SCANNING MOBILITY PARTICLE SIZER™ (SMPS™) SPECTROMETER

APPLICATION: BASIC RESEARCH & NUCLEATION STUDIES

2006

Destaillats 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 0.04–200×10^5 molecules/cm^3 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 $(2.8 \pm 0.4)\ 10^6$. 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). 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^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.

2005

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^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), 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^2 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^3 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$_2$SO$_4$), 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). 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^{TM} (SMPS^{TM}) 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.

Hermsdorf, D.; Bonnamy, A.; Suhm, M. A.; Signorell, R., 2004, "Infrared spectra of phenanthrene particles generated by pulsed rapid expansion of CO$_2$ 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$_2$. 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 can not 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).

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-m3 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 (H2SO4), 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. 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^3 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 β-Pinene with O3," *Aerosol Science & Technology*, **37**(11):877–891

The formation of secondary organic aerosol (SOA) from reactions of O_3 with β-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[TM] (SMPS[TM]) 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). 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 (H_2SO_4), and led to higher aerosol yields, than when H_2SO_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 -Caryophyllene with Ozone," *Journal of Atmospheric Chemistry*, **45**(3):261–287

Gas and particulate reaction products from the ozonolysis of β-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^3). 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 β-caryophyllene (**I**) carbon. Measurements show that a number of reaction products with low vapor pressure (e.g., β-caryophyllone aldehyde (**IV**), β-norcaryophyllone aldehyde (**V**), β-caryophyllonic acid (**VIII**), β-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[TM] (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.; Zhau, Q. F., 2003, "Direct measurement of attachment of 220Rn 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 α-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}Rn and ^{220}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. 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 β-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 α-pinene and Δ^3-carene. However, when the reactant concentrations of α-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 α-and β-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., β-pinene) or intramolecularly (e.g., α-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 Cl_2 molecules in the presence of toluene and oxygen, at levels of $\sim 10^{14}$ radicals/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 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 $Cl + C_6H_5CH_3$ 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 NO and 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 $k_4 = (8 \pm 2) \times 10^{-12}$ cm^3 s^{-1} for the benzylperoxy self-reaction, and $k_6 = (2.7 \pm 0.5) \times 10^{-11}$ cm^3 s^{-1} for its reaction with 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 be79.47nm and the growth ratio was 77.94%, when the initial size of aerosol was 101.82nm. 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 142.7nm 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/NO_x or toluene/air/CH_3ONO/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, 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 m3 Teflon film bags under darkness in the presence of background seed aerosols. The aldehydes used were as follows: glyoxal, butanol, 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^{TM} (TSI-SMPS^{TM}), 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) 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 (NH4)2SO4-H2SO4-H2OIII. 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. α-Fe$_2$O$_3$, α-Al$_2$O$_3$, Al$_6$Si$_2$O$_{13}$, and amorphous SiO$_2$) coated by (NH$_4$)$_2$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. 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 NHSUB4NOSUB3 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). 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 (NH4)2SO4-H2SO4-H$_2$O aerosols internally mixed with metal oxide particles," *Journal of Aerosol Science*, **30**(S1):S827–S828

Aerosols composed of a water soluble (NH$_4$)$_2$SO$_4$-H$_2$SO$_4$-H$_2$O components and a water insoluble TiO$_2$, Al$_2$O$_3$, or 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_x, pinonaldehyde, (87±20)%; total nitrates (18±9)%; formaldehyde, (23±9)%; acetone (9±6)%; for the alpha -pinene experiments in the absence of NO_x: 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^{13} molecule cm^{-3} 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_x 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_4NH_3, and NH_4SO_4. 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) 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.

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 H2SO4 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 H_2SO_4 aerosol with NH3 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 μm 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 as 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 0.1μm 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.

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 filed 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 m3 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.