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

Timothy E. Lanea, Robert W. Pinderb, Manish Shrivastavac, Allen L. Robinsonb,c, Spyros N. Pandisa,b,d,*

aDepartment of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA
bDepartment of Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, PA 15213, USA
cDepartment of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA
dDepartment of Chemical Engineering, University of Patras, Patra 26500, Greece

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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.

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1. Introduction

Atmospheric particles with diameters less than 2.5 μm (PM2.5) have adverse effects on human health and visibility and contribute to global climate changes by scattering light and serving as cloud
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
(Environ, 2003) to simulate horizontal and vertical advection, horizontal and vertical dispersion, wet and dry deposition, and gas-phase chemistry. Three aerosol modules have been implemented in PMCAMx to describe inorganic and organic aerosol dynamics and aerosol–cloud interactions using an operator-splitting approach (Gaydos et al., 2007). The order of the processes is: emission, horizontal advection, vertical advection, vertical dispersion, horizontal dispersion, wet deposition, gas-phase chemistry, aerosol processes (nucleation, coagulation, inorganic aerosol condensation/evaporation), secondary organic aerosol (SOA) formation, and aqueous phase-chemistry. This approach is further described in Gaydos et al. (2007). PMCAMx tracks 13 different aerosol species: sulfate, nitrate, ammonium, aerosol water content, four SOA species, sodium, chloride, primary organic aerosol, primary EC, and all primary inert material. Each aerosol species has 10 size sections, ranging from 40 nm to 40 μm.

For this study, PMCAMx is applied to a 17 day pollution episode in the eastern US starting on 12 July 2001. The modeling domain covers a 3492 × 3240 km region in the eastern US with 36 × 36 km grid resolution with 14 different levels up to 6 km (Gaydos et al., 2007). The initial and boundary conditions for primary PM$_{2.5}$ OM are 2.0 and 1.0 μg m$^{-3}$, respectively, while zero initial and boundary conditions are used for EC. Inputs to the model include horizontal wind components, temperature, pressure, water vapor, vertical diffusivity, clouds, and rainfall, all created using the meteorological model MM5 (Grell et al., 1995).

Eight source-resolved inventories are used as the emissions inputs in PMCAMx when applied to the eastern US. The primary OM and EC concentrations for each of the eight sources over the eastern US are simulated for the 17 day episode. For each source-resolved simulation, the boundary conditions for primary OM and EC are set to zero and the initial conditions are set to a very small non-zero value to avoid numerical problems (0.006 μg m$^{-3}$). There are two more cases simulated as well: the initial conditions case and the boundary conditions case. For these two cases, the emissions for primary OM and EC within the domain are set at zero. For the boundary condition case, the initial conditions were set to 0.006 μg m$^{-3}$; similarly, the boundary conditions are set to zero for the initial conditions case. The base case with entire emission inventory was also simulated so that the sum of the primary OM and EC for the sub-inventories can be compared to primary OM and EC from the inventory to evaluate the quality of the solution.

3. Emission inventories

The LADCO BaseE inventory generated using EMS-2003 (LADCO, 2003), is used to generate source-resolved inventories. Emissions are derived primarily from the U.S. EPA’s National Emission Inventory (NEI) 1999 Version 2.0 (U.S. EPA, 2002a), with the following changes: on-road transportation sources are from the U.S. EPA’s MOBILE6 (U.S. EPA, 2002b); non-road sources are from U.S. EPA’s NONROAD (U.S. EPA, 2002c). The temporal profiles for electric power utility point sources are from an analysis of continuous emission monitors (Janssen, 2003). Ammonia emissions are from the CMU ammonia emission inventory (Goebes et al., 2003; Pinder et al., 2004). Biogenic emissions are from BIOME3 (Wilkinson and Janssen, 2001). A different emission inventory is used for weekdays, Saturdays, and Sundays.

