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Seasonal distribution and modeling of diesel particulate matter in the Southeast US

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ABSTRACT

The fine and ultra fine size of diesel particulate mater (DPM) are of great health concern and significantly contribute to the overall cancer risk. In addition, diesel particles may contribute a warming effect on the planet's climate. The composition of these particles is composed principally of elemental carbon (EC) with adsorbed organic compounds, sulfate, nitrate, ammonia, metals, and other trace elements. The purpose of this study was to depict the seasonality and modeling of particulate matter in the Southeastern US produced by the diesel fueled sources (DFSs). The modeling results came from four one-month cases including March, June, September, and December to represent different seasons in 2003 by linking Models-3/CMAQ and SMOKE. The 1999 National Emissions Inventory Version 3 (NEI99) was used in this analysis for point, area, and non-road sources, whereas the National Mobile Inventory Model (NMIM) was used to create the on-road emissions. Three urban areas, Atlanta, Birmingham, and Nashville were selected to analyze the DPM emissions and concentrations. Even though the model performance was not very strong, it could be considered satisfactory to conduct seasonal distribution analysis for DPM. Important hourly DPM seasonality was observed in each city, of which higher values occurred at the morning traffic rush hours. The EC contributions of primary DPM were similar for all three sites (~74%). The results showed that there is no significant daily seasonality of DPM contribution to $PM_{2.5}$ for any of these three cities in 2003. The annual DPM contribution to total $PM_{2.5}$ for Atlanta, Nashville, and Birmingham were 3.7%, 2.5%, and 2.2%, respectively.

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1. Introduction

DPM is currently a topic of considerable concern from a number of potentially interrelated standpoints: 1) fuel/energy issues; 2) climate change; and 3) public health impact, (Arlt, 2005; Geller et al., 2006; Hesterberg et al., 2006; Jacobson, 2002, 2005; Monforton, 2006; See et al., 2006; USEPA, 2002). Over the past decade, the United States Environmental Protection Agency (USEPA) has promulgated and implemented new and strict regulations on DPM emissions (USEPA, 2001a). Although specific output depends on equipment operating conditions, the largest single component of DPM emissions is carbonaceous soot produced by the incomplete combustion of diesel fuel. An intensive research effort is currently underway devoted to reducing the level and toxicity of DPM emissions. These efforts include reducing the diesel fuel sulfur content, making the diesel engine combustion process more efficient, as well as removing particles from the exhaust stream (e.g., use of particle traps and catalytic converters) (Burtscher, 2005; USEPA, 2001a, 2004). DPM is part of a complex mixture. The sizes of DPM are in the categories of ultra fine and nano particles, representing a potential source of significant human harm. They penetrate more deeply to the respiratory and cardiovascular systems. The mixture of these particles is composed mainly of elemental carbon (EC) with adsorbed compounds, such as organic carbon (OC), sulfate, nitrate, metals, and other trace elements (Kleeman et al., 2000). Many carcinogenic and mutagenic compounds have been measured in the organic fraction of DPM, such as polycyclic aromatic hydrocarbons (PAHs) and nitroarenes (Shah et al., 2004). DPM initially consists of an agglomeration of EC spheres coated with organic and inorganic compounds that are adsorbed or absorbed at the surface of this agglomerate (Kim et al., 2002; Vouitsis et al., 2005). DPM lose its identity rapidly as it coagulate with other particles and act as condensation sites for secondary aerosols (Ning et al., 2004; Seigneur et al., 2003). The DPM chemical composition is variable, and typically has a composition of 25–60% EC (Moosmuller et al., 2001; Shi et al., 2000) (with estimates ranging from 5 to 90%) and 20–50% OC (Shi et al., 2000). Sulfate may account up to 4% of the total mass, depending on the sulfur diesel content and vehicles type (Saiyasitpanich et al., 2005). This composition and emissions quantity is strongly dependent on the diesel fuel sulfur content (Saiyasitpanich et al., 2005). From a climate change point of view, the differences in sulfur content of coal power plants and DFS, the obvious distinction in color

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(black for diesel soot versus white for power plant emissions) may have a harmful effect on climate by somewhat offsetting the albedo effect of light reflecting particles from power plants (Flanner et al., 2007; Jacobson, 2004; Reddy and Boucher, 2007).

Increased mortality and morbidity in sites with elevated DPM concentrations has been reported by a variety of studies (Carlsten et al., 2007; Tomaru et al., 2007; Toren et al., 2007). Adverse effects also are observed when breathing airborne diesel exhaust particles in controlled acute human exposure studies, including cough, respiratory symptoms of asthmatics, and reduced lung function (Scapellato and Lotti 2007; Urch et al., 2005; Utell and Frampton 2000). A systemic marker of inflammation, c-reactive protein, is thought to be increased in production by the liver and found in higher blood concentrations in exposed individuals (Roux et al., 2006; Ruckerl et al., 2007, 2006; Sullivan et al., 2007; Zeka et al., 2006).

