AUTHOR: Allen L. Robinson, Spyros N. Pandis, Eric Lipsky, Charles Stanier

INSTITUTION: Carnegie Mellon University
Dept. of Mechanical Engineering, Scaife Hall
Pittsburgh, PA 15213
(412) 268 – 3657; (412) 268 – 3348 (fax)
e-mail: alr@andrew.cmu.edu

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Abstract

A dilution sampler has been designed and manufactured to simulate the effects of dilution processes on particulate matter emissions from coal-fired power plants and other combustion systems. The sampler allows independent control of the dilution ratio and residence time. Experiments were performed to examine the effects of the sampling conditions on the particulate emissions of a pilot-scale pulverized coal combustor burning a low sulfur bituminous coal. Measurements included the particle size distribution in the range from 0.003 to 20 μm and the $PM_{2.5}$ mass emission rate. The residence time and dilution ratio do not influence the particle mass emission rate, but have a significant effect on the size distribution and total number emission rate. Increasing the residence time dramatically decreases the total particle number concentration, and shifts the particle mass to larger sizes. Increasing the dilution ratio increases the concentration of ultrafine particles. The effects of residence time can be explained quantitatively by the coagulation of the emitted particles; however the effects of dilution ratio are more complex because dilution ratio influences both the coagulation rate and gas-to-particle conversion.

Table of Contents

Executive Summary

During this year, significant effort went into improving the design of the dilution sampler. The current design allows independent control of the dilution ratio and residence time. Experiments were performed to examine the effects of the sampling conditions on the particulate emissions of a pilot-scale pulverized coal combustor, the Combustion and Environmental Research Facility (CERF) at the National Energy Technology Laboratory. During this year the measurements focused on the effects of sampling on the particle size distribution, particularly on ultrafine particles. Measurements included the particle size distribution in the range from 0.003 to 20 μm and the $PM_{2.5}$ mass emission rate. The CERF was operated at steady conditions firing an Eastern Bituminous coal during these experiments. The residence time and dilution ratio do not influence the particle mass emission rate, but have a significant effect on the size distribution and total number emission rate. Increasing the residence time dramatically decreases the total particle number concentration, and shifts the particle mass to larger sizes. Increasing the dilution ratio increases the concentration of ultrafine particles. The effects of residence time can be explained quantitatively by the coagulation of the emitted particles; however the effects of dilution ratio are more complex because dilution ratio influences both the coagulation rate and gas-to-particle conversion.

Introduction

Ambient particulate matter is a complex mixture of multi-component particles whose size distribution, composition, and morphology can vary significantly in space and time[1]. Atmospheric aerosol size ranges from a few nanometers to tens of micrometers. Major components include sulfates, nitrate, ammonium, and hydrogen ions; trace elements (including toxic and transition metals); organic material; elemental carbon (or soot); crustal components and water. PM is emitted directly from sources (primary PM) and is also formed in the atmosphere from gaseous precursors (secondary PM). In 1997, the U.S. Environmental Protection Agency (EPA) revised the National Ambient Air Quality Standards (NAAQS) to address ambient air concentrations of particulate matter (PM) with an aerodynamic diameter of 2.5 micrometers or less ($PM_{2.5}$). Emissions from coal-fired utility boilers contribute to the ambient concentrations of $PM_{2.5}$. Utility boilers are a source for both primary fine particles and gaseous precursors that react in the atmosphere to create secondary aerosol.

Fine particles play an important role in climate forcing because of their ability to scatter and absorb light and also because they act as cloud condensation nuclei[1,2]. The total radiative effect of anthropogenic aerosols is negative, so these particles tend to cool the earth's surface, but the exact magnitude of this aerosol effect remains uncertain. To reduce this uncertainty, it is necessary to improve our understanding of the physical and chemical properties of atmospheric particles including their size distribution, their number concentration, and their composition.

Measurements of particulate emissions from coal-fired power plants have been reported in a number of studies[2-10]. Most of the available data are for the emissions of total particle mass. For the studies reporting the particle size distributions, the measurements were often limited by the available instrumentation to particles larger than 20-30 nm.

The emissions after the boiler have a bimodal distribution; the majority of the mass occurs as larger particles with a mean mode around 20 microns. These particles are formed from the fusion and coalescence of mineral matter in coal, and are effectively removed by existing control technologies such as bag houses and electrostatic precipitators. Vaporization and condensation of inorganic constituents produce the finer particles in the submicron size range, which dominate the emissions on a number basis. The peak of the fine mode varies between 50 and 150 nm[5,7]. This variability is not well understood. Kauppinen and Pakkanen[7] hypothesize that this variation may be due to dynamical processes changing the particle size distributions for measurements made on diluted flue gases. Zhuang and Biswas[10] show that formation of the sub-micrometer peak can be suppressed by sorbent injection.

Sampling PM emissions from coal-boilers is challenging because of the elevated temperatures and high moisture content of the exhaust gases in combination with the inherent complexity of PM. Upon exiting the stack, aerosol processes such as coagulation, condensation, and nucleation often change the size and composition of the

PM emissions as the combustion products are rapidly cooled and diluted with ambient air; for example, semi-volatile compounds are still in the gas phase at typical stack temperatures, but then condense or nucleate to form fine particulate matter when cooled. These transformations are not captured by traditional stack sampling methodologies such as EPA Method 5[11], which pass hot combustion products directly through a heated filter assembly. Dilution sampling is a technique that has been developed to examine the influence of the rapid cooling and dilution on PM emissions from combustion systems[12,13]. A dilution sampler rapidly mixes hot exhaust gases with a specified amount of conditioned air, and better simulates processes such as nucleation, condensation, and coagulation that occur in the first few seconds of the emitted aerosol lifetime. Such local processes cannot be captured by atmospheric chemical transport models and need to be estimated by an emission-processing module. The dilution sampler results can be used for the development of such a module.