From the overall emission inventory described above, eight source-resolved emission inventories have been developed. The primary OM and EC in each source-resolved inventory consist of the carbonaceous aerosol emissions from that source, while the rest of the inventory remains the same. Table 1 lists the emission source categories and the fraction of the total emissions of EC and organic carbon that each category contributes. The sources are sorted based on source classification codes from the NEI database. The “other” source category consists of all source codes not mapped to one of the other seven source categories.

Table 1 shows the contribution of each source to the overall inventory for each day. The total emissions for the entire domain decrease on weekends; for example, the EC emissions decrease from 1.09 ktons day$^{-1}$ on a weekday to 0.89 ktons day$^{-1}$. There is also a change in the distribution of sources. For instance, the major difference between weekdays and weekends is that gasoline exhaust increases from 13% of the overall primary OM emissions to 22% of the primary OM on the weekends. Fig. 1 shows the total emissions for primary OM and EC on a weekday across the eastern US. As expected, there are higher emission rates for OM and EC in urban areas. The highest daily emission rate for OM in a cell is 19 tons day$^{-1}$ over southern New Jersey,
east of Philadelphia, and the highest emission rate for EC is 19.5 tons day\(^{-1}\) in Nashville.

### 4. Model results

#### 4.1. Base case predictions and measurements

![Fig. 1. Daily emission rates (tons day\(^{-1}\)) for primary OM and EC from the weekday emission inventory.](image)

Table 1

<table>
<thead>
<tr>
<th>Name</th>
<th>Description</th>
<th>Elemental carbon (%)(a)</th>
<th>Primary organic matter (%)(a)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>W</td>
<td>Sa</td>
</tr>
<tr>
<td>Gasoline</td>
<td>On-road, non-road, and stationary gasoline combustion</td>
<td>4</td>
<td>5</td>
</tr>
<tr>
<td>On-road diesel</td>
<td>On-road transportation diesel combustion</td>
<td>26</td>
<td>25</td>
</tr>
<tr>
<td>Off-road diesel</td>
<td>Non-road and stationary diesel combustion</td>
<td>53</td>
<td>51</td>
</tr>
<tr>
<td>Natural gas</td>
<td>Natural gas combustion</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Wood</td>
<td>Residential fireplaces, wood stoves, wood-fired boilers</td>
<td>7</td>
<td>8</td>
</tr>
<tr>
<td>Biomass</td>
<td>Open agricultural burning, land-clearing, residential yard waste</td>
<td>4</td>
<td>4</td>
</tr>
<tr>
<td>Dust</td>
<td>Soil and road dust</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Other</td>
<td>All other sources</td>
<td>6</td>
<td>7</td>
</tr>
<tr>
<td>Total</td>
<td>Total emissions for entire modeling domain (kt tons day(^{-1}))</td>
<td>1.09</td>
<td>0.93</td>
</tr>
</tbody>
</table>

\(a\)W—Weekday, Sa—Saturday, Su—Sunday.

Contribution of source categories to primary OM and EC emissions across the entire domain

The 24-hour average model predictions are compared to the measurements from the EPA Speciation Trends Network (STN) and the Interagency Monitoring of Protected Visual Environments (IMPROVE). As described by Gaydos et al. (2007), the predicted primary OM was divided by concentrations are: 13.4 \(\mu g \cdot m\(^{-3}\) over New York City for total carbonaceous aerosols, 2.9 \(\mu g \cdot m\(^{-3}\) over Arkansas for SOA, 8.3 \(\mu g \cdot m\(^{-3}\) over southern New Jersey for primary OM, and 5.4 \(\mu g \cdot m\(^{-3}\) over Boston for EC. The domain average concentrations for total carbonaceous aerosol, SOA, primary OM, and EC predicted over the eastern US are 2.01, 0.73, 1.03 and 0.25 \(\mu g \cdot m\(^{-3}\).
1.2 to convert to primary OC and the predicted SOA is divided by 1.8 to convert to secondary organic carbon. Fig. 3 shows the comparison of the predicted total carbon (TC), OC, and EC concentrations with STN and IMPROVE.

