of multiply charged fibrous particles with a larger diameter. These test aerosols were challenged to a medium performance fibrous filter at various filtration velocities. As a result, fibrous particles were captured by fibrous filter at a higher collection efficiency than the spherical particles with the same mobility. By analyzing the single fiber capturing efficiency, interception incorporating the rotation of fibrous particles is found to be the dominant capturing mechanism for the fibrous particles in the studied size range.

## 2009

Kulkarni, P., Deye, G. J., and Baron, P. a., 2009, "Bipolar diffusion charging characteristics of single-wall carbon nanotube aerosol particles," *Journal of Aerosol Science*, **40**(2): 164–179

Bipolar diffusion charging characteristics of airborne single-wall carbon nanotube (SWCNT) agglomerates were investigated in the mobility diameter range of 100–1000nm. Neutral fractions of three types of SWCNT aerosols following bipolar charge equilibrium in a radioactive source were experimentally measured to infer their electrical charging characteristics. Significant deviation from Boltzmann and Fuchs stationary charge equilibrium was observed, with neutral fractions of SWCNT particles lower by 30–53% compared to that of spherical particles of the same mobility. Particles with mobility diameter larger than 400nm showed high electrical charging efficiencies compared to that of mobility-equivalent spherical particles. Higher charging efficiencies of SWCNT particles were attributed to their higher electrical capacitance resulting from complex nonspherical morphologies. Numerical calculations using idealized fiber geometries confirmed the qualitative trend in the experimental data. The electrical capacitance of nanotubes particles deduced from experimentally measured neutral fractions were also found to be higher by a factor ranging from 1.6 to 4.6 compared to that of mobility-equivalent spherical particles, indicating high charge carrying capacity. The charging-equivalent diameters of nanotube particles were computed and were found to be higher than their mobility diameter by a factor of 2.85–4.34.

## 2007

Hsu, Li-Yeh; Chein, Hung-Min, 2007, "Evaluation of nanoparticle emission for TiO<sub>2</sub> nanopowder coating materials," *Journal of Nanoparticle Research*, **9**(1):157–163

In this study, nanoparticle emission of TiO<sub>2</sub> nanopowder coated on different substrates including wood, polymer, and tile, was evaluated in a simulation box and measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer for the first time. The coating process for the substrate followed the instructions given by the supply company. In the simulation box, UV light, a fan, and a rubber knife were used to simulate the sun light, wind, and human contacting conditions. Among the three selected substrates, tile coated with TiO<sub>2</sub> nanopowder was found to have the highest particle emission (22 #/cm<sup>3</sup> at 55 nm) due to nanopowder separation during the simulation process. The UV light was shown to increase the release of particle below 200 nm from TiO<sub>2</sub> nanopowder coating materials. The results show that, under the conditions of UV lamps, a fan and scraping motion, particle number concentration or average emission rate decreases significantly after 60 and 90 min for TiO<sub>2</sub>/polymer and TiO<sub>2</sub>/wood, respectively. However, the emission rate continued to increase after 2 h of testing for TiO<sub>2</sub>/tile. It is suggested that nanoparticle emission evaluation is necessary for products with nanopowder coating.

Zhao, Bin; Uchikawa, Kei; Wang, Hai, 2007, "A comparative study of nanoparticles in premixed flames by Scanning Mobility Particle Sizer, small angle neutron scattering, and transmission electron microscopy," *Proceedings of the Combustion Institute*, **31**(1):851–860

Scanning Mobility Particle Sizer (SMPS) spectrometer and transmission electron microscopy (TEM) studies were conducted for TiO<sub>2</sub> and soot particles. The TiO<sub>2</sub> particles were produced from a premixed stagnation ethylene–oxygen–argon flame ( $\Phi = 0.36$ ) doped with titanium tetraisopropoxide. Soot was generated from a burner-stabilized premixed ethylene–oxygen–argon flame ( $\Phi = 2.5$ ). The close agreement among SMPS, TEM, and X-ray diffraction results for TiO<sub>2</sub> nanoparticles demonstrates that the probe sampling/mobility measurement technique is accurate for on-line analysis of the size distribution of particles as small as 3 nm in diameter. In the case of soot, notable disagreement between the SMPS and TEM sizes was found and attributable to the fact that the soot taken from the flame studied herein is liquid-like and that upon deposition on the TEM grid, the primary particles do not retain their sphericity. This interpretation is supported by measurements with photo ionization aerosol mass spectrometry, small angle neutron scattering, and thermocouple particle densitometry.

## 2006

Afergan, Eyal; Epstein, Hila; Golomb, Gershon; Moise, Tamar; Richter, Yoram; Rudich, Yinon, 2006, "Number-concentration of nanoparticles in liposomal and polymeric multiparticulate preparations: Empirical and calculation methods," *Biomaterials*, **27**(4):651–659

The actual number of particles in formulations of nanoparticles (NP) is of importance for quality assurance, comprehensive physicochemical characterization, and pharmacodynamics. Some calculation methods that have been previously employed are limited because they rely on several assumptions and are not applicable for certain preparations. Currently there are no validated experimental methods for determining the particle number-concentration ( $N_c$ ) of liposomal and polymeric nanoparticulate preparations (500 nm). This study examines a new empirical method for counting the number of particles in nanoparticulate formulations including drug-containing liposomes and polymeric NP. In the new method, suspended NP are nebulized to form aerosol droplets which are dried and counted using a Scanning Mobility Particle Sizer (SMPS) spectrometer. Experiments were conducted with three different preparations, empty liposomes (200 and 400 nm), drug-loaded liposomes (200 nm), and polymeric NP (150 nm). It was verified that no detrimental morphological or structural changes of the formulations have been induced by the SMPS technique, and that the obtained  $N_c$  values represent the original particles. It is concluded that nano-formulations with concentrations of up to  $10^7$  particles per 1 cm<sup>3</sup> air, corresponding to approximately  $10^{12}$  particles per 1 ml solution, can be directly counted within the size range of 30–900 nm. The measured values are compared to newly developed theoretical calculations to assess the viability of these calculations. 32 Refs.

Cho, Kuk; Biswas, Pratim, 2006, "Sintering Rates for Pristine and Doped Titanium Dioxide Determined Using a Tandem Differential Mobility Analyzer System," *Aerosol Science & Technology*, **40**(5):309–319

Sintering rates of pristine and V-doped TiO<sub>2</sub> were obtained using a tandem DMA system. A range of experiments were conducted to first map out the variation of mobility size of a monodisperse (by mobility) agglomerate with time at three fixed temperatures. Using relationships of the surface area to the mobility size, the sintering equation was solved to determine the activation energy and pre-exponential factor. The value of the activation energy was 236 ( $\pm 46$ ) kJ/mol for pristine TiO<sub>2</sub> and 363 ( $\pm 1$ ) kJ/mol for V-doped TiO<sub>2</sub>. The corresponding pre-exponential factors were  $7.22 \times 10^{19}$  and  $2.22 \times 10^{12}$  s/m<sup>4</sup>K, respectively. These values were then used to predict changes in mobility diameter at different temperatures, and good agreement was obtained with measurements. Possible reasons for faster sintering rates of V-TiO<sub>2</sub> relative to pristine TiO<sub>2</sub> were conjectured.

Epstein, Hila; Afergan, Eyal; Moise, Tamar; Richter, Yoram; Rudich, Yinon; Golomb, Gershon, 2006, "Number-concentration of nanoparticles in liposomal and polymeric multiparticulate preparations: Empirical and calculation methods," *Biomaterials*, **27**(4):651–659

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have been induced by the SMPS technique, and that the obtained  $N_c$  values represent the original particles. It is concluded that nano-formulations with concentrations of up to  $10^7$  particles per  $1\text{ cm}^3$  air, corresponding to approximately  $10^{12}$  particles per 1 ml solution, can be directly counted within the size range of 30–900 nm. The measured values are compared to newly developed theoretical calculations to assess the viability of these calculations.

Hyun, Jung Eun; Lee, Tai Gyu, 2006, "Structural effect of the in situ generated titania on its ability to oxidize and capture the gas-phase elemental mercury," *Chemosphere*, **62**(1):26–33

Structural effect of the in situ generated  $\text{TiO}_2$  sorbent particle was examined for its ability to capture elemental mercury under UV irradiation in a simulated combustion flue gas. Titania particles were prepared by thermal gas-phase oxidation of Titanium (IV) isopropoxide (TTIP) using a high temperature electric furnace reactor. The structural characteristics of the in situ generated  $\text{TiO}_2$  at various synthesis temperatures were investigated; size distribution and the geometric mean diameter were measured using a Scanning Mobility Particle Sizer, while fractal dimension and radius of gyration were evaluated from the transmission electron microscopy images. Results from the  $\text{Hg}^0$  capture experiment show that with increasing titania synthesis temperature, the overall aggregate size increases and the morphology becomes more open-structured to gas-phase  $\text{Hg}^0$  and UV light, resulting in the improved mercury removal capability.

Ku, Bon Ki; Maynard, Andrew D., 2006, "Generation and investigation of airborne silver nanoparticles with specific size and morphology by homogeneous nucleation, coagulation and sintering," *Journal of Aerosol Science*, **37**(4):452–470

An aerosol generation facility has been characterized to produce well-defined silver nanoparticles for use as a test aerosol when evaluating instrument response to different particle morphologies within a range of sizes. The generator consists of two in-series laminar tube furnaces to produce and subsequently sinter particles, and is capable of generating spherical or agglomerated fractal-like silver particles, with corresponding projected surface (two-dimensional) fractal dimensions from 1.58 to 1.94. The morphologies of generated particles as well as size distributions at different sintering temperatures were characterized using a transmission electron microscope (TEM) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. Mean diameters measured were significantly reduced for sintering temperatures from 100 to 300°C, but showed little variation for sintering temperatures above 500°C. TEM analysis indicated that this phenomenon was caused by sintering, followed by partial and complete coalescence of fractal-like agglomerates into spheres. Agglomerate restructuring from classical completely sintered agglomerates 500°C to spherical particles 700°C did not lead to a change in the particle mobility size distribution. The temperature at which complete sintering occurred was higher than that predicted by theory, but was in reasonable agreement with previously published experimental data. For monodisperse particles in the size range from 20 to 100 nm, a simple exponential model related sintering temperature to the diameter of coalesced spherical particles.

Lee, Tai Gyu; Hyun, Jung Eun, 2006, "Structural effect of the in situ generated titania on its ability to oxidize and capture the gas-phase elemental mercury," *Chemosphere*, **62**(1):26–33

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## 2005

Choi, Jong Hyun; Koshland, Catherine P.; Lucas, Donald; Sawyer, Robert F., 2005, "Photochemical interaction of polystyrene nanospheres with 193 nm pulsed laser light," *Journal of Physical Chemistry B*, **109**(50):23905–23910

The photochemical interaction of 193 nm light with polystyrene nanospheres is used to produce particles with a controlled size and morphology. Laser fluences from 0 to  $0.14\text{ J/cm}^2$  at 10 and 50 Hz photofragment nearly monodisperse 110 nm spherical polystyrene particles. The size distributions before and after irradiation are measured with a Scanning Mobility Particle Sizer (SMPS) spectrometer, and the morphology of the irradiated particles is examined with a transmission electron microscope (TEM). The results show that the irradiated particles have a smaller mean diameter (similar to 25 nm)

and a number concentration more than an order of magnitude higher than nonirradiated particles. The particles are formed by nucleation of gas-phase species produced by photolytic decomposition of nanospheres. A nondimensional parameter, the photon-to-atom ratio (PAR), is used to interpret the laser-particle interaction energetics. 39 Refs.

Choi, Jong Hyun; Koshland, Catherine P.; Lucas, Donald; Sawyer, Robert F.; Stipe, Christopher B., 2005, "NaCl particle interaction with 193-nm light: Ultraviolet photofragmentation and nanoparticle production," *Journal of Applied Physics*, **97**(12):1-9

The interaction of nanoscale NaCl particles with 193-nm photons is studied to better understand particle disintegration and production by ultraviolet photofragmentation. The particles are irradiated in a constrained air stream with laser fluences from 0.08 to 0.23 J cm<sup>2</sup> with single and multiple pulses striking the particles. The resulting particle size distributions are measured with a Scanning Mobility Particle Sizer and the morphology is analyzed qualitatively by scanning electron microscopy (SEM). Photofragmentation of NaCl particles at 193 nm produces gas-phase species as well as small solid-phase fragments without significantly heating the particles or creating a plasma. The irradiated particles have a mean diameter from 20 to 55 nm (depending on the photon energy) and a number concentration an order of magnitude higher than the 118 nm mean diameter nonirradiated particles. The SEM images before and after 193-nm irradiation reveal that the irradiated particles are less fractal and more spherical. 52 Refs.

Girshick, S.L.; Hafiz, J.; Heberlein, J.; McMurry, P.H.; Mukherjee, R.; Renault, T.; Wang, X., 2005 "System for in situ characterization of nanoparticles synthesized in a thermal plasma process," *Plasma Chemistry and Plasma Processing*, **25**(5):439-453

We have designed a particle diagnostic system that is able to measure particle size and charge distributions from low stagnation pressure and high temperature (2000-4000 K) environments in near real time. This system utilizes a sampling probe interfaced to an ejector to draw aerosol from the low pressure chamber. Particle size and charge distributions are measured with a Scanning Mobility Particle Sizer spectrometer. A hypersonic impactor is mounted in parallel with the Scanning Mobility Particle Sizer to collect particles for off-line microscopic analysis. This diagnostic system has been used to measure size and charge distributions of nanoparticles (Si, Ti, Si-Ti-N, etc.) synthesized with our thermal plasma reactor. We found that the mean particle size increases with operating pressure and reactant flow rates. We also found that most particles from our reactor are neutral for particles smaller than 20nm, and that the numbers of positively and negatively charged particles are approximately equal. Inc. 33 Refs.

Ku, Bon Ki; Maynard, Andrew D., 2005, "Comparing aerosol surface-area measurements of monodisperse ultrafine silver agglomerates by mobility analysis, transmission electron microscopy and diffusion charging," *Journal of Aerosol Science*, **36**(9):1108-1124

Three methods - Scanning Mobility Particle Sizer (SMPS) spectrometer, transmission electron microscopy (TEM), and diffusion charging (DC) - for estimating aerosol surface area were evaluated and compared. The aerosol used was monodisperse silver particles, having morphologies that range from spherical to agglomerated particles, with corresponding fractal dimensions from 1.58 to 1.94. For monodisperse silver particle agglomerates smaller than 100 nm, the DC response was proportional to the mobility diameter squared, regardless of morphology. For particle sizes from 80 to 200 nm, the DC response varied as the mobility diameter to the power 1.5. The projected surface area of agglomerates analyzed by TEM agreed well with that estimated from particle mobility diameters for particles smaller than 100 nm. The surface area of monodisperse particles, measured by DC and SMPS, was comparable to the geometric surface area below 100 nm, but in the size range of 100-200 nm, the methods used underestimated the geometric surface area. SMPS, TEM, and DC-based measurements of surface area were in good agreement with one another for monodisperse aerosol particles smaller than 100 nm. 24 Refs.

