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Aerosol size distribution modeling with the Community Multiscale Air Quality modeling system in the Pacific Northwest: 3. Size distribution of particles emitted into a mesoscale model

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[1] In order to improve the Community Multiscale Air Quality modeling system (CMAQ) performance for ultrafine particle concentrations in the Pacific Northwest, CMAQ v4.4 was modified so that particles are input to the model with an appropriate size distribution. CMAQ's default emission size distributions are based on outdated measurements which underrepresent ultrafine particles. At the same time, the size distribution must represent the results of all processes between the point of emission and the smallest resolvable spatial scale of the model. As a computationally efficient and simple solution, size distributions were compiled from published modern observations for traffic-dominated urban areas, power plants, and marine sources at the typical mesoscale air quality model spatial resolution of 4–15 km. CMAQ was modified so that all chemical species are input according to the emission size distribution of traffic-dominated urban areas because this source emits the majority of each aerosol species in the Pacific Northwest. For a summer 2001 case study based around field measurements, CMAQ v4.4 underpredicts the observable aerosol number concentrations by 1–2 orders of magnitude, while CMAQ with new emission size distributions underpredicts by ~1 order of magnitude. The modeled size distributions have improved properties in terms of more distinct Aitken and accumulation modes and a more prominent Aitken mode. Errors remain, especially in the accumulation mode.

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1. Introduction

[2] Regional-scale modeling studies with the Community Multiscale Air Quality modeling system (CMAQ) [*Byun and Schere*, 2006] are unable to reproduce sufficient aerosol particle concentrations, especially for ultrafine particles with diameters less than 100 nm [*Elleman and Covert*, 2009a; *Park et al.*, 2006; *Fan et al.*, 2006; Y. Zhang et al., Predicting aerosol number and size distribution with CMAQ: Homogeneous nucleation algorithms and process analysis, paper presented at 4th Annual CMAS Models-3 Users' Conference, Community Modeling and Analysis System Center, Chapel Hill, N.C., 26–28 September 2005]. Insufficient particle nucleation is one possible cause, but incorporation of ternary NH_3-H_2SO_4-H_2O nucleation and a nucleation parameterization does not consistently resolve these errors [*Elleman and Covert*, 2009b]. Another possible cause is the size distribution of emitted particles because the number underpredictions were observed to vary little with air mass aging, time of day, meteorology, and particle chemistry. CMAQ uses emission size distributions based on outdated measurements and uses an inappropriate spatial scale that underrepresent ultrafine particles. This work examines the emission size distributions in CMAQ, proposes new ones for CMAQ consistent with an efficient mesoscale air quality model, and tests them for a case in the Puget Sound region of the Pacific Northwest for which detailed airborne and surface aerosol measurements are available.

2. Justification for Mesoscale Emission Size Distributions

[3] CMAQ's particle emission size distributions come from a review by *Whitby* [1978] which compiled measurements from field campaigns in the 1960s and 1970s. Two representative emission size distributions, one for organic and elemental carbon and another for all other species, were hard-coded into CMAQ in a manner consistent with the model's treatment of aerosols as three overlapping modes for the Aitken (20–90 nm), accumulation (90 nm to 1 μm), and coarse modes (1–10 μm) [*Binkowski and Roselle*, 2003]. Emission source characteristics or meteorological conditions

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do not play a role in determining the emission size distribution other than through the source's relative amount of emitted organic and elemental carbon versus other species.

[4] Most if not all of the distributions given by *Whitby* [1978] were measured with the Whitby Aerosol Analyzer (WAA). This instrument typically measured 8 size bins less than 100 nm with centers of 8.75, 12.5, 17.5, 25.0, 35.0, 50.0, 70.0, and 90.0 nm. There is considerable uncertainty in the Aitken mode median diameter as measured by the WAA, and the WAA measured a nearly monodisperse aerosol as having a modal standard deviation of typically 1.3. The charging and counting efficiency of the instrument became quite poor for particle diameters less than 20 nm [*Whitby et al.*, 1972]. Counter to this, modern sizing instruments such as the Scanning Mobility Particle Sizer (SMPS) typically have 30–60 size bins below 100 nm [e.g., *Hogrefe et al.*, 2006] and accurately count particles typically down to 10 nm or as low as 2.5 nm [*Heim et al.*, 2004]. The SMPS and other modern instruments improve our understanding of the Aitken mode and the size distribution of emitted particles. Especially because of the better counting sensitivity below 20 nm, modern instruments will estimate a much larger contribution to the overall size distribution from the Aitken mode and will, for the same emitted mass, produce more particles.

[5] Once the search has begun for appropriate size distributions, there is the question of what the term "emission" size distribution means for a mesoscale air quality model. *K. M. Zhang et al.* [2005], *Ketzel and Berkowicz* [2004], *Gidhagen et al.* [2005], and *Jacobson and Seinfeld* [2004] have all recognized that the term emission, especially for particulate matter, is dependent on the time and space scales of interest. For example, as automobile exhaust travels from the combustion source in the engine through the tailpipe, catalytic converter, and muffler, the mixture of solids, liquids, and gases undergo significant chemical and physical processes: nucleation of elemental carbon chains, cooling and condensation of semivolatile species, particle fragmentation, deposition on the walls of the exhaust system, and chemical reaction in the tailpipe and catalytic converter [*Kittelson*, 1998; *Burtscher*, 2005; *Harris and Maricq*, 2002]. Measurements downstream of the tailpipe combine combustion and processing to the 1 to 10 m spatial scale. This is a convenient scale since it lumps together all the processes that do not depend on atmospheric conditions, other than the effect of temperature on engine and catalytic converter operating efficiency. Although mobile sources are used as an example here, a similar discussion applies for combustion sources of all types.

[6] This emission scale is not, however, appropriate for a mesoscale model because a mesoscale air quality model such as CMAQ cannot directly treat spatial scales below its grid resolution. The CMAQ grid resolution is chosen by the user sometimes as fine as 1 km but more often 4–15 km or even greater because of limitations in emissions and meteorology inputs or limitations in computing resources. The 4–15 km grid resolution is used as an example here since it is a typical high resolution for current mesoscale studies. CMAQ takes the inventory of emissions in a grid cell per time step and spreads them throughout the entire cell. Hence, emissions on a spatial scale of a few meters are immediately transformed into emissions on the scale of several kilometers. Any processing that would alter the plume as it dilutes and ages from the tens of meters to the 4–15 km scale is ignored. This can be an important omission for emitting the correct aerosol composition and mass [e.g., *Volkamer et al.*, 2006], but it is critical for the size distribution and especially the ultrafine number concentration since multiple processes act to modify submicron particles on these spatial scales. Particle nucleation often occurs in plumes from point sources and roadways on the tens of meters to 1 km scale [e.g., *Brock et al.*, 2002; *Wilson and McMurry*, 1981; *Hewitt*, 2001]. For mobile emissions of ultrafine particles, processes between the scale of a tailpipe and the scale of a mesoscale model include: dilution, cooling, coagulation, and deposition in the roadway environment [*Zhang and Wexler*, 2004; *Gidhagen et al.*, 2004]; and dilution, condensation, evaporation, coagulation, and fragmentation on the scale of hundreds of meters beyond the roadside environment [*Zhu et al.*, 2002; *Shi et al.*, 1999; *Zhang et al.*, 2004; *Gramotnev and Gramotnev*, 2005; *Jacobson et al.*, 2005; *Kerminen et al.*, 2007]. In all cases, these processes significantly affect the number, median diameter, and geometric standard deviation of the Aitken mode in particular but also the accumulation mode.

[7] Aerosol chemistry and dynamics between the 1 to 10 m scale and the 4 to 15 km scale can be accounted for either by modeling them as part of atmospheric processing or by incorporating them into the emission inventory. Modeling solutions include nested domains, plume models, and a general spatial and temporal aging model. However, nested domains cannot capture these scales because it is unrealistic to create a nested domain with a grid resolution of meters to tens of meters for an entire urban area, parameterizations and assumptions for atmospheric dynamics and thermodynamics in mesoscale models are not appropriate for scales below 1 km, and emission inventories have not been compiled on this spatial scale for entire urban areas.

[8] Plume models have effectively simulated pollution plumes on the roadside, near-field (100 m to 1 km), and far-field (1–50 km) scales. As examples, at the roadside scale *Gidhagen et al.* [2004] used an aerosol dynamics model to simulate ultrafine aerosol in four size sections in a Stockholm, Sweden street canyon; at the near-field scale *K. M. Zhang et al.* [2005] link the CALINE4 line source roadway dispersion model [*Benson*, 1992] and the UCD 2001 plume source roadway dispersion model [*Held et al.*, 2003] to a simple aerosol dilution scheme in order to investigate processing downwind of a Los Angeles freeway; and far-field, plume-in-grid models integrate aerosol chemistry and dynamics for large point sources within a regional grid [*Karamchandani et al.*, 2002; J. M. Godowitch, Development and application of the CMAQ plume-in-grid-model, paper presented at 4th Annual CMAS Models-3 Users' Conference, Community Modeling and Analysis System Center, Chapel Hill, N.C., 26–28 September 2005; R. Morris et al., Use of hybrid plume/grid modeling and the St. Louis super site data to model $PM_{2.5}$ concentrations in the St. Louis area, paper presented at 6th Annual CMAS Conference, Community Modeling and Analysis System Center, Chapel Hill, N.C., 1–3 October 2007]. Although these tools have successfully modeled the transformation of a plume to a larger scale, it is unfeasible to model each of the thousands of sources in an urban area and

all of their plume interactions. Each vehicle or vehicle type on a freeway produces its own plume and would need to be modeled separately until it merged with other vehicle plumes and other nearby sources. Modeling each plume in one road segment and the consolidation process of all the plumes in the segment could easily involve a computational demand equal to modeling the entire mesoscale Eulerian domain. Even if the emissions were first parameterized to the road segment level, the number of road segments and other small-scale stationary sources in an urban mesoscale grid cell of typically 4–15 km quickly overwhelms current computing capabilities.

[9] One alternative is a general spatial and temporal aging model. The most general version of the model would use off-line calculations to determine how a plume from an average source (traffic, coal power plant, gas power plant, refinery, etc.) ages over the typical transition to the mesoscale for typical meteorological conditions like day/night, stagnant air, clear and sunny, cloudy and windy, etc. Aerosol processing under various regimes could be applied to the emission sources before these emissions are input to CMAQ. A more spatially and temporally specific version of the model would be to run the temporal aging model for a few different classes of sources for the specific meteorological conditions in each grid cell every hour. In this case, the advantage in speed over a full plume-in-grid model is that only a few realizations would be needed per grid cell rather than three-dimensional modeling for the specific tens, hundreds, or thousands of plumes in each cell. This technique would bridge the gap in scale between traditional emissions and the mesoscale model and would account for meteorology but would do so without an unrealistic computational demand.