The TC predictions appear to have little bias but significant scatter, with concentrations in many locations greatly over or underpredicted. The mean bias between the predicted total carbonaceous aerosol and IMPROVE and STN measurements is 0.11 and −0.92 μg C m⁻³, respectively. The STN data in Fig. 3 are not blank corrected while the IMPROVE measurements already include this correction. Without a blank correction, the carbon measurements from the two networks are in poor agreement. A blank correction of around 1 μg m⁻³ needed to bring the STN measurements into agreement with the IMPROVE measurements (Graham, 2004), also improves the agreement between the model predictions for TC and the STN measurements.

For OC concentrations, the model tends to overpredict at low observed levels of OC and underpredicts at high observed levels of OC. Observed OC concentrations of less than 1 μg C m⁻³ are consistently overpredicted by the model. Overall, the model predictions compared to IMPROVE for OC have little bias, yet many sites
are over or underpredicted. However, the model consistently underpredicts the STN measurements. The STN sites are located in more populated areas than the IMPROVE sites and therefore generally have higher OC levels. The mean bias for the predicted OC compared to STN is $-1.8 \mu gC m^{-3}$. Even assuming a blank correction of around $1 \mu g m^{-3}$, the STN measurements for OC are still underpredicted by the model.

The predicted EC concentrations are about 3 times higher than the STN observed EC concentrations (Fig. 3) and have little bias when compared to the IMPROVE measurements. This suggests that the EC emissions in cities may be overestimated. For example, the model predicts over $5 \mu g m^{-3}$ of EC in New York City which is significantly greater than $0.2–1.5 \mu g m^{-3}$ measured at STN sites in Queens and the Bronx. The mean bias of the model compared to the IMPROVE and STN measurements is 0.17 and 0.88 $\mu g m^{-3}$, respectively (Gaydos et al., 2007). There is better agreement with the IMPROVE measurements because the inventories for most of the major sources of carbonaceous material were constructed using the TOR protocol (Bhave, 2004; Gaydos et al., 2007). The STN measurements use the NIOSH TOT protocol for measuring EC and OC (NIOSH, 1999); while the IMPROVE measurements used the TOR protocol to analyze the filters (Chow et al., 1993). The consistent model overprediction probably indicates problems with the EC emission inventory. Although the predicted primary OC and EC concentrations do not match the observed concentrations, the source-resolved model will help to predict which sources contribute the most to primary OC and EC and will also provide insight into which parts of the inventory need improvement.

4.2. Source-resolved mass balance test

The sum of the predicted primary OM and EC concentrations from each source-resolved simulation

Fig. 3. Comparison of predicted PM$_{2.5}$ total carbon, organic carbon (primary OM divide by 1.2 and SOA divided by 1.8), and elemental carbon concentrations to STN and IMPROVE measurements. Dashed lines represent ±30%.
should equal the primary OM and EC concentration predicted in the base case simulation. For the full simulation, the sum of the primary OM from each source category is on average within 0.001 μg m$^{-3}$ (or 0.6%) of the predicted base case primary OM over the whole domain. The maximum absolute error and relative error of any grid cell at any time step is 0.78 μg m$^{-3}$ (relative error of 10% over New York City) and 24% (absolute error of $10^{-6}$ μg m$^{-3}$ over the Atlantic Ocean north of Miami), respectively.

On average, the sum of the EC concentrations from each source is also within 0.001 μg m$^{-3}$ of the base case EC concentration over the whole domain. Although the absolute error is very small, the relative error can be large in remote areas where the EC concentration is close to zero (e.g., over the Atlantic ocean).

These differences are small for all practical purposes and are due to numerical issues related to the transport and removal algorithms used in PMCAMx. Overall, the source-resolved method works well in predicting the source contributions for primary OM.