Despite the progress in understanding the number and mass distribution of aerosol emitted by combustion sources such coal-fired boilers a number of issues have yet to be elucidated. These include the importance of the sampling conditions (temperature, dilution ratio, and residence time in the sampling device) on the size distribution and the mass concentration of the emitted particles; the distribution of particles smaller than 20 nm; the changes to these size distributions as the particles are mixed with ambient air.

This paper aims to improve our understanding of the influence of dilution processes on the size distribution of PM emitted from coal-fired boilers. A dilution sampler was designed and constructed to allow independent control of dilution ratio and residence time. This sample was installed upstream and downstream of the bag house of a pilot-scale pulverized coal combustor. Measurements of particle size distributions from 0.003 μm – 20 μm were made across a range of dilution ratios and residences times. Filter samples of PM$_{2.5}$ were also collected for gravimetric analysis. The data are compared to predictions from coagulation theory to investigate the influence of aerosol processes on the particle size distribution.

Dilution Sampler Design and Characterization

Figure 1 shows a schematic of the dilution sampling system developed for this investigation. The design is based on the Caltech sampler[12], with improvements that allow for the independent control of dilution ratio and residence time to allow investigation of the effects of sampling on the emissions. England et al.[13] outline the design criteria for dilution samplers and review previous designs.

The CMU sampler consists of three major components: the sample inlet line, the dilution tunnel, and the residence time tank. Combustion products are isokinetically drawn from the exhaust duct through the sample inlet line, and then turbulently mixed with conditioned ambient air in the dilution tunnel, a 15-cm-diameter, 2.3-m-long stainless steel tube. At the end of the dilution tunnel, a slipstream of this mixture is drawn continuously through the residence time tank. Ambient air used to dilute the combustion products is first passed through an activated carbon filter to remove organic

vapors, and a HEPA filter to remove particles in the air. All of the parts of the sampler in contact with the exhaust sample and the diluted mixture passes are made off stainless steel with Teflon gaskets to reduce contamination. Sampling ports for aerosol characterization instrumentation and filter packs are located at the end of the dilution tunnel and at various locations on the residence time tank. The performance specifications for the sampler are listed in Table 1.

Table 1: Performance characteristics of dilution sampler.

Parameter	Range
Dilution ratio	20-200
Sample flow	10-30 lpm
Dilution air flow	350-1,400 lpm
Residence time	0-12 min
Tunnel Re	3,000-12,000

The flow through the system is controlled with blowers and valves located before and after the dilution tunnel. A slight imbalance between the flows into and out of the tunnel draws the combustion products through the inlet sample line and into the dilution tunnel. The flow rate through the sample inlet line is measured directly using a venturi flow meter. The flow rate of the dilution air into the tunnel is monitored using an orifice plate. The dilution ratio is defined as the ratio of volumetric flow rate of the dilution air into the tunnel to the volumetric flow rate of combustion products through the inlet sample line – all flows are corrected to standard conditions, 20 °C and 1 atm.

A computer based data acquisition system automatically records flow rate, temperature, and relative humidity measurements. The temperature and relative humidity of the dilution air entering the tunnel, the temperatures of the combustion products entering and leaving the tunnel and the temperature, relative humidity, and pressure of mixture inside the tank are continuously monitored.
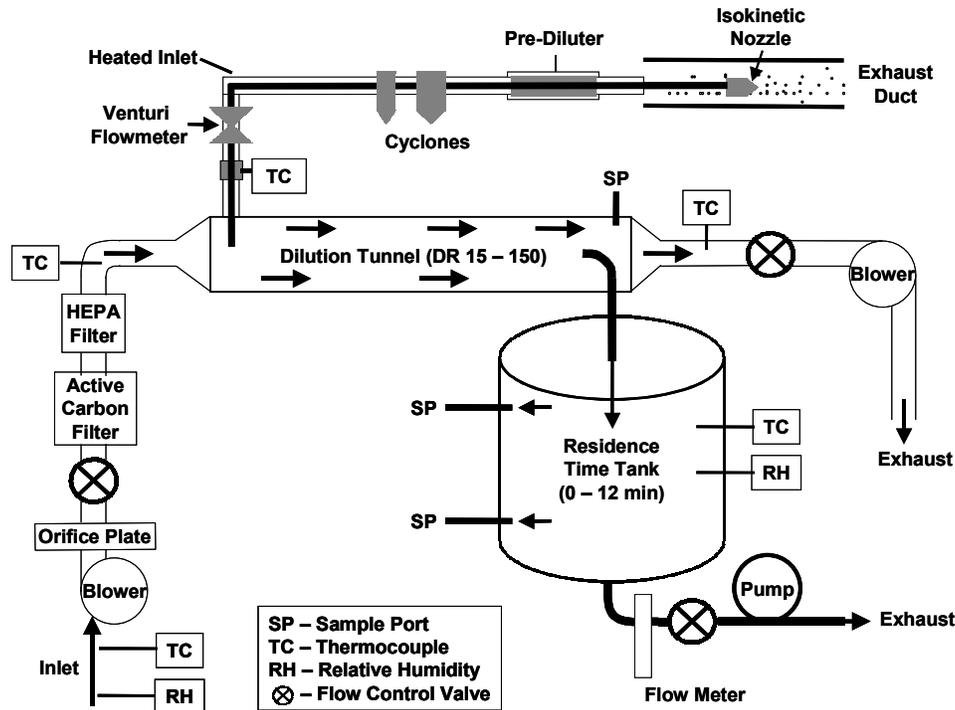
Figure 1. Schematic of dilution sampler.

Mixing experiments using carbon dioxide were performed to verify that the combustion products and ambient air are well mixed by the end of the tunnel across the full range of operational conditions. The mixing rate depends on the tunnel flow rate. At a tunnel Re of 3000 the sample is well mixed 2.2-m downstream of the inlet, ~ 9 s after entering the tunnel; at a tunnel Re of 10000, the sample is well mixed 0.75-m downstream of the inlet, ~ 0.33 s after entering the tunnel.