[10] Another avenue is to parameterize every process that occurs on a scale below the resolution of the air quality model grid as "emissions" based on measurements and empirical models. Scale parameterization concepts similar to this are used routinely in cloud and boundary layer modeling where cloud processes and boundary layer turbulence have spatial scales much smaller than the typical model grid resolution. Moreover, such parameterization assumptions are already implicit in traditional emissions estimates for processes occurring between the combustion chamber and the end of the tailpipe or smoke stack. The task in this paradigm is to find high-quality size distribution measurements that represent the 4–15 km scale for each of the many sources of interest. The difficulty is that a measurement near one source and at one location may be contaminated by other sources, may not be representative of all sources of this kind, or may not even be representative of the same source at a different time under different atmospheric or operating conditions. However, one clear advantage is that these measurements exist and can be relatively easily incorporated into a modeling system now with little or no degradation in model speed, whereas even a simplified modeling solution would require significant development and testing and would make the overall modeling system less efficient.

[11] Either a modeling or measurement solution would be an improvement over the current strategy of ignoring the problem entirely. This project chooses to update the emission size distribution with parameterizations from measurements. If the CMAQ results are highly improved by the changes, then the results would motivate further study with a generalized aging model. The measurement parameterizations might still be preferred over a generalized aging model because they would not affect model efficiency and are likely to be routinely used by all CMAQ modelers. If this technique can achieve most of the benefit of improved emission size distributions with no impact on model speed, it is a reasonable choice that balances performance, efficiency, and usability. Thus the solution employed here is to parameterize all processes below the 4–15 km grid as emissions and use existing measurements on this scale as the size distributions of emitted primary aerosol.

3. Selection of Mesoscale Emission Size Distributions

[12] This study contains published and unpublished size distribution measurements that represent major urban sources at the 4–15 km scale. The focus is on improving size distribution modeling generally for industrialized, urban areas. Measurements representing this type of environment around the world are treated equally. The mix of sources categorized in this study are representative of the major sources in most industrialized, urban areas, including those in our Pacific Northwest case study: (1) an urban core dominated by transportation, light industry, and residences; (2) coal-fired power plants; and (3) large, marine transport. The criteria for including published results were that the data had to be measured with instruments capable of characterizing the Aitken mode and that the measurements had to represent source emissions on the 4–15 km scale, or at least had to provide an upper or lower bound on size distribution properties on the 4–15 km scale. In all, 53 published studies and one unpublished study reported size distributions that meet the criteria.

[13] Ideally, the data sets adequately represent the full range of aerosol size distributions from midsized to large, industrialized, developed urban areas and adequately average emissions across operating conditions, seasons, and common weather patterns. In the reality of not being able to design a comprehensive monitoring program for this purpose, a compilation of existing measurement campaigns will inevitably have biases. In some cases one journal article contains measurements from multiple distinct locations. Sometimes several journal articles represent data from the same site, even the same exact data sets, but either subset them differently, analyze them differently, or process different time periods. In general, unless two articles display the same data analyzed in the same manner, the methodology here was to treat the data set in each journal article as separate data sets as well as to treat each location within one journal article as separate data sets. Other biases include: data sets biased toward the neighborhoods which have been studied rather than an adequately representative number of locations within each city; limited measurement periods not fully representative of seasonal and meteorological conditions at the location; locations not representative of the urban background; and analyses not specifically designed to assess size distribution characteristics. This study averaged all data sets with equal weight so long as they met the minimum criteria for a well-instrumented urban background

Table 1. Summary of Data Sets Used to Derive Emission Size Distribution for Urban Areas

Location	Reference	Period of Observation
Alkmaar, Netherlands	*Ruuskanen et al.* [2001]	4 months
Atlanta, GA	*Woo et al.* [2001]	13 months
Atlanta	*McMurry and Woo* [2002]	25 months
Atlanta	*Rhoads et al.* [2003]	3 weeks
Baltimore, MD	*Tolocka et al.* [2005]	9 months
Brisbane, Australia	*Morawska et al.* [1999]	1.75 years
Brisbane	*Morawska et al.* [1998]	1.75 years
Copenhagen, Denmark	*Ketzel and Berkowicz* [2004]	months of data over 2 years
Copenhagen	*Ketzel et al.* [2003]	11 weeks over 6 months
Copenhagen	*Ketzel et al.* [2004]	4 weeks over 3 months
Copenhagen	*Wåhlin et al.* [2001]	1.5 months
Erfurt, Germany	*Ruuskanen et al.* [2001]	4 months
Erfurt	*Tuch et al.* [1997]	6 months
Göteborg, Sweden	*Janhäll et al.* [2006]	2 months
Graz, Austria	*Sturm et al.* [2003]	<1 month
Helsinki, Finland	*Hussein et al.* [2004]	6 years
Helsinki	*Hussein et al.* [2005]	12 weeks over 2 years
Helsinki	*Laakso et al.* [2003]	3 years
Helsinki	*Ruuskanen et al.* [2001]	4 months
Kawasaki City, Japan	*Fushimi et al.* [2008]	13 months
Kuopio, Finland	*Kikas et al.* [1996]	1–2 months
Leipzig, Germany	*Putaud et al.* [2003]	4 years
Leipzig	*Tuch et al.* [2003]	5 years
Leipzig	*Wehner and Wiedensohler* [2002]	4 years
Leipzig	*Wehner et al.* [2002]	3 months
Leipzig	*Wehner et al.* [2002] (self-processed)	3 months
Leipzig	*Wiedensohler et al.* [2002]	4.5 years
London, UK	*Putaud et al.* [2003]	1.5 years
Los Angeles, CA	*Kim et al.* [2002]	5 months
Los Angeles	*Sardar et al.* [2005]	2 months
Los Angeles	*K. M. Zhang et al.* [2005]	5 months
Milano-Bresso, Italy	*Putaud et al.* [2003]	2 months
Nagoya, Japan	*Minoura and Takekawa* [2005]	2 months
New York City, NY	*Hogrefe et al.* [2006]	4 weeks
Pittsburgh, PA	*Bein et al.* [2005]	1 year
Pittsburgh	*Cabada et al.* [2004]	1 year
Pittsburgh	*Stanier et al.* [2004]	1 year
Pittsburgh	*Q. Zhang et al.* [2005]	3 weeks
Pittsburgh	*Zhou et al.* [2005a]	5 days
Pittsburgh	*Zhou et al.* [2005b]	13 months
Santiago, Chile	*Trier* [1997]	2.5 years
Seattle, WA	*Larson et al.* [2006]	1.5 years
Stockholm, Sweden	*Gidhagen et al.* [2005]	1 month
Tallinn, Estonia	*Kikas et al.* [1996]	1–2 months
Tartu, Estonia	*Kikas et al.* [1996]	1–2 months

site. Then, the data set averages were modified to be better consistent with data sets that are very well understood and trusted by these authors [e.g., *Wehner et al.*, 2002; *Larson et al.*, 2006; *Brock et al.*, 2002].

[14] Thirty-eight of the qualifying journal articles contain 45 distinct urban data sets (Table 1). Many of these are claimed to represent traffic emissions at the kilometer scale but were not specifically segregated from other contributing sources. Though the sampling site may in fact be dominated by traffic emissions, only three publications presented distributions segregated by sources [*Larson et al.*, 2006; *Zhou et al.*, 2005a, 2005b], and one publication [*Zhang and Wexler*, 2004] sampled on the kilometer scale but near such a concentration of freeway traffic as to be sufficiently dominated by mobile emissions. The sample duration varied from 5 days [*Zhou et al.*, 2005a] to 6 years [*Hussein et al.*, 2004] with an average of 14 months, and the locations ranged from rooftop sites [e.g., *Ketzel et al.*, 2004] to sites in city parks surrounded by a variety of streets, arterial routes, and freeways [e.g., *Zhou et al.*, 2005a]. Most data sets come from midlatitude American and European cities that are no longer dominated by industrial emissions. In the United States, the cities sampled were Atlanta, Baltimore, Los Angeles, Pittsburgh, Rochester, and Seattle. The European cities tend not to be the large, polluted capitals; instead, they are Central and Northern Europe urban areas where universities have dedicated resources to aerosol sampling. Other continents are represented by Nagoya, Japan; Santiago, Chile; and Brisbane, Australia. Notably absent at this point are megacities.

[15] Thirteen studies representing 17 coal-fired power plant data sets met the needs of this study (Table 2). Few studies have sampled power plants on a 4 to 15 km scale. Much of the airborne plume research effort has gone into characterizing aerosol mass and sulfate gas-to-particle conversion rates because of the need to address $PM_{2.5}$, acid deposition, and regional haze and because of the difficulty in sampling transient, heterogeneous plumes from a moving aircraft. To expand the number of data sets in this analysis,

Table 2. Summary of Data Sets Used to Derive Emission Size Distribution for Coal-Fired Power Plants[a]

Location	Reference	Period of Observation
Centralia, WA	*Hobbs et al.* [1983]	4 plumes on 4 days
Chamber study	*Lipsky et al.* [2004]	NA
Chamber study	*Teinemaa et al.* [2002]	NA
Conesville, OH	C. A. Brock (unpublished data, 2006)	a few hours
Cumberland, TN	*Brock et al.* [2002]	2 passes
Finland	*Dekati Ltd.* [2003]	1 plume on 1 day
Four Corners	*Hobbs et al.* [1983]	3 plumes on 3 days
Gallatin, TN	*Brock et al.* [2002]	1 pass
Hong Kong	*Yao et al.* [2006]	1 summer and 1 winter plume
Johnsonville, TN	*Brock et al.* [2002]	3 passes
Labadie, MO	*Cantrell and Whitby* [1978]	1 pass
Nanticoke, ON	*Cho* [2005]	21 passes over 5 winter/summer flights
Navajo, AZ	*McMurry et al.* [1981]	1 pass
Navajo, AZ	*Wilson and McMurry* [1981]	2 plumes on one morning
Paradise, KY	*Mueller and Imhoff* [1994]	NA
Three Plants	*Ylatalo* [2006]	NA
Thomas Hill, MO	*Brock et al.* [2002]	1 pass

[a]NA, not applicable.