### 4.3. Source-resolved OM

In order to correctly predict the primary OM concentration for each source, the primary OM concentrations predicted from the boundary and initial condition cases must be factored into the source-resolved predictions for each source. Fig. 4 shows the average contributions of the initial and boundary conditions for the last 13 days of the episode for the primary OM concentrations. The boundary conditions have a major impact on areas with low OM emissions, which are located over the oceans and along the boundaries in the Northern Plains and Canada. In these areas, the predicted average OM concentrations are around 1 μg m$^{-3}$ with the boundary condition contributing almost 100% of the primary OM. Away from the boundary in the eastern US, the boundary conditions contribute between 0.1 and 0.6 μg m$^{-3}$ (10–20% in urban areas and closer to 30% in rural areas) to the total primary OM concentrations. Excluding the initial four day spin up, the initial conditions on average only contribute 8% of the total primary OM in the eastern US. Over Arkansas, northern and eastern Louisiana, northeast Texas, and southern Missouri, the initial conditions contribute around 0.25 μg m$^{-3}$ (less than 30%) to the primary OM concentrations. The initial conditions have little impact on the primary OM in other areas of the eastern US after the first four simulation days.

The OM contributions from the aerosol that pre-existed in the model domain before the beginning of the simulation (initial conditions) and from sources outside the modeling domain (boundary conditions) are split into the eight source categories by assuming the same source distribution of the weekday inventory. For example, in areas where the boundary condition contribution is close to 100% of the total primary OM, the source contributions to the

**Fig. 4.** Predicted average ground-level PM$_{2.5}$ primary OM concentrations (μg m$^{-3}$) from the boundary and initial conditions for the eastern US between 16 July and 28 July 2001.
primary OM are: gasoline 13%, on-road diesel 7%, off-road diesel 14%, natural gas 23%, wood 15%, biomass 6%, dust 4%, and other 18%. The total contribution of each source is calculated by combining concentrations predicted using the source-resolved inventory plus the contribution of the boundary and initial condition concentrations.

As expected, the domain average contributions are similar to the emissions shown in Table 1; however, there are temporal and spatial variations in the contribution of each source category. Temporal variations during the 17 day pollution episode for Pittsburgh, PA are shown in Figs. 5 and 6. Fig. 5 shows that peak primary OM concentrations occur during morning rush hour and then decrease to a minimum in the early afternoon before the evening traffic increases the OM concentrations. This cycle can be seen in all urban areas across the whole domain. In Pittsburgh, the predicted peak hourly primary OM concentration is 5.3 µg m⁻³, whereas the low is 0.9 µg m⁻³. The average predicted primary OM concentration for Pittsburgh during the episode is 2.6 µg m⁻³. The day-to-day variability in the 24-hour average source contributions for Pittsburgh are shown in Fig. 6. The small fluctuations in the source contributions...
for Pittsburgh are due to the change in wind patterns, temperature, and emission inventories from weekdays to weekends. Across the entire domain, predicted temporal variations are not significant because of the small temporal variability of the inventory. Therefore, the focus in the rest of this study will be on the average contribution of each source and its spatial variations.

Fig. 7 shows the spatially resolved 13 day average (the first four days are not included in the average) contribution of each source category to the total primary \( \text{PM}_{2.5} \) OM concentrations. The scale is the same for each source to allow for comparisons of relative contributions of the different categories. The source that surprisingly contributes the most primary OM in the eastern US according to the inventory is the natural gas category. Diesel exhaust has a significant impact on primary OM concentrations as well. Overall, diesel exhaust is predicted to contribute an average of 20\% of the total primary OM in the eastern US. Non-road diesel contributes more primary OM to the Midwest and North Plains associated with farming activities. There are also high contributions of non-road diesel (20–25\%) to primary OM along the Mississippi river caused by river boats. Non-road diesel also contributes significant amounts of primary OM in eastern cities (Fig. 7). On-road diesel contributes mostly to urban primary OM.