Lee, Gyo Woo; Lee, Seung Bok; Jung, Jong Soo; Bae, Gwi Nam, 2005, "Effect of flame temperature on the characteristics of flame synthesized TiO<sub>2</sub> nanoparticles," *Transactions of the Korean Society of Mechanical Engineers. B*, **29**(9):1013-1021

In this work, TiO<sub>2</sub> nanoparticles were synthesized using a N<sub>2</sub>-diluted hydrogen coflow diffusion flame. The effects of flame temperature on the crystalline structure and the size of formed nanoparticles were investigated. The maximum centerline temperature of the flame ranged from 1,920K for H<sub>2</sub>-only flame to 863K for 81% N<sub>2</sub>-diluted flame. The morphology and the crystal structure of TiO<sub>2</sub> nanoparticles were analyzed by a TEM and a XRD, respectively. The particle size distribution was also measured by using a Scanning Mobility Particle Sizer (SMPS) spectrometer. The mean particle diameter was calculated from the TEM images depended on the flame temperature, having minimum at about 1,200K. Based on the SMPS measurements, the mean particle diameter of TiO<sub>2</sub> nanoparticles at flame temperatures @1,300K was smaller than that at flame temperatures @ 1,300K. From the XRD analysis, it was evident that the

anatase fraction increased with decreasing the flame temperature. The portion of anatase phase in TiO<sub>2</sub> nanoparticles might be greater than 80% when the flame temperature was lower than 1,000K.

Moravec, P.; Smolík, J.; Levdansky, V.V., 2005, "Preparation of Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> fine particles by CVD method in tube flow reactor," *Powder Technology*, **153**(3):159-165

The aim of this study was the synthesis of mixed or coated multicomponent alumina-silica particles by chemical vapor deposition method in a tube flow reactor. The particles were produced by simultaneous thermal decomposition of aluminum tri-*sec*-butoxide and tetraethylorthosilicate in one reactor. The particle production was monitored by Differential Mobility Particle Sizer, composition of particles was analyzed by energy dispersive X-ray analysis, morphology by scanning/transmission electron microscopy and crystallinity by selected area electron diffraction. In dependence on experimental conditions, the particles produced were either alumina particles with intermixture of silica, or they were coated by silica, or it was a mixture of particles of various compositions. The particles were often agglomerates of the primary nanoparticles and were partially crystalline.

Vasiliou, J., 2005, "An evaluation of a scanning mobility particles sizer with NIST-traceable particle size standards," *2005 NSTI Nanotechnology Conference and Trade Show, Anaheim, CA*

A Scanning Mobility Particle Sizer (SMPS) spectrometer (TSI Model 3936-Series) was evaluated using Duke Scientific NIST-traceable particle size standards and Standard Reference Materials from the National Institute of Standards and Technology (NIST SRMs). The importance of instrument setup, electrospray operation and sample preparation for polystyrene spheres are discussed as well as the results from 14 different size reference standards. Correlations between the SMPS system and established electron microscopy and dynamic light scattering methods are also shown in tabular and graphical forms. Results show that with proper operation, the SMPS results fall within the uncertainty of the NIST traceable diameters in the range that was evaluated - 20 to 100 nanometers.

Wang, Yanmin; Forssberg, Eric, 2006, "Production of carbonate and silica nano-particles in stirred bead milling," *International Journal of Mineral Processing*, **81**(1):1-14

This paper presents the experimental results of the mechanical production of silica and carbonate colloidal particles below 100 nm using two types of stirred bead mills (i.e., DCP Superflow 12 and PML H/V). It is shown that the stirred bead mill with very small beads can be used as an efficient equipment for the production of the colloidal particles in nanoscale from the feed materials of several microns in sizes at high energy consumptions. The DCP Superflow mill with high power densities is superior for the effective size reduction and production rate, compared to the conventional PML H/V mill with lower power densities. The smaller particles could be produced by the DCP Superflow mill at the same level of high energy inputs as from the PML H/V mill. The "grinding limit" for the processes in the mills has been discussed.

The nanoparticle sizes of the ground products obtained in the mills were determined by a Scanning Mobility Particle Sizer (SMPS) spectrometer, an acoustic particle sizer (APS-100), a nitrogen gas adsorption method (BET), and transmission electron microscopy (TEM). The surface mean particle diameters on the same sample obtained with the different methods for the nanoparticle size analysis are varying due to the different characterization techniques. The TEM micrographs show a large amount of the primary nanoparticles below 100 nm produced after milling.

In addition, the surface and structure of the original and the ground colloidal products have been investigated with some particle/surface characterization techniques such as X-ray diffraction (XRD), nuclear magnetic resonance (<sup>29</sup>Si NMR) and Fourier transform infrared spectroscopy (FTIR). It is indicated that an intense comminution of carbonate minerals in the stirred bead mills leads to a progressive loss in crystallinity of the basal planes of the crystal structure. An intensive mechanical treatment of silica gives the structural changes and the amorphisation.

Wang, X.; Hafiz, J.; Mukherjee, R.; Renault, T.; Heberlein, J.; Girshick, S.; McMurry, P., 2005, "System for In Situ Characterization of Nanoparticles Synthesized in a Thermal Plasma Process," *Plasma Chemistry and Plasma Processing*, Volume **25**(5):439-453

We have designed a particle diagnostic system that is able to measure particle size and charge distributions from low stagnation pressure ( $\approx 746$  Pa) and high temperature (2000-4000 K) environments in near real time. This system utilizes a sampling probe interfaced to an ejector to draw aerosol from the low pressure chamber. Particle size and charge distributions are measured with a Scanning Mobility Particle Sizer spectrometer. A hypersonic impactor is mounted in parallel with the Scanning Mobility Particle Sizer to collect particles for off-line microscopic analysis. This diagnostic system has been used to measure size and charge distributions of nanoparticles (Si, Ti, Si-Ti-N, etc.) synthesized with our thermal plasma reactor. We found that the mean particle size increases with

operating pressure and reactant flow rates. We also found that most particles from our reactor are neutral for particles smaller than 20 nm, and that the numbers of positively and negatively charged particles are approximately equal.

Zhao, Bin; Uchikawa, Kei; McCormick, John R.; Ni, Chao Ying; Chen, Jingguang G.; Wang, Hai, 2005, "Ultrafine anatase TiO<sub>2</sub> nanoparticles produced in premixed ethylene stagnation flame at 1 atm," *Proceedings of the Combustion Institute*, **30**(1):1441–1448

Particle size distribution functions (PSDFs) of incipient soot formed in laminar premixed 24.2% ethylene–37.9% oxygen-diluent (nitrogen and/or argon) flames with an equivalence ratio of 1.92 were studied by online sampling and Scanning Mobility Particle Sizer spectrometer. Two series of flames were studied to quantify the effect of flame temperature on the characteristics of PSDFs. In the first series, the variation of the flame temperature was accomplished by varying the cold gas velocity. Temperature in the second series of flames was manipulated by the diluent composition from argon to nitrogen. The results show that for flames with the maximum temperature ( $T_{\max}$ ) around 1800 K the soot PSDFs were distinctively bimodal. As the flame temperature was increased to  $\sim$  1850 K, bimodality faded away. The distribution was unimodal for  $T_{\max} > 1900$  K. The variation of the characteristics of the PSDF as a function of the flame temperature is consistent with the theoretical explanation that bimodality is the result of competition between persistent particle nucleation and particle–particle coagulation in low-temperature flames.

## 2004

Carter, C.B.; Deneen, J.; Gerberich, W.W.; Girshick, S.L.; Hafiz, J.; Heberlein, J.V.R.; McMurry, P.H.; Mook, W.; Mukherjee, R.; Perrey, C.R.; Wang, X., 2004, "Hypersonic plasma particle deposition of Si-Ti-N nanostructured coatings," *Surface and Coatings Technology*, 188–189(1-3 SPEC.ISS.):364–370

Si-Ti-N coatings with various compositions were deposited on molybdenum substrates using hypersonic plasma particle deposition (HPPD). In this method, vapor phase precursors (TiCl<sub>4</sub>, SiCl<sub>4</sub> and NH<sub>3</sub>) are dissociated in a DC plasma arc and the hot gas is quenched in a rapid nozzle expansion to nucleate nanoparticles. These nanoparticles are then accelerated in hypersonic flow, causing them to deposit by ballistic impaction on a substrate placed downstream of the nozzle. Films of 10–25  $\mu$ m thickness were deposited at rates of 2–10  $\mu$ m/min, depending on reactant flow rates, at substrate temperatures ranging from 200 to 850°C. When the reactant gases were premixed the coatings consisted of nc-TiN, nc-TiSi<sub>2</sub>, nc-Ti<sub>5</sub>Si<sub>3</sub> and amorphous Si<sub>3</sub>N<sub>4</sub>. For the unpremixed reactants case, the coatings consisted of free Si, nc-TiN and amorphous Si<sub>3</sub>N<sub>4</sub>. Hardness of as-deposited films was evaluated by nanoindentation of polished film cross-sections. Measured hardness values, averaged over 10–15 locations for each film, ranged from 16–24 GPa. In separate experiments with the same conditions, particle size distributions were measured by placing a sampling probe at the same location as the film substrate. The sampled aerosol was rapidly diluted and delivered to a Scanning Mobility Particle Sizer (SMPS) spectrometer. In-situ particle size distribution measurements confirmed that the coatings were formed by impaction of nanoparticles in the 5–15 nm range, with higher reactant flow rates producing larger particles. Focused ion beam (FIB) milling was used to observe film cross-section and porosity. For as-deposited films containing pores, in-situ plasma sintering was used to densify the film without grain growth.

Heszler, P.; Landstrom, L.; Marton, Z., 2004, "Early stage of the material removal during ArF laser ablation of graphite," *Applied Physics A: Materials Science and Processing*, **79**(3):579–585

Material removal during ArF excimer laser ablation of graphite at atmospheric pressure was investigated by two independent methods; 1) by observation of the propagating properties of the shock wave generated by the carbonaceous ejecta and 2) by in situ measurement of the size distribution of carbon nanoparticles condensing in the ablation plume. This latter was carried out by a Scanning Mobility Particle Sizer system based on a differential mobility analyzer. The performed measurements indicate that the material removal during ArF laser ablation consists of two steps at fluences above the threshold fluence. First, a thin layer of carbon (of the order of 1 nm) is removed by a quick desorption process, leading to shockwave formation. This process takes place in a ns time scale, and desorption rate estimations reveal that this cannot be explained by thermal surface evaporation. Since to our knowledge there is no thermal process that could account for the estimated desorption rate, it is argued that this is a fast photochemical (i.e., non-thermal) process. The size distribution of the condensed nanoparticles related to this step shows a rising edge at diameters below 10 nm. At fluences above the ablation threshold, the majority of the material is ejected in the second phase, resulting in condensation of carbon nanoparticles, peaking at similar to 50 nm diameters in the size spectrum. Both shockwave formation and material removal are also detected well below the ablation threshold fluence, which is attributed to the photochemical process. 39 Refs.



Márton, Z.; Landström, L.; Heszler, P., 2004, "Early stage of the material removal during ArF laser ablation of graphite," *Applied Physics A*, **79**(3):579–585

Material removal during ArF excimer laser ablation of graphite at atmospheric pressure was investigated by two independent methods; 1) by observation of the propagating properties of the shock wave generated by the carbonaceous ejecta and 2) by in situ measurement of the size distribution of carbon nanoparticles condensing in the ablation plume. This latter was carried out by a Scanning Mobility Particle Sizer system based on a differential mobility analyzer. The performed measurements indicate that the material removal during ArF laser ablation consists of two steps at fluences above the threshold fluence. First, a thin layer of carbon (of the order of 1 nm) is removed by a quick desorption process, leading to shockwave formation. This process takes place in a ns time scale, and desorption rate estimations reveal that this cannot be explained by thermal surface evaporation. Since to our knowledge there is no thermal process that could account for the estimated desorption rate, it is argued that this is a fast photochemical (i.e., non-thermal) process. The size distribution of the condensed nanoparticles related to this step shows a rising edge at diameters below 10 nm. At fluences above the ablation threshold, the majority of the material is ejected in the second phase, resulting in condensation of carbon nanoparticles, peaking at ~50 nm diameters in the size spectrum. Both shockwave formation and material removal are also detected well below the ablation threshold fluence, which is attributed to the photochemical process.

Young, Li-Hao; Keeler, G. J., 2004, "Characterization of Ultrafine Particle Number Concentration and Size Distribution During a Summer Campaign in Southwest Detroit," *Journal of the Air & Waste Management*, **54**(9)

This paper presents results from a study conducted in southwest Detroit from July 20 to July 30, 2002, to characterize ambient ultrafine particles ( $d_p < 0.1 \mu\text{m}$ ), and to examine the effect of local sources and meteorological parameters on the ultrafine number concentration and size distribution. The number concentrations of ambient particles in the size range of 0.01–0.43  $\mu\text{m}$  were obtained from a Scanning Mobility Particle Sizer (SMPS) spectrometer. Meteorological parameters including ambient temperature, relative humidity, wind speed, wind direction, rainfall, and solar radiation flux were also monitored concurrently atop a 10-m tower. On average, ultrafine particles ranged from  $1.4 \times 10^4$  to  $2.5 \times 10^4 \text{ cm}^{-3}$ , with significant diurnal and daily variations, and accounted for 89% of the total number concentration ( $0.01 < d_p < 0.43 \mu\text{m}$ ). Time-series plots of the 5-min number concentrations revealed that peak concentrations often occurred during morning rush hour and/or around solar noon when photochemical activity was at a maximum. The morning traffic-related peak coincided with the NO<sub>x</sub> peak, whereas the photochemical-related peak correlated with solar radiation flux. On some days, the noon peak concentration was many times higher than the morning peak concentration. Although the number size distribution varied considerably over the course of the study, it typically exhibited one to three modes, with diameters around 0.01, 0.05, and 0.09  $\mu\text{m}$ . Analysis of the influence of wind direction indicated that stationary sources could be one of the contributors to elevated ultrafine particle concentration. Overall, the data indicated that fossil fuel combustion and atmospheric gas-to-particle conversion of precursor gases are the major sources of ultrafine particles in the southwest Detroit area during the summer.