Modeling has assumed an increasingly important role in establishing current and future risks of climate, weather, and air pollution incidences. The CMAQ air quality model, in conjunction with SMOKE's advanced inventory capabilities are heavily employed in air quality work. Use of 1999 inventories for point, area, and nonroad emissions in the 36 km × 36 km modeling domain centered over the metropolitan areas of concern brings to bear the ability to discern

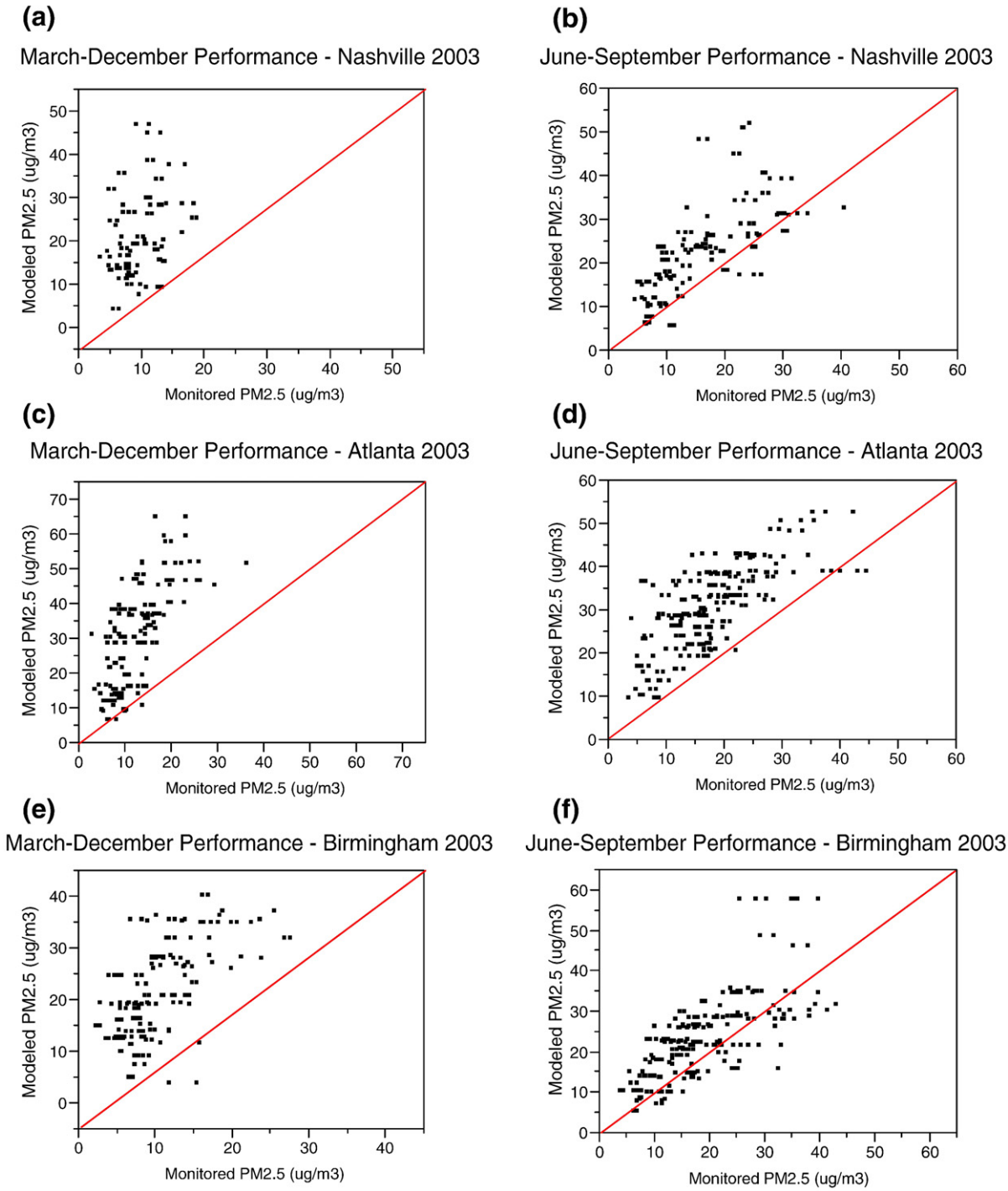

Fig. 1. Modeling performance — cold versus hot months (March and December versus June and September.

Table 1
Normalized bias (USEPA, 2001b).

City	Normalized bias		Cold–hot ratio
	Cold seasons (March and December)	Hot seasons (June and September)	
Nashville	1.326	0.487	2.7
Atlanta	1.546	0.922	1.7
Birmingham	1.278	0.323	4.0

finer geospatial resolution. In the case of Atlanta, with its major population center (estimated 4.5 million in 2003), health impacts contributed by diesel would be strikingly higher, based on both concentration and population factors, than in adjoining rural areas.

Among the potential ways that DPM may act adversely in health are: priming allergens such as pollen to trigger immune dysfunction such as heighten airways reactivity (i.e., asthma), promoting inflammatory processes due to their propensity to create a burden of oxidative free radicals within the bloodstream or at specific locations of harm via metal contaminants, behaving like the estrogen disrupting agent bisphenol A to increase this nation's rampant obesity rate, including among children (Matsumoto et al., 2005; Wilson et al., 2003, 2007). Not just total particulate matter per se, but speciation into more toxic components thus becomes a tool for improved scientific decision-making.

USEPA and other agencies performing climate modeling, such as NASA's Goddard Institute for Space Studies have begun to question whether heat- and light-reflective particles from power plant emissions, but not dark, soot particles from diesel engines may differentially act to decrease the greenhouse effect (Jacobson, 2002, 2005). A model such as ours, can contribute to increased understanding of how future changes in diesel emissions inventories, coupled with atmospheric conditions and changes in ozone levels, moisture, meteorological patterns, etc. in the troposphere, act to offset otherwise anticipated improvements in planetary warming or cooling potential.

Because DPM is often the major source of EC in the atmosphere, this is used a surrogate for DPM (Schauer, 2003). The ability to accurately use EC as a tracer for DPM critically relies on a clear understanding of the relative contributions of other sources to EC concentrations. This approximation has a great deal of uncertainty since studies used EC to estimate the DPM concentration found the EC contribution varying between 50 and 80%. In addition, EC is not a unique tracer for ambient DPM and efforts to utilize EC as an indicator of DPM must properly address other sources of EC such as gasoline vehicles, heating wood combustion, restaurant kitchens, agriculture biomass burning, and forest wildfires (Mysliwiec and Kleeman, 2002). Furthermore a consistent measurement technique for EC must be utilized when comparing source and ambient EC measurements to avoid significant biases.