Table 3. Summary of Data Sets Used to Derive Emission Size Distribution for Marine Vessels

Location	Reference	Period of Observation
Bremen Express	*Frick and Hoppel* [2000]	3 passes
California coast ship 1	*Chen et al.* [2005]	2 passes
California coast ship 2	*Chen et al.* [2005]	2 passes
Fremo Scorpius	*Hobbs et al.* [2000]	1 pass
MSC Giovanna	*Sinha et al.* [2003]	1 pass
NYK Sunrise	*Hobbs et al.* [2000]	2 passes
Royal Sphere	*Sinha et al.* [2003]	1 pass
Sea Pearl	*Frick and Hoppel* [2000]	4 passes
Slocan Park, Vancouver, BC	*Lu et al.* [2006]	30 min average
Star Livorno	*Hobbs et al.* [2000]	2 passes
Tai He	*Hobbs et al.* [2000]	1 pass
Unidentified	*Frick and Hoppel* [2000]	2 passes
USS Mt. Vernon	*Frick and Hoppel* [2000]	3 passes

three laboratory studies [*Teinemaa et al.*, 2002; *Dekati Ltd.*, 2003; *Lipsky et al.*, 2004] were included where conditions of dilution and residence time represented the mesoscale. Two of the laboratory studies and one ambient study were conducted in Europe, while 12 ambient data sets and the remaining laboratory data set were collected in North America. Because a power plant's downwind effluent can depend on the type of fuel burned, on air pollution control equipment, on atmospheric mixing, and on UV radiation, averaging size distributions from several power plants will average power plant conditions and fuel types. This average will represent the mix of study locations and times in the scientific literature rather than individual plants or an appropriate average of power plants, fuels, and atmospheric conditions.

[16] Only five journal articles report measurements of the aerosol size distribution from large marine vessels on the scale of interest (Table 3). Ship traffic has not been typically viewed as a major source of air pollution in urban areas until very recently when other urban sources became dramatically cleaner and when marine trade and cruise ship traffic increased. Early in situ measurements of ship plumes focused on reporting the number of particles that might act as cloud condensation nuclei rather than the aerosol size distribution [e.g., *Radke et al.*, 1989]. Starting in 1994, real-world ship emissions of aerosol size distributions were measured as part of the Monterey Area Ship Track (MAST) [*Frick and Hoppel*, 2000; *Hobbs et al.*, 2000], the Intercontinental Transport and Chemical Transformation (ITCT) [*Chen et al.*, 2005], and the Safari 2000 [*Sinha et al.*, 2003] aircraft campaigns. Sampling times varied from 15 s to several minutes. *Lu et al.* [2006] identified ship plumes originating in Burrard Inlet and received at the Slocan Park Pacific 2001 site in central Vancouver, BC. The aerosol size distribution displayed by *Lu et al.* [2006] represents the only known measured ship emission size distribution in an urban area, from a ground location, or with a long averaging time.

4. Application of Updated Mesoscale Emission Size Distributions to CMAQ

[17] The source-specific size distributions must be converted into chemical species-specific, lognormal modes that CMAQ expects for emissions input. First, to form lognormal modes each size distribution data set was analyzed for five parameters: the Aitken mode median diameter, Aitken mode geometric standard deviation, accumulation mode median diameter, accumulation mode geometric standard deviation, and the mass fraction split of $PM_{2.5}$ into Aitken and accumulation modes. Auxiliary material provides the results of this analysis.[1]

[18] From the 45 urban size distribution data sets, these five parameters were obtained as reported values in the journal article for 17 data sets, by numerically fitting lognormal distributions to differential data for two data sets, and by estimation from journal figures for 31 data sets. Parameters for some data sets came from a combination of techniques and are counted in the tally for each technique. All the values for each parameter were averaged because there is no reason to apply weights to each measurement. However, a minority of data sets are known to be of the highest quality and applicable to this study. The averages for each parameter are checked against these known data sets and are subjectively adjusted toward them in some cases. For instance, the average of the Aitken median diameter is 30 nm, but 25 nm was used to better conform to *Larson et al.* [2006] and *Wehner et al.* [2002]. The accumulation mode geometric standard deviation was adjusted from 1.8 to 1.7 to conform with *Larson et al.* [2006]. Self-processing of the *Wehner et al.* [2002] data set was not used to anchor the accumulation mode geometric standard deviation because the accumulation mode numerical fit was relatively unstable and less reliable.

[19] For power plants, numerical fitting, table or text values, and estimation from journal figures provided size distribution information for 1, 4 and 12 data sets, respectively. With fewer data sets for each parameter than for urban areas, there is more uncertainty in the values and more opportunity for a spurious data set to affect the average. The Aitken median diameter, Aitken geometric standard deviation, accumulation mode geometric standard deviation, and Aitken/accumulation $PM_{2.5}$ split were all adjusted toward the data sets of the highest known quality (Conesville from C. A. Brock (unpublished data, 2006) and the data sets given by *Brock et al.* [2002]).

[20] No marine vessel data sets were fit numerically, four studies gave specific size distribution information, and parameters were estimated from figures for 13 data sets. The information for the Aitken median diameter was simply averaged. For the accumulation median diameter and Aitken/accumulation $PM_{2.5}$ split, the Slocan Park organic carbon size distribution was heavily weighted and the Slocan Park sulfate size distribution was deemphasized because much more organic mass than sulfate was present in the plumes identified by *Lu et al.* [2006] as marine vessel emissions. None of the data sets provides an Aitken or accumulation mode geometric standard deviation. For these two parameters, the power plant values were used since marine vessels are more similar to power plants than to urban areas in that they are both large, relatively sulfate-rich combustion sources that mix with only a few (or no) other similar sources on the 4–15 km scale. It is only an approximate solution since power plants typically have

[1] Auxiliary materials are available in the HTML. doi:10.1029/2009JD012401.

Table 4. Emission Lognormal Parameters for Urban Area, Power Plant, and Marine Vessel Sources as Well as Species-Specific Emission Lognormal Parameters From the CMAQ Base Case[a]

Mode	Species or Source	% Mass	D_g (nm)	D_{gv} (nm)	σ_g
Aitken	CMAQ sulfate, nitrate, other fine	0	13	30	1.7
	CMAQ OC and EC	0.1	13	30	1.7
	urban	*10*	*25*	*60*	*1.7*
	power plant	*15*	*25*	*85*	*1.9*
	marine vessel	*25*	*60*	*200*	*1.9*
Accumulation	CMAQ sulfate, nitrate, other fine	100	70	300	2.0
	CMAQ OC and EC	99.9	70	300	2.0
	urban	*90*	*120*	*280*	*1.7*
	power plant	*85*	*150*	*250*	*1.5*
	marine vessel	*75*	*150*	*250*	*1.5*
Coarse	CMAQ	–	929	6000	2.2

[a]Urban area, power plant, and marine vessel sources are italic. "% mass" refers to the percentage of emitted $PM_{2.5}$ mass apportioned to mode. % mass has no meaning for the coarse mode. D_g is number median diameter, D_{gv} is volume median diameter, and σ_g is geometric standard deviation. "OC and EC" indicates organic carbon and elemental carbon.

pollution control devices that limit sulfur dioxide and soot emissions while marine vessels do not.

[21] Because most of the size distribution parameters were not explicitly recorded in publications but instead had to be estimated from figures and other information provided, there is considerable error and uncertainty in some of the derived parameters. However, the literature analysis revealed a similar level of variability in the data sets from location to location, and sometimes equivalent variability for data sets using the same fundamental data source. For example, the three journal articles from the same site in Copenhagen [*Ketzel et al.*, 2003, 2004; *Ketzel and Berkowicz*, 2004] gave averaged Aitken median diameters of 28 to 39 nm. Even numerical fitting to high-quality data sets has similar variability when different fitting parameters are used.

[22] The source-specific size distributions on the 4–15 km scale differ substantially from the CMAQ emission size distributions (Table 4 and Figure 1). The biggest difference is that the new distributions have a more prominent Aitken mode. CMAQ's elemental and organic carbon distribution has an Aitken mode that is less prominent than its accumulation mode, and CMAQ's distribution for sulfate, nitrate, and unspeciated mass has no Aitken mode at all. All of the new sources have distributions with significantly more $PM_{2.5}$ apportioned to the Aitken mode, up to 25% for marine vessels. *Park et al.* [2006] also updated the emission size distribution in CMAQ and focused on the split of emitted $PM_{2.5}$ into the Aitken and accumulation modes. They concluded from tunnel measurements of traffic emissions [*Venkataraman et al.*, 1994] that 15% of the emitted elemental and organic carbon should be emitted into the Aitken mode. Our study arrived at a similar conclusion based on a more extensive analysis. The Aitken mode

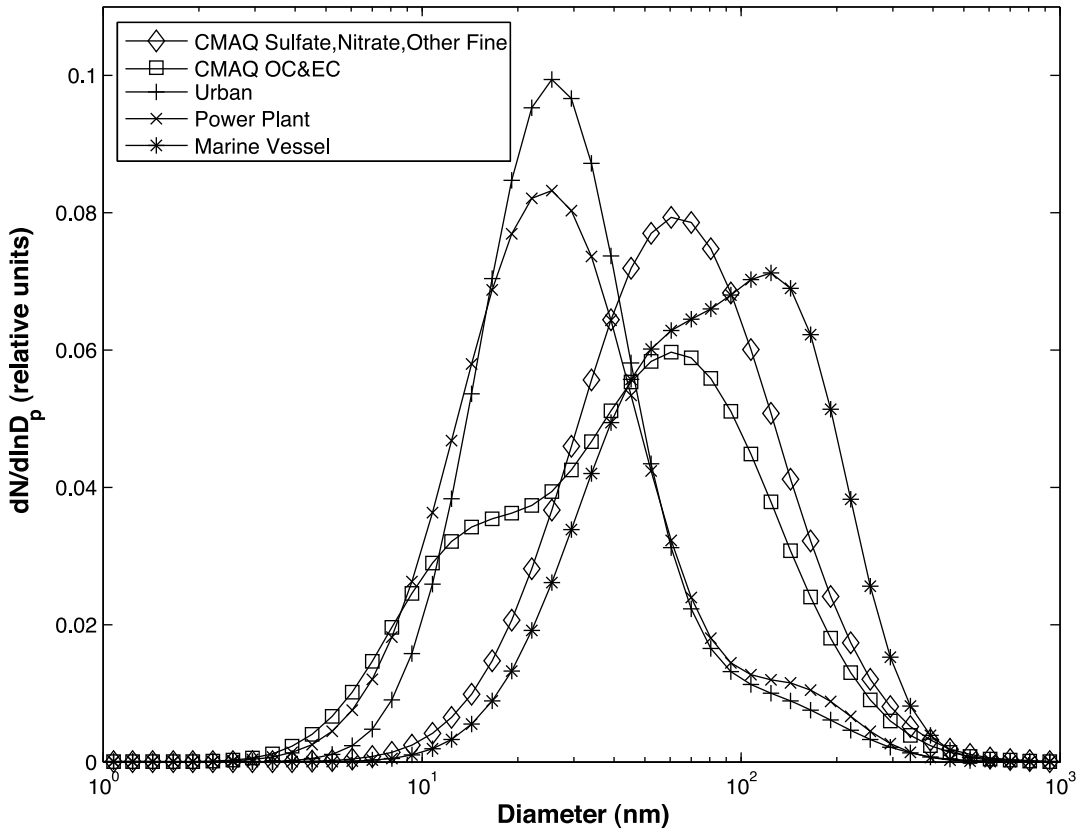

Figure 1. Emission number size distributions on 4–15 km scale for urban area, power plant, and marine vessel sources, as well as species-specific emission size distributions from the CMAQ base case.

number median diameter (D_g) increases from 13 nm in CMAQ to 25 nm for urban areas and power plants and to 60 nm for marine vessels, while the Aitken mode geometric standard deviation increases from 1.7 in CMAQ to 1.9 for power plants and marine sources. The accumulation mode number median diameter also increases by 50–80 nm, and the mode's geometric standard deviation decreases from 2.0 to 1.7 for urban areas and to 1.5 for power plants and marine vessels. These changes represent a large departure from what CMAQ has been using. An example of the effect on total particle number concentration is presented below.