## 2003

Boman, M.; Heszler, P.; Landstrom, L.; Marton, Z., 2003, "A comparative study of size distribution of nanoparticles generated laser ablation of graphite and tungsten," *Material Science and Engineering C*, **23**(1-2):225–228

Nanoparticles (NPs) were generated by ArF excimer laser ablation of graphite and tungsten targets in N<sub>2</sub> ambient at atmospheric pressure. The size distribution of the particles was monitored in situ by a Scanning Mobility Particle Sizer (SMPS) system, based on differential mobility analyzer. The experimental conditions made possible to record the size distributions in the 7–133-nm diameter range and results are presented for different laser fluences, repetition rates and ablated areas, respectively. Material analysis was performed by photoelectron spectroscopy (XPS), Raman spectroscopy, X ray diffraction and SEM. 17 Refs.

## 2001

Ahn, K. H.; Lee, J. S.; Yu, J. H., 2001, "In situ characterization of TiO<sub>2</sub> nanoparticle in chemical vapor condensation reactor," *Scripta Materialia*, **44**(8-9):2213–2217

The effects of oxygen flow rate on the particle coagulation in the chemical vapor condensation (CVC) reactor were investigated using Scanning Mobility Particle Sizer spectrometer. With a decrease of oxygen flow rate, the titanium dioxide particle size increased and the size distribution broadened with a longer residence time and a higher particle concentration. These characteristics in size were dominantly controlled by the ratio of oxygen flow rate in the CVC reactor. 8 Refs.

Ahn K.H.; Jung C.H.; Choi M.1; Lee J.S., 2001, "Particle Sampling and Real Time Size Distribution Measurement in H<sub>2</sub>/O<sub>2</sub>/TEOS Diffusion Flame," *Journal of Nanoparticle Research*, **3**(2-3):161-170

Growth characteristics of silica particles have been studied experimentally using in situ particle sampling technique from H<sub>2</sub>/O<sub>2</sub>/Tetraethylorthosilicate (TEOS) diffusion flame with carefully devised sampling probe. The particle morphology and the size comparisons are made between the particles sampled by the local thermophoretic method from the inside of the flame and by the electrostatic collector sampling method after the dilution sampling probe. The Transmission Electron Microscope (TEM) image processed data of these two sampling techniques are compared with Scanning Mobility Particle Sizer (SMPS) spectrometer measurement. TEM image analysis of two sampling methods showed a good agreement with SMPS measurement. The effects of flame conditions and TEOS flow rates on silica particle size distributions are also investigated using the new particle dilution sampling probe. It is found that the particle size distribution characteristics and morphology are mostly governed by the coagulation process and sintering process in the flame. As the flame temperature increases, the effect of coalescence or sintering becomes an important particle growth mechanism which reduces the coagulation process. However, if the flame temperature is not high enough to sinter the aggregated particles then the coagulation process is a dominant particle growth mechanism. In a certain flame condition a secondary particle formation is observed which results in a bimodal particle size distribution.

Hansen, J. P.; Jensen, J. R.; Johannessen, T.; Livbjerg, H., 2001, "Synthesis of ZnO particles in a quench-cooled flame reactor," *AIChE Journal*, **47**(11):2413-2418

The quench cooling of a flame by injection of cold air was studied in a flame reactor for the formation of ZnO particles in a premixed flame with a precursor jet. A rapid temperature drop downstream from the temperature peak is advantageous for the attainment of a large specific surface area. Computational fluid dynamics simulations were used to design a quench ring with nozzles directed slightly upward and at a small tangential angle from the direct line to the center. This novel design avoids distortion of the flow pattern below the quenching plane and effectively cools the flame immediately above. At the highest tested production rate, the specific surface area of the ZnO particles increases from 20 to 60 m<sup>2</sup>/g when quenching is employed. The particles are characterized by BET surface area measurements, TEM images, and the size distributions of particle aggregates are measured by a Scanning Mobility Particle Sizer spectrometer.

Nakaso, Koichi; Fujimoto, Toshiyuki; Seto, Takafuimi; Shimada, Manabu; Okuyama, Kikuo; Lunden, Melissa M., 2001, "Size Distribution Change of Titania Nano-Particle Agglomerates Generated by Gas Phase Reaction, Agglomeration, and Sintering," *Aerosol Science & Technology*, **35**(5):929-947

In the manufacturing of nanometer-sized material particulates by aerosol gas-to-particle conversion processes, it is important to analyze how the gas-phase chemical reaction, nucleation, agglomeration, and sintering rates control the size distribution and morphology of particles. In this study, titania particles were produced experimentally by the thermal decomposition of titanium tetraisopropoxide (TTIP) and oxidation of titanium tetrachloride (TiCl<sub>4</sub>) using a laminar flow aerosol reactor. The effect of reaction temperature on the size and morphology of the generated particles was investigated under various conditions. The size distributions of agglomerates were measured using a DMA/CNC system. The size distributions of primary particles were measured using TEM pictures of the agglomerates sampled by a thermophoretic aerosol sampler. In order to model the growth of both agglomerates and primary particles simultaneously, a two-dimensional discrete-sectional representation of the size distribution was employed, solving the aerosol general dynamic equation for chemical reaction, agglomeration, and sintering. Qualitative agreement between the experimentally observed results and the simulation are satisfactory for the large variations in reactor temperature explored.

## 2000

Martin, S. T.; Han, J.-H., 2000, "An aerosol chemical reactor for coating metal oxide particles with (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O. Manipulation of the sulfate coating," *Journal of Crystal Growth*, **219**(3):290-299

One important chemical and morphological class of atmospheric particles consists of mineral dusts contained as inclusions in crystalline and aqueous sulfate particles. The mineral dust surface plays an important role in regulating the sulfate phase by providing a surface for the heterogeneous nucleation of the crystal from supersaturated aqueous sulfate phases. In principle, the efficacy of mineral dusts in promoting phase transitions can be investigated through laboratory process studies. To do so, however, a particle generator is necessary. The present work describes the generation of metal oxide particles (viz.  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>,  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, Al<sub>6</sub>Si<sub>2</sub>O<sub>13</sub>, and amorphous SiO<sub>2</sub>) coated by (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The diameter of the oxide core and the thickness of the sulfate coating are regulated from 30 to 400 nm and 13 to 81 nm, respectively. Sizing is based upon aerosol particles collected by electrostatic precipitation and subsequently imaged by transmission electron microscopy. Depending on reactor conditions, the fraction of particles containing a

metal oxide core varies from 0.01 to 0.86, as inferred from number size distributions measured with a Scanning Mobility Particle Sizer spectrometer. The single most important reactor variable affecting fraction of particles containing a metal oxide core is the total sulfate loading. No dependencies on the effects of different chemical surface properties of the metal oxide particles on the coating process have been observed in our laboratory to date. Rather, the number size distribution of metal oxide core particles appears important without regard to chemical surface properties.

Signorell, R.; Kunzmann, M. K.; Suhm, M. A., 2000, "FTIR investigation of non-volatile molecular nanoparticles," *Chemical Physics Letters*, **329**(1-2):52-60

A new approach for the spectroscopic investigation of non-volatile molecular nanoparticles with diameters in the range between 10 and 100 nm is proposed. The nanoparticles are produced in an electrospray with subsequent solvent evaporation. The number size distribution of the generated aerosol is determined with a Scanning Mobility Particle Sizer spectrometer. Fourier transform infrared spectroscopy is used to study the vibrational dynamics of the nanoparticles. As an example, we have investigated sucrose nanoparticles with six different diameters between 36 and 82 nm. From a comparison with different bulk phase spectra, we conclude that sucrose aerosols are formed in a solid amorphous state.

## 1994

Somers, J.; Caperan, Ph.; Richter, K.; Fourcaudot, S., 1994, "Agglomeration of a titanium dioxide aerosol in a high intensity sound field," *Journal of Aerosol Science*, **25**(S1):355-56

The paper investigates the influence of a high intensity field on the agglomeration of droplets in the 0.02-0.5  $\mu\text{m}$  size range. Results show that acoustic treatment could enhance the rate of decrease in concentration of droplets in the size range 0.08-0.5  $\mu\text{m}$ . Measurements of the influence of acoustic waves on a solid aerosol were necessary to confirm these findings. Also results corroborate the measurements on droplet coagulation in this size range and the unexpected efficiency of acoustic agglomeration for particles with diameter as small as 0.08  $\mu\text{m}$  is confirmed.

## 1991

Akhtar, M. Kamal; X., Yun; P., Sotiris E., 1991, "Vapor synthesis of titania powder by titanium tetrachloride oxidation," *AIChE Journal*, **37**(10):1561-570

Formation of titania particles by vapor-phase oxidation of titanium tetrachloride was studied in an aerosol reactor between 1,200 and 1,723 K. The effect of process variables (reactor residence time, temperature, and reactant concentration) on powder size and phase characteristics was investigated using the differential Scanning Mobility Particle Sizer, electron microscopy, and X-ray diffraction. Titania particles were primarily anatase though the rutile weight fraction increased with increasing reactor temperature. The geometric number average diameter of the particles was between 0.13 and 0.35  $\mu\text{m}$ , and the geometric standard deviation of the particle size distribution was about 1.4. The average particle size increased with increasing temperature, inlet  $\text{TiCl}_4$  concentration, and residence time. The observed changes in the particle size distribution were compared with those predicted by solving the aerosol dynamic equation by a sectional method and accounting for coagulation and first-order chemical reaction. While variations in the process variables resulted in discernible changes in the size of the particles, the spread of the distribution remained rather unaffected.

Graham, G.M.; Alam, M.K., 1991, "Experimental Study of the Outside Vapor Deposition Process," *Aerosol Science & Technology*, **15**:69-6

Outside vapor deposition has been studied experimentally, and the deposition process parameters have been examined. The system consisted of an industrial-type burner and a cylindrical bait rod. The following aspects of this process were studied: 1) the temperature profile of the bait rod surface, 2) the deposition efficiency measured in terms of the mass deposited  $\text{SiO}_2$ , and 3) the size distribution of the deposited particles.

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## Occupational Health/Indoor Studies

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### 2013

Asimakopoulou, A., Daskalos, E., Lewinski, N., Riediker, M., Papaioannou, E., and Konstandopoulos, A. G., 2013, "Development of a dose-controlled multiculture cell exposure chamber for efficient delivery of airborne and engineered nanoparticles," *Journal of Physics: Conference Series*, **429**(012023):

In order to study the various health influencing parameters related to engineered nanoparticles as well as to soot emitted by Diesel engines, there is an urgent need for appropriate sampling devices and methods for cell exposure studies that simulate the respiratory system and facilitate associated biological and toxicological tests. The objective of the present work was the further advancement of a Multiculture Exposure Chamber (MEC) into a dose-controlled system for efficient delivery of nanoparticles to cells. It was validated with various types of nanoparticles (Diesel engine soot aggregates, engineered nanoparticles for various applications) and with state-of-the-art nanoparticle measurement instrumentation to assess the local deposition of nanoparticles on the cell cultures. The dose of nanoparticles to which cell cultures are being exposed was evaluated in the normal operation of the in vitro cell culture exposure chamber based on measurements of the size specific nanoparticle collection efficiency of a cell free device. The average efficiency in delivering nanoparticles in the MEC was approximately 82%. The nanoparticle deposition was demonstrated by Transmission Electron Microscopy (TEM). Analysis and design of the MEC employs Computational Fluid Dynamics (CFD) and true to geometry representations of nanoparticles with the aim to assess the uniformity of nanoparticle deposition among the culture wells. Final testing of the dose-controlled cell exposure system was performed by exposing A549 lung cell cultures to fluorescently labeled nanoparticles. Delivery of aerosolized nanoparticles was demonstrated by visualization of the nanoparticle fluorescence in the cell cultures following exposure. Also monitored was the potential of the aerosolized nanoparticles to generate reactive oxygen species (ROS) (e.g. free radicals and peroxides generation), thus expressing the oxidative stress of the cells which can cause extensive cellular damage or damage on DNA.

Buonanno, G., Fuoco, F. C., Morawska, L., and Stabile, L., 2013, "Airborne particle concentrations at schools measured at different spatial scales," *Atmospheric Environment*, **67**: 38–45

Potential adverse effects on children health may result from school exposure to airborne particles. To address this issue, measurements in terms of particle number concentration, particle size distribution and black carbon (BC) concentrations were performed in three school buildings in Cassino (Italy) and its suburbs, outside and inside of the classrooms during normal occupancy and use. Additional time resolved information was gathered on ventilation condition, classroom activity, and traffic count data around the schools were obtained using a video camera. Across the three investigated school buildings, the outdoor and indoor particle number concentration monitored down to 4 nm and up to 3 mm ranged from  $2.8 \times 10^4$  part  $\text{cm}^{-3}$  to  $4.7 \times 10^4$  part  $\text{cm}^{-3}$  and from  $2.0 \times 10^4$  part  $\text{cm}^{-3}$  to  $3.5 \times 10^4$  part  $\text{cm}^{-3}$ , respectively. The total particle concentrations were usually higher outdoors than indoors, because no indoor sources were detected. I/O measured was less than 1 (varying in a relatively narrow range from 0.63 to 0.74), however one school exhibited indoor concentrations higher than outdoor during the morning rush hours. Particle size distribution at the outdoor site showed high particle concentrations in different size ranges, varying during the day; in relation to the starting and finishing of school time two modes were found. BC concentrations were 5 times higher at the urban school compared with the suburban and suburban-to-urban differences were larger than the relative differences of ultrafine particle concentrations.

Chen, S.-C., Wang, J., Fissan, H., and Pui, D. Y. H., 2013, "Exposure assessment of nanosized engineered agglomerates and aggregates using Nuclepore filter," *Journal of Nanoparticle Research*, **15**(10): 1955

Nuclepore filter collection with subsequent electron microscopy analysis for nanosized agglomerates (20-500 nm in mobility diameter) was carried out to examine the feasibility of the method to assess the personal engineered nanoparticle exposure. The number distribution of the nanoparticles collected on the filter surface was obtained by visual counting and converted to the distribution in the air using validated capillary tube models. The model was validated by studying the overall penetrations of nanoparticles (Ag and soot) with different agglomeration degrees through 1  $\mu\text{m}$  pore diameter Nuclepore filters at different face velocities (2–15 cm/s). In the model, the effects of the maximum length of agglomerates on interception deposition and the dynamic shape factor on impaction deposition were taken into account. Results showed that the data of the overall penetration were in very good agreement with the properly applied models. A good agreement of filter surface collection between the validated

model and the SEM analysis of this study was obtained, indicating a correct particle number distribution in the air can be converted from the Nuclepore filter surface collection and this method can be applied for quantitative engineered nanoparticle exposure assessment.