In order to better manage air quality, it is important to know the sources or source categories that contribute to the concentrations of DPM at a particular area. Receptor models have been used for $PM_{2.5}$ source apportionment; however, they do not fully take into account the chemical reactions involved in the formation of secondary fine particles (Lloyd and Cackette, 2001; Mysliwiec and Kleeman, 2002), and so far there is not an available method to measure ambient DPM.

The uncertainty to use EC as a tracer for DPM was reduced in the present study, even though the model performance on $PM_{2.5}$ was not very strong and similar to the performance seen by other researchers (Ching et al., 2004). In effect, it could be considered satisfactory to do seasonal distribution analysis for DPM, since the analysis approach considers the comparison in mass concentrations among the proposed sites. The model might not get better due to uncertain in emission inventory and model itself based on the hourly or daily average, for example, some emission temporal profiles can be one factor which is no available data to support (Diaz-Robles et al., 2008). From a health point of view, the useful concentration should be the annual concentration, although the CMAQ over-predicted this concentration. EC and DPM were calculated from a model instead of ambient monitoring results. Both EC and DPM were modeled temporally and spatially over an urban to a regional area to predict emissions and aerosol concentrations of primary and secondary aerosols and their differences that come from diesel fueled sources (DFSs) in the Southeastern US.

2. Methodology

2.1. Conceptual model's development

Diesel aerosol concentrations were predicted using the advanced air quality model Community Multi-scale Air Quality (CMAQ) version 4.3 (Byun and Ching, 1999). The predicted concentrations are based on emissions that were temporally and spatially allocated using the advanced emissions model Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System version 2.0 (UNC, 2004). Emissions from on-road sources were predicted using NMIM for the whole modeling domain (USEPA, 2005a), whereas the NEI99 (USEPA, 2005b) was used for point, area, and non-road sources in the 36-km by 36-km modeling domain of the Southeastern US. This analysis was focused in Nashville and the urban areas of Atlanta and Birmingham. The meteorological variables were generated for March, June, September, and December of 2003 through the mesoscale model (MM5) version 3.7 developed by the National Center for Atmospheric Research (NCAR) at the Pennsylvania State University, PSU (NCAR/PSU, 2005) and processed by the meteorology–chemistry interface processor (MCIP) version 2.2. For this study, each month was set to start 5 days earlier to avoid the initial concentration effects. Finally, daily total $PM_{2.5}$ was used to analyze the modeling performance. To do that, the normalized biases (NB) of $PM_{2.5}$ concentrations were estimated for all sites using the Eq. (1).

$$\mathrm{NB} = \frac{1}{n}\sum\left[\frac{\text{Predicted} - \text{Observed}}{\text{Observed}}\right] \quad (1)$$

where n is the number of days with monitored data. The predicted concentrations that were used in the equation were actually the average 24-hour concentration predicted by the models CMAQ 4.3. For this case, the USEPA guidance (USEPA, 2001b, 2007) recommends that the normalized bias has to be less than or equal to 25%, and less or equal to 30% for normalized gross error.

2.2. Description of the modeling domain

The grid size of the domain of 36-km by 36-km was selected due to the unavailability of the computer sources required and the necessary input data. To run 12 km by 12 km or 4 km by 4 km grid for 4 months would have required more computer resources than we had available. Because the results of this study were focused principally in the Nashville metropolitan area, the domain was selected such that Nashville was approximately at the center of the domain surrounded by 23 other states to minimize the effect of boundary conditions and

Table 2
Correlation coefficient.

City	Correlation coefficient	
	Cold seasons (March and December)	Hot seasons (June and September)
Nashville	0.115	0.507
Atlanta	0.482	0.531
Birmingham	0.361	0.516

