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Sources of ultrafine particles in the Eastern United States

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HIGHLIGHTS

- New size and source-resolved particle number emissions for Eastern US.
- Gasoline cars are the most important source of primary particle number.
- Contributions of industrial sources, on- and non-road diesel are significant.
- Results are consistent with limited available source-attribution studies.

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ABSTRACT

Source contributions to ultrafine particle number concentrations for a summertime period in the Eastern U.S. are investigated using the chemical transport model PMCAMx-UF. New source-resolved number emissions inventories are developed for biomass burning, dust, gasoline automobiles, industrial sources, non-road and on-road diesel. According to the inventory for this summertime period in the Eastern U.S., gasoline automobiles are responsible for 40% of the ultrafine particle number emissions, followed by industrial sources (33%), non-road diesel (16%), on-road diesel (10%), and 1% from biomass burning and dust. With these emissions as input, the chemical transport model PMCAMx-UF reproduces observed ultrafine particle number concentrations ($N_{3\text{-}100}$) in Pittsburgh with an error of 12%. For this summertime period in the Eastern U.S., nucleation is predicted to be the source of more than 90% of the total particle number concentrations. The source contributions to primary particle number concentrations are on average similar to those of their source emissions contributions: gasoline is predicted to contribute 36% of the total particle number concentrations, followed by industrial sources (31%), non-road diesel (18%), on-road diesel (10%), biomass burning (1%), and long-range transport (4%). For this summertime period in Pittsburgh, number source apportionment predictions for particles larger than 3 nm in diameter (traffic 65%, other combustion sources 35%) are consistent with measurement-based source apportionment (traffic 60%, combustion sources 40%).

1. Introduction

Ultrafine particles (less than 100 nm in diameter) play a role in climate change and may be relevant to human health. Since these smaller particles typically dominate aerosol number distributions, they can greatly contribute to the number of cloud condensation nuclei (CCN) through growth by condensation (Pierce and Adams, 2009). CCN affect the albedo (reflectance) of a cloud, with greater numbers increasing the cloud's albedo (Seinfeld and Pandis, 2006). The change in reflectance of sunlight off clouds during the last two centuries (known as the aerosol indirect effect) is currently the most uncertain factor in the global radiative balance of the earth (IPCC, 2007). In addition, multiple studies highlighted in the review by Delfino et al. (2005) indicate a possible link between exposure to ultrafine particles and cardiovascular disease in humans.

Most previous work on particle source apportionment has focused on mass concentrations, both using observations (Zhou et al., 2005; Aiken et al., 2009; Heo et al., 2009) and models (Marmur et al., 2006; Lane et al., 2007; Wang et al., 2009). Since ultrafine particles dominate number concentrations in polluted areas but contribute little to mass, mass-based source

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Table 1
Mass boundary conditions for July 2001 in PMCAMx-UF.

Modeled chemical species	Boundary concentration ($\mu g\ m^{-3}$)
Side boundary conditions	
Organic aerosol	0.80
Primary elemental carbon	0.10
Ammonium	0.37
Nitrate	0.10
Sulfate	0.90
Sodium ion [eastern boundary, only]	0.39
Chloride [eastern boundary, only]	0.61
Top boundary conditions	
Organic aerosol	0.50
Ammonium	0.14
Sulfate	0.36

apportionment does not accurately describe the sources of particles in the ultrafine range. In this study, N_x denotes the number concentration of particles with diameters greater than x nm, e.g. N_3 is the number concentration of particles with diameters larger than 3 nm. $N_{x\text{-}y}$ denotes the number concentration of particles between x and y nm in diameter, e.g. $N_{0.8\text{-}3}$ is the number concentration of particles with diameters between 0.8 and 3 nm. There has been some observation-based work that has focused on particle number source apportionment of N_{13} in Spain (Pey et al., 2009) and N_3 in Pittsburgh (Zhou et al., 2004). For July 2001, Zhou et al. (2004) used advanced factor analysis and size distribution measurements to estimate that 58% of primary N_3 came from local traffic, 29% from stationary combustion, and 13% was related to remote combustion sources and secondary PM.

Study of the sources of ultrafine particles using a chemical transport model (CTM) requires accurate simulation of the two main pathways through which these particles enter the atmosphere: nucleation and direct emissions. The contribution of nucleation to particle number concentrations has been explored in several studies (Spracklen et al., 2006; Elleman and Covert, 2009; Jung et al., 2010). Other studies have broadly investigated the accurate representation of number emissions and their contribution to particle number concentrations. Spracklen et al. (2006) considered the contribution of primary emissions to global particle number concentrations. Elleman and Covert (2010) applied source-specific number distributions to three general sources in order to improve model-measurement agreement for number concentrations. In this study, we both develop a detailed number emissions inventory for six source types, each with a source-specific number size distribution derived from the literature, and then investigate the contribution of each source to particle number concentrations in several size ranges.

Previous work using PMCAMx-UF for Pittsburgh (Jung et al., 2008) and the Eastern U.S. (Jung et al., 2010) focused on nucleation as a source of particle number concentrations. The authors compared the results of using several nucleation rate parameterizations in a box model to observations in Pittsburgh for July 2001, and the ternary sulfuric acid-water-ammonia nucleation rate parameterization of Napari et al. (2002) was found to correctly reproduce the lack or occurrence of nucleation on all days tested (Jung et al., 2008). Since this nucleation rate parameterization tended to overpredict particle number concentrations in Pittsburgh, however, the authors scaled the rate parameterization using a 10^{-5} nucleation tuner and implemented it in the nucleation routine of PMCAMx-UF (Jung et al., 2010). Since this previous application of PMCAMx-UF was focused on nucleation, the authors did not develop a source-resolved number emissions inventory; instead, they assumed a single source number distribution for each particulate species. For all primary carbonaceous emissions, the authors used a traffic number distribution based on the measurements of Stanier et al. (2004) taken during the Pittsburgh Air Quality Study (PAQS). All other emitted species were given size distributions recommended by AeroCom (Dentener et al., 2006). In order to perform a detailed study of the sources of emitted particle number, it is necessary to develop a source-resolved number emissions inventory with careful attention to individual source number distributions.

We first describe the development of a new source-resolved number emissions inventory for a summertime period in the Eastern U.S. and then introduce a number-focused zero-out method for source apportionment of ultrafine particle number concentrations. We evaluate the performance of the model against measurements in Pittsburgh and then investigate the source contributions to ultrafine particle number concentrations over the Eastern U.S. Finally, the sensitivity of our results to the nucleation rate parameterization is investigated.

Table 2
Number size distributions for the boundary and initial conditions in PMCAMx-UF.

	Aitken mode			Accumulation mode			Coarse mode		
	Number (cm^{-3})	Mean diameter (μm)	σ	Number (cm^{-3})	Mean diameter (μm)	σ	Number (cm^{-3})	Mean diameter (μm)	σ
Initial, northern, southern, and western	1600	0.020	1.45	580	0.12	1.65	0.10	1.80	2.40
Eastern	133	0.008	4.54	66.6	0.27	1.62	1.55	0.58	2.49
All above 6 km	129	0.007	4.42	59.7	0.25	1.79	–	–	–

Table 3
Average source emissions composition over the Eastern U.S. for July 2001.

	PM_{10} composition (% mass)					
	Biomass	Dust	Gasoline	Industrial	Non-road diesel	On-road diesel
Elemental carbon	20	0	9	9	31	18
Organic aerosol	44	6	45	30	33	11
Crustal	36	94	46	5	36	71
Nitrate	0	0	0	0	0	0
Sulfate	0	0	0	56	0	0