Zhuang, Z., Bergman, M. S., Eimer, B. C., and Shaffer, R. E., 2013, "Laboratory face seal leakage evaluation of n95 filtering facepiece respirators against nanoparticles and "all size" particles.," *Journal of occupational and environmental hygiene*, **10**(9): 496–504

National Institute for Occupational Safety and Health (NIOSH)-certified N95 filtering facepiece respirators (FFRs) are used for respiratory protection in some workplaces handling engineered nanomaterials. Previous NIOSH research has focused on filtration performance against nanoparticles. This article is the first NIOSH study using human test subjects to compare N95 FFR face seal leakage (FSL) performance against nanoparticles and "all size" particles. In this study, estimates of FSL were obtained from fit factor (FF) measurements from nine test subjects who participated in previous fit-test studies. These data were analyzed to compare values obtained by: 1) using the PortaCount® Plus Respirator Fit Tester (8020A, TSI, Inc., MN, USA) alone (measurable particle size range 20 nm to > 1,000 nm, hereby referred to as the "all size particles test"), and 2) using the PortaCount Plus fit tester with N95-Companion™ accessory (8095, TSI Inc., Minn.) accessory (negatively charged particles, size range ~40 to 60 nm, hereby referred to as the "nanoparticles test"). Log-transformed FF values were compared for the "all size particles test" and "nanoparticles test" using one-way analysis of variance tests (significant at  $P < 0.05$ ). For individual FFR models, geometric mean (GM) FF using the "nanoparticles test" was the same or higher than the GM FFs using "all size particles test." For all three FFR models combined, GM FF using the "nanoparticles test" was significantly higher than the GM FF using "all size particles test" ( $P < 0.05$ ). These data suggest that FSL for negatively charged ~40-60 nm nanoparticles is not greater than the FSL for the larger distribution of charged and uncharged 20 to >1,000 nm particles.

## 2012

Pawar, A. A., Chen, D.-R., and Venkataraman, C., 2012, "Influence of precursor solvent properties on matrix crystallinity and drug release rates from nanoparticle aerosol lipid matrices.," *International journal of pharmaceutics*, **430**(1-2): 228–37

The crystallinity of drug-loaded lipid nanoparticles is believed to affect drug release rates; however, effective control over lipid crystallinity has not been achieved by current lipid nanoparticle preparation methods. The present study investigates control over the crystallinity of drug-loaded nanoparticle aerosol lipid matrices (NALM) through differences in evaporation rates of precursor solution drops and the subsequent control over drug release rates from these matrices. Gefitinib-loaded NALM were synthesized in an aerosol reactor using precursor solutions of gefitinib and stearic acid at a ratio of 1:4 w/w in organic solvents with high (dichloromethane) and low (ethyl acetate and chloroform) vapor pressures. Mean mobility diameter measured using a Scanning Mobility Particle Sizer was in the range of 123-132 nm with a unimodal distribution and a geometric standard deviation of 1.6-1.9. A layered particle structure was observed using transmission electron microscopy, which suggests partial drug enrichment in the surface layer. Higher drug loading (20% w/w) and uniform entrapment efficiencies (~100%) were achieved. The initial drug to lipid ratio (1:4 w/w) of the precursor solution was preserved in the synthesized lipid matrices. The crystallinity of the gefitinib-loaded lipid matrix was measured using X-ray diffraction and differential scanning calorimetry. In vitro drug release from gefitinib-loaded NALM in phosphate buffered saline (pH 7.2) over 10 days showed an initial fast release period followed by a prolonged sustained release period with varying release rates. Gefitinib-loaded NALM synthesized at higher evaporation rates exhibited lower degrees of crystallinity and faster drug releases. These results suggest the determinant role of lipid crystallinity manipulated by differing evaporation rates during aerosol synthesis on drug releases from nanometer-sized lipid matrices.

Rim, D., Green, M., Wallace, L., Persily, A., and Choi, J.-I., 2012, "Evolution of Ultrafine Particle Size Distributions Following Indoor Episodic Releases: Relative Importance of Coagulation, Deposition and Ventilation," *Aerosol Science and Technology*, **46**(5): 494–503

Indoor ultrafine particles (UFP, <100 nm) undergo aerosol processes such as coagulation and deposition, which alter UFP size distribution and accordingly the level of exposure to UFP of different sizes. This study investigates the decay of indoor UFP originated from five different sources: a gas stove and an electric stove, a candle, a hair dryer, and power tools in a residential test building. An indoor aerosol model was developed to investigate differential effects of coagulation, deposition, and ventilation. The coagulation model includes Brownian, van der Waals, and viscosity forces, and also fractal geometry for particles of >24 nm. The model was parameterized using different values of the Hamaker constant for predicting the coagulation rate. Deposition was determined for two different conditions: central fan on versus central fan off. For the case of a central fan running, deposition rates were measured by using a nonlinear solution to the mass balance equation for the whole building. For the central fan off case, an empirical model was used to estimate deposition rates. Ventilation was measured continuously using an

automated tracer gas injection and sampling system. The study results show that coagulation is a significant aerosol process for UFP dynamics and the primary cause for the shift of particle size distribution following an episodic high-concentration UFP release with no fans operating. However, with the central mechanical fan on, UFP deposition loss is substantial and comparable to the coagulation loss. These results suggest that Received

Rim, D., Wallace, L., Nabinger, S., and Persily, A., 2012, "Reduction of exposure to ultrafine particles by kitchen exhaust hoods: the effects of exhaust flow rates, particle size, and burner position.," *The Science of the total environment*, **432**: 350–6

Cooking stoves, both gas and electric, are one of the strongest and most common sources of ultrafine particles (UFP) in homes. UFP have been shown to be associated with adverse health effects such as DNA damage and respiratory and cardiovascular diseases. This study investigates the effectiveness of kitchen exhaust hoods in reducing indoor levels of UFP emitted from a gas stove and oven. Measurements in an unoccupied manufactured house monitored size-resolved UFP (2 nm to 100 nm) concentrations from the gas stove and oven while varying range hood flow rate and burner position. The air change rate in the building was measured continuously based on the decay of a tracer gas (sulfur hexafluoride, SF<sub>6</sub>). The results show that range hood flow rate and burner position (front vs. rear) can have strong effects on the reduction of indoor levels of UFP released from the stove and oven, subsequently reducing occupant exposure to UFP. Higher range hood flow rates are generally more effective for UFP reduction, though the reduction varies with particle diameter. The influence of the range hood exhaust is larger for the back burner than for the front burner. The number-weighted particle reductions for range hood flow rates varying between 100 m<sup>3</sup>/h and 680 m<sup>3</sup>/h range from 31% to 94% for the front burner, from 54% to 98% for the back burner, and from 39% to 96% for the oven.

Sze-to, G. N., Wu, C. L., Chao, C. Y. H., Wan, M. P., and Chan, T. C., 2012, "Exposure and cancer risk toward cooking-generated ultrafine and coarse particles in Hong Kong homes," *HVAC&R Research*, **18**(204): 204–216

Particulate matters generated during cooking contain various carcinogens. These particles consist of both ultrafine particles (nanoparticle) and coarse particles. Exposure and risk-assessment studies often use particle mass concentration as dosimetry, which ignores the impact of ultrafine particles due to their insignificant mass compared to coarse particles. This study aims at characterizing the cancer risk toward cooking-generated particulate matter using Hong Kong homes as an example. A risk-assessment scheme modified from an existing risk model was developed to consider the cancer risk contributed from both fine ultrafine and coarse particles. Exposure assessment was conducted based on particle concentration data measured in 16 Hong Kong homes. The predicted cancer risk was then compared to the cancer incidence rate in Hong Kong to examine its appropriateness. It was found that the ultrafine particles contribute a much higher risk than that of coarse particles, and the modified risk-assessment scheme gives an estimate much closer to the incidence rate than the conventional scheme. Use of grease extractor cannot completely contain the particles, and a significant portion of particles can be transported from kitchens to other regions of the homes. The modified risk assessment scheme can serve as a tool in assessing environmental quality as well as setting up design and ventilation guidelines and exposure standards on particulate matters.

## 2011

Bari, M. A., Baumbach, G., Brodbeck, J., Struschka, M., Kuch, B., Dreher, W., and Scheffknecht, G., 2011, "Characterisation of particulates and carcinogenic polycyclic aromatic hydrocarbons in wintertime wood-fired heating in residential areas," *Atmospheric Environment*, **45**(40): 7627–7634

Wood as a renewable and worldwide available fuel is used for residential heating in small-scale firings during winter. This wood combustion can cause very high emissions of inhalable particles resulting in short and long-term health effects. The target of this study was to characterise particulate matter, emissions of polycyclic aromatic hydrocarbons (PAHs) and their size distribution and to show that those emissions can be found in the ambient air of residential areas with wood-fired heating. Emission samples were collected from pellet stove and log wood boiler under different combustion conditions. Ambient PM<sub>10</sub> sampling was performed during two winter seasons at two rural residential areas near Stuttgart in Germany. Samples were extracted using toluene with ultrasonic bath and analysed by gas chromatography mass spectrometry (GC-MS). Twenty-one PAH compounds including nine carcinogenic ones were detected and quantified. It was found that emission concentrations of carcinogenic PAHs were higher during incomplete combustion compared to complete combustion. Significant amounts of ambient PAHs were found in the residential villages, where the contribution of carcinogenic PAHs was 44% of total PAHs in the ambient air during winter 2009. The morphology and elemental analysis of ambient particles were also investigated. The findings indicate a rising concern to reduce emissions from wood-fired heating during winter in residential areas and underline the importance of using good wood combustion technologies to improve the air quality.

Cho, H.-W., Yoon, C.-S., Lee, J.-H., Lee, S.-J., Viner, A., and Johnson, E. W., 2011, "Comparison of pressure drop and filtration efficiency of particulate respirators using welding fumes and sodium chloride.," *The Annals of occupational hygiene*, **55**(6): 666–80

Respirators are used to help reduce exposure to a variety of contaminants in workplaces. Test aerosols used for certification of particulate respirators (PRs) include sodium chloride (NaCl), dioctyl phthalate, and paraffin oil. These aerosols are generally assumed to be worst case surrogates for aerosols found in the workplace. No data have been published to date on the performance of PRs with welding fumes, a hazardous aerosol that exists in real workplace settings. The aim of this study was to compare the performance of respirators and filters against a NaCl aerosol and a welding fume aerosol and determine whether or not a correlation between the two could be made. Fifteen commercial PRs and filters (seven filtering facepiece, two replaceable single-type filters, and six replaceable dual-type filters) were chosen for investigation. Four of the filtering facepiece respirators, one of the single-type filters, and all of the dual-type filters contained carbon to help reduce exposure to ozone and other vapors generated during the welding process. For the NaCl test, a modified National Institute for Occupational Safety and Health protocol was adopted for use with the TSI Model 8130 automated filter tester. For the welding fume test, welding fumes from mild steel flux-cored arcs were generated and measured with a SIBATA filter tester (AP-634A, Japan) and a manometer in the upstream and downstream sections of the test chamber. Size distributions of the two aerosols were measured using a Scanning Mobility Particle Sizer spectrometer. Penetration and pressure drop were measured over a period of aerosol loading onto the respirator or filter. Photos and scanning electron microscope images of clean and exposed respirators were taken. The count median diameter (CMD) and mass median diameter (MMD) for the NaCl aerosol were smaller than the welding fumes (CMD: 74 versus 216 nm; MMD: 198 versus 528 nm, respectively). Initial penetration and peak penetration were higher with the NaCl aerosol. However, pressure drop increased much more rapidly in the welding fume test than the NaCl aerosol test. The data and images clearly show differences in performance trends between respirator models. Therefore, general correlations between NaCl and weld fume data could not be made. These findings suggest that respirators certified with a surrogate test aerosol such as NaCl are appropriate for filtering welding fume (based on penetration). However, some respirators may have a more rapid increase in pressure drop from the welding fume accumulating on the filter. Therefore, welders will need to choose which models are easier to breathe through for the duration of their use and replace respirators or filters according to the user instructions and local regulations.

## 2010

Buonanno, G., Morawska, L., Stabile, L., and Viola, A., 2010, "Exposure to particle number, surface area and PM concentrations in pizzerias," *Atmospheric Environment*, **44**(32): 3963–3969

The aim of this work was to quantify exposure to particles emitted by wood-fired ovens in pizzerias. Overall, 15 microenvironments were chosen and analyzed in a 14-month experimental campaign. Particle number concentration and distribution were measured simultaneously using a Condensation Particle Counter (CPC), a Scanning Mobility Particle Sizer (SMPS) spectrometer, an Aerodynamic Particle Sizer (APS). The surface area and mass distributions and concentrations, as well as the estimation of lung deposition surface area and PM<sub>1</sub> were evaluated using the SMPS-APS system with dosimetric models, by taking into account the presence of aggregates on the basis of the Idealized Aggregate (IA) theory. The fraction of inhaled particles deposited in the respiratory system and different fractions of particulate matter were also measured by means of a Nanoparticle Surface Area Monitor (NSAM) and a photometer (DustTrak DRX monitor), respectively. In this way, supplementary data were obtained during the monitoring of trends inside the pizzerias. We found that surface area and PM<sub>1</sub> particle concentrations in pizzerias can be very high, especially when compared to other critical microenvironments, such as the transport hubs. During pizza cooking under normal ventilation conditions, concentrations were found up to 74, 70 and 23 times higher than background levels for number, surface area and PM<sub>1</sub>, respectively. A key parameter is the oven shape factor, defined as the ratio between the size of the face opening in respect to the diameter of the semicircular oven door, and particular attention must also be paid to hood efficiency

Sleiman, M., Destailats, H., Smith, J. D., Liu, C.-L., Ahmed, M., Wilson, K. R., and Gundel, L. a., 2010, "Secondary organic aerosol formation from ozone-initiated reactions with nicotine and secondhand tobacco smoke," *Atmospheric Environment*, **44**(34): 4191–4198

We used controlled laboratory experiments to evaluate the aerosol-forming potential of ozone reactions with nicotine and secondhand smoke. Special attention was devoted to real-time monitoring of the particle size distribution and chemical composition of SOA as they are believed to be key factors determining the toxicity of SOA. The experimental approach was based on using a vacuum ultraviolet photon ionization time-of-flight aerosol mass spectrometer (VUV-AMS), a Scanning Mobility Particle Sizer (SMPS) spectrometer and off-line thermal desorption coupled to mass spectrometry (TD-GC-MS) for gas-phase byproducts analysis. Results showed that exposure of SHS to ozone induced the formation of ultrafine particles (<100 nm) that contained high molecular weight nitrogenated species (m/z 400-500),

which can be due to accretion/acid-base reactions and formation of oligomers. In addition, nicotine was found to contribute significantly (with yields 4-9%) to the formation of secondary organic aerosol through reaction with ozone. The main constituents of the resulting SOA were tentatively identified and a reaction mechanism was proposed to elucidate their formation. These findings identify a new component of thirdhand smoke that is associated with the formation of ultrafine particles (UFP) through oxidative aging of secondhand smoke. The significance of this chemistry for indoor exposure and health effects is highlighted.