[23] The second step to using the measured emission size distributions in CMAQ is to resolve the disconnect between the source-specific size distributions on the 4–15 km scale and the way CMAQ normally applies chemical species-specific size parameters. Standard emissions inventories provide particle emissions as $PM_{2.5}$ and PM_{10} by major chemical species for specific sources. An emissions processor such as SMOKE maps the source-specific emissions into gridded species-specific emissions for the period of simulation, losing source information in the process. Until this point, particle size is no more specific than $PM_{2.5}$ or PM_{10}. CMAQ then assumes a size distribution of emitted particles for each species. To properly incorporate the source-specific size distributions on the 4–15 km scale, SMOKE must be modified so that it applies a size distribution profile in addition to a chemical species profile when processing sources and must be modified so that it outputs species-specific size distributions for each grid cell at each time. CMAQ would then need modifications to accept this input. This is the best solution in the long run, but it requires coordinating major changes to two very different models.

[24] Most of the improvement in emission size distributions can be achieved without major changes to SMOKE and CMAQ because the difference between the source-specific distributions on the 4–15 km scale and what is currently in CMAQ is much greater than that among the emitted size distributions for the major urban, power plant, and ship sources (Table 4). Instead of adding a size distribution profile to SMOKE, it is possible to take each species (elemental carbon, organic carbon, nitrate, sulfate, and unspeciated mass) and apply the emission size distribution from the source that emits the most of that species. Analysis of the emission inventory for the Puget Sound region, including the Seattle metropolitan area, showed that the urban areas dominate marine vessels and power plants for emissions of organic carbon and elemental carbon. The region's sole coal-fired power plant at the southern edge of the Puget Sound region dominates the unspeciated $PM_{2.5}$ emissions, but as the dumping ground for uncharacterized emissions, this category is highly uncertain and much of this mass is likely organic carbon. When unspeciated $PM_{2.5}$ mass is assumed to be organic carbon, the urban areas still emit much more organic carbon than the power plant. The urban area and power plant emit a similar amount of sulfate in this 2001 inventory. For this study, the urban size distribution is applied to sulfate because the focus is more on the urban and urban-influenced areas than areas downwind of the power plant. Some regions of the eastern United States with many coal-fired power plants may want to include a separate emission size distribution for sulfate based on the power plant size distributions. Nitrate was ignored in this study since little nitrate is emitted as primary mass in the inventory. In light of these results and considerations, CMAQ v4.4 was modified to use the urban emission size distribution for all $PM_{2.5}$ species: sulfate, nitrate, elemental carbon, organic carbon, and unspeciated $PM_{2.5}$. The emission size distribution for coarse mode aerosol species was not modified. Although this size distribution input is representative for the Puget Sound region, similar considerations likely apply to a majority of urban areas.

[25] Given the discussion above, two simulations of the case study were run with emission aerosol size distributions different from that in CMAQ v4.4 (Table 5): an improved-emission scenario where all $PM_{2.5}$ emissions are emitted with the urban size distribution in Table 4, and a high-emission scenario that emits more particles while remaining within the bounds of the studies in Table 1. For the high-emission scenario, the Aitken number mode median diameter was lowered from the improved-emission value of 25 nm to 21 nm, the lower end of the range from the urban data sets. The percentage of $PM_{2.5}$ emissions apportioned to the Aitken mode was increased from 10% to 20% based on the standard deviation of the values from urban data sets. The standard deviation and not the upper end of the range was used since the upper end is diffuse and not clustered around one value. Only these two parameters of the size distribution were changed for the high-emission scenario because they have by far the largest effect on the number of emitted particles. The improved-emission and high-emission scenarios were implemented in CMAQ, applied for a period of August 2001 when detailed airborne and surface measurements were available from the coordinated Pacific Northwest 2001 (PNW2001) and Pacific 2001 field campaigns, and compared to results from the standard CMAQ v4.4 (base case). Although CMAQ v4.4 was used so that results could be compared to previous analyses, the most current version of CMAQ, v4.7, uses the same emission size distributions and has a similar aerosol scheme. See *Elleman and Covert* [2009a] for more information on the field campaigns, CMAQ modeling, and modeling domain. When modeled number concentrations are compared to observations, only the modeled number in the observable size range with diameters greater than 7 or 9 nanometers (depending on the site) are compared to the observed number concentrations.

5. CMAQ Results With Updated Emission Size Distributions

[26] Before comparing CMAQ model results with different emission size distributions, it is instructive to examine the change in emitted particles resulting from modifications to the emission size distributions. Starting with organic and elemental carbon, when the mass-based percentage of $PM_{2.5}$ emissions apportioned to the Aitken mode was increased from 0.1% (base case) to 10% (improved emission) and to 20% (high emission) while holding the other size distribution parameters at the base case values, the number of emitted particles increases by a factor of 30 and 59. When the Aitken mode volume median diameter (D_{gv}) was changed along with the Aitken mode apportionment (from 30 nm and 0.1% to 60 nm and 10% for improved emission

Table 5. Summary of Aitken and Accumulation Mode Parameters for Improved-Emission and High-Emission Scenarios in CMAQ[a]

Mode	Scenario	% Mass	D_g (nm)	D_{gv} (nm)	σ_g
Aitken	improved emission	10	25	60	1.7
	high emission	20	21	50	1.7
Accumulation	improved emission	90	120	280	1.7
	high emission	80	120	280	1.7

[a]Coarse mode parameters remained unchanged from CMAQ v4.4. "% mass" refers to the percentage of emitted $PM_{2.5}$ mass apportioned to mode. D_g is number median diameter, D_{gv} is volume median diameter, and σ_g is geometric standard deviation.

and to 50 nm and 20% for high emission), the number of emitted particles increases over the base case by only a factor of 4.3 and 13.1. Changes to the accumulation mode median diameter and geometric standard deviation have little effect on the number of emitted particles. When all the modifications to the emission size distributions are made, the improved-emission and high-emission distributions increase the number of emitted particles by a factor of 3.9 and 12.8. For sulfate, nitrate, and unspeciated emissions, the parameters display similar sensitivity: the number of emitted particles increases by a factor of 5.6 and 18.0 for the improved-emission and high-emission scenarios. Even though this study proposes the same improved- and high-emission scenarios for all species, the change in emitted particles from the base CMAQ is different for organic and elemental carbon than it is for other species because CMAQ emits 0.1% of $PM_{2.5}$ mass into the Aitken mode for carbon species and 0% for all other species. When the emission size distribution changes were made to the mix of species emitted from the entire PNW2001/Pacific 2001 domain, the Puget Sound only, and Seattle metropolitan area only (where organic/elemental carbon represents 50%, 90%, and 93% of the total emitted $PM_{2.5}$, respectively), the improved-emission and high-emission scenarios increase the number of emitted particles by a factor of 4 to 5 and a factor of 13 to 15 (Table 6).

[27] About half of the enhancement in the number of emitted particles is preserved as an increase in number concentrations after CMAQ meteorological, dynamical, and chemical processing. The other half is lost via coagulation with the surviving particles, lost to the Earth's surface through dry deposition, or advected out of the domain. Over land, the changes in emissions always increase the number of modeled particles. The improved-emission scenario produces about three times more particles in the urban areas than the base case, and in regions impacted by sulfate emissions, it produces up to four times more particles. Enhancements greater than a factor of four occur in the immediate vicinity of sulfate sources. The improved-emission number concentrations are correlated with the base case number concentrations with an $R^2 = 0.97$ domain-wide and $R^2 = 0.98$ for urban and urban-influenced areas. There are no cases where the improved-emission scenario creates high number concentrations where they did not already exist in the base case. The high-emission scenario increases the number of particles in urban, sulfate-influenced, and sulfate-dominated areas by another multiple of three over the improved-emission scenario, and it is also very highly correlated with the base case number concentrations. Thus, the mesoscale updates to the emission size distributions increase the number of particles modeled by CMAQ by a factor of 2 to 3 downwind of urban areas, by a factor of 3 to 4 in urban areas, and up to a factor of 4 near sulfate sources. The largest though still realistic increase in the number of particles is a factor of 12 immediately next to sulfate sources such as pulp mills. This is consistent with *Park et al.* [2006] who found an increase by a factor of 4.3 and 6.0 at an urban location for emitting 10% and 15% of the elemental and organic carbon into the Aitken mode.

[28] The changes in CMAQ performance can be investigated through the change in number concentrations at the time and location of observations, number concentrations in particular environments along the PNW2001 flight paths, and average size distributions measured at the Pacific 2001 ground stations. *Elleman and Covert* [2009a] found that CMAQ underpredicted the total number of observable particles by 1–2 orders of magnitude in the urban areas regardless of air mass aging, chemistry, time of day, and meteorology. The updated emission chemical size distributions noticeably increase the CMAQ modeled number concentrations and reduce the number underprediction relative to observations (Table 7). An example result is shown in Figure 2 for the PNW2001 flight on the afternoon of 26 August. For the improved-emission case the observed number concentrations (plotted in blue) are generally underpredicted by 1 order of magnitude, and for the high-emission case (plotted in cyan) they are underpredicted by about 0.5 orders of magnitude. The Pacific 2001 surface sites see an increase in number concentrations by 0.5 and 1 orders of magnitude for the improved-emission and high-emission cases at all hours of the day. The improvement with updated emissions is more pronounced where CMAQ models higher concentrations, regardless of source, chemical aging, day, or time of day. The features in the number concentrations along the flight path and in the time series at the ground stations remain unchanged. Such behavior indicates that these emission size distribution changes are linear changes to the CMAQ results and are not large enough to appreciably affect other aerosol processes such as nucleation and coagulation, which act nonlinearly on the number of particles and depend on the preexisting amount of aerosol.