A cyclone is installed on the inlet line to remove particles larger than 2.5 microns from the system. Experiments performed by Hildemann et al.[12] revealed significant losses of large particles in the sample inlet line and venturi flow meter of the Caltech dilution sampler. The inlet line is heat traced to maintain the exhaust sample at the stack temperature, and minimize losses due to thermophoresis.   For high dilution ratio experiments, a first stage diluter is added to the inlet line to dilute the combustion products with conditioned ambient air heated to the exhaust temperature.

The residence time tank is a 76-cm diameter, 91-cm tall stainless steel cylindrical tank. The flow through the tank is controlled independently using a separate pump and valve system to allow for the diluted sample to remain in the tank for a specified time ranging from 0.5 to 12 minutes. Two different residence tank configurations were used in this study. The first configuration consists of an inlet through the top of the tank, and a set of sample ports located at the bottom of the tank. The flow path was lengthened in the

second configuration by installing a 38-cm diameter stainless steel tube inside the tank. The diluted sample enters through the center of the top of the tank, passes downward through the inner cylinder, and then flows upwards outside the annulus between the inner cylinder and the tank wall. In the second configuration sample ports are installed at both the top and bottom of the tank to allow for samples to be taken simultaneously at one-half and the full residence time.

Tracer experiments were performed using carbon dioxide to characterize the sample transit time through the residence time tank. For a given flow pattern, the median residence time is defined as the time it takes for 50% of the sample to travel between the tank inlet and the sampling port. The results, such as those shown in Figure 2, reveal a distribution of transit times through the tank. The performance of the first configuration is relatively poor – a fast path causes a portion of the sample rapidly passing through the tank. A set of experiments on the pilot-scale coal combustor was performed with this first configuration. The second configuration has improved performance, much closer to that of theoretical predictions assuming parabolic laminar flow through a circular tube[14].
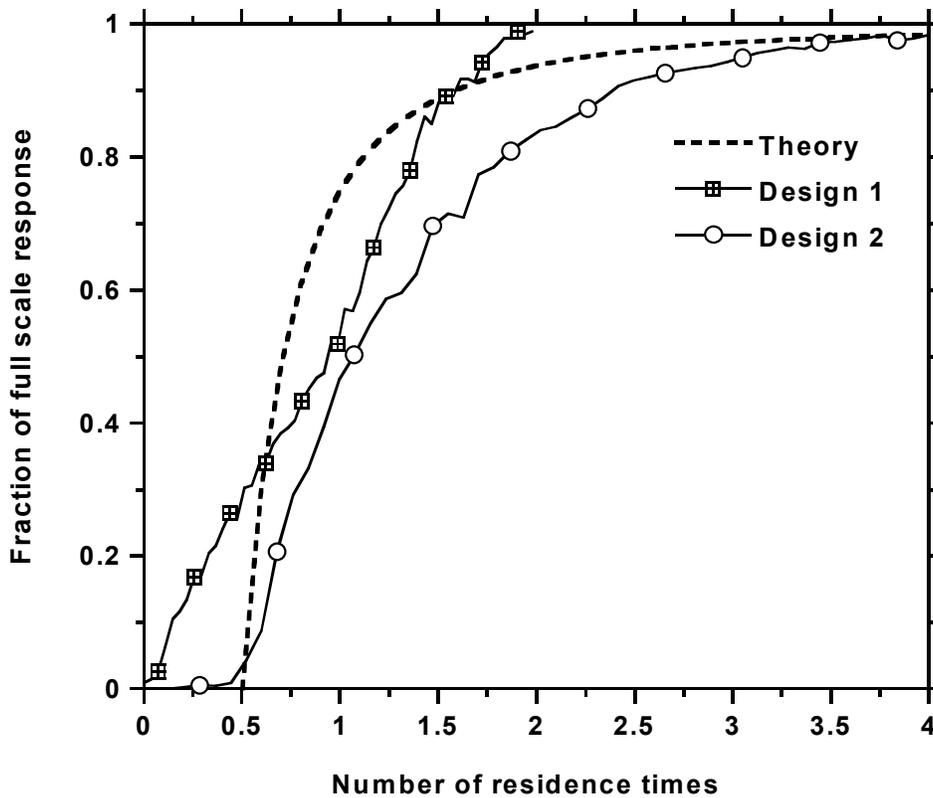

Figure 2. Measurements of transit time distribution of flow through the residence time tank. The theoretical calculations assume fully developed laminar flow through a circular tube.

Experimental Set-up and Procedure

Measurements were performed on the exhaust of the Combustion and Environmental Research Facility (CERF) at the Department of Energy National Energy Technology Laboratory. The CERF is a pilot-scale pulverized coal combustor that simulates the time-temperature history of a commercial coal boiler. At full load it consumes 50 lbs of pulverized coal per hour, roughly 150 kW when operating on a bituminous coal. Coal is injected through a swirl-stabilized burner at the top of a 3-m tall and 45-cm diameter refractory lined combustion zone. Combustion products then flow into a horizontal convective section, through two flue gas coolers, heat-traced piping, and into a bag-house. The CERF was operated on Prater Creek coal, an eastern bituminous coal with low ash and low sulfur content. Results from standard analysis of the coal are shown in Table 2.

Table 2. Composition of Prater Creek Coal.

Proximate Analysis (wt %, as rcvd)		Ash (wt %, ash basis)	
Moisture	2.0	SiO_2	38.4
Volitale Matter	38.7	N_2O_3	25
Fixed Carbon	54.5	Fe_2O_3	23.5
Ash	4.7	TiO_2	1.0
HHV (BTU/lb)	14,167	P_2O_5	0.1
		CaO	3.8
Ultimate Analysis (wt %, dry basis)		MgO	2.1
Hydrogen	5.4	Na_2O	0.3
Carbon	78.3	K_2O	2.2
Nitrogen	2.3	SO_3	4.6
Sulfur	0.8		
Oxygen	8.4		
Ash	4.7		

Experiments were conducted to examine the effects of dilution ratio and residence time on the PM size distribution and PM mass emission rate while the CERF operated at constant conditions. Combustion products were isokinetically sampled from the exhaust duct before and after the bag-house. Two types of experiments were performed: (1) the residence time was held constant while the dilution ratio was varied, and (2) the dilution ratio was held constant while varying the residence time. One week of particle size distribution measurements were made before the bag house using the first residence tank configuration. This configuration requires resetting the flows to change the residence time in the system. Two weeks of testing were conducted with the second tank configuration, one week of particle size measurements after the bag house, and one week of filter samples before the bag house. This second configuration allows measurement of multiple residence times without adjusting the flows, which reduces the effects of operational variability on the data.