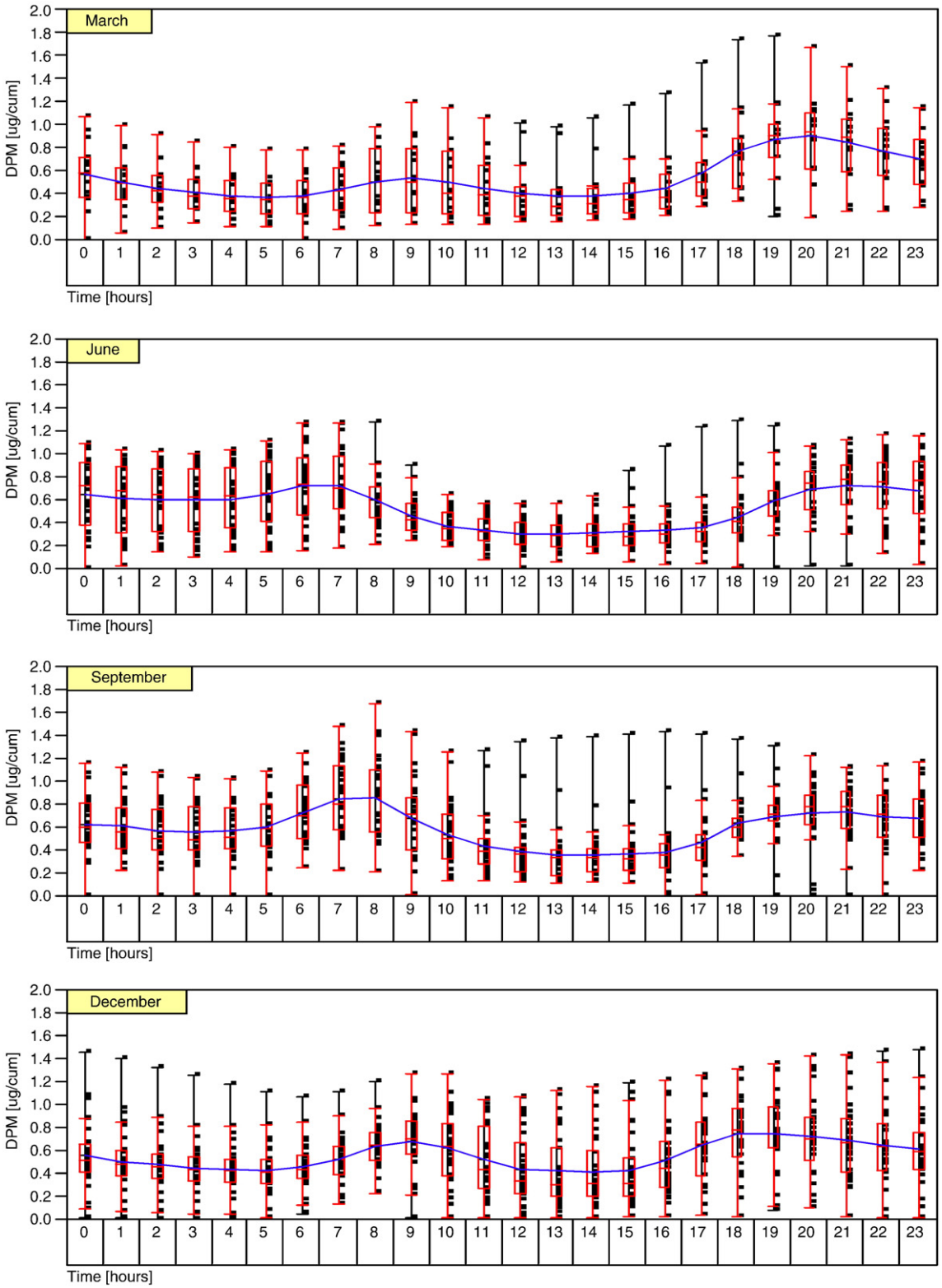

Fig. 2. Box-plots of the modeled hourly average DPM concentration at Nashville, TN, for March, June, September, and December 2003.

pollution transport (Doraiswamy et al., 2007). This domain included most of the central eastern region of the U.S.

2.3. Emissions scenarios

The methodology consisted of running the SMOKE 2.0 and CMAQ 4.3 models with and without diesel fueled sources (DFSs). The base case was run with all sources included. The scenario without DFS was estimated through control matrices for the corresponding source (s). Those source categories were eliminated using the source classification codes (SCC) through a control matrix for each scenario in SMOKE 2.0. The difference between the base case scenario and the scenario without DFS were the DPM emissions and concentrations for the base case run.

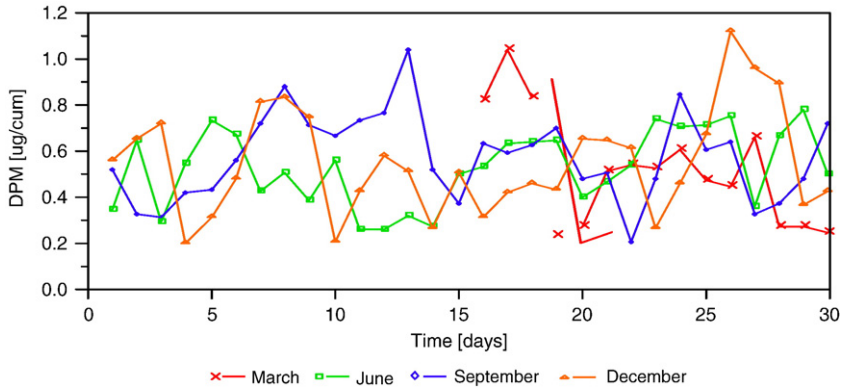

Fig. 3. Modeled daily DPM concentration in Nashville, TN for March, June, September, and December 2003.

The DPM emissions were estimated considering the addition of following $PM_{2.5}$ species defined in SMOKE2.0: elemental carbon (EC), primary fine particulate matter (PMFINE), primary nitrate (PNO3), primary organic aerosols (POA), and primary sulfate (PSO4) (UNC, 2004). This approach did not account for the PM sources that are apportioned between primary and secondary sulfate and nitrate aerosol concentrations, because there currently is no way to determine how much of the sulfate and nitrate in the Aitken and accumulative modes of CMAQ 4.3 are primary and how much are secondary (Byun and Ching, 1999). However, the sulfate and nitrate concentrations in the DPM emissions modeled by Diaz-Robles et al. (2008) for Atlanta, Birmingham, Nashville, Memphis, and Knoxville for the summer of 1999 were generally very low, averaging 1.82% and 0.16%, respectively (Diaz-Robles et al., 2008).