[29] The improved-emission scenario improves the overall character of the modeled aerosol size distributions. The base case produced an Aitken mode at Langley (Figure 3) and Sumas (Figure 4) that appears as little more than the lower tail of the accumulation mode. The improved-emission scenario creates a more prominent Aitken mode with a lower median diameter that is closer to observations. The average daytime Langley and Sumas Aitken modes still

Table 6. Factor Increase in Aerosol Number Emissions Relative to the Base Case for the Improved-Emission Size Distribution and for the High-Emission Size Distribution, as Calculated for the Domain, Puget Sound, and Seattle/Bellevue Metropolitan Area

	Domain (50% OC and EC)	Puget Sound (90% OC and EC)	Seattle/Bellevue (93% OC and EC)
Improved emission	5.0	4.4	4.3
High emission	15.3	13.5	13.4

Table 7. Average Observable Number Bias Relative to Observations for the PNW2001 Flights and for the Two Pacific 2001 Sites Where Aerosol Number Concentration Measurements Are Available[a]

Scenario	26 August, P.M.	27 August, A.M.	27 August, P.M.	Langley	Sumas (Daytime)
Base case	−6,500	−4,000	−14,800	−11,700	−14,100
Improved emission	−5,800	−3,400	−13,800	−8,600	−11,000
High emission	−4,000	−2,000	−11,000	−300	−2,100

[a]The Sumas number concentrations are limited to daytime hours. Unit is $1/cm^3$. A.M. indicates morning, and P.M. indicates afternoon.

peak at an unrealistically high diameter and are not sufficiently distinct from the accumulation mode, but the number underprediction is now uniform for sizes below 150 nm. The improved-emission averaged nighttime distribution at Langley has an Aitken mode that is more prominent than the accumulation mode, while the observations have an Aitken mode that is smaller than the accumulation mode. The error is due to a weak accumulation mode in the model rather than due to errors in the Aitken mode. The high-emission distributions at Langley and Sumas accentuate the changes from the base case to the improved-emission scenario to such an extent that the Aitken mode dominates the accumulation mode, the two modes are visually distinct, the Aitken median diameter is close to the observed value, and the Aitken mode is overstated, especially at night. Updating the emission size distribution resolves the underrepresentation of the Aitken mode relative to the accumulation mode, but it does not distinguish the two modes unless the high-emission size distribution is used, it does not solve the overall number underprediction below 150 nm during the day, it overemphasizes the importance of the Aitken mode at night, and it does not improve accumulation mode performance day or night.

[30] While updates to the emission size distribution do not alter emitted aerosol mass, they do have a small effect on the amount of modeled $PM_{2.5}$. The improved-emission scenario increases $PM_{2.5}$ at Washington surface sites by on average 0.5 $\mu g/m^3$ or 5%. The Pacific 2001 sites experience similar increases in modeled $PM_{2.5}$ with the largest increase at the site whose air mass is the most processed, Sumas, and the smallest increase at the urban site, Slocan Park. The high-emission scenario produces the same pattern of $PM_{2.5}$ increase but with a 0.7 $\mu g/m^3$ increase at Washington sites and a higher Sumas to Slocan Park spread at the Pacific 2001 sites. The emission size distribution affects the modeled aerosol mass more when CMAQ models a stable, shallow boundary layer. This occurs on the evening of 25 and 27 August for all three Pacific 2001 sites. It occurs additionally for Slocan Park overnight on 26–27 August and on the late afternoon of 26 August when the MM5 meteorological simulation brings a sea breeze front into Vancouver, BC. Nitrate accounts for a large amount of the mass increase because it comprises a high proportion of modeled $PM_{2.5}$ at night when boundary layers are stable.

[31] Several aerosol processes depend on the size distribution. Condensation of gas phase sulfate and organic compounds is proportional to the aerosol surface area

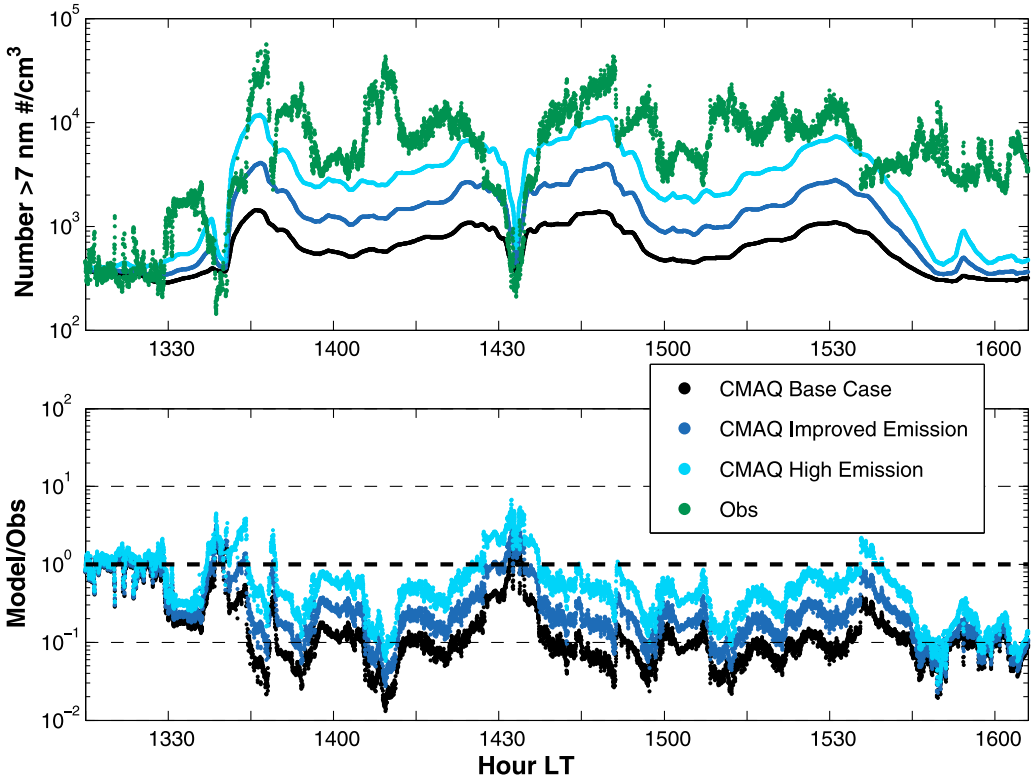

Figure 2. Comparison of CMAQ number concentrations to observations for particles with diameter greater than 7 nm for the PNW2001 flight on the afternoon of 26 August. Local time (LT) is equivalent to Pacific standard time.

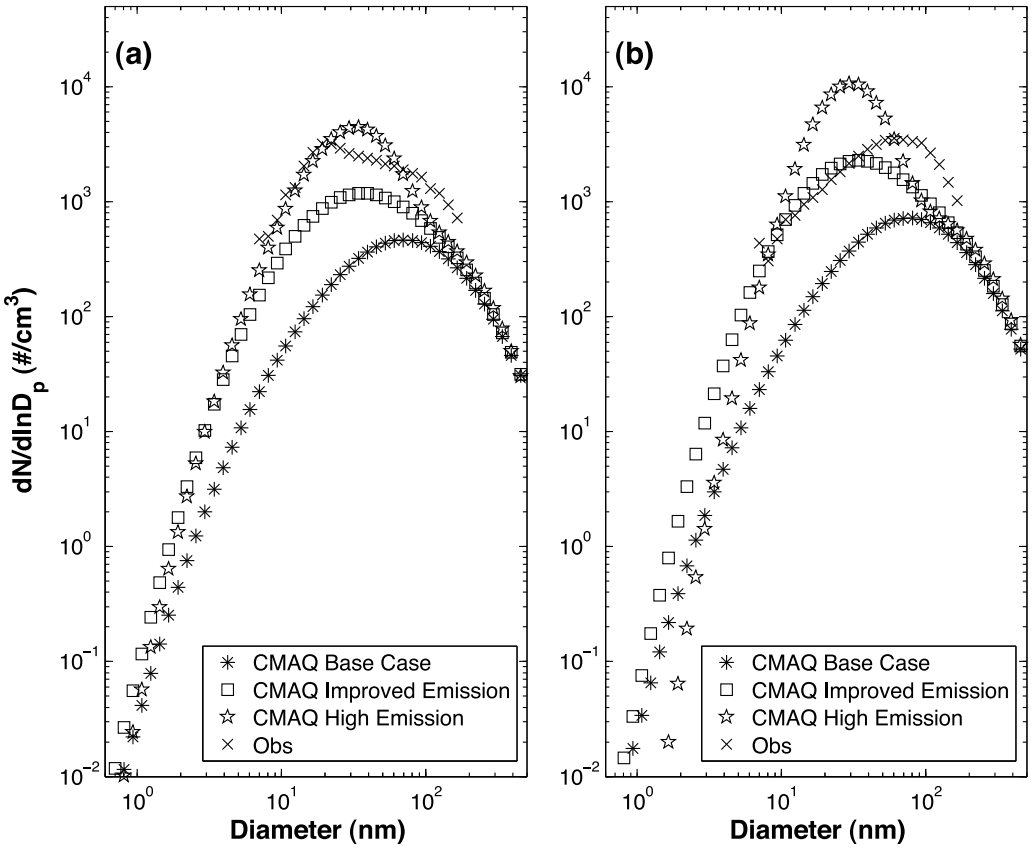

Figure 3. (a) Day and (b) night average particle number-size distributions at Langley as observed and for the base case, improved-emission, and high-emission cases.

which, for constant mass, increases as the distribution is weighted toward smaller particles; as the surface area to volume ratio increased by 25% in urban areas on average for the simulation period, condensation is more effective for the improved-emission scenario than for the base case. Both wet and dry deposition remove fewer aerosols from the atmosphere when the distribution has a more prominent Aitken mode. For wet deposition, only particles larger than a critical size will act as cloud condensation nuclei and potentially end up in raindrops that remove aerosol from the atmosphere. For dry deposition, smaller particles have a higher surface area to volume ratio and thus have a lower deposition velocity. A stable boundary layer increases the importance of dry deposition by confining the aerosol close to the surface, and it promotes aerosol condensation through its relatively cold surface layer. Indeed, the nighttime improved-emission dry deposition fluxes are 15% lower in the urban areas than for the base case. Since no rain fell during the 3 day simulation, it is likely that emitting aerosol preferentially into smaller sizes for the updated emission size distribution increases modeled $PM_{2.5}$ through higher aerosol condensation and lower dry deposition during stable boundary layer conditions.