Measurements of particle size distribution from 0.003 μm – 20 μm are made using sizing instrumentation connected to the sample ports on the dilution tunnel and residence time tank. One Scanning Mobility Particle Sizer (SMPS) measures particles between 0.003 μm and 0.075 μm (TSI model 3085 Electrostatic Classifier with a TSI model 3025 CPC). A second SMPS measures particles from 0.015 μm - 0.65 μm (TSI model 3081 Electrostatic Classifier with a TSI model 3010 CPC). An Aerodynamic Particle Sizer (APS) measures particles in the range of 0.5 μm – 20 μm, overlapping with the second SMPS (TSI model 3320). Two neutralizers are used in series before both of the SMPS analyzers to ensure a known charge on the particles. Further dilution of the sample flow was required under certain conditions to lower concentrations of the particles below maximum detection levels of the CPC.

Filter samples were collected to examine the effect of dilution ratio and residence time on PM mass emission rates. The samples are collected on 47-mm, 2-μm Teflo filters (Gelman Corp). The filters are equilibrated for at least 24 hours and weighed in a temperature (21-23 °C) and relative humidity (30-40%) controlled glove box following standard EPA protocols.

All of the results are reported on a unit exhaust volume basis of the pilot-scale combustor at standard temperature and pressure. This allows direct evaluation of the effects of dilution and residence time on actual emissions, and facilitates comparison of data collected at different dilution ratios.

Results

Figure 3 presents size distributions measured before and after the bag house at a residence time of 1.5 min and a dilution ratio 70. The emissions on a mass basis are dominated by particles greater than 1 μm; the emissions on a number basis are dominated by particles smaller than 100 nm. Comparing the distributions measured before and after the bag house reveals that the bag house significantly reduces the emissions of both number and mass of particles less than 2.5 μm.

Three modes can be seen in the particle size distribution measured before the bag house; only two modes are present in the distribution measured after the bag house. A coarse mode that peaks around 2 μm is present before and after the bag house. These are residual fly ash particles formed from the fusion and coalescence of mineral matter in the coal. Note that the experiments before the bag house only measure the lower end of the residual fly ash mode because the cyclone on the inlet line removes particles greater than 2.5 μm – in an uncontrolled boiler the residual fly ash mode typically peaks around 20 μm. A fine mode around 0.05 to 0.1 μm can also be seen before and after the bag house, which is formed from the condensation and nucleation of metals volatilized during the combustion process[15]. The number distributions measured before the bag house also contain an ultrafine mode at around 0.01 μm. This mode is probably a sulfuric acid fume, which formed from the reaction of SO_3 and H_2O and the condensation and nucleation of H_2SO_4 as the combustion products rapidly cool in the dilution tunnel[16]. This ultrafine mode is not observed after the bag house – only one brief nucleation event was recorded

during the week of sampling after the bag house. We suspect that the absence of the ultrafine mode after the bag house is due to the removal of the SO_3 in the exhaust as the flue gas passes through the bag; in addition, the removal efficiency to the bag should be high for these ultrafine particles.
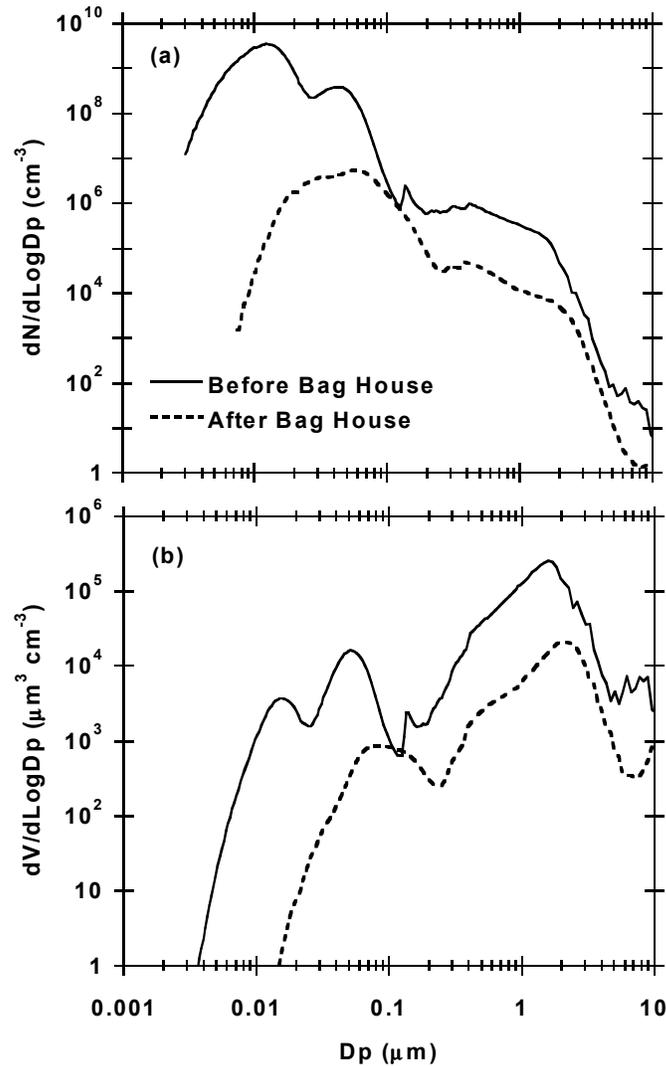

Figure 3. Typical particle (a) number and (b) volume distributions measured before and after the bag house at a dilution ratio of 70 and a residence time of 1.5 min. Concentrations are reported on a unit exhaust volume basis of the pilot-scale combustor corrected to standard temperature and pressure.