## 2009

Buonanno, G., Morawska, L., and Stabile, L., 2009, "Particle emission factors during cooking activities," *Atmospheric Environment*, **43**(20): 3235–3242

Exposure to particles emitted by cooking activities may be responsible for a variety of respiratory health effects. However, the relationship between these exposures and their subsequent effects on health cannot be evaluated without understanding the properties of the emitted aerosol or the main parameters that influence particle emissions during cooking. Whilst traffic-related emissions, stack emissions and concentrations of ultrafine particles (UFPs, diameter < 100 nm) in urban ambient air have been widely investigated for many years, indoor exposure to UFPs is a relatively new field and in order to evaluate indoor UFP emissions accurately, it is vital to improve scientific understanding of the main parameters that influence particle number, surface area and mass emissions. The main purpose of this study was to characterise the particle emissions produced during grilling and frying as a function of the food, source, cooking temperature and type of oil. Emission factors, along with particle number concentrations and size distributions were determined in the size range 0.006–20 µm using a Scanning Mobility Particle Sizer (SMPS) spectrometer and an Aerodynamic Particle Sizer (APS). An infrared camera was used to measure the temperature field. Overall, increased emission factors were observed to be a function of increased cooking temperatures. Cooking fatty foods also produced higher particle emission factors than vegetables, mainly in terms of mass concentration, and particle emission factors also varied significantly according to the type of oil used.

Demou, E., Tran, L., and Housiadas, C., 2009, "Effective biological dose from occupational exposure during nanoparticle synthesis," *Journal of Physics: Conference Series*, **151**: 012016

Nanomaterial and nanotechnology safety require the characterization of occupational exposure levels for completing a risk assessment. However, equally important is the estimation of the effective internal dose via lung deposition, transport and clearance mechanisms. An integrated source-to-biological dose assessment study is presented using real monitoring data collected during nanoparticle synthesis. Experimental monitoring data of airborne exposure levels during nanoparticle synthesis of CaSO<sub>4</sub> and BiPO<sub>4</sub> nanoparticles in a research laboratory is coupled with a human lung transport and deposition model, which solves in an Eulerian framework the general dynamic equation for polydisperse aerosols using particle specific physical-chemical properties. Subsequently, the lung deposition model is coupled with a mathematical particle clearance model providing the effective biological dose as well as the time course of the biological dose build-up after exposure. The results for the example of BiPO<sub>4</sub> demonstrate that even short exposures throughout the day can lead to particle doses of 1.10·E+08 #/(kg-bw·8h-shift), with the majority accumulating in the pulmonary region. Clearance of particles is slow and is not completed within a working shift following a 1 hour exposure. It mostly occurs via macrophage activity in the alveolar region, with small amounts transported to the interstitium and less to the lymph nodes.

## 2008

Waring, M. S., Siegel, J. a., and Corsi, R. L., 2008, "Ultrafine particle removal and generation by portable air cleaners," *Atmospheric Environment*, **42**(20): 5003–5014

Portable air cleaners can both remove and generate pollutants indoors. To investigate these phenomena, we conducted a two-phase investigation in a 14.75 m<sup>3</sup> stainless steel chamber. In the first phase, particle size-resolved (12.6–514 nm diameter) clean air delivery rates (CADR) and efficiencies were determined, as were ozone emission rates, for two high-efficiency particle arresting (HEPA) filters, one electrostatic precipitator with a fan, and two ion generators without fans. The two HEPA air cleaners had count average CADR (standard deviation) of 188 (30) and 324 (44) m<sup>3</sup> h<sup>-1</sup>; the electrostatic precipitator 284 (62) m<sup>3</sup> h<sup>-1</sup>; and the two ion generators 41 (11) and 35 (13) m<sup>3</sup>h<sup>-1</sup>. The electrostatic precipitator emitted ozone at a rate of 3.8 ± 0.2 mg h<sup>-1</sup>, and the two ion generators 3.3 ± 0.2 and 4.3 ± 0.2mg h<sup>-1</sup>. Ozone initiates reactions with certain unsaturated organic compounds that produce ultrafine and fine particles, carbonyls, other oxidized products, and free radicals. During the second phase, five different ion generators were operated separately in the presence of a plug-in liquid or solid air freshener, representing a strong terpene source. For air exchange rates of between 0.49 and 0.96 h<sup>-1</sup>, three ion generators acted as steady-state net particle generators in the entire measured range of 4.61–157 nm, and two generated particles in the range of approximately 10 to 39-55 nm. Terpene and aldehyde



concentrations were also sampled for one ion generator, and concentrations of terpenes decreased and formaldehyde increased. Given these results, the pollutant removal benefits of ozone-generating air cleaners may be outweighed by the generation of indoor pollution.

## 2007

Chang, Yu-Chen; Lee, Hsiu-Wei; Tseng, Huan-Hsiung, 2007, "The formation of incense smoke," *Journal of Aerosol Science*, **38**(1):39-51

The formation of incense smoke generated from four different types of incense sticks, three manufactured in Taiwan and one in Japan, was investigated in a small controlled chamber. The Scanning Mobility Particle Sizer and the quartz crystal microbalance were used for particle size analyses. The count median diameter (CMD) was found to rise swiftly along the path of the incense smoke. Consequently, a representative sampling location was selected for all measurements performed thereafter. All four types of incense smoke were shown to exhibit characteristic size distributions, CMDs, and mass median aerodynamic diameters (MMADs). Electron microscopy depicted liquid and solid nature of Taiwan and Japan incense smoke, respectively. The different physical states of the particles were suspected to be a result of different smoke-generating ingredients used by different cultures. Finally, the formation mechanisms of both liquid and solid incense smoke were discussed.

Fromme, H.; Twardella, D.; Dietrich, S.; Heitmann, D.; Schierl, R.; Liebl, B.; Rden, H., 2007, "Particulate matter in the indoor air of classrooms—exploratory results from Munich and surrounding area," *Atmospheric Environment*, **41**(4):854-866

Numerous epidemiological studies have demonstrated the association between particle mass (PM) concentration in outside air and the occurrence of health related problems and/or diseases. However, much less is known about indoor PM concentrations and associated health risks. In particular, data are needed on air quality in schools, since children are assumed to be more vulnerable to health hazards and spend a large part of their time in classrooms.

On this background, we evaluated indoor air quality in 64 schools in the city of Munich and a neighbouring district outside the city boundary. In winter 2004–2005 in 92 classrooms, and in summer 2005 in 75 classrooms, data on indoor air climate parameters (temperature, relative humidity), carbon dioxide (CO<sub>2</sub>) and various dust particle fractions (PM<sub>10</sub>, PM<sub>2.5</sub>) were collected; for the latter both gravimetric and continuous measurements by laser aerosol spectrometer (LAS) were implemented. In the summer period, the particle number concentration (PNC), was determined using a Scanning Mobility Particle Sizer (SMPS) spectrometer. Additionally, data on room and building characteristics were collected by use of a standardized form. Only data collected during teaching hours were considered in analysis. For continuously measured parameters the daily median was used to describe the exposure level in a classroom.

The median indoor CO<sub>2</sub> concentration in a classroom was 1603 ppm in winter and 405 ppm in summer. With LAS in winter, median PM concentrations of 19.8 µg m<sup>-3</sup> (PM<sub>2.5</sub>) and 91.5 µg m<sup>-3</sup> (PM<sub>10</sub>) were observed, in summer PM concentrations were significantly reduced (median PM<sub>2.5</sub>=12.7 µg m<sup>-3</sup>, median PM<sub>10</sub>=64.9 µg m<sup>-3</sup>). PM<sub>2.5</sub> concentrations determined by the gravimetric method were in general higher (median in winter: 36.7 µg m<sup>-3</sup>, median in summer: 20.2 µg m<sup>-3</sup>) but correlated strongly with the LAS-measured results. In explorative analysis, we identified a significant increase of LAS-measured PM<sub>2.5</sub> by 1.7 µg m<sup>-3</sup> per increase in humidity by 10%, by 0.5 µg m<sup>-3</sup> per increase in CO<sub>2</sub> indoor concentration by 100 ppm, and a decrease by 2.8 µg m<sup>-3</sup> in 5–7th grade classes and by 7.3 µg m<sup>-3</sup> in class 8–11 compared to 1–4th class. During the winter period, the associations were stronger regarding class level, reverse regarding humidity (a decrease by 6.4 µg m<sup>-3</sup> per increase in 10% humidity) and absent regarding CO<sub>2</sub> indoor concentration. The median PNC measured in 36 classrooms ranged between 2622 and 12,145 particles cm<sup>-3</sup> (median: 5660 particles cm<sup>-3</sup>).

The results clearly show that exposure to particulate matter in school is high. The increased PM concentrations in winter and their correlation with high CO<sub>2</sub> concentrations indicate that inadequate ventilation plays a major role in the establishment of poor indoor air quality. Additionally, the increased PM concentration in low level classes and in rooms with high number of pupils suggest that the physical activity of pupils, which is assumed to be more pronounced in younger children, contributes to a constant process of resuspension of sedimented particles. Further investigations are necessary to increase knowledge on predictors of PM concentration, to assess the toxic potential of indoor particles and to develop and test strategies how to ensure improved indoor air quality in schools.

## 2006

Destallats H.; Lunden M.M.; Singer B.C.; Coleman B.K.; Hodgson A.T.; Weschler C.J.; Nazaroff W.W., 2006, "Indoor secondary pollutants from household product emissions in the presence of ozone: A bench-scale chamber study," *Environmental Science and Technology*, **40**(14): 4421-4428

Ozone-driven chemistry is a source of indoor secondary pollutants of potential health concern. This study investigates secondary air pollutants formed from reactions between constituents of household products and ozone. Gas-phase product emissions were introduced along with ozone at constant rates into a 198-L Teflon-lined reaction chamber. Gas-phase concentrations of reactive terpenoids and oxidation products were measured. Formaldehyde was a predominant oxidation byproduct for the three studied products, with yields for most conditions of 20-30% with respect to ozone consumed. Acetaldehyde, acetone, glycolaldehyde, formic acid, and acetic acid were each also detected for two or three of the products. Immediately upon mixing of reactants, a Scanning Mobility Particle Sizer detected particle nucleation events that were followed by a significant degree of secondary particle growth. The production of secondary gaseous pollutants and particles depended primarily on the ozone level and was influenced by other parameters such as the air-exchange rate. Hydroxyl radical concentrations in the range  $0.04\text{-}200 \times 10^5$  molecules/cm<sup>3</sup> were determined by an indirect method. OH concentrations were observed to vary strongly with residual ozone level in the chamber, which was in the range 1-25 ppb, as is consistent with expectations from a simplified kinetic model. In a separate chamber study, we exposed the dry residue of two products to ozone and observed the formation of gas-phase and particle-phase secondary oxidation products.

## 2005

Chen, Chih-Chieh; Huang, Sheng-Hsiu; Kuo, Yu-Mei; Lin, Wane-Yun; Shih, Tung-Sheng; Weng, Yi-Mei, 2005, "Development of a size-selective inlet-simulating ICRP lung deposition fraction," *Aerosol Science and Technology*, **39**(5):437-443

A size-selective inlet made of polyurethane filter foam was designed and fabricated to simulate a portion of the ICRP respiratory deposition curve. A downstream aerosol measuring device then could be used to generate aerosol concentration data that represented the fraction reaching the respiratory system. This article introduces useful knowledge about porous foam penetration for particle size ranges below those reported in the previous studies. Different porosities of polyurethane foam filters were tested for aerosol penetration. Among the parameters operated in this work were (1) foam porosity (ppi), (2) filter thickness, (3) face velocity, and (4) packing density of the filter foams. Di-octyl ph-thalate was used as the test agent. A constant output atomizer and an ultrasonic atomizing nozzle were used to generate polydisperse submicrometer- and micrometer-sized particles, respectively. Aerosol concentrations and size distributions upstream and downstream of the filter foams were monitored by using a Scanning Mobility Particle Sizer (for particles with diameters smaller than 0.7  $\mu\text{m}$ ) and an aerodynamic particle sizer (for particles larger than 0.7  $\mu\text{m}$ ). The aerosol output was neutralized by a radioactive source. A lognormal-distribution curve with a mode of 0.25  $\mu\text{m}$  and a GSD of 6.2 was set as the primary target curve simulating the light-work ICRP deposition model. The results showed that the most penetrating size (also referred to as collection minimum) of the filter foams decreased upon increasing the foam porosity, packing density, and face velocity. In this work, the highest foam porosity and packing density we could acquire were 100 ppi and 0.2, respectively. By adjusting the face velocity, the most penetrating size was moved to 0.25  $\mu\text{m}$ , which happened to be the most penetrating size for ICRP light-work criterion. The whole aerosol penetration curve could further fit to the modified ICRP curve by adjusting the filter thickness. There are numerous ways to match the ICRP definition. This size-selective inlet becomes even more versatile if the auxiliary detector and vacuum system are operated under different flow rates to simulate light-to-heavy workloads. 19 Refs.