[32] If changes in the emission size distribution can have a small effect on the modeled aerosol mass through condensation and deposition, then it is also possible for them to affect the concentration of gas-phase species. Some of the higher aerosol mass in the improved-emission and high-emission cases is due to a transfer of gaseous sulfate, nitrate, and organics to the aerosol phase. It is possible this would modify the gas-phase concentration of sulfur, nitrogen, and organic carbon compounds and potentially modify the entire chemical environment. However, the CMAQ concentrations of major species involved in air pollution and aerosol chemistry such as sulfur dioxide, nitrogen oxides, ozone, and ammonia, change by less than 1% for both the improved-emission and high-emission cases. Nitric acid increases by 1%–2% for both emission scenarios, but even this change is very small. Updates to the emission size distribution have a negligible effect on the gaseous chemical environment and only a modest effect on the modeled aerosol mass.

6. Summary and Conclusions

[33] In the goal to improve CMAQ's modeled aerosol size distributions, the emission size distributions in the model were updated to reflect current measurements and to address the spatial and temporal scales important for aerosols. The emission size distribution used for an Eulerian grid must either empirically incorporate all processes between combustion and the smallest resolvable spatial scale, or another modeling step must account for the scale between the emissions measurement and the grid scale of the regional model. If this is not done, then important aerosol processes occurring on this missing spatial scale will be ignored.

[34] This study culled observations of size distributions for urban areas, power plants, and marine vessels on the 4–

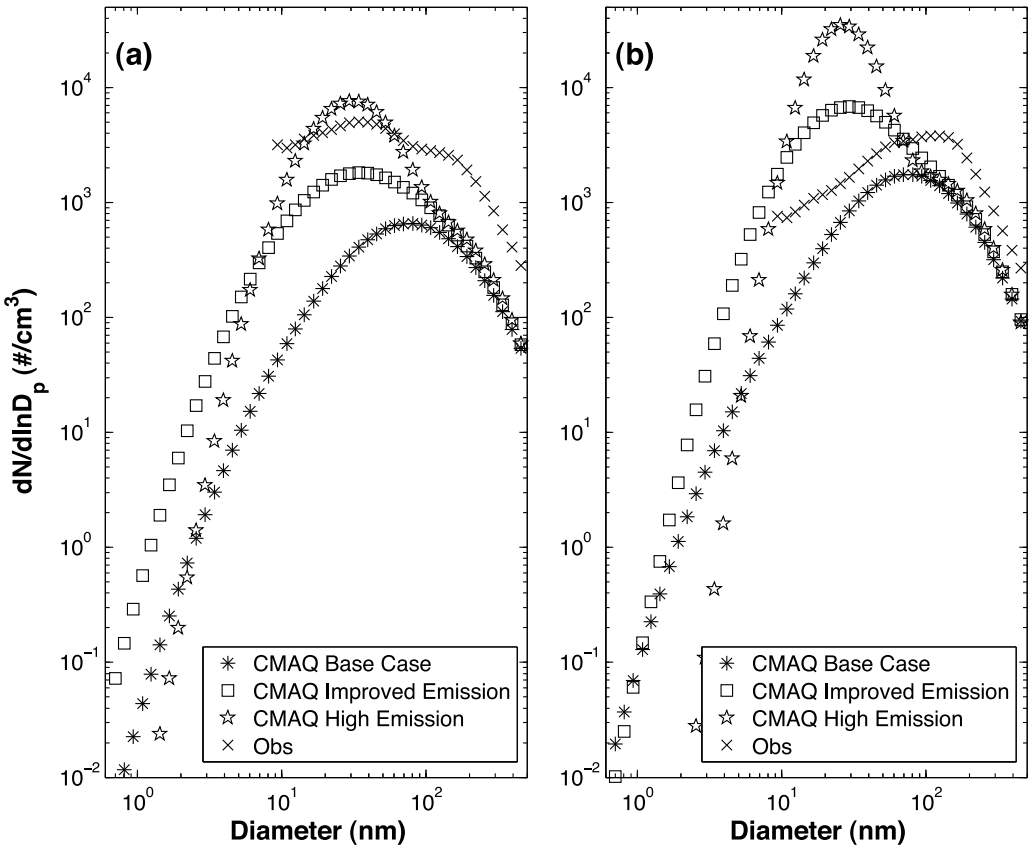

Figure 4. (a) Day and (b) night average particle number-size distributions at Sumas as observed and for the base case, improved-emission, and high-emission cases.

15 km scale from a survey of published and unpublished articles. The data sets disproportionately favor locations where many measurements have been taken rather than provide a representative distribution of measurements appropriate for most CMAQ domains. Nonetheless, they provide a large sample for calculating average emission size distributions. The new emission size distributions for urban areas, power plants, and marine vessels apportion a much larger fraction of aerosol mass to the Aitken mode than what is currently in CMAQ. Instead of 0% or 0.1%, now 10%, 15%, and 25% of the emitted $PM_{2.5}$ go to the Aitken mode for urban areas, power plants, and marine vessels. Even when combined with the larger Aitken mode median diameters suggested by this study, the new distributions emit many more particles, especially in the ultrafine range, than CMAQ currently does.

[35] To fit with the CMAQ modeling paradigm, the source-specific size distributions from the literature survey had to be modified in two ways. First, they were transformed into lognormal Aitken and accumulation modes to obtain modal median diameters, modal geometric standard deviations, and the apportionment of mass emitted into the Aitken and accumulation modes. Second, CMAQ expects the emission size distributions to be segregated by chemical species instead of by source, but this was seldom available in the literature survey. Because the urban source dominates the emissions of organic carbon and elemental carbon and emits as much sulfate as the major power plants in the Puget Sound, the urban source Aitken and accumulation mode characteristics were applied to all emitted species. The CMAQ results are specific to this case study but have valuable implications for most CMAQ modeling studies because all current CMAQ studies use the older and less accurate emission size distributions [*Elleman and Covert*, 2009a] and because the changes made to the emission size distributions in this study are relevant to most developed, urban areas.

[36] CMAQ was run using the urban source size distribution as the improved-emission scenario and was run for a high-emission scenario with an emission size distribution that emits the highest number of particles within bounds of the compiled urban size distributions. Of all the changes from the base case emission distributions, the revised apportionment of $PM_{2.5}$ into the Aitken and accumulation modes has the biggest effect on the number of emitted particles. For the PNW2001/Pacific 2001 domain, the improved-emission and high-emission size distributions increase the number of emitted particles by a factor of 4–5 and 13–15, respectively. When CMAQ is run using the improved-emission size distributions, the modeled number of particles increases by a factor of 2 to 3 downwind of urban areas, a factor of 3–4 in urban areas, and up to a factor of 4 near sulfate sources. Results from CMAQ using the high-emission size distributions show that changes in emission size distributions can, at most, increase the number of modeled particles by typically a factor of 5 to 10. The largest change relative to the base case occurs in areas with already high number concentrations and in sulfate-rich

regions, but in general the time and spatial pattern of number concentrations is not affected. $PM_{2.5}$ increases 5% due to higher condensational gas-to-particle conversion and lower dry deposition as aerosol mass is marginally shifted to smaller-sized particles. No significant change is seen in gas-phase species.

[37] While increased particle number concentrations are only a partial step toward solving the underprediction by 1 to 2 orders of magnitude, the new emission size distributions produce modeled size distributions that more closely match the observed distributions in terms of modes and modal parameters. The daytime distributions have fairly distinct Aitken and accumulation modes, and the overall shape is closer to observations despite being uniformly underpredicted by a factor of 5 below 150 nm. Nighttime distributions at Langley are well modeled in the Aitken mode but negatively biased in the accumulation mode. The inconsistent model performance from day to night either reveals additional problems with the model simulation or suggests that the emission size distribution may be different during the day than at night. The updated emission size distributions modestly improve the number concentration performance and better model the Aitken mode particles in the ultrafine range, which is not included in CMAQ as a specific mode.

[38] The modeled size distributions using updated emission size distributions moderately reduce but do not eliminate the underprediction in particle concentrations. This result suggests that emission size distributions are a relatively small factor in the size distribution errors and helps focus efforts on other potential causes. Our understanding of aerosols, especially in the ultrafine range, is not yet complete. Certainly the role of various chemical species in particle nucleation and in growing nucleation mode particles to 10–20 nm are unresolved scientific questions and the focus of intense research [e.g., *Elleman and Covert*, 2009b]. Mesoscale emissions of particles below the general instrument detection limit of 3 to 10 nm may be a substantial source of aerosol. Since the accumulation mode number below 250 nm is consistently underpredicted while $PM_{2.5}$ mass is well modeled, CMAQ's mode merging, competing gas phase condensation, or particle coagulation could be removing particles from the Aitken mode and low end of the accumulation mode and stacking them in the high end of the accumulation mode. A modeling technique for representing aerosol processing on the neighborhood scale is a necessary step beyond the observation-based parameterization in this study. Regardless of these open research questions, this study shows that after model improvements CMAQ is able to produce size distributions with the appropriate major features but still underpredicts number concentrations by $\sim$1 order of magnitude. The updated science that makes this possible was chosen to improve the model in the Pacific Northwest but applies generally to simulations in all midlatitude regions influenced by urban emissions.

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References

Bein, K. J., Y. Zhao, A. S. Wexler, and M. V. Johnston (2005), Speciation of size-resolved individual ultrafine particles in Pittsburgh, Pennsylvania, *J. Geophys. Res.*, *110*, D07S05, doi:10.1029/2004JD004708.

Benson, P. E. (1992), A review of the development and application of the CALINE3 and CALINE4 models, *Atmos. Environ.*, *26*, 379–390.

Binkowski, F. S., and S. J. Roselle (2003), Models-3 Community Multiscale Air Quality (CMAQ) model aerosol component: 1. Model description, *J. Geophys. Res.*, *108*(D6), 4183, doi:10.1029/2001JD001409.

Brock, C. A., et al. (2002), Particle growth in the plumes of coal-fired power plants, *J. Geophys. Res.*, *107*(D12), 4155, doi:10.1029/2001JD001062.

Burtscher, H. (2005), Physical characterization of particulate emissions from diesel engines: A review, *J. Aerosol Sci.*, *36*, 896–932, doi:10.1016/j.jaerosci.2004.12.001.