3. Results and discussion

3.1. Modeling performance

Modeled daily $PM_{2.5}$ concentrations were compared with daily average of ambient $PM_{2.5}$ concentrations obtained from the available AQS monitoring sites in Nashville, Atlanta, and Birmingham for 2003, as shown in Fig. 1. Although a wide 36-km domain was used, the modeled daily $PM_{2.5}$ values compared reasonably well against the observed values for hot months (June and September 2003) at Birmingham, with an overestimation bias of approximately 30% (USEPA, 2001b, 2007), even though the USEPA recommends that the normalized bias has to be less than or equal to 25%, and less or equal to 30% for normalized gross error. For Atlanta and Nashville the normalized biases were over 30% for hot months, which could be explained due to emission inventories uncertainty. On the other hand, the model did not perform well for cold months (March and December 2003) with important overestimation bias, as shown in Table 1 and correlation coefficients (R^2) of Table 2. Birmingham and Nashville showed a cold season bias 4.0 and 2.7 times higher than the hot season bias, respectively. Although limited monitoring data was available for each site, the best correlation

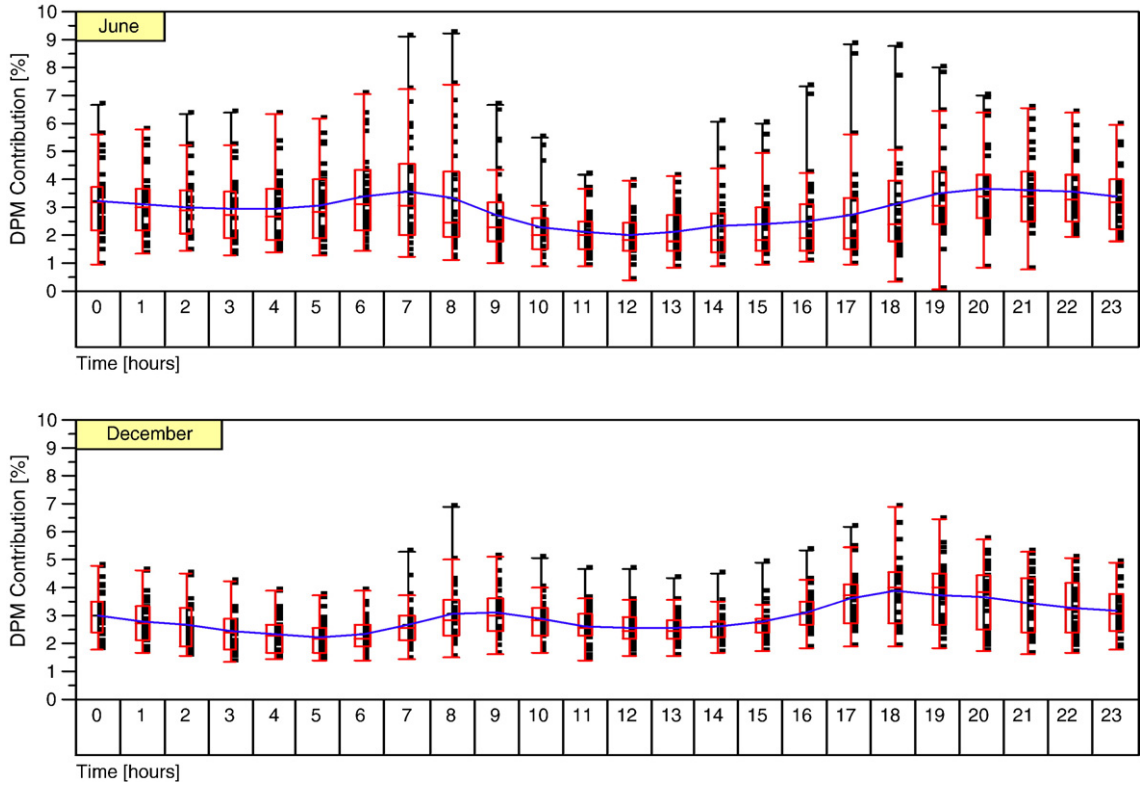

Fig. 4. Modeled hourly DPM contribution to total $PM_{2.5}$ in Nashville, TN, June and December 2003.

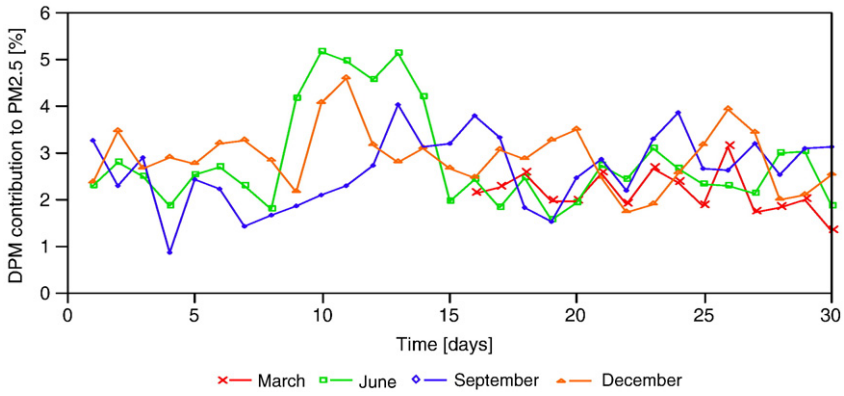

Fig. 5. Modeled daily DPM contribution to total $PM_{2.5}$ in Nashville, TN, for March, June, September, and December 2003.

coefficient was for Atlanta during hot months of 2003 with a value of R^2 equal to 0.531. Finally, the model data is reasonably consistent in pattern. Although our results were overestimated for summer and winter time, those in winter were more overestimated than in summer. These results are comparable with those in Morris et al. (2005), who analyzed CAMx, CMAQ, and REMSAD models (Morris et al., 2005). These authors found that all three of the models exhibited winter overestimation and summer underestimation bias mainly due to secondary aerosol formation. The results could indicate that CMAQ4.3 does not properly simulate $PM_{2.5}$ during cold seasons and better improvements have to be made within the model. Modeling MM5 and CMAQ for 36 km grid resolution produces inaccurate meteorology, dispersion fields, spatial, and temporal variability of the pollutants; however, this assumption is good enough to analyze the proposed emission scenarios and seasonal distribution on ground level DPM concentrations. In fact, the analysis approach involves considering the comparison in mass concentrations among the proposed sites. This assumes that the factors that contributed to the under and over prediction of those EC concentrations for March, June, September, and December would contribute relatively similar in all the sites