Gasoline exhaust, similar to on-road diesel exhaust, contributes large amounts of primary OM in cities across the eastern US and is more of an urban problem (Figs. 7 and 8). Wood burning is predicted to contribute a domain average of 15\% of the total primary OM, with most of the primary OM from wood burning located in the North, especially the Northeast. Biomass burning according to at least to the inventory only has a significant impact in Texas with a relatively small contribution (~6\%) over the rest of the domain (Fig. 8). Road dust contributes little primary OM across the eastern US.

The contribution of other sources is relatively high around Chicago, Philadelphia, and Canada (northwest of Ottawa and Montreal). In Canada, the corresponding concentration is predicted to be 0.77 \( \mu \)g m\(^{-3}\) (42\%). However, the mobile gasoline and diesel emissions are included in the other sources category in Canada.

### 4.4. Source-resolved EC

Since EC has no initial concentration or boundary condition, the sum of the sub-categories is the sum of the eight sources. The domain average EC concentration for the eastern US is predicted to be 0.25 \( \mu \)g m\(^{-3}\). The average source contributions for the whole domain are: gasoline 4\%, non-road diesel 54\%, on-road diesel 24\%, natural gas 0\%, wood burning 8\%, biomass burning 4\%, road dust 0\%, and other 6\%. Similar to the primary OM, the
Fig. 7. Predicted average PM$_{2.5}$ primary OM concentrations (µg m$^{-3}$) from each source for the eastern US between 16 July and 28 July 2001.
Fig. 8. Predicted average contribution of each source to the total PM$_{2.5}$ primary OM concentrations for the eastern US between 16 July and 28 July 2001.
domain averages for EC are comparable to the emissions of EC shown in Table 1.

The daily fluctuations in the EC concentrations follow the same pattern as the OM concentrations, shown in Fig. 5 for Pittsburgh, PA. On average, the predicted EC concentrations are a factor of two higher than the measurements in Pittsburgh (Gaydos et al., 2007). The discrepancies are larger at night, with the model considerably overpredicting the observed concentrations by approximately 1 μg m⁻³ for most nights. Similar to primary OM, the EC source contributions exhibit little day-to-day variability. The day-to-day variations in source contributions to ambient EC are due to changes in wind patterns and temperature along with the difference in emissions from weekdays and weekends (Fig. 6). Therefore, the focus will be on the 13 day average contribution of each source and the spatially variations.

Fig. 9 shows the average contribution of each source to the total EC predicted from the base case. Diesel exhaust is predicted to contribute an average of 75% of the EC in the eastern US. In the Midwest/ Northern Plains the EC concentrations are dominated by non-road diesel emissions (Fig. 10). According to the emission inventories, non-road diesel exhaust is both an urban and rural problem for EC concentrations while on-road diesel is more of an urban problem. Non-road diesel dominates (according to the inventory) in the Midwest and the Northeastern seaboard while its contribution is much smaller in the Southeast.

As shown in Fig. 10, gasoline exhaust contributed roughly 5–10% of the EC across the eastern US. A large fraction of the predicted EC concentration over Maine comes from wood burning (60–80%), similar to primary OM. Over the coast of Maine, the EC concentration from wood burning contributes 0.5–1.8 μg m⁻³. Except in the Northern Plains (close to zero contribution) and Texas, biomass burning contributes around 5% of the EC which represents around 0.02–0.05 μg m⁻³ of the EC across the eastern US. The road dust and natural gas sources are predicted to have little impact on the EC concentrations. Combined, these sources contribute less than 1% of the EC in the eastern US.

The average predicted contribution of the other sources is around 6% of the EC. As seen with primary OM, the other source EC contributions are higher in Chicago, Philadelphia, and Canada. East of Philadelphia, the EC from other sources is predicted to be 0.5 μg m⁻³ (26%) and in northern Illinois 0.1 μg m⁻³ (5%). Over parts of Canada, the EC concentration contributed by the other sources category is 1.2 μg m⁻³. A large fraction of these emissions in Canada is actually the mobile gasoline and diesel exhaust.