Gilbert, Dale; He, Congrong; Morawska, Lidia, 2005, "Particle deposition rates in residential houses," *Atmospheric Environment*, **39**(21): 3891-3899

As part of a large study investigating indoor air in residential houses in Brisbane, Australia, the purpose of this work was to quantify the particle deposition rate of size classified particles in the size range from 0.015 to 6  $\mu\text{m}$ . Particle size distribution resulting from cooking, repeated under two different ventilation conditions in 14 houses, as well as changes to particle size distribution and PM<sub>2.5</sub> concentration as a function of time, were measured using a Scanning Mobility Particle Sizer (SMPS) spectrometer, an aerodynamic particle sizer (APS), and a DustTrak monitor. Deposition rates were determined by regression fitting of the measured size-resolved particle number and PM<sub>2.5</sub> concentration decay curves, and accounting for air exchange rate. The measured deposition rates were shown to be particle size dependent and they varied from house to house. The lowest deposition rates were found for particles in the size range from 0.2 to 0.3  $\mu\text{m}$  for both minimum (air exchange rate:  $0.61 \pm 0.45$  /h) and normal (air exchange rate:  $3.00 \pm 1.23$  /h) ventilation conditions. The results of statistical analysis indicated that ventilation condition (measured in terms of air exchange rate) was an important factor affecting deposition rates for particles in the size range from 0.08 to 1.0  $\mu\text{m}$ , but not for particles smaller than 0.08  $\mu\text{m}$  or larger than 1.0  $\mu\text{m}$ . Particle coagulation was assessed to be negligible compared to the two other

processes of removal: ventilation and deposition. This study of particle deposition rates, the largest conducted so far in terms of the number of residential houses investigated, demonstrated trends in deposition rates comparable with studies previously reported, usually for significantly smaller samples of houses (often only one). However, the results compare better with studies which, similarly to this study, investigated cooking as a source of particles (particle sources investigated in other studies included general activity, cleaning, artificial particles, etc). 35 Refs.

Hitchins-Loveday, Jane; Hofmann, Werner; Mengersen, Kerrie; Morawska, Lidia; Swanson, Cheryl, 2005, "Experimental study of the deposition of combustion aerosols in the human respiratory tract," *Journal of Aerosol Science*, **38**(8): 939-957

The total deposition of environmental tobacco smoke (ETS), diesel and petrol smoke in the respiratory tract of 14 non-smokers between the ages of 20 and 30 was determined experimentally. A Scanning Mobility Particle Sizer (SMPS) spectrometer measuring a size range of 0.016-0.626 $\mu$ m was used to characterise the inhaled and exhaled aerosol during relaxed nasal breathing over a period of 10 min. The ETS, diesel, and petrol particles had average count median diameter (and geometric standard deviation) of 0.183  $\mu$ m (1.7), 0.125 $\mu$ m (1.7), and 0.069 $\mu$ m (1.7), respectively. The average total number deposition of ETS was 36% (standard deviation 10%), of diesel smoke 30% (standard deviation 9%), and of petrol smoke 41% (standard deviation 8%). The analysis of the deposition patterns as a function of particle size for the three aerosols in each individual showed that there is a significant difference between each aerosol for a majority of individuals (12 out of 14). This is an important result as it indicates that differences persist regardless of inter-subject variability. 13 Refs.

Smolík, Jiří; Lazaridis, Mihalis; Moravec, Pavel; Schwarz, Jaroslav; Zaripov, Shamil; Dímal, Vladimír, 2005, "Indoor Aerosol Particle Deposition in an Empty Office," *Water, Air, and Soil Pollution*, **165**(1-4):301-312

The size- and time-resolved indoor/outdoor aerosol concentration relationships were studied experimentally in an empty office without internal particle sources. Two Scanning Mobility Particle Sizer (SMPS) spectrometers and an Aerodynamic Particle Sizer (APS) spectrometer sampled alternately from indoor and outdoor, together covering the size range 3–10,000 nm. The results showed that the indoor aerosol concentration depends mainly on the air exchange and deposition rates, and the outdoor concentration. At higher air exchange rates the indoor aerosol concentration approaches the outdoor one. This was observed for the accumulation mode particles. The size-dependent deposition rates were estimated using a simple aerosol dynamics experiment. It was based on equilibration of the indoor/outdoor concentrations by opening windows and analyzing the concentration decay curves after the windows were closed. For this purpose a simple mass balance model was used. The overall loss rate and the asymptotic value of indoor concentration were found by applying a non-linear least squares method on the time dependence of the indoor concentrations. The air exchange rate was estimated from the overall loss rate and the steady state values of the indoor/outdoor concentration of the accumulation mode particles.

## 2004

Brouwer, D.H.; Gijsbers, J.H.; Lurvink, M.W., 2004, "Personal exposure to ultrafine particles in the workplace: Exploring sampling techniques and strategies," *Annals of Occupational Hygiene*, **48**(5):439-453

Recently, toxicological and epidemiological studies on health effects related to particle exposure suggest that 'ultrafine particles' (particles with an aerodynamic diameter of <100 nm) may cause severe health effects after inhalation. Although the toxicological mechanisms for these effects have not yet been explained, it is apparent that measuring exposures against mass alone is not sufficient. It is also necessary to consider exposures against surface area and number concentration. From earlier research it was hypothesized that results on number concentration and particle distributions may vary with distance to the source, limiting the reliability of estimates of personal exposure from results which were obtained using static measurement equipment. Therefore, a workplace study was conducted to explore the performance of measurement methods in a multi-source emission scenario as part of a sampling strategy to estimate personal exposure. In addition, a laboratory study was conducted to determine possible influences of both distance to source and time course on particle number concentration and particle size distribution. In both studies different measurement equipment and techniques were used to characterize (total) particle number concentration. These included a condensation particle counter (CPC), a Scanning Mobility Particle Sizer (SMPS) spectrometer and an electrical low pressure impactor (ELPI). For the present studies CPC devices seemed to perform well for the identification of particle emission sources. The range of ultrafine particle number concentration can be detected by both SMPS and ELPI. An important advantage of the ELPI is that aerosols with ultrafine sizes can be collected for further analysis. Specific surface area of the aerosols can be estimated using gas adsorption analysis; however, with this technique ultrafine particles cannot be distinguished from particles with non-ultrafine sizes. Consequently, estimates based on samples collected from the breathing zone and scanning electron

microscopic analysis may give a more reliable estimate of the specific surface area of the ultrafine particles responsible for personal exposure. The results of both the experimental and the workplace study suggest both spatial and temporal variation in total number concentration and aerosol size distribution. Therefore, the results obtained from static measurements and grab sampling should be interpreted with care as estimates of personal exposure. For evaluation of workplace exposure to ultrafine particles it is recommended that all relevant characteristics of such exposure are measured as part of a well-designed sampling strategy.

Fine, P. M.; Misra, C.; Singh, M.; Sioutas, C., 2004, "Development and Evaluation of A Compact Facility for Exposing Humans to Concentrated Ambient Ultrafine Particles," *Aerosol Science & Technology*, **38**(1):27-35

This article presents the development and evaluation of a very compact facility for exposing humans to concentrated ambient ultrafine particles ( $d_a < 0.15 \mu\text{m}$ ). The human ultrafine particle concentrator (UFPC) operates at an intake flow rate of 1200 liters per minute (LPM). The concentrator is preceded by an ultrafine impactor which separates the accumulation mode from ultrafine mode particles under a very low pressure drop (1.5 kPa), a feature that is essential in enabling human inhalation studies of ultrafine concentrated ambient particulates (CAP). A key feature of the UFPC is a new cooling system, consisting of a programmable refrigerated circulator, which produces the supersaturation that is necessary to grow ultrafine PM to supermicrometer sizes so that they can be concentrated by means of conventional virtual impaction. The new cooling system allows for entirely automated operation of the UFPC. The UFPC was characterized in field experiments, in which the concentration enrichment of ultrafine particles was determined based on their number and mass concentration as well as on chemical composition including elemental carbon (EC), inorganic ions (sulfate and nitrate), and polycyclic aromatic hydrocarbons (PAH). Tests were conducted at minor-to-total flow ratios varying from 2.5-5% (hence at minor flow rates between 30-60 LPM). Measurements with the Scanning Mobility Particle Sizer (SMPS) spectrometer showed a near-ideal increase in number concentrations (corresponding to the ratio of total-to-minor flow rate) of ultrafine particles after enrichment. The concentration enrichment was uniform across the entire particle diameter range of 15-660 nm. Similar results were obtained for EC and PAH concentrations (measured by an Aethalometer). Time-integrated filter-based tests, conducted to characterize the system for ultrafine PM mass and inorganic ion concentrations showed that the average enrichment factor was very close to the ideal values, indicating near-perfect collection efficiency with minimal particle losses. 21 Refs.

He, C.; Morawska, L.; Hitchins, J.; Gilbert, D., 2004, "Contribution from indoor sources to particle number and mass concentrations in residential houses," *Atmospheric Environment*, **38**(21):3405-3415

As part of a large study investigating indoor air in residential houses in Brisbane, Australia, the purpose of this work was to quantify emission characteristics of indoor particle sources in 15 houses. Submicrometer particle number and approximation of  $\text{PM}_{2.5}$  concentrations were measured simultaneously for more than 48 h in the kitchen of all the houses by using a condensation particle counter (CPC) and a photometer (DustTrak monitor), respectively. In addition, characterizations of particles resulting from cooking conducted in an identical way in all the houses were measured by using a Scanning Mobility Particle Sizer (SMPS) spectrometer, an aerodynamic particle sizer (APS) and a DustTrak monitor. All the events of elevated particle concentrations were linked to indoor activities using house occupants diary entries, and catalogued into 21 different types of indoor activities. This enabled quantification of the effect of indoor sources on indoor particle concentrations as well as quantification of emission rates from the sources. For example, the study found that frying, grilling, stove use, toasting, cooking pizza, cooking, candle vaporizing eucalyptus oil and fan heater use, could elevate the indoor submicrometer particle number concentration levels by more than five times, while  $\text{PM}_{2.5}$  concentrations could be up to 3, 30 and 90 times higher than the background levels during smoking, frying and grilling, respectively.

Kittelson, D.; Watts, W.; Johnson, J.; Remerowki, M.; Ische, E.; Oberdörster, G.; Gelein, R.; Elder, A.; Hopke, P.; Kim, E.; Zhao, W.; Zhou, L.; Jeong, C.-H., 2004, "On-Road Exposure to Highway Aerosols. 1. Aerosol and Gas Measurements," *Inhalation Toxicology*, **16**(S1):31-39

On-road experiments were conducted to determine the sensitivities of rats to real-world aerosol. This article summarizes the on-road aerosol and gas measurements and provides background information for the companion paper on the rat exposures. Measurements were carried out over 10 days, 6 h/day, driving a route from Rochester to Buffalo. Aerosol instrumentation used in this study included two Scanning Mobility Particle Sizer (SMPS) spectrometers to determine the aerosol size distribution from 10 to 300 nm, 2 stand-alone condensation particle counters to determine the total aerosol number concentration, and an electrical aerosol detector to determine the aerosol length concentration. A thermal denuder (TD) was used with one of the SMPS instruments to determine the size distribution of the non-volatile fraction. Filter samples were collected and analyzed for elemental carbon, and gas analyzers measured ambient levels of CO, CO<sub>2</sub>, and NO. Average daily total aerosol number concentration ranged from 200,000 to

560,000 particles/cm<sup>3</sup>. Past studies on urban highways have measured total number concentrations ranging between 10<sup>4</sup> and 10<sup>6</sup>particles/cm<sup>3</sup>. The average daily NO concentration ranged from 0.10 to 0.24 ppm and the corresponding CO<sub>2</sub>concentration ranged from 400 to 420 ppm. The average daily geometric number mean particle size determined by the SMPS ranged from 15 to 20 nm. The TD reduced the average SMPS number concentration between 87 and 95%and the SMPS volume between 54 and 83%, suggesting that most of the particles consisted of volatile material. The TD also increased the geometric number mean diameter from 15 to 20 nm to 30 to 40 nm.

Kim, C. S.; Leith, D.; Rosati, J. A., 2003, "Monodisperse and polydisperse aerosol deposition in a packed bed," *Aerosol Science & Technology*, **37**(6):528-535

Although polydisperse aerosols in ambient and occupational settings have been associated with adverse health effects, researchers have mostly used monodisperse aerosols to investigate particle deposition in the human lung. The objective of this work was to determine whether the deposition of a series of monodisperse aerosols is reasonable to simulate the deposition of a polydisperse aerosol. A packed bed was used as an approximate surrogate to the human lung. Polydisperse and monodisperse sebacate aerosols were generated by nebulizers and a Monodisperse Aerosol Generator (MAGE), respectively. A Harvard respirator pump "inhaled" and "exhaled" aerosol through the packed bed. Complete size distributions of inhaled and exhaled aerosol were measured by an Aerodynamic Particle Sizer (APS) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. The deposition for discrete sections of polydisperse aerosols was compared with the deposition of monodisperse aerosols. Also, the total deposition of a polydisperse aerosol was compared with the deposition of a series of monodisperse aerosols that formed the same size distribution as the polydisperse aerosol. Experiments were run with and without a charge neutralizer. With the neutralizer, no difference in deposition occurred between the discrete sections of polydisperse and monodisperse aerosols. Thus, total deposition was the same for both a polydisperse and a series of monodisperse aerosols, indicating that a series of monodisperse aerosols is reasonable to represent a polydisperse aerosol. Without a charge neutralizer, discrete sections of polydisperse aerosols with particle diameters of 3 µm or greater deposited no differently than they did with a neutralizer. However, for particle diameters of 1 µm or less, deposition of discrete sections of polydisperse aerosols was greater than that of monodisperse aerosols. Thus, total deposition was greater for polydisperse aerosols than for a series of monodisperse aerosols, indicating the need to condition aerosol with a neutralizer after the nebulization of a sebacate/alcohol solution. 21 Refs.

Rosati J.A.; Brown J.S.; Peters T.M.; Leith D.; Kim C.S., 2002, "A polydisperse aerosol inhalation system designed for human studies," *Journal of Aerosol Science*, **33**(10): 1433-1446

A polydisperse aerosol inhalation system has been developed to measure particle deposition in the lungs of human subjects. Nebulizers are used to generate aerosols with mass median aerodynamic diameters from 0.3 to 3 µm, and geometric standard deviations of 1.8-2.0. Inspired aerosol is drawn from a holding bag, passes through a sliding valve and a pneumotachograph, and enters a heated mouthpiece. Exhaled aerosol passes from the mouthpiece and pneumotachograph, through a second sliding valve and is collected in a heated sample bag. Inhalation and exhalation valves trigger automatically with change in flow direction through the pneumotachograph. Complete size distributions of inhaled and exhaled aerosol are measured by an aerodynamic particle sizer and a Scanning Mobility Particle Sizer spectrometer. Fractional particle deposition of a test aerosol in the lung is determined by comparing inhaled and exhaled aerosol size fractions. Total deposition is determined from the sum of the fractional depositions. This new system precludes the need for monodisperse aerosol series to simulate polydisperse aerosol data, thus substantially reducing both study length and subject exposure.