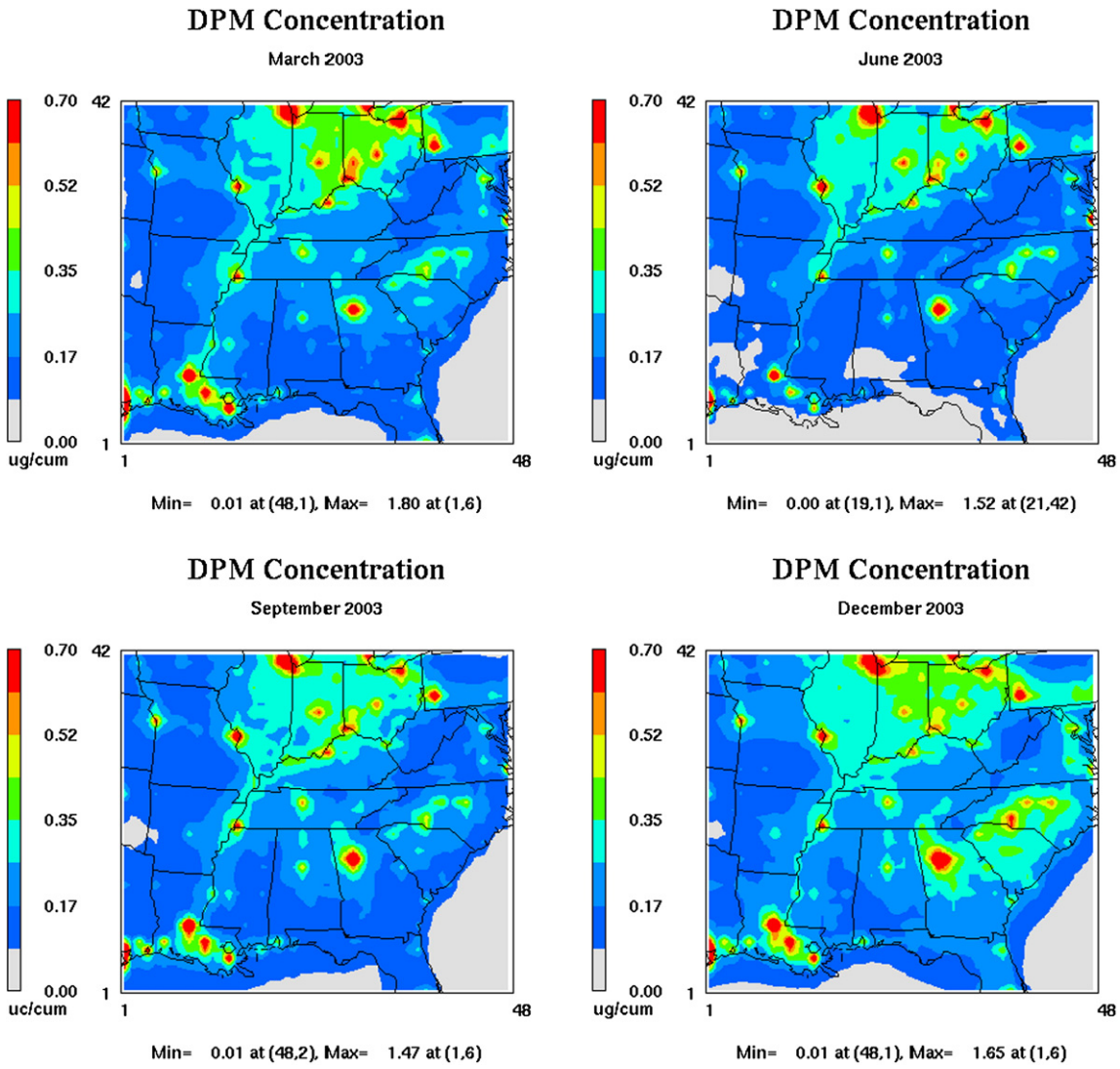

Fig. 6. Modeled monthly DPM concentrations for the modeling domain.

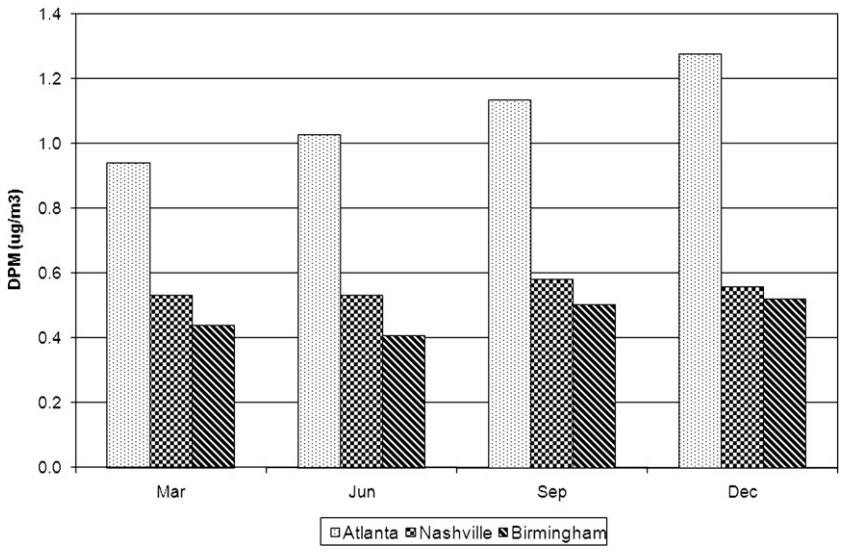

Fig. 7. Monthly DPM concentrations in urban areas of the Southeastern US for 2003.

considered in the analysis, with uncertainty. The differences and this uncertainty among the sites would depend mainly from each emission inventory quality, local meteorological conditions, monitored data quality, and terrain complexity (Tesche et al., 2006). Future simulations could include running fine-scale modeling on DPM and for the whole year simulation to better capture spatial and temporal variability, as well as, concentration magnitudes, which could help identify and characterize the hot spots of DPM from an exposure point of view. However, working with fine-scale modeling on CMAQ does not improve the modeling performance at all (Jimenez-Guerrero et al., 2008).