4.5. Comparison with CMB

Fig. 11 shows the source apportionment predicted by the source-resolved model compared to the CMB results for Pittsburgh in July 2001 (Subramanian et al., 2006, 2007; Robinson et al., 2006a–c). The high and low CMB results represent the source apportionment calculated using two different sets of source profiles. The calculation of the “best estimate” CMB results is described in Subramanian et al. (2007).

In Pittsburgh, the predicted primary OC concentration from the base case, which was calculated by dividing the primary OM by 1.4, is higher than the primary OC predicted by CMB. Looking at individual source categories, large discrepancies exist for natural gas, wood combustion, and diesel exhaust when compared to the CMB results. The gasoline source falls within the predicted high and low CMB predictions; while the others and road dust sources are slightly less than the CMB lower bounds. However, the others category for PMCAMx is dominated by industrial sources whereas the CMB others category is largely meat cooking emissions.

The predicted EC concentrations in Pittsburgh are on average two times higher than the measurements taken during the Pittsburgh air quality study and three times higher than the summed CMB predictions (Gaydos et al., 2007; Subramanian et al., 2006, 2007; Robinson et al., 2006a–c). This is mainly due an overestimation of the diesel emission inventory (Fig. 11). In Pittsburgh, the source-resolved model predicts that non-road diesel contributes 0.9 μg m⁻³ of EC and on-road diesel contributes 0.4 μg m⁻³ compared to measured values of around 0.6 μg m⁻³. Clearly the non-road diesel emissions are too high; however, the on-road diesel emissions may also be overestimated.

Fig. 12 shows the comparison of the predicted primary OM concentrations located at the eight different sites from the SEARCH network for the source-resolved model and the CMB model (Zheng et al., 2002). The CMB results are from 1999, whereas the source-resolved results are for 2001. However, the measured average organic aerosol
Fig. 9. Predicted average PM$_{2.5}$ EC concentrations (µg m$^{-3}$) from each source for the eastern US between 16 July and 28 July 2001.
Fig. 10. Predicted average contribution of each source to the total PM$_{2.5}$ EC concentrations for the eastern US between 16 July and 28 July 2001.
concentrations during the two periods are within 20% of each other for all eight sites. It is also important to note that the Atlanta supersite (Jefferson St.) was located close to a series of diesel combustion sources (Solomon et al., 2003). The other SEARCH sites were pairs of urban and rural sites located in Alabama (Birmingham and Centreville), Mississippi (Gulfport and Oak Grove), Florida (Pensacola and OLF#8) and Georgia (Yorkville).

The estimates of the primary organic aerosol concentrations by the two methods are in reasonably good agreement. PMCAMx predicts much higher contributions from gasoline vehicles and lower from diesel combustion sources compared to CMB in all SEARCH sites. It is interesting to note that the predicted sum of these two sources is in general consistent between these two sources suggesting that the problem could be in the corresponding split either in the emissions inventory or in the CMB analysis.

The PMCAMx-predicted contribution of natural gas combustion is too high compared to CMB in the Southeast too, suggesting that this is a general problem in the inventory used. The two methods are in agreement regarding the contribution of wood combustion in most sites, with the exception of the Florida sites where the inventory suggests much lower contributions than CMB. The predicted contributions of road dust emissions and other sources by the two approaches are quite consistent in most sites.

PMCAMx does a reasonable job in reproducing the measured EC concentrations in seven SEARCH sites while it underpredicts the Birmingham levels by 50%. In these areas, the non-road diesel contribution to EC is predicted to be much smaller than in the northeast. Zheng et al. (2002) did not perform a detailed source-attribution of EC in their study, but combining their total PM and organic carbon source contributions it appears that the EC attributed to diesel exhaust is higher than the measured EC in several sites. The source-resolved diesel EC concentration is comparable to the CMB results for Atlanta, both the CMB and the source-resolved model results are most likely overpredicting the corresponding EC concentration. The CMB prediction is too high and not representative of the rest of the city because the measurement site was located near a series of EC sources.

