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Source contributions to primary organic aerosol: Comparison of the results of a source-resolved model and the chemical mass balance approach

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Abstract

A source-resolved model has been developed to predict the contribution of different sources to primary organic aerosol concentrations. The model was applied to the eastern US during a 17 day pollution episode beginning on 12 July 2001. Primary organic matter (OM) and elemental carbon (EC) concentrations are tracked for eight different sources: gasoline vehicles, non-road diesel vehicles, on-road diesel vehicles, biomass burning, wood burning, natural gas combustion, road dust, and all other sources. Individual emission inventories are developed for each source and a three-dimensional chemical transport model (PMCAMx) is used to predict the primary OM and EC concentrations from each source. The source-resolved model is simple to implement and is faster than existing source-oriented models. The results of the source-resolved model are compared to the results of chemical mass balance models (CMB) for Pittsburgh and multiple urban/rural sites from the Southeastern Aerosol Research and Characterization (SEARCH) network. Significant discrepancies exist between the source-resolved model and the CMB model predictions for some of the sources. There is strong evidence that the organic PM emissions from natural gas combustion are overestimated. It also appears that the OM and EC emissions from wood burning and off-road diesel are too high in the Northeastern US. Other similarities and discrepancies between the source-resolved model and the CMB model for primary OM and EC are discussed along with problems in the current emission inventory for certain sources.

Keywords: Source apportionment; Organic matter; Elemental carbon; PMCAMx; Eastern US

1. Introduction

Atmospheric particles with diameters less than 2.5 μm ($PM_{2.5}$) have adverse effects on human health and visibility and contribute to global climate changes by scattering light and serving as cloud

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condensation nuclei. To limit their effects on human health and visibility, the United States Environmental Protection Agency (US EPA) has set daily and annual average National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$. According to the US EPA, all or part of 225 counties nationwide, many of which are in the eastern US, are not in attainment of the standards.

$PM_{2.5}$ is comprised of sulfate, nitrate, ammonia, organic matter (OM), elemental carbon (EC), crustal species, and other compounds. OM represents a major fraction of the total $PM_{2.5}$ concentrations across the United States. OM has both primary and secondary components while EC is only emitted from sources. Primary OM is emitted from various sources such as gasoline and diesel vehicles, biomass burning, industrial sources, and other forms of combustion. Dominant sources of EC are on-road and off-road diesel vehicles and biomass burning. Implementation of effective organic $PM_{2.5}$ control strategies requires the quantification of the contribution of each source to the ambient OM and EC concentrations.

Chemical mass balance (CMB) receptor modeling methods can determine the source contributions to primary OM concentrations using organic compounds as tracers (Rogge et al., 1993a; Schauer et al., 1996). In the CMB method, the total concentration of each organic tracer in the ambient sample is reconstructed from a linear combination of the source emissions profiles (Watson et al., 1990). CMB receptor models have been used to determine the source apportionment of $PM_{2.5}$ primary OM in Los Angeles (Schauer et al., 1996), the San Joaquin Valley (Schauer and Cass, 2000), the southeastern US (Zheng et al., 2002), and Pittsburgh (Subramanian et al., 2006, 2007; Robinson et al., 2006a–c). The results of these CMB applications suggest that gasoline and diesel vehicles along with wood burning are the major primary OM emission sources in the investigated areas.

A second approach used for source apportionment is source-oriented modeling. Unlike receptor-oriented modeling, source-oriented models predict the pollutant concentrations by using a regional chemical transport model and emissions data as input. Source-oriented models separately track PM emissions from different source categories in the model instead of combining them into a single species. Both a 1D Lagrangian and a 3D Eulerian source-oriented model have been applied to Los Angeles and the San Joaquin Valley (Kleeman et al., 1997, 1999; Kleeman and Cass, 1998, 2001; Held et al., 2004) predicting the source contribution to the size and composition distribution of PM_{10}. In the 3D Eulerian model, Kleeman and Cass (2001) separately tracked the particles emitted by 10 different source classes. Instead of one primary OM species Kleeman and Cass (2001) used 10 different primary OM species, one for each source class. Although source-oriented models are numerically accurate, the corresponding simulations require considerable computational resources. The required CPU time can be reduced significantly if a source-resolved model is used. These models still simulate only one primary OM and one EC species corresponding to a single source, so the total CPU time is linearly proportional to the number of sources modeled. A major disadvantage of source-resolved models is that they can describe only primary aerosol species that do not interact with the rest of the components of the particles.

The current study applies a source-resolved three-dimensional chemical transport model to the eastern US in order to critically evaluate the primary OM and EC emission inventories. The overall performance of PMCAMx for the July 2001 episode using the current emission inventory has been evaluated by Gaydos et al. (2007). PMCAMx is used to predict contributions of eight different source categories to primary OM and EC: on-road diesel, off-road diesel, gasoline engines, natural gas, wood burning, biomass burning, dust, and all other sources. The primary organic aerosol from each source category is modeled individually by splitting the emissions inventories into sub-categories. By having individual source emission inventories, only one primary OM and EC species are required in the model instead of eight species for both primary OM and EC.

The predicted primary OM and EC concentrations for each source category are then compared to published CMB results at different locations (Pittsburgh, Atlanta, Birmingham, etc.). Agreement between the source-resolved inventory results and the CMB results can increase our confidence in the source apportionment for primary OM and EC. Disagreement can point to problems in individual source emission inventories.

2. Description of PMCAMx

PMCAMx is a three-dimensional chemical transport model which uses the framework of CAMx