## 2003

Kim, C. S.; Leith, D.; Rosati, J. A., 2003, "Monodisperse and polydisperse aerosol deposition in a packed bed," *Aerosol Science & Technology*, **37**(6):528-535

Although polydisperse aerosols in ambient and occupational settings have been associated with adverse health effects, researchers have mostly used monodisperse aerosols to investigate particle deposition in the human lung. The objective of this work was to determine whether the deposition of a series of monodisperse aerosols is reasonable to simulate the deposition of a polydisperse aerosol. A packed bed was used as an approximate surrogate to the human lung. Polydisperse and monodisperse sebacate aerosols were generated by nebulizers and a Monodisperse Aerosol Generator (MAGE), respectively. A Harvard respirator pump "inhaled" and "exhaled" aerosol through the packed bed. Complete size distributions of inhaled and exhaled aerosol were measured by an Aerodynamic Particle Sizer (APS) and a Scanning Mobility Particle Sizer (SMPS) spectrometer. The deposition for discrete sections of polydisperse aerosols was compared with the deposition of monodisperse aerosols. Also, the total deposition of a polydisperse aerosol was compared with the deposition of a series of monodisperse aerosols that formed the same size distribution as the polydisperse aerosol. Experiments were run with and without a charge neutralizer. With the neutralizer, no difference in deposition occurred between the

discrete sections of polydisperse and monodisperse aerosols. Thus, total deposition was the same for both a polydisperse and a series of monodisperse aerosols, indicating that a series of monodisperse aerosols is reasonable to represent a polydisperse aerosol. Without a charge neutralizer, discrete sections of polydisperse aerosols with particle diameters of 3  $\mu\text{m}$  or greater deposited no differently than they did with a neutralizer. However, for particle diameters of 1  $\mu\text{m}$  or less, deposition of discrete sections of polydisperse aerosols was greater than that of monodisperse aerosols. Thus, total deposition was greater for polydisperse aerosols than for a series of monodisperse aerosols, indicating the need to condition aerosol with a neutralizer after the nebulization of a sebacate/alcohol solution.

Klepeis, Neil E.; Apte, Michael G.; Gundel, Lara A.; Sextro, Richard G.; Nazaroff, William W., 2003, "Determining Size-Specific Emission Factors for Environmental Tobacco Smoke Particles," *Aerosol Science & Technology*, **37**(10):780-790

Because size is a major controlling factor for indoor airborne particle behavior, human particle exposure assessments will benefit from improved knowledge of size-specific particle emissions. We report a method of inferring size-specific mass emission factors for indoor sources that makes use of an indoor aerosol dynamics model, measured particle concentration time series data, and an optimization routine. This approach provides--in addition to estimates of the emissions size distribution and integrated emission factors--estimates of deposition rate, an enhanced understanding of particle dynamics, and information about model performance. We applied the method to size-specific environmental tobacco smoke (ETS) particle concentrations measured every minute with an 8-channel optical particle counter (PMS-LASAIR; 0.1-2  $\mu\text{m}$  diameters) and every 10 or 30 min with a 34-channel differential mobility particle sizer (TSI-DMPS; 0.01-1  $\mu\text{m}$  diameters) after a single cigarette or cigar was machine-smoked inside a low air-exchange-rate 20  $\text{m}^3$  chamber. The aerosol dynamics model provided good fits to observed concentrations when using optimized values of mass emission rate and deposition rate for each particle size range as input. Small discrepancies observed in the first 1-2 h after smoking are likely due to the effect of particle evaporation, a process neglected by the model. Size-specific ETS particle emission factors were fit with log-normal distributions, yielding an average mass median diameter of 0.2  $\mu\text{m}$  and an average geometric standard deviation of 2.3 with no systematic differences between cigars and cigarettes. The equivalent total particle emission rate, obtained by integrating each size distribution, was 0.2-0.7  $\text{mg}/\text{min}$  for cigars and 0.7-0.9  $\text{mg}/\text{min}$  for cigarettes.

Palmgren, F.; Waahlin, P.; Kildesoe, J.; Afshari, A.; Fogh, C.L., 2003, "Characterisation of particle emissions from the driving car fleet and the contribution to ambient and indoor particle concentrations," *Physics and Chemistry of the Earth, Parts A/B/C*, **28**(8):327-334

The population is mainly exposed to high air pollution concentrations in the urban environment, where motor vehicle emissions constitute the main source of fine and ultrafine particles. These particles can penetrate deep into the respiratory system, and studies indicate that the smaller the particle, the larger the health impacts. The chemical composition, surface reactivity and physical properties are also important. However, the knowledge about chemical and physical properties of particles and the temporal and spatial variability of the smallest particles is still very limited. The present study summarises the first results of a larger project with the aims to improve the knowledge. The concentration and the emissions of ultrafine particles from petrol and diesel vehicles, respectively, have been quantified using Scanning Mobility Particle Sizer of ultrafine particles in the size range 6-700 nm and routine monitoring data from urban streets and urban background in Denmark. The quantification was carried out using receptor modelling. The number size distributions of petrol and diesel emissions showed a maximum at 20-30 nm and non-traffic at about 100 nm. The contribution of ultrafine particles from diesel vehicles is dominating in streets. The same technique has been applied on  $\text{PM}_{10}$ , and about 50 percent contribution from non-traffic. The technique has also been introduced in relation to elemental and organic carbon, and the first data showed strong correlation between traffic pollution and elemental carbon. The outdoor air quality has a significant effect on indoor pollution levels, and we spend most of the time indoors. Knowledge about the influence of ambient air pollution on the concentrations in the indoor environment is therefore crucial for assessment of human health effects of traffic pollution. The results of our studies will be included in air quality models for calculation of human exposure. Preliminary results from our first campaign showed, that the deposition rate of particles in the apartment is negligible in the particle size range 100-500 nm. In the size range below 100 nm the deposition rate increases with decreasing particle diameter to a value of approximately 1/h at 10 nm. The penetration efficiency shows a maximum of 60 percent at 100 nm. More detailed studies of exchange of particles in outdoor/indoor air and the transformation are planned to take place during three next campaigns.

Stephenson, D; Seshadri, G; Veranth, J M, 2003 "Workplace exposure to submicron particle mass and number concentrations from manual arc welding of carbon steel," *AIHA Journal*, **64**:516-521

Particle emissions from manual shielded metal arc welding of carbon steel were sampled in a typical industrial maintenance and metal fabrication workplace environment. Particle number measurements over the size range from 14 nm to 10 $\mu\text{m}$  using a Scanning Mobility Particle Sizer and an optical particle counter showed that

welding produced an approximately lognormal particle mode with a 120 nm count median and a geometric standard deviation of 2.07. This study produced welding particle number concentrations on the order of  $2 \times 10^5/\text{cm}^3$  in the building air 8.5 m away from the welding. Workplace exposure samples were below the current 8-hour American Conference of Governmental Industrial Hygienists mass concentration threshold limit value of  $5 \text{ mg}/\text{m}^3$ . Submicron particles comprised 80% of the total aerosol mass collected by a cascade impactor during welding. The concentration of larger particles was indistinguishable from indoor background. Microscopy showed that the welding emissions are dominated by clusters formed from  $<0.1 \mu\text{m}$  primary spheres. These data on the particles resulting from aerosol transformation by natural dilution inside an industrial building can be compared with laboratory-scale studies of welding particulate. The particle number characteristics observed in this study are significant because toxicological hypotheses suggest that number or surface area may be a better metric than mass when evaluating the health effects of fine particles.

## 2002

Hitchins, J.; Morawska, L.; Gilbert, D.; Jamriska, M., 2002, "Dispersion of particles from vehicle emissions around high- and low-rise buildings," *Indoor Air*, **12**(1):64-71

Understanding the distribution of outdoor pollutants around a building envelope, generated by sources located in its vicinity, is important when choosing the location of building ventilation system intakes, as well as for quantifying the exposure of people living or working in the building. A systematic experimental characterisation of the number concentration of submicrometre particles was undertaken around the envelope of six buildings (both low- and high-rise) at different distances from a road (the main pollution source). The concentrations were measured using two TSI Scanning Mobility Particle Sizing.  $\text{PM}_{2.5}$  concentrations were also monitored around the low-rise buildings using two TSI DustTrak monitors. For the three high rise buildings the concentration of fine and ultra-fine particles decreased in most cases to about 50-60% from the approximate ground level readings (between heights of 0 to 6 m), to full building height (from 24 to 33 m above the ground). Measurements of submicrometre particle number concentrations as well as  $\text{PM}_{2.5}$  fraction in the envelope around low-rise isolated buildings did not show any significant trends from the front to the rear of the building. The sensitivity of  $\text{PM}_{2.5}$  measurements to a small number of larger particles, possibly from sources other than vehicle emissions, was observed.

Sarnat, J. A.; Coull, B. A.; Koutrakis, P.; Long, C. M.; Schwartz, J.; Suh, H. H., 2002, "Using sulfur as a tracer of outdoor fine particulate matter," *Environmental Science & Technology*, **36**(24):5305-5314

Six homes in the metropolitan Boston area were sampled between 6 and 12 consecutive days for indoor and outdoor particle volume and mass concentrations, particle elemental concentrations, and air exchange rates (AERs). Indoor/outdoor (I/O) ratios of nighttime (i.e., particle nonindoor source periods) sulfur,  $\text{PM}_{2.5}$  and the specific particle size intervals were used to provide estimates of the effective penetration efficiency. Mixed models and graphical displays were used to assess the ability of the I/O ratios for sulfur to estimate corresponding I/O ratios for  $\text{PM}_{2.5}$  and the various particle sizes. Results from this analysis showed that particulate sulfur compounds were primarily of outdoor origin and behaved in a manner that was representative of total  $\text{PM}_{2.5}$  in Boston, MA. These findings support the conclusion that sulfur can be used as a suitable tracer of outdoor  $\text{PM}_{2.5}$  for the homes sampled in this study. Sulfur was more representative of particles of similar size (0.06–0.5  $\mu\text{m}$ ), providing evidence that the size composition of total  $\text{PM}_{2.5}$  is an important characteristic affecting the robustness of sulfur-based estimation methods.

## 2001

Morawska, L.; He, Congrong; Hitchins, J.; Gilbert, D.; Parappukkaran, S., 2001, "The relationship between indoor and outdoor airborne particles in the residential environment," *Atmospheric Environment*, **35**(20):3463-3473

The relationship between indoor and outdoor airborne particles was investigated for 16 residential houses located in a suburban area of Brisbane, Australia. The submicrometer particle numbers were measured using the Scanning Mobility Particle Sizer, the larger particle numbers using the Aerodynamic Particle Sizer and an approximation of  $\text{PM}_{2.5}$  was also measured using a DustTrak monitor. The measurements were conducted for normal and minimum ventilation conditions using simultaneous and non-simultaneous measurement methods designed for the purpose of the study. Comparison of the ratios of indoor to outdoor particle concentrations revealed that while temporary values of the ratio vary in a broad range from 0.2 to 2.5 for both lower and higher ventilation conditions, average values of the ratios were very close to one regardless of ventilation conditions and of particle size range. The ratios were in the range from 0.78 to 1.07 for submicrometer particles, from 0.95 to 1.0 for supermicrometer particles and from 1.01 to 1.08 for  $\text{PM}_{2.5}$  fraction. Comparison of the time series of indoor to outdoor particle concentrations shows a clear positive relationship existing for many houses under normal ventilation conditions (estimated to be about and above  $2\text{h}^{-1}$ , but not under minimum ventilation conditions

(estimated to be about and below  $1\text{h}^{-1}$ ). These results suggest that for normal ventilation conditions, outdoor particle concentrations could be used to predict instantaneous indoor particle concentrations but not for minimum ventilation, unless air exchange rate is known, thus allowing for estimation of the "delay constant".

Osunsanya, T.; Prescott, G.; Seaton, A., 2001, "Acute respiratory effects of particles: mass or number?" *Occupational and Environmental Medicine*, **58**(3):154-159

To determine whether associations might be found, in patients with chronic airflow obstruction, between symptoms, peak flow rate (PEF), and particle mass and numbers, and to assess which measure was most closely associated with changes in health. Epidemiological studies have shown associations between particulate air pollution and cardiovascular and respiratory disease, and it has been proposed that these may be mediated by particles of nm size (ultrafine). Relations were investigated between symptom scores, PEF, and bronchodilator use in 44 patients aged  $\geq 50$  years with chronic obstructive pulmonary disease, and daily measurements of both mass of ambient particles of aerodynamic diameter less than  $10\ \mu\text{m}$  ( $\text{PM}_{10}$ ) and numbers of ultrafine particles ( $<100\ \text{nm}$ ), allowing for meteorological variables. Symptom scores, bronchodilator use, and PEF were recorded daily for 3 months. Counts of ultrafine particles were made by the TSI model 3934 Scanning Mobility Particle Sizer (SMPS) spectrometer and  $\text{PM}_{10}$  measurements by the tapered element oscillating microbalance (TEOM). Ultrafine particle counts indoors and outdoors were significantly correlated, those indoors being about half of those outdoors. No associations were found between actual PEF and  $\text{PM}_{10}$  or ultrafine particles. However, there was a 19% increase in the rate of 10% decrements in daytime PEF with increases in  $\text{PM}_{10}$  from 10 to  $20\ \mu\text{g}/\text{m}^3$  which was of borderline significance ( $p=0.05$ ). A change in  $\text{PM}_{10}$  from 10 to  $20\ \mu\text{g}/\text{m}^3$  was significantly associated with a 14% increase in the rate of high scores of shortness of breath ( $p=0.003$ ). A similar change in  $\text{PM}_{10}$  as a moving average of the same day and 2 previous days was associated with a 31% increase in the rate of high scores for cough ( $p=0.02$ ). Cough symptoms were also associated with lower temperatures ( $p=0.02$ ). Higher use of medicines was also associated with higher  $\text{PM}_{10}$ , but the increases were very small in clinical terms. Evidence was not found to support the hypothesis that the component of particulate pollution responsible for effects on respiratory symptoms or function resides in the fraction below  $100\ \text{nm}$  diameter. The consistent associations between symptoms and  $\text{PM}_{10}$  suggest that a contribution of the coarser fraction should not be dismissed. Further studies will be needed before the conclusions of this specific project may be generalised.