3.2. DFS sources contribution

Modeled hourly DPM concentrations at Nashville produced seasonal patterns for the 24-hour time periods for each month, as shown in Fig. 2. In general, the maximum modeled DPM concentrations occurred between 6 and 9 PM, and the minimum DPM concentrations occurred between 1 and 3 PM. The wood burning contribution in the urban areas of the Southeastern US is important (about 27%) (Marmur et al., 2006), however, the modeled DPM concentration was obtained running a withholding DFS category (domain-wide exclusion). In other words, the DPM concentrations are just from DFS.

A second maximum occurred between 6 and 10 AM. It should be noted that DPM are generated as primary pollutants. The temporal results shown in Fig. 2 were obtained using SMOKE2.0 that utilizes a default on-road temporal profile for all vehicles emissions, including diesel trucks. This, in all likelihood, is not accurate, because the maximum volume of trucks occurs around mid-afternoon PM for the most days, including weekends (Miller, 2005). This more realistic temporal profile for trucks could be incorporated in SMOKE to produce better temporal hourly emissions and concentrations of DPM at places close to highways; however, the percentage of trucks of these urban areas is not very significant. As in urban areas not only there are heavy-duty diesel trucks, but also there are diesel buses, diesel SUVs, and light diesel cars on the roads with temporal profiles that USEPA defines, the default SMOKE2.0 on-road temporal emissions profile was considered as most appropriate for this study.

The main factor causing dilution of the emitted primary DPM is the wind during afternoon hours. Fig. 2 shows that an important amount of DPM emitted during morning rush-hour traffic were dispersed by about 2 PM as a result of the higher wind speeds during afternoon hours. This particulate matter behavior has been described for Atlanta, GA, (Weber, 2003).

At the end of the daytime hours and at night, DPM emitted during afternoon rush-hour traffic dropped and dispersed slowly until new DPM is emitted during morning traffic congestion as a new diurnal emissions cycle commence. The daily DPM concentration was similar for each month and was around 0.5 $\mu g/m^3$, as shown in Fig. 3, ranging from 0.2 $\mu g/m^3$ to 1.6 $\mu g/m^3$ during June and December.

According to Fig. 4, the mean DPM contribution to the total hourly $PM_{2.5}$ concentration in Nashville was higher during morning and afternoon rush-hour traffic,

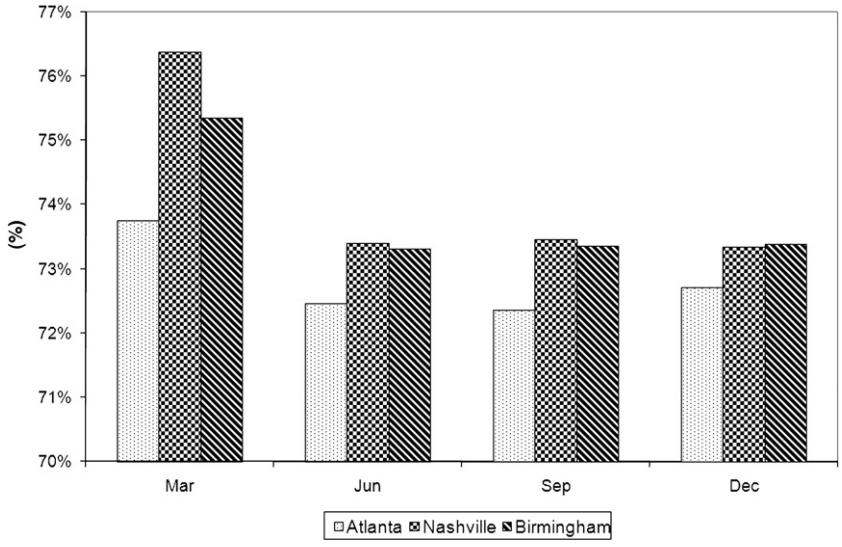

Fig. 8. EC Contribution to Primary DPM for Urban Areas in the Southeastern US for 2003.

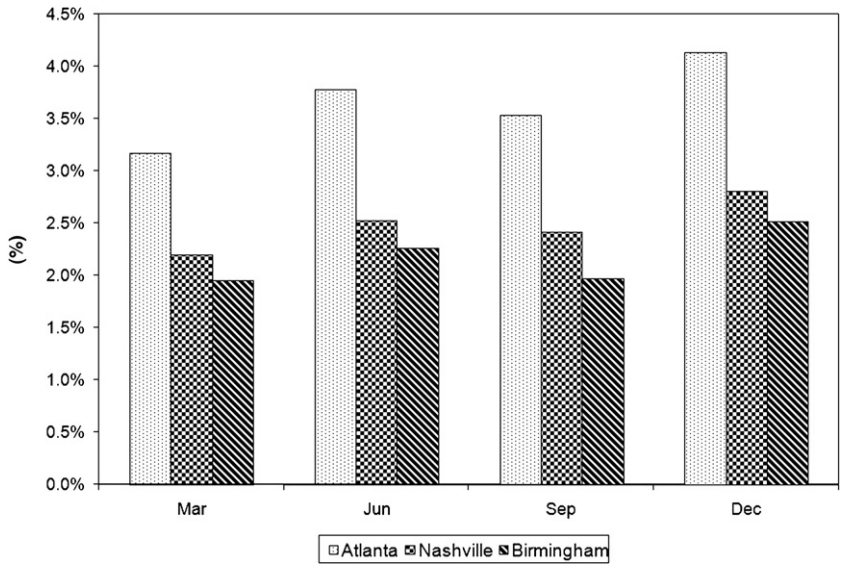

Fig. 9. DPM contribution to total monthly $PM_{2.5}$ in the Southeastern US for 2003.