Byun, D. W., and K. L. Schere (2006), Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multiscale Air Quality (CMAQ) modeling system, *Appl. Mech. Rev.*, *59*, 51–77, doi:10.1115/1.2128636.

Cabada, J. C., S. Rees, S. Takahama, A. Khlystov, S. N. Pandis, C. I. Davidson, and A. L. Robinson (2004), Mass size distribution and size resolved chemical composition of fine particulate matter at the Pittsburgh supersite, *Atmos. Environ.*, *38*, 3127–3141, doi:10.1016/j.atmosenv.2004.03.004.

Cantrell, B. K., and K. T. Whitby (1978), Aerosol size distributions and aerosol volume formation for a coal-fired power plant plume, *Atmos. Environ.*, *12*, 323–333, doi:10.1016/0004-6981(78)90214-7.

Chen, G., et al. (2005), An investigation of the chemistry of ship emission plumes during ITCT 2002, *J. Geophys. Res.*, *110*, D10S90, doi:10.1029/2004JD005236.

Cho, S.-H. (2005), Detailed microphysical modeling study of particle size distributions in an industrial plume, Ph.D. dissertation, York Univ., Toronto, Ont., Canada.

Dekati Ltd. (2003), Applying a fine Particle sampler for combustion studies, technical note, Tampere, Finland.

Elleman, R. A., and D. S. Covert (2009a), Aerosol size distribution modeling with the Community Multiscale Air Quality modeling system in the Pacific Northwest: 1. Model comparison to observations, *J. Geophys. Res.*, *114*, D11206, doi:10.1029/2008JD010791.

Elleman, R. A., and D. S. Covert (2009b), Aerosol size distribution modeling with the Community Multiscale Air Quality modeling system in the Pacific Northwest: 2. Parameterizations for ternary nucleation and nucleation mode processes, *J. Geophys. Res.*, *114*, D11207, doi:10.1029/2009JD012187.

Fan, J., R. Zhang, D. Collins, and G. Li (2006), Contribution of secondary condensable organics to new particle formation: A case study in Houston, Texas, *Geophys. Res. Lett.*, *33*, L15802, doi:10.1029/2006GL026295.

Frick, G. M., and W. A. Hoppel (2000), Airship measurements of ship's exhaust plumes and their effect on marine boundary layer clouds, *J. Atmos. Sci.*, *57*, 2625–2648, doi:10.1175/1520-0469(2000)057<2625:AMOSSE>2.0.CO;2.

Fushimi, A., S. Hasegawa, K. Takahashi, Y. Fujitani, K. Tanabe, and S. Kobayashi (2008), Atmospheric fate of nuclei-mode particles estimated from the number concentrations and chemical composition of particles measured at roadside and background sites, *Atmos. Environ.*, *42*, 949–959, doi:10.1016/j.atmosenv.2007.10.019.

Gidhagen, L., C. Johansson, J. Langner, and G. Olivares (2004), Simulation of NO_x and ultrafine particles in a street canyon in Stockholm, Sweden, *Atmos. Environ.*, *38*, 2029–2044, doi:10.1016/j.atmosenv.2004.02.014.

Gidhagen, L., C. Johansson, J. Langner, and V. L. Foltescu (2005), Urban scale modeling of particle number concentration in Stockholm, *Atmos. Environ.*, *39*, 1711–1725.

Gramotnev, D. K., and G. Gramotnev (2005), Modeling of aerosol dispersion from a busy road in the presence of nanoparticle fragmentation, *J. Appl. Meteorol.*, *44*, 888–899, doi:10.1175/JAM2238.1.

Harris, S. J., and M. M. Maricq (2002), The role of fragmentation in defining the signature size distribution of diesel soot, *J. Aerosol Sci.*, *33*, 935–942, doi:10.1016/S0021-8502(02)00045-9.

Heim, M., G. Kasper, G. P. Reischl, and C. Gerhart (2004), Performance of a new commercial electrical mobility spectrometer, *Aerosol Sci. Technol.*, *38*(12), suppl. 2, 3–14, doi:10.1080/02786820490519252.

Held, T., D. P. Y. Chang, and D. A. Niemeier (2003), UCD 2001: An improved model to simulate pollutant dispersion from roadways, *Atmos. Environ.*, *37*, 5325–5336, doi:10.1016/j.atmosenv.2003.09.015.

Hewitt, C. N. (2001), The atmospheric chemistry of sulphur and nitrogen in power station plumes, *Atmos. Environ.*, *35*, 1155–1170, doi:10.1016/S1352-2310(00)00463-5.

Hobbs, P. V., M. W. Eltgroth, D. A. Hegg, and L. F. Radke (1983), Particle formation and growth in power plant plumes. Volume 1: Field observations and theoretical studies of the evolution of particles in the plumes from coal-fired electric power plants, *EA-3105 Res. Proj. 330-1*, 109 pp., Electr. Pow. Res. Inst., Palo Alto, Calif.

Hobbs, P. V., et al. (2000), Emissions from ships with respect to their effects on clouds, *J. Atmos. Sci.*, *57*, 2570–2590, doi:10.1175/1520-0469(2000)057<2570:EFSWRT>2.0.CO;2.

Hogrefe, O., G. G. Lala, B. P. Frank, J. J. Schwab, and K. L. Demerjian (2006), Field evaluation of a TSI 3034 Scanning Mobility Particle Sizer in New York City: Winter 2004 intensive campaign, *Aerosol Sci. Technol.*, *40*, 753–762, doi:10.1080/02786820600721846.

Hussein, T., A. Puustinen, P. P. Aalto, J. M. Mäkelä, K. Hämeri, and M. Kulmala (2004), Urban aerosol number size distributions, *Atmos. Chem. Phys.*, *3*, 391–411.

Hussein, T., K. Hämeri, P. P. Aalto, P. Paatero, and M. Kulmala (2005), Modal structure and spatial-temporal variations of urban and suburban aerosols in Helsinki—Finland, *Atmos. Environ.*, *39*, 1655–1668, doi:10.1016/j.atmosenv.2004.11.031.

Jacobson, M. Z., and J. H. Seinfeld (2004), Evolution of nanoparticle size and mixing state near the point of emission, *Atmos. Environ.*, *38*, 1839–1850, doi:10.1016/j.atmosenv.2004.01.014.

Jacobson, M. Z., D. B. Kittelson, and W. F. Watts (2005), Enhanced coagulation due to evaporation and its effect on nanoparticle evolution, *Environ. Sci. Technol.*, *39*, 9486–9492, doi:10.1021/es0500299.

Janhäll, S., K. F. G. Olofson, P. U. Andersson, J. B. C. Pettersson, and M. Hallquist (2006), Evolution of the urban aerosol during winter temperature inversion episodes, *Atmos. Environ.*, *40*, 5355–5366, doi:10.1016/j.atmosenv.2006.04.051.

Karamchandani, P., C. Seigneur, K. Vijayaraghavan, and S.-Y. Wu (2002), Development and application of a state-of-the-science plume-in-grid model, *J. Geophys. Res.*, *107*(D19), 4403, doi:10.1029/2002JD002123.

Kerminen, V.-M., et al. (2007), Development of particle number size distribution near a major road in Helsinki during an episodic inversion situation, *Atmos. Environ.*, *41*, 1759–1767, doi:10.1016/j.atmosenv.2006.10.026.

Ketzel, M., and R. Berkowicz (2004), Modelling the fate of ultrafine particles from exhaust pipe to rural background: An analysis of time scales for dilution, coagulation, and deposition, *Atmos. Environ.*, *38*, 2639–2652, doi:10.1016/j.atmosenv.2004.02.020.

Ketzel, M., P. Wåhlin, A. Kristensson, E. Swietlicki, R. Berkowicz, and F. Palmgren (2003), Particle and trace gas emission factors under urban driving conditions in Copenhagen based on street and roof-level observations, *Atmos. Environ.*, *37*, 2735–2749, doi:10.1016/S1352-2310(03)00245-0.

Ketzel, M., P. Wåhlin, A. Kristensson, E. Swietlicki, R. Berkowicz, O. J. Nielsen, and F. Palmgren (2004), Particle size distribution and particle mass measurements at urban, near-city and rural level in the Copenhagen area and southern Sweden, *Atmos. Chem. Phys.*, *4*, 281–292.

Kikas, Ü., A. Mirme, E. Tamm, and T. Raunemaa (1996), Statistical characteristics of aerosol in Baltic Sea region, *J. Geophys. Res.*, *101*, 19,319–19,327, doi:10.1029/95JD03758.

Kim, S., S. Shen, C. Sioutas, Y. Zhu, and W. C. Hinds (2002), Size distribution and diurnal and seasonal trends of ultrafine particles in source and receptor sites of the Los Angeles Basin, *J. Air Waste Manage. Assoc.*, *52*, 297–307.

Kittelson, D. B. (1998), Engines and nanoparticles: A review, *J. Aerosol Sci.*, *29*, 575–588, doi:10.1016/S0021-8502(97)10037-4.

Laakso, L., T. Hussein, P. Aarnio, M. Komppula, V. Hiltunen, Y. Viisanen, and M. Kulmala (2003), Diurnal and annual characteristics of particle mass and number concentrations in urban, rural and Arctic environments in Finland, *Atmos. Environ.*, *37*, 2629–2641, doi:10.1016/S1352-2310(03)00206-1.

Larson, T. V., D. S. Covert, E. Kim, R. Elleman, A. B. Schreuder, and T. Lumley (2006), Combining size distribution and chemical species measurements into a multivariate receptor model of $PM_{2.5}$, *J. Geophys. Res.*, *111*, D10S09, doi:10.1029/2005JD006285.

Lipsky, E. M., N. J. Pekney, G. F. Walbert, W. J. O'Dowd, M. C. Freeman, and A. Robinson (2004), Effects of dilution sampling on fine particle emissions from pulverized coal combustion, *Aerosol Sci. Technol.*, *38*, 574–587, doi:10.1080/02786820490479851.

Lu, G., J. R. Brook, M. R. Alfarra, K. Anlauf, W. R. Leaitch, S. Sharma, D. Wang, D. R. Worsnop, and L. Phinney (2006), Identification and characterization of inland ship plumes over Vancouver, BC, *Atmos. Environ.*, *40*, 2767–2782, doi:10.1016/j.atmosenv.2005.12.054.

McMurry, P. H., and K. S. Woo (2002), Size distributions of 3–100-nm urban Atlanta aerosols: Measurement and observations, *J. Aerosol Med.*, *15*, 169–178, doi:10.1089/089426802320282293.

McMurry, P. H., D. J. Rader, and J. L. Stith (1981), Studies of aerosol formation in power plant plumes—I. Growth laws for secondary aerosols in power plant plumes: Implications for chemical conversion mechanisms, *Atmos. Environ.*, *15*, 2315–2327, doi:10.1016/0004-6981(81)90262-6.