Figure 4 shows the evolution of the size distribution as a function of residence time. These distributions were measured before the bag house at a dilution ratio of 70, and residence times of 0, 1.5, and 12 minutes. The initial (zero time) residence time distribution is measured at the end of the tunnel before the diluted sample enters the residence time tank. The data indicate that changing the residence time alters the size distribution of the submicron particles, but has no effect on the distribution of particles greater than 1 μm. Increasing the residence time reduces the particles in the ultrafine 0.01 μm mode; this reduction is especially dramatic during the first minute. Increasing the residence also shifts 0.05 μm mode towards larger particle sizes.

Changing the dilution ratio also alters the size distribution of the submicron particles. Figure 5 presents size distributions measured before the bag house at residence time and dilution ratios of 15, 70, and 150. Increasing the dilution ratio increases the particle number, especially in the ultrafine 0.01-μm mode. Changing the dilution ratio does not effect the size distribution of particles greater than 1 μm.

Figure 6 presents $PM_{2.5}$ mass emission rate data determined from gravimetric analysis of the filter samples collected before the bag house. For each experiment filter samples were collected simultaneously at three different residence times; the data are then normalized by the average mass emission rate for a given experiment to facilitate comparison between experiments. The average $PM_{2.5}$ mass emission rate before bag house was 1.9 gm/kg coal.

The dilution ratio and residence time do not influence $PM_{2.5}$ mass emission rate, indicating that there is not significant transfer of mass via nucleation or condensation occurring within the residence time tank. This is not unexpected for this system because of the very low organic emissions from coal-based power plants[17-20]. Plume data does suggest gas-to-particle conversion of semi-volatile trace metals such as selenium and arsenic[21,22]; however, these species are not presented in sufficient quantities to alter the $PM_{2.5}$ mass emission rate.
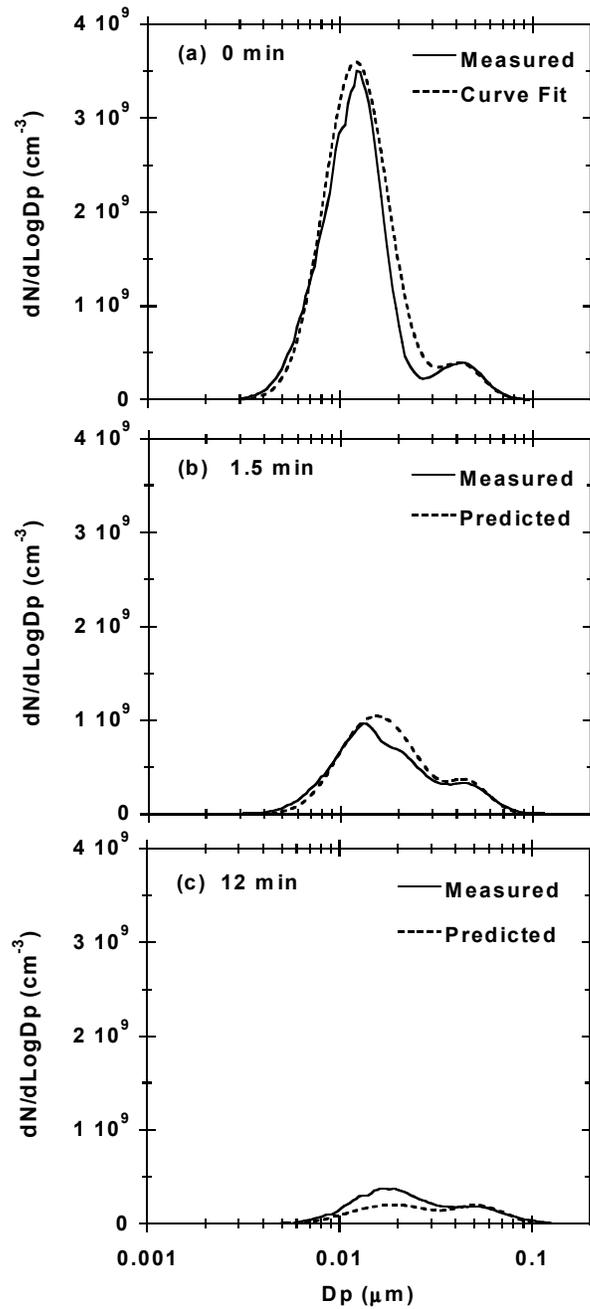
Figure 4. Change in particle size distribution with residence time at a dilution ratio of 70. Predictions based on coagulation theory using a curve fit of the zero residence time measurements as the initial condition and account for the distribution of transit times through the residence time tank. Concentrations are reported on a unit exhaust volume basis of the pilot-scale combustor corrected to standard temperature and pressure. All the distributions are plotted on the same scale to underscore the impact of residence time on the size distribution.
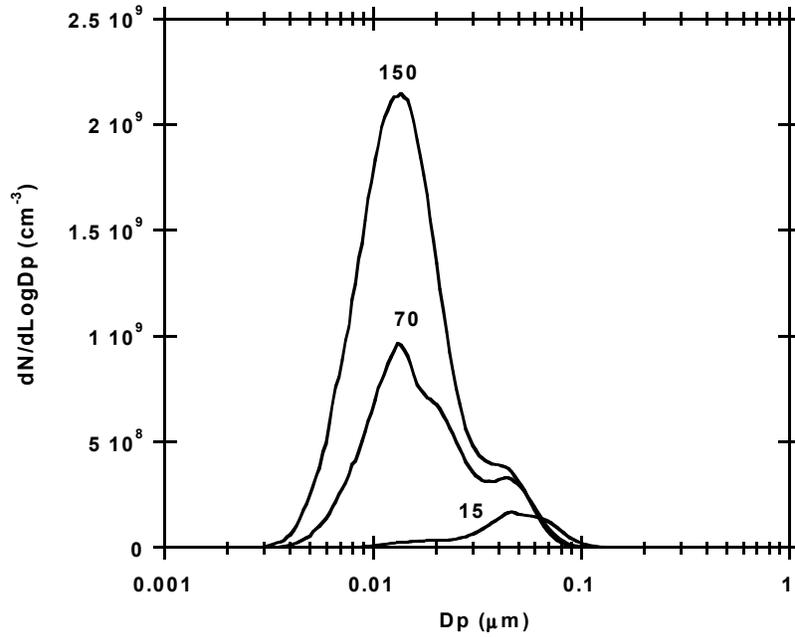
Figure 5. Particle number distributions measured at dilution ratios of 150, 70, and 15 at a residence time of 1.5 min. Concentrations are reported on a unit exhaust volume basis of the pilot-scale combustor corrected to standard temperature and pressure.
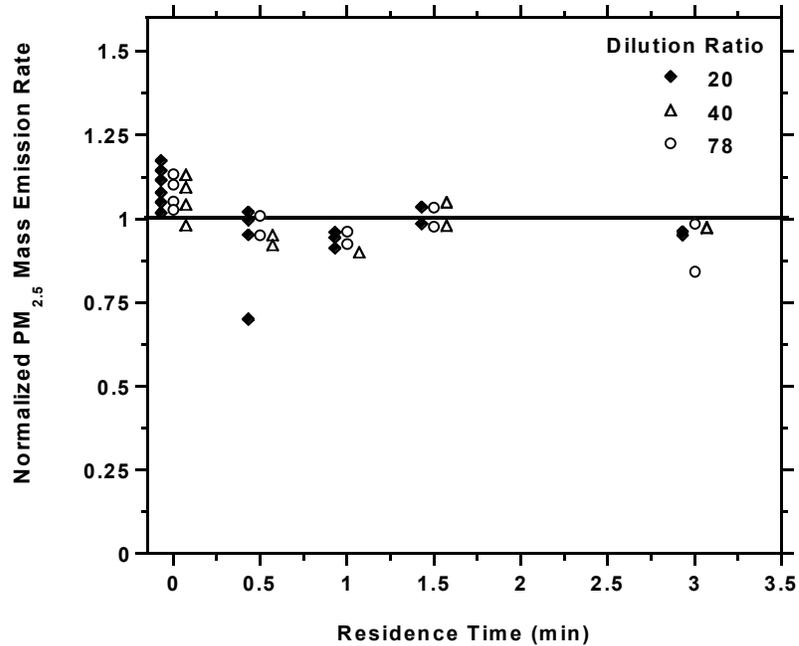