i.e., around 7 AM and 5 PM. The minimum DPM contribution to the total hourly $PM_{2.5}$ concentration was produced around 1 PM. The highest hourly contribution was as high as 9.3% on June 13th at 8 AM (see Fig. 4). The hourly contribution was higher and more variable in June than in December, since in the summer season there is more variability in the secondary aerosol component of $PM_{2.5}$ due to the higher temperatures. The daily DPM contribution to the total $PM_{2.5}$ was similar for each month and was around 2.5%, as shown in Fig. 5, ranging from 1% to 5% during June and December.

3.3. Spatial distribution of modeled DPM concentrations

The four plots in Fig. 6 show the spatial variation of modeled DPM for March, June, September, and December 2003. It must be noted that these plots of monthly average concentrations were generated for 5 AM GMT, which corresponds to midnight in the central daylight savings time zone (CDT).

In general, the tile plots show that higher DPM concentrations occurred in southeastern urban areas, principally in Atlanta, followed by Nashville, Birmingham, Raleigh, and Memphis. Concentrations of DPM were comparatively higher at the Atlanta urban site than in rural locations. More significant temporal fluctuations were seen at the urban areas. The rural areas seem less significantly affected by local emissions of DPM.

The area around the Mississippi river showed the impact of diesel marine engines on modeled DPM concentrations. Atlanta showed higher modeled DPM concentrations in December than other months and it increased from March to December, as shown in Fig. 7. Nashville and Birmingham did not show significant seasonal DPM concentration variability during 2003.

Analyzing the primary DPM species given by CMAQ4.3, we can plot the EC contribution of primary DPM for three southeastern urban areas (Fig. 8). The sources that use diesel contributed approximately 75% of the EC concentrations at Nashville for March, June, September, and December, 2003. This contribution is close to the values obtained by Zheng et al. (2002), who employed a molecular marker chemical mass balance model to apportion the sources of atmospheric particulate matter in eight cities in the Southeastern US for one-month of each season between the spring of 1999 and the winter of 2000 (Zheng et al., 2002). Their calculated values, for January, April, July, and October were 74%, 84%, 92%, and 85% respectively, with the results demonstrating the seasonal impact of wood smoke on EC concentrations. Fig. 8 shows much higher EC contribution to primary DPM in March because during this cold month there were more frequent atmospheric inversions.

The differences between our results and those obtained by Zheng et al. (2002) could be due to the fact that Zheng et al. did not consider the photochemical reaction's effect on the Secondary Organic Aerosols (SOA) formation, while our results could be affected by inaccurate emission inventory provided by the U.S. EPA. Our estimates of the EC contribution to primary DPM for Birmingham and Atlanta were somewhat similar to that for Nashville.

Fig. 9 shows the DPM contribution to total monthly $PM_{2.5}$ for Atlanta, Nashville, and Birmingham. The results showed that there is no significant seasonality of DPM contribution to $PM_{2.5}$ for any of these three cities in 2003. December showed slightly higher DPM contributions than the other three time periods. The annual DPM contribution to total $PM_{2.5}$ for Atlanta, Nashville, and Birmingham were 3.7%, 2.5%, and 2.2%, respectively.

4. Conclusions

Even though the model performance was not very strong, it could be considered satisfactory to do seasonal distribution analysis for DPM, since the analysis approach considers the comparison in mass concentrations among the proposed sites. The CMAQ model over-predicted the $PM_{2.5}$ concentration throughout the monitoring sites of each site of each city, mainly during cold months. The maximum hourly DPM concentrations are predicted to occur between 6 and 9 PM and the minimum concentrations between 1 and 3 PM. A second maximum is predicted to occur between 6 and 10 AM. The mean DPM contribution to the total hourly $PM_{2.5}$ was higher during morning and afternoon rush-hour traffic (i.e., around 7 AM and 5 PM) in Nashville. The minimum was produced around 1 PM. Future simulations could include running fine-scale modeling on DPM to better capture spatial and temporal variability, as well as, performance and concentration magnitudes, which could help identify and characterize the hot spots of DPM from an exposure point of view.

A large amount of DPM emitted during morning rush-hour traffic was dispersed at around 2 PM. At the end of the daytime hours and at night, the DPM emitted during afternoon rush-hour traffic dropped and dispersed slowly. Concentrations of DPM were typically a factor of 11 higher at the Atlanta urban site than at the rural sites. More significant temporal fluctuations were seen at the urban areas. The rural areas seem less significantly affected by local emissions of DPM.

The EC contribution to primary DPM was similar at all three sites, averaging approximately 74%. The results showed no significant seasonality of DPM contribution to $PM_{2.5}$ for any of these three cities in 2003. The annual DPM contribution to total $PM_{2.5}$ for Atlanta, Nashville, and Birmingham were 3.7%, 2.5%, and 2.2%, respectively. A likely rationale for the absence of DPM seasonality can be because the vehicle miles traveled (VMTs) are similar along the year (USEPA, 2005b) and relative weak impact the meteorology has vs. strong effect of the primary DPM concentrations.

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