There are a number of explanations for this difference in space. The first is that the EC emissions inventory has different biases in the northeast (too high) and in the southeast (too low). The second is uncertainties in the CMB approaches used by the different groups in these two different studies. Finally, the model predicts the average concentration over a relatively large area, while the measurements are at given points in the domain. Depending on the location of the measurement stations this would introduce discrepancies which for the relatively coarse grid used here would be underpredictions for the stations close to sources of EC and overpredictions for stations further away from these sources. Using data from the Pittsburgh Supersite (Tang et al., 2004) for the period of the simulation these errors can be estimated at least for that area. For July 2001 the four measurement stations inside Pittsburgh (including a downwind suburb) reported average EC concentrations (measurements were collected every day) of approximately 0.9, 0.6, 0.6
Fig. 12. Predicted average primary OM concentrations from each source for 16 July to 28 July 2001 compared to CMB results from Zheng et al. (2002). The left column shows the comparison for the urban sites and the right column shows the corresponding rural site comparison. Diesel = onroad and offroad diesel; and wood combustion = biomass + wood.
and 0.7 \mu g m^{-3}. The predicted value for that period in the corresponding computational cell was around 1.6 \mu g m^{-3}. The maximum difference between sites is 0.3 \mu g m^{-3}. The difference between measurements and prediction far exceeds the variability inside the grid cell and supports strongly the conclusion that the EC emissions are overpredicted in this area in the inventory.

5. Conclusions

The source-resolved model for primary OM and EC is simple to implement compared to source-oriented models. No species need to be added to PMCAMx in order to separately track the concentration of primary OM and EC from different sources. Splitting the emission inventory for primary OM and EC and running each source inventory is a fast way to accomplish the source apportionment of primary components. The CPU time required is linearly proportional to the number of sources modeled.

Even in cases where the OM results from PMCAMx for the BaseE inventory case are in reasonable agreement with the results from IMPROVE and STN, the source-resolved model and the CMB results have significant differences.

The model predicts EC concentrations that are much higher than measurements from the STN network. Non-road diesel, according to the emission inventory, is predicted to contribute more to EC in urban areas than on-road diesel. This overprediction suggests that the non-road diesel emission inventory is currently too high. While the non-road diesel inventory should be reduced, the on-road diesel emission inventory may also need to be reduced.

Natural gas, wood burning and biomass burning are other sources that have emission inventory problems. Clearly, the natural gas emissions are overestimated in the BaseE inventory used in this study. Rogge et al. (1993b) found that the emission of OM from natural gas combustion is low (48.5 ng kJ^{-1} \pm 17.4) and that natural gas contributed only around 0.1% of the total primary OM concentrations over Los Angeles in 1982 (Hildemann et al., 1991). The primary OM emission inventory for natural gas should be reduced by at least 50 times the current value. The contribution of primary OM and EC from wood burning is too high along the northeastern coast, predicting average primary OM concentrations greater than 0.5 \mu g m^{-3}. In July, there should not be much residential wood burning. Primary OM contributions from biomass burning are overestimated in Texas.

The model predicts reasonable values for road dust and others for both primary OM and EC. The predicted primary OM and EC concentrations for these source categories in Pittsburgh and the eight SEARCH locations are in agreement with the CMB results.

If the natural gas inventory is lowered by 95% and the wood burning emissions are also reduced in July for primary OM, then predicted OC concentrations from PMCAMx would be even less than the STN and IMPROVE measurements. If the underprediction of the OC is due to the primary OM emissions, then one or more of the remaining source emission inventories would need to be increased or the secondary contribution should be much higher.

Overall, the source-resolved model is a quick and easy technique to predict the source contributions of primary OM and EC. Clearly, the model has shown that there are significant problems in the emission inventories.

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