Figure 6. Normalized PM-2.5 mass emissions as a function of residence time. Measurements were made at four different residence times 0, 0.5, 1, 1.5, and 3 min. The residence times of the symbols indicating the dilution ratio 20 and 40 data have been shifted slightly to improve the visual clarity of the figure.

Both dilution ratio and residence time have a significant impact on total number emissions. Figure 7 shows measurements of number concentration as a function of residence time and dilution ratio. Increasing the residence time reduces the number concentration; for example, increasing the residence time from 0 to 12 minutes reduces the particle number by a factor 7 for experiments conducted with a dilution ratio of 70. Increasing the dilution ratio increases the total number concentration; for example increasing the dilution ratio from 15 to 100 increases the number concentration by a factor of 15 for experiments conducted with a residence time of 1.5 min.
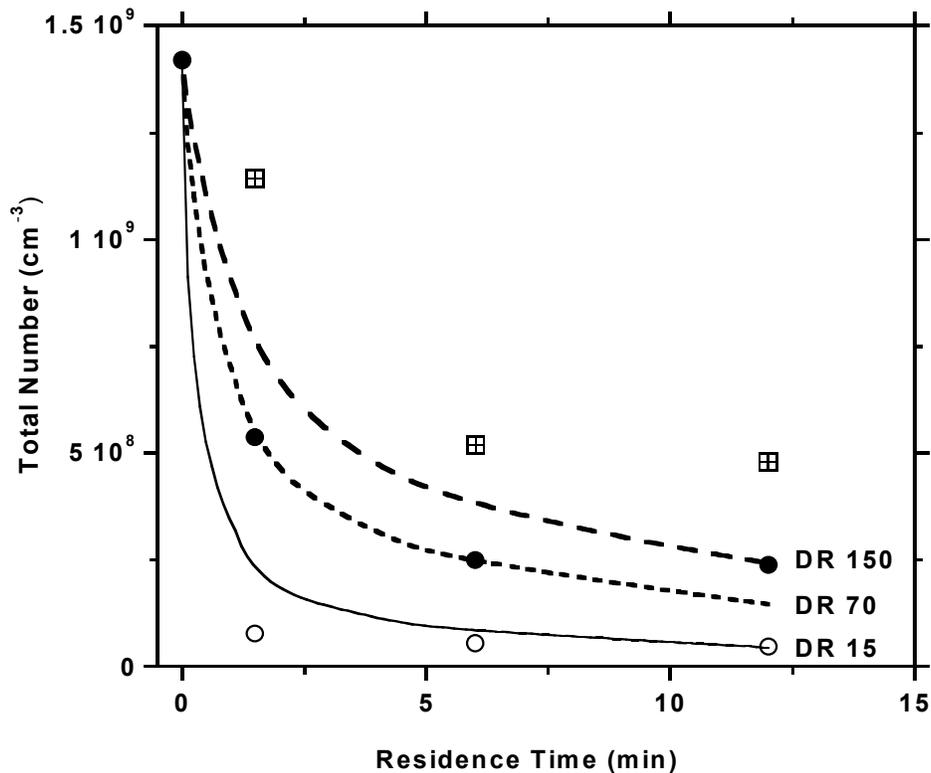

Figure 7. Comparison of measured and predicted total number concentration as a function of residence time. The squares, solid circles, and open circles indicate measured values at dilution ratios of 150, 70, and 15, respectively. The lines represent model calculations at dilution ratios of 15, 70, and 150, as indicated by the text on the figure. Concentrations are reported on a unit exhaust volume basis of the pilot-scale combustor corrected to standard temperature and pressure.