Minoura, H., and H. Takekawa (2005), Observation of number concentrations of atmospheric aerosols and analysis of nanoparticle behavior at an urban background area in Japan, *Atmos. Environ.*, *39*, 5806–5816, doi:10.1016/j.atmosenv.2005.06.033.

Morawska, L., S. Thomas, N. Bofinger, D. Wainwright, and D. Neale (1998), Comprehensive characterization of aerosols in a subtropical urban atmosphere: Particle size distribution and correlation with gaseous pollutants, *Atmos. Environ.*, *32*, 2467–2478, doi:10.1016/S1352-2310(98)00023-5.

Morawska, L., S. Thomas, M. Jamriska, and G. Johnson (1999), The modality of particle size distributions of environmental aerosols, *Atmos. Environ.*, *33*, 4401–4411, doi:10.1016/S1352-2310(99)00217-4.

Mueller, S. F., and R. E. Imhoff (1994), Estimates of particle formation and growth in coal-fired boiler exhaust—I. Observations, *Atmos. Environ.*, *28*, 595–602, doi:10.1016/1352-2310(94)90035-3.

Park, S.-K., A. Marmur, S. B. Kim, D. Tian, Y. Hu, P. H. McMurry, and A. G. Russell (2006), Evaluation of fine particle number concentrations in CMAQ, *Aerosol Sci. Technol.*, *40*, 985–996, doi:10.1080/02786820600907353.

Putaud, J.-P., et al. (2003), A European aerosol phenomenology: Physical and chemical characteristics of particulate matter at kerbside, urban, rural, and background sites in Europe, *Rep. 20411EN*, Eur. Comm. Jt. Res. Cent., Ispra, Italy.

Radke, L. F., J. A. Coakley Jr., and M. D. King (1989), Direct and remote sensing observations of the effects of ships on clouds, *Science*, *246*, 1146–1149, doi:10.1126/science.246.4934.1146.

Rhoads, K. P., D. J. Phares, A. S. Wexler, and M. V. Johnston (2003), Size-resolved ultrafine particle composition analysis: 1. Atlanta, *J. Geophys. Res.*, *108*(D7), 8418, doi:10.1029/2001JD001211.

Ruuskanen, J., et al. (2001), Concentrations of ultrafine, fine, and $PM_{2.5}$ particles in three European cities, *Atmos. Environ.*, *35*, 3729–3738, doi:10.1016/S1352-2310(00)00373-3.

Sardar, S. B., P. M. Fine, and C. Sioutas (2005), Seasonal and spatial variability of the size-resolved chemical composition of particulate matter (PM_{10}) in the Los Angeles Basin, *J. Geophys. Res.*, *110*, D07S08, doi:10.1029/2004JD004627.

Shi, J. P., A. A. Khan, and R. M. Harrison (1999), Measurements of ultrafine particle concentration and size distribution in the urban atmosphere, *Sci. Total Environ.*, *235*, 51–64, doi:10.1016/S0048-9697(99)00189-8.

Sinha, P., P. V. Hobbs, R. J. Yokelson, T. J. Christian, T. W. Kirchstetter, and R. Bruintjes (2003), Emissions of trace gases and particles from two ships in the southern Atlantic Ocean, *Atmos. Environ.*, *37*, 2139–2148, doi:10.1016/S1352-2310(03)00080-3.

Stanier, C. O., A. Y. Khlystov, and S. N. Pandis (2004), Ambient aerosol size distributions and number concentrations measured during the Pittsburgh Air Quality Study (PAQS), *Atmos. Environ.*, *38*, 3275–3284, doi:10.1016/j.atmosenv.2004.03.020.

Sturm, P. J., et al. (2003), Roadside measurements of particulate matter size distribution, *Atmos. Environ.*, *37*, 5273–5281, doi:10.1016/j.atmosenv.2003.05.006.

Teinemaa, E., U. Kirso, M. R. Strommen, and R. M. Kamens (2002), Atmospheric behavior of oil-shale combustion fly ash in a chamber study, *Atmos. Environ.*, *36*, 813–824, doi:10.1016/S1352-2310(01)00520-9.

Tolocka, M. P., D. A. Lake, and M. V. Johnston (2005), Size-resolved fine and ultrafine particle composition in Baltimore, Maryland, *J. Geophys. Res.*, *110*, D07S04, doi:10.1029/2004JD004573.

Trier, A. (1997), Submicron particles in an urban atmosphere: A study of optical size distributions—I, *Atmos. Environ.*, *31*, 909–914, doi:10.1016/S1352-2310(96)00271-3.

Tuch, T., P. Brand, H. E. Wichmann, and J. Heyder (1997), Variation of particle number and mass concentration in various size ranges of ambient aerosols in Eastern Germany, *Atmos. Environ.*, *31*, 4193–4197, doi:10.1016/S1352-2310(97)00260-4.

Tuch, T. M., B. Wehner, M. Pitz, J. Cyrys, J. Heinrich, W. G. Kreyling, H. E. Wichmann, and A. Wiedensohler (2003), Long-term measurements of size-segregated ambient aerosol in two German cities located 100 km apart, *Atmos. Environ.*, *37*, 4687–4700, doi:10.1016/j.atmosenv.2003.07.010.

Venkataraman, C., J. M. Lyons, and S. K. Friedlander (1994), Size distributions of polycyclic aromatic hydrocarbons and elemental carbon. 1. Sampling, measurement methods, and source characterization, *Environ. Sci. Technol.*, *28*, 555–562, doi:10.1021/es00053a005.

Volkamer, R., J. L. Jimenez, F. San Martini, K. Dzepina, Q. Zhang, D. Salcedo, L. T. Molina, D. R. Worsnop, and M. J. Molina (2006), Secondary organic aerosol from anthropogenic air pollution: Rapid and higher than expected, *Geophys. Res. Lett.*, *33*, L17811, doi:10.1029/2006GL026899.

Wåhlin, P., F. Palmgren, and R. Van Dingenen (2001), Experimental studies of ultrafine particles in streets and the relationship to traffic, *Atmos. Environ.*, *35*, suppl. 1, 63–69, doi:10.1016/S1352-2310(00)00500-8.

Wehner, B., and A. Wiedensohler (2002), Long term measurements of submicrometer urban aerosols: Statistical analysis for correlations with meteorological conditions and trace gases, *Atmos. Chem. Phys.*, *2*, 1699–1733.

Wehner, B., W. Birmili, T. Gnauk, and A. Wiedensohler (2002), Particle number size distributions in a street canyon and their transformation into the urban-air background: Measurements and a simple model study, *Atmos. Environ.*, *36*, 2215–2223, doi:10.1016/S1352-2310(02)00174-7.

Whitby, K. T. (1978), The physical characteristics of sulfur aerosols, *Atmos. Environ.*, *12*, 135–159, doi:10.1016/0004-6981(78)90196-8.

Whitby, K. T., B. Y. H. Liu, R. B. Husar, and N. J. Barsic (1972), The Minnesota Aerosol-Analyzing System used in the Los Angeles Smog Project, in *Aerosols and Atmospheric Chemistry*, edited by G. M. Hidy, pp. 189–217, Academic, New York.

Wiedensohler, A., B. Wehner, and W. Birmili (2002), Aerosol number concentrations and size distributions at mountain-rural, urban-influenced rural, and urban-background sites in Germany, *J. Aerosol Med.*, *15*, 237–243, doi:10.1089/089426802320282365.

Wilson, J. C., and P. H. McMurry (1981), Studies of aerosol formation in power plant plumes—II. Secondary aerosol formation in the Navajo Generating Station plume, *Atmos. Environ.*, *15*, 2329–2339, doi:10.1016/0004-6981(81)90263-8.

Woo, K. S., D. R. Chen, D. Y. H. Pui, and P. H. McMurry (2001), Measurement of Atlanta aerosol size distributions: Observations of ultrafine particle events, *Aerosol Sci. Technol.*, *34*, 75–87, doi:10.1080/027868201300082049.

Yao, X., N. T. Lau, M. Fang, and C. K. Chan (2006), Use of stationary and mobile measurements to study power plant emissions, *J. Air Waste Manage. Assoc.*, *56*, 144–151.

Ylatalo, S. (2006), Studies on aerosol particle emission, sampling, and authenticity, Ph.D. thesis, 69 pp., Univ. of Helsinki, Helsinki.

Zhang, K. M., and A. S. Wexler (2004), Evolution of particle number distribution near roadways—Part I: Analysis of aerosol dynamics and its implications for engine emission measurement, *Atmos. Environ.*, *38*, 6643–6653, doi:10.1016/j.atmosenv.2004.06.043.

Zhang, K. M., A. S. Wexler, Y. F. Zhu, W. C. Hinds, and C. Sioutas (2004), Evolution of particle number distribution near roadways—Part II: The 'Road-to-Ambient' process, *Atmos. Environ.*, *38*, 6655–6665, doi:10.1016/j.atmosenv.2004.06.044.

Zhang, K. M., A. S. Wexler, D. A. Niemeier, Y. F. Zhu, W. C. Hinds, and C. Sioutas (2005), Evolution of particle number distribution near roadways. Part III: Traffic analysis and on-road size resolved particulate emission factors, *Atmos. Environ.*, *39*, 4155–4166, doi:10.1016/j.atmosenv.2005.04.003.

Zhang, Q., M. R. Canagaratna, J. T. Jayne, D. R. Worsnop, and J.-L. Jimenez (2005), Time- and size-resolved chemical composition of submicron particles in Pittsburgh: Implications for aerosol sources and processes, *J. Geophys. Res.*, *110*, D07S09, doi:10.1029/2004JD004649.

Zhou, L., P. K. Hopke, C. O. Stanier, S. N. Pandis, J. M. Ondov, and J. P. Pancras (2005a), Investigation of the relationship between chemical composition and size distribution of airborne particles by partial least squares and positive matrix factorization, *J. Geophys. Res.*, *110*, D07S18, doi:10.1029/2004JD005050.

Zhou, L., E. Kim, P. K. Hopke, C. Stanier, and S. N. Pandis (2005b), Mining airborne particulate size distribution data by positive matrix factorization, *J. Geophys. Res.*, *110*, D07S19, doi:10.1029/2004JD004707.

Zhu, Y., W. C. Hinds, S. Kim, S. Shen, and S. Sioutas (2002), Study of ultrafine particles near a major highway with heavy-duty diesel traffic, *Atmos. Environ.*, *36*, 4323–4335, doi:10.1016/S1352-2310(02)00354-0.

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