## 2000

Abt, E.; Suh, H. H.; Catalano, P.; Koutrakis, P., 2000, "Relative contribution of outdoor and indoor particle sources to indoor concentrations," *Environmental Science and Technology*, **34**(17):3579-3587

The effect of indoor particle sources on indoor particle size distributions and concentrations was previously investigated using real-time indoor and outdoor particle size distribution data collected in four homes in Boston in 1996. These data demonstrated the importance of indoor sources (i.e., cooking, cleaning, and movement of people) and air exchange rates on observed indoor concentrations. As part of the continued analyses of these data, a simple physical model was used to determine the source emission and infiltration rates for specific particle sizes. Decay rates were also estimated. Cooking, cleaning, and indoor work (characterized by movement of people) significantly increased  $\text{PM}_{(0.7-10)}$  concentrations by  $0.27$ ,  $0.27$ , and  $0.25\ \mu\text{m}^3\ \text{cm}^{-3}\ \text{min}^{-1}$ , respectively. Cooking was the only variable significantly associated with generation of particles less than  $0.5\ \mu\text{m}$  in diameter. Outdoor particles ( $0.02$ - $0.5$  and  $0.7$ - $10\ \mu\text{m}$ ) were found to contribute significantly to indoor particle levels. Effective penetration efficiencies ranged from  $0.38$  to  $0.94$  for  $0.02$ - $0.5\ \mu\text{m}$  particles and from  $0.12$  to  $0.53$  for  $0.7$ - $10\ \mu\text{m}$  particles. Estimates for  $0.7$ - $10\ \mu\text{m}$  particles decreased with increasing particle size, reflecting the influence of deposition losses from gravitational settling. The real-time particle size distribution data in conjunction with time-activity information provides valuable information on the origin and fate of indoor particles.

Chang, Y. C.; Tseng, H. H.; Hong, G. I.; Wang, K. T., 2000, "Measurement of smoke properties of incenses made in three countries," *Journal of Aerosol Science*, **31**(S1):S488-S489

Smoke properties of incenses made in different countries are investigated. The system used consists of a flow control/conditioning console, a smoke chamber ( $0.035\ \text{m}^3$ ), an electrical resistance type of ignition control unit, and particle measuring instrumentation. The results showed that the smoke properties such as CMD and  $\text{MMAD}_{50}$  were very similar for all three incense samples. The maximum total mass and the total number concentrations differ from each other. 3 Refs.

Ristovski, Z. D.; Tass, I.; Morawska, L.; Saxby, W., 2000, "Investigation into the emission of fine particles, formaldehyde, oxides of nitrogen and carbon monoxide from natural gas heaters," *Journal of Aerosol Science*, **31**(S1):S490-S491

The range of concentration levels of submicrometer particles, formaldehyde (HCHO), oxides of nitrogen ( $\text{NO}_x$ ), and carbon monoxide (CO) that arises from the standard operation of four typical flueless natural



gas heaters are investigated. The results showed that both fine particle mass and number emission rates are low and the mass ones are close to 0.046 ng/J. This showed that the emission of particles during the combustion of natural gas in domestic heaters do not contribute to the indoor particle concentration. Natural gas heaters are a larger contributor of organic compounds such as formaldehyde and NO<sub>2</sub>.

Wallace L., 2000, "Real-Time Monitoring of Particles, PAH, and CO in an Occupied Townhouse," *Applied Occupational and Environmental Hygiene*, **15**(1):39-47

Beginning in October 1996, indoor and sometimes outdoor air at an occupied house in a suburban area of Virginia has been monitored continuously for particles, PAH, and CO. Two Climet monitors have been used to count particles in six size ranges between 0.3 and > 10 µm, with 1-minute averages being collected every 5 minutes. Two Ecochem PAH monitors have been used to sample for particle-bound PAHs once every minute. Also, two Langan CO monitor-data loggers have measured CO once each minute while logging the PAH data. Two Aethalometers measure black carbon. A single Scanning Mobility Particle Sizer (SMPS) spectrometer measures ultrafine particles. The pairs of monitors are set up either to provide an indoor/outdoor or an upstairs office/downstairs kitchen comparison. Air exchange is occasionally measured using a Bruel & Kjaer 1302 SF6 monitor, as a parameter necessary for estimating deposition rates for particles and PAH. Results from the first 16 months of monitoring (10M observations) include: neighborhood woodburning and morning rush hour traffic are the most important sources of PAH and black carbon outdoors; candles, matches, incense, and frying, sauteeing, broiling, deep-frying, and stir-frying are additional important indoor sources of PM. One citronella candle was an extremely powerful PAH source. Neither woodburning nor vehicles appears to be an important source of particles indoors, but frying, grilling, and sauteeing are extremely strong indoor sources, together with combustion events such as use of matches and candles. Physical movement was an important source of coarse but not fine particles. Use of the gas stove for extended periods of time led to increased CO concentrations—vehicles and woodburning were relatively minor sources in comparison. The gas oven, gas burners, and electric toaster oven were important sources of ultrafine particles (< 0.1 µm). A source-proximity effect was noted, with the kitchen monitor reading two to five times higher than the upstairs monitor for particles from kitchen events, while the upstairs monitor often read higher than the kitchen monitor for events caused by physical activity alone.

## 1999

Morawska, L.; Barron, W.; Hitchins, J., 1999, "Experimental deposition of environmental tobacco smoke submicrometer particulate matter in the human respiratory tract," *American Industrial Hygiene Association Journal*, **60**(3):334-339

Measurements of 15 nonsmokers and 3 smokers breathing environmental tobacco smoke (ETS), were conducted to study particle deposition within the human respiratory tract. The subjects inhaled ETS of count median diameter (CMD) of about 0.2 µm and geometric standard deviation (GSD) of 1.7. The particle size distribution in the submicrometer range in the inhaled and exhaled air from the subjects was measured using a Scanning Mobility Particle Sizer (SMPS) spectrometer. A deposition of 56.0 plus or minus 15.9% was observed for nonsmokers while breathing ETS through the nose and 48.7 plus or minus 11.6% while breathing ETS through the mouth. One individual tested four times gave an average deposition of 57.4 plus or minus 11.5%, providing an indication of intra-individual variation. Such a variation is expected since the breathing rate was not controlled in order that an indication of the deposition experienced on a day-to-day basis could be obtained. For nonsmokers the deposition while breathing through the mouth was lower than through the nose and the variability within the measurements was also lower for mouth breathing. The latter could be due to the variation in individual size and shape of the nasal passage. Smokers had, on average, a higher rate of deposition but also a higher inter-individual variability making it difficult to draw conclusions with respect to the effect of smoking on ETS particle deposition. The average deposition of the three smokers was 65.3 plus or minus 24.1% for nasal breathing and 66.1 plus or minus 17.6% for mouth breathing.

## 1998

Walton, W. Henry; Vincent, James H., 1998, "Aerosol Instrumentation in Occupational Hygiene: An Historical Perspective," *Aerosol Science and Technology*, **28**(5)417-438

Occupational hygiene has always been very influential in aerosol science - and vice versa. This paper gives an historical overview of this interaction, in particular how aerosol measurement instrumentation has evolved for the measurement of workers' exposures to aerosols in the occupational setting. It shows how health-related criteria for aerosol measurement have shifted from ones based on airborne particulate mass to ones based on particle count concentration, and then back again, depending on the aerosol science knowledge that was available at the time. It also draws the distinction between instrumentation based on time weighted-average sampling and direct-reading measurement, and the factors that govern

how the choice of type of measuring instrument was made in the past, the way it is made now, and the way it might be made in the future.

## 1997

Morawska, L.; Jamriska, M.; Bofinger, N. D., 1997, "Size characteristics and ageing of the environmental tobacco smoke," *Science of the Total Environment*, **196**(1):43-55

The work presented here is a study of the behaviour of the particulate phase of ETS under controlled laboratory conditions and in real indoor environments with the aim of providing information for assessment of human exposure to ETS. This paper reports investigations of the size distribution of ETS and changes to the distribution with time under a range of environmental conditions. Measurements were performed using two instruments, the Scanning Mobility Particle Sizer and the Aerodynamic Particle Sizer, which enabled the determination of the precise locations of ETS peaks at frequent short time intervals. While total particle concentrations or changes in concentrations are not specific markers of ETS, peaks related to ETS in the spectral distribution of atmospheric particles, for a properly designed experiment, are. The presence and locations of these peaks are characteristic of ETS in indoor environments and are clearly distinguishable from the background particle distribution. It is demonstrated that an initial ETS size distribution in an indoor environment about 10 min after generation by a human smoker has a major peak in the submicron range between 60 and 90 nm. The location of the peak does not depend on the relative humidity, but does depend on the way the cigarette is smoked. An increase in particle size in the range of 20 to 50%, takes place in the first 30 to 60 min after ETS generation and then remains unchanged for the duration of the experiment. A decrease in particle size (shrinkage) was not observed during these experiments.

## 1996

Spanne, M.; Olsson, B. O.; Gudmundsson, A.; Bohgard, M., 1996, "Characteristics of particle emissions at paint manufacturing," *Journal of Aerosol Science*, **27**(S1):S437-S438

The handling of pigments and powders in paint manufacturing cause emissions of airborne particles. A great variety of substances occur. Previous studies of simulations of pouring of pigments, which occur in small scale weighing procedures, showed typically bimodal size distribution including a sub-micron and a super-micron mode. The objective of the study was to get detailed knowledge on the relation between different source processes (depending on production technology, equipment and the manual handling routines) and aerosol characteristics. Super-micron particles were continuously measured with a time-of-flight instrument (Aerodynamic Particle Sizer (APS) spectrometer, TSI Inc.) and sub-micron particles with an electric mobility spectrometer (Scanning Mobility Particle Sizer (SMPS) spectrometer, TSI Inc.). In addition filter sampling techniques for standardized work-place sampling (so-called total dust filter cassettes, a cyclone sampler for respirable dust an IOM-samplers for inhalable dust) have been used for idealized exposure/are measurements using a rotating mannequin as a carrier of these samplers. The filters were analyzed gravimetrically and with Particle Induced X-ray Emission Analysis.

## 1994

Lin, Rong-Hwa; Wang, Chrong-Reen; Li, Chih-Shan, 1994, "Size Distributions and Lung Deposition of Submicrometer Particles from Metered Dose Inhalers," *Environment International*, **20**(2):161-167

The size distributions of the submicrometer particles generated from ten metered dose inhalers (MDIs) were determined by a high resolution particle sizer, which could measure the particles in the size range of 0.01  $\mu\text{m}$  to 1  $\mu\text{m}$ . The particle sizer contains a differential mobility analyzer (TSI 3071) and a condensation particle counter (TSI 3022). The median diameters ranged from 0.06–0.10  $\mu\text{m}$  with a geometric standard deviation of 2. In addition, the surface median diameters and volume median diameters were found to be 0.30  $\mu\text{m}$  and 0.45  $\mu\text{m}$ , respectively. Moreover, none of the size distributions of the generated submicrometer particles fits log-normal distributions. The deposition probabilities of the submicrometer particles in the airways were evaluated. It was observed that the average deposition percentages of the particles in the alveolar, tracheobronchial, and extrathoracic regions are 32%, 4%, and 4%, respectively. Understanding the deposition of submicrometer particles from MDIs should benefit the clinical practice in inhalation therapy.

## 1995

Wampler, David A.; Miller-Leiden, Shelly; Nazaroff, William W.; Gadgil, Ashok J.; Litvak, Andres; Mahanama, K.R.R.; Nematollahi, Matty, 1995, "Effectiveness of smokeless ashtrays," *Air and Waste Management Association*, **45**:494-500

Most environmental tobacco smoke (ETS) issues from the tips of smoldering cigarettes between puffs. Smokeless ashtrays are designed to reduce ETS exposure by removing particulate and/or gas-phase

contaminants from this plume. This paper describes an experimental investigation of the effectiveness of four smokeless ashtrays: two commercial devices and two prototypes constructed by the authors. In the basic experimental protocol, one or more cigarettes was permitted to smolder in a room. Particulate or gas-phase pollutant concentrations were measured in the room air over time. Device effectiveness was determined by comparing pollutant concentrations with the device in use to those obtained with no control device. A lung deposition model was applied to further interpret device effectiveness for particle removal. The commercial ashtrays were found to be substantially ineffective in removing ETS particles because of the use of low-quality filter media and/or the failure to draw the smoke through the filter. A prototype ashtray using HEPA filter material achieved better than 90% particle removal efficiency. Gas-phase pollutant removal was tested for only one prototype smokeless ashtray, which employed filters containing activated carbon and activated alumina. Removal efficiencies for the 18 gas-phase compounds measured (above the detection limit) were in the range of 70 to 95%.

### 1993

Li, Chih-Shan; Lin, Wen-Hai; Jenq, Fu-Tein, 1993, "Characterization of Outdoor Submicron Particles and Selected Combustion Sources of Indoor Particles," *Atmospheric Environment*, **27B**(4):413-424

Environmental tobacco smoke, mosquito-coil smoke, and joss stick smoke are the major indoor combustion sources in Asian countries. Field evaluations of the size distributions of outdoor submicron particles and selected combustion sources of indoor particles were conducted in an apartment in Taipei urban area. The size distributions of submicron aerosols were determined by a high resolution particle sizer, which could measure the particles in the size range of 0.017-0.886  $\mu\text{m}$ . The particle sizer contains a differential mobility analyser (TSI 3071) and a condensation particle counter (TSI 3022). The number concentrations of the indoor and outdoor submicron particles varied from 14,000 to 150,000  $\text{cm}^{-3}$  and from 10,000 to 45,000  $\text{cm}^{-3}$ , respectively. The changes of the size distributions and the number concentrations of submicron aerosols before, during, and after the aerosol generations were compared. The average number median diameters of environmental tobacco smoke, smoldering cigarettes, mosquito-coil smoke, joss stick smoke, the indoor typical conditions, and the outdoor typical conditions were 0.090, 0.085, 0.094, 0.084, 0.091 and 0.054  $\mu\text{m}$  respectively. Regarding the surface area-weighted size distributions, the average surface median diameters of these conditions were 0.229, 0.219, 0.282, 0.188, 0.224 and 0.221  $\mu\text{m}$ , respectively. In addition, the average volume median diameters were 0.338, 0.332, 0.398, 0.289, 0.330 and 0.340  $\mu\text{m}$ , respectively. These indoor combustion sources did generate a significant number of the ultrafine and submicron particle which have higher deposition probabilities in the respiratory tract. Further health evaluations of the submicron particles from these combustion sources are needed.

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