Analysis and Discussion

To better understand the effects of dilution and residence time on the measured size distributions, calculations were performed to examine the changes in the particle size distribution over time due to the coagulation. The model assumes the collision of

particles is due to Brownian motion[1]. The input to the simulations was the three-mode lognormal size distribution fit to the measurements made at the end of the dilution tunnel (0 residence time). Two types of simulations were performed – the first assuming ideal plug flow through the residence time tank, the second accounting for the distribution of sample transit times through the tank. To account for the varying amounts of time spent in the residence time tank by different air parcels, the results for 10 separate plug flow coagulation simulations (each at a different residence time) were combined in a weighted average corresponding to the measured residence time distribution.

For a given dilution ratio, the model reproduces the changes in the particle size distribution with increasing in residence time (Figure 4). Figure 7 compares the model predictions to the measured total number concentration as a function of residence time at a dilution ratio of 70. The model-measurement agreement suggests that the main process occurring within the residence time tank for these conditions is Brownian coagulation of the emitted particles.

Model predictions are shown in Figures 7 and 8 to examine whether the observed effects of dilution ratio on the total number concentration and size distribution can be explained by changes in the coagulation rate of the particles. Dilution dramatically alters the coagulation rate, which varies with the square of the particle number concentration. If coagulation is the dominant process in determining the effects of dilution then we should be able to scale a size distribution measured at 0 residence time to any dilution ratio and then use the coagulation model to predict the time evolution of the size distribution at the new dilution ratio. If true, this would significantly simplify sampling requirements because measurements at one dilution ratio could be used to predict emissions at another dilution ratio. To test this hypothesis we scale the particle concentrations of the measured size distribution at 0 residence time and a dilution ratio of 70 to a dilution ratio of 15 and 150, and use the model to predict the evolution of the total number concentration (Figure 7) and the size distributions (Figure 8) at the new dilution ratios.

Comparing the measurements and model predictions in Figures 7 and 8 reveals that coagulation alone is not sufficient to explain the effects of dilution ratio on the size distribution. For the dilution-ratio-150 case the model underpredicts the number of ultrafine particles and the total number concentration; for the dilution-ratio-15 case the model overpredicts the number of ultrafine particles and the total number concentration. For the dilution-ratio-15 case the predictions of total number concentration become more accurate at longer residence time.
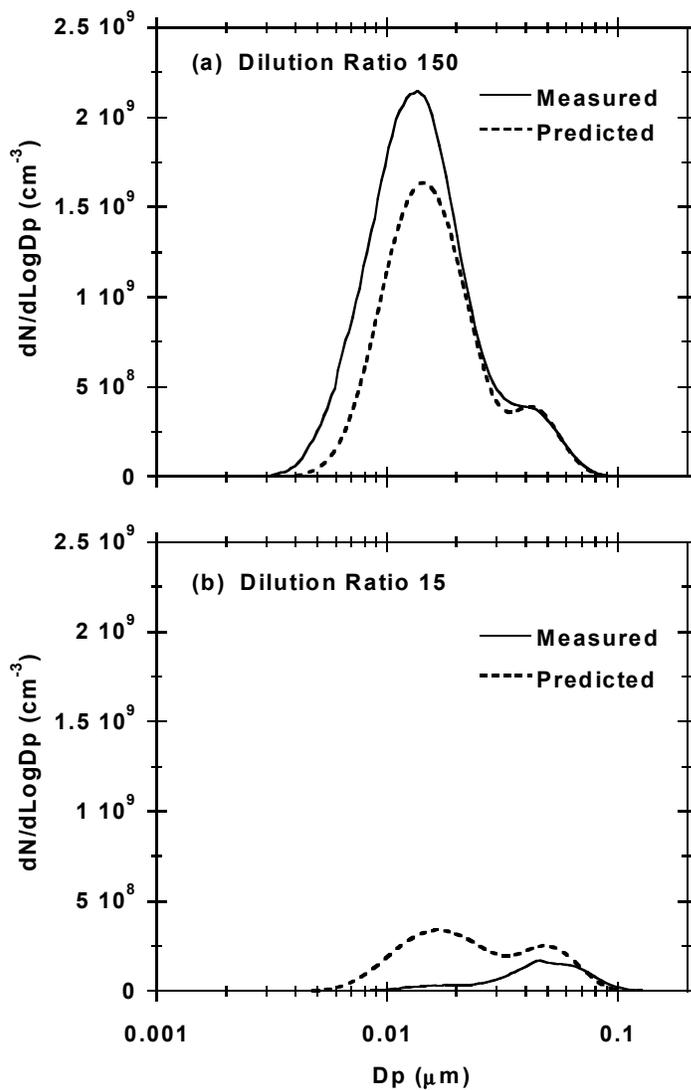
Figure 8. Comparison of measured and predicted size distributions at a residence time of 1.5 min. Concentrations are reported on a unit exhaust volume basis of the pilot-scale combustor corrected to standard temperature and pressure. All the distributions are plotted on the same scale to underscore the impact of dilution ratio on the size distribution.

While the simulation does explain qualitatively the observed trend of increasing concentrations with increasing dilution, the predicted distributions differ from the measured ones. This indicates that while coagulation is important another process affects the size distribution of the emitted ultrafine particles. Comparisons of the measured distributions at zero residence time indicate that this additional process is not taking place in the residence time tank but rather in the dilution tunnel. The rapid cooling of the combustion products creates a supersaturation of sulfuric acid. Whether or not this material condenses or nucleates depends on the available surface area. The lower the dilution ratio the greater amount of surface area per unit volume and the more likely that the sulfuric acid will condense onto existing particles reducing the production of new particles in the ultrafine mode. High dilution ratios reduce the amount of available surface area, which increases nucleation and the number of particles in the ultrafine mode. Therefore, one must account for both coagulation and particle formation by homogeneous nucleation when accounting the effects of dilution ratio on the particle size distribution. Similar interactions were observed in studies of diesel exhaust particles[23]. The details of the homogeneous nucleation processes in these systems are not well understood.

The simulations indicate that for coal fired boiler exhaust the effect of exhaust aging after the initial dilution can accurately be predicted accounting for coagulation only. This is useful because it reduces the number of samples required to establish the number distribution as a function of exhaust age. Furthermore, it shows that significant (>10%/minute) changes in number concentrations can be expected when ultrafine particles are present in concentrations greater than approximately 2 million particles/cc (actual diluted concentration). However, the simulations also indicate that the number emissions of particles depend nonlinearly on the dilution ratio, and that the competition between nucleation and condensation must be considered for the first few seconds of interaction of the plume with the atmosphere. Therefore, results at one dilution ratio cannot be used to predict the number concentration at other dilution ratios without accounting for changes in gas-to-particle conversion.

Conclusions

Particle size distributions from 0.003 to 20 μm were measured using a dilution sampler before and after the bag house of pilot-scale pulverized coal combustor. Experiments were performed to examine the influence of dilution ratio and residence time on the size distribution and the total number and mass emissions.

The residence time and dilution ratio do not influence the particle mass emission rate but change the size distribution and total number emissions. Increasing the residence time dramatically decreases the total particle number concentration, and shifts the particle mass to larger sizes. Increasing the dilution ratio increases the concentration of ultrafine particles. The effects of residence time can be explained with coagulation theory; however the effects of dilution ratio are more complex because dilution ratio influences both the coagulation rate and gas-to-particle conversion. The sensitivity of the particle size distribution and total number emissions to changes in dilution ratio and residence

time underscore the challenges associated with measuring and simulating the particle size distribution in the plume of a coal-fired power plant.

Acknowledgments

Publications and Presentations

E. Lipsky, S.N. Pandis, A.L. Robinson, and M. Freeman, "Effect of sampling conditions on primary particulate matter emissions from a pilot-scale coal combustor," AAAR (American Association for Aerosol Research) 2000, St. Louis, MO, November 6-10, 2000.

References

(1)   Seinfeld, J. H.; Pandis, S. N. *Atmospheric chemistry and physics: From air pollution to climate change*; John Wiley & Sons Inc.: New York, 1998.

(2)   Wehner, B.; Bond, T. C.; Mirmili, W.; Heintzenberg, J.; Wiedensohler, A.; Charlson, R. J. *Environmental Science & Technology* **1999**, *33*, 38881-3886.

(3)   Cantrell, B. K.; Whitby, K. T. *Atmospheric Environment* **1978**, *12*, 323-333.

(4)   McElroy, M. W.; Carr, R. C.; Ensor, D. S.; Markowski, G. R. *Science* **1982**, *215*, 13-19.

(5)   Markowski, G. G.; Ensor, D. S.; Hooper, R. G.; Carr, R. C. *Environmental Science and Technology* **1980**, *14*, 1400-1402.

(6)   Linak, W. P.; Peterson, T. W. *Aerosol Science and Technology* **1984**, *3*, 77-96.

(7)   Kauppinen, E. I.; Pakkanen, T. A. *Environmental Science & Technology* **1990**, *24*, 1811-1818.

(8)   Lind, T.; Kauppinen, E. I.; Maenhaut, W.; Shah, A.; Huggins, F. *Aerosol Science and Technology* **1996**, *24*, 135-150.

(9)   Mohr, M.; Ylatalo, S.; Klippel, N.; Kauppinen, E. I.; Riccius, O.; Burtscher, H. *Aerosol Science and Technology* **1996**, *24*, 191-204.

(10)   Zhuang, Y.; Biswas, P. *Energy & Fuels* **2001**, *15*, 510-516.

(11)   EPA, U. S. In *Federal Register 42(160):41776*, 1977, pp 24888-24890.

(12)   Hildemann, L.; Cass, G. R.; Markowski, G. R. *Aerosol Science and Technology* **1989**, *10*, 193-204.

(13)   England, G.; Toby, B.; Zilinska, B. "Critical review of source sampling and analysis methodologies for characterizing organic aerosol and final particulate source

emission profiles," Rep. No. `API 344,` American Petroleum Institute, `Washington D.C., 1998.`

(14)   Folger, H. S. *Elements of Chemical Reaction Engineering*; 2nd ed.; Prentice Hall: Englewood Cliffs, NJ, 1992.

(15)   Linak, W. P.; Wendt, J. O. L. *Fuel Processing Technology* **1994**, *39*, 173-198.

(16)   Flagan, R. C.; Seinfeld, J. H. *Fundamentals of Air Pollution Engineering*; Prentice-Hall: Englewood Cliffs, NJ, 1988.

(17)   Weber, G. F.; Erickson, T. A.; Hassett, D. J.; Hawthorne, S. B.; Katrinak, K. A.; Lousi, P. K. K. "A comprehensive assessment of toxic emissions from coal-fired power plants: Phase I results from the U.S. Department of Energy study," Energy & Environmental Research Center, `Grand Forks, ND, 1996.`

(18)   Garcia, J. P.; Beyne-Masclet, S.; Mouvier, G.; Masclet, P. *Atmospheric Environment* **1992**, *26A*, 1589-1597.

(19)   Miller, C. A.; Srivastava, R. K.; Ryan, J. V. *Environmental Science and Technology* **1994**, *28*, 1150-1158.

(20)   EPRI "Electric utility trace substances synthesis report," Rep No. TR-104614; Electric Power Research Institute, Palo Alto, CA, 1994.

(21)   Ondov, J.; Choquette, C.; Zoller, W.; Gordon, G.; Biermann, A.; Heft, R. *Atmospheric Environment* **1989**, *23*, 2193-2204.

(22)   Eatough, D.; Eatough, M.; Lewis, L.; Lewis, E.; Tomlinson, E.; Gordon, J.; Eatough, N. *Atmospheric Environment* **1996**, *30*, 283-294.

(23)   Abdul-Khalek, I.; Kittelson, D.; Brear, F. *SAE Tech. Pap. Ser.* **1999**, *No. 1999-01-1142*, 563-571.