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Source apportionment of $PM_{2.5}$ in the southeastern United States using receptor and emissions-based models: Conceptual differences and implications for time-series health studies

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Abstract

Elevated levels of fine particulate matter ($PM_{2.5}$) have been associated with adverse effects on human health, but whether specific components of $PM_{2.5}$ are responsible for specific health effects is still under investigation. A complementary approach to examining species-specific associations is to assess associations between health outcomes and sources contributing to $PM_{2.5}$. This approach could help target and regulate the sources that contribute most to adverse health effects. Various techniques have been developed to quantify source impacts on air quality, allowing examination of their health impacts. We compare two conceptually different approaches to source apportionment (SA): a receptor model and an emissions-based air-quality model. Daily source impacts for July 2001 and January 2002 at four sites in the southeastern US were calculated using CMB-LGO, an extended chemical mass balance receptor model incorporating the Lipschitz global optimizer, and EPA's Models-3 emissions-based air-quality modeling system (MM5–SMOKE–community multiscale air-quality (CMAQ)). The receptor model captured more of the temporal variation in source impacts at a specific receptor site compared to the emissions-based model. Driven by data at a single site, receptor models may have some significant shortcomings with respect to spatial representativeness (unless a reduced study area is used or data from multiple sites are available). SA results from emissions-based models, such as CMAQ, may be more spatially representative as they represent an average grid-cell value. Limitations in the ability to model daily fluctuations in emissions, however, lead to results being driven mainly by regional meteorological trends, likely underestimating the true daily variations in local source impacts. Using results from either approach in a health study would likely introduce an attenuation of the observed association, due to limited spatial representativeness in receptor modeling results and to limited temporal representativeness in emissions-based models results.

Keywords: CMAQ; CMB; LGO; Source apportionment; Health effects; $PM_{2.5}$

1. Introduction

Particulate matter, especially particles $<2.5\,\mu m$ in diameter ($PM_{2.5}$), has been associated with adverse

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health effects and mortality in studies covering >150 cities (Dockery et al., 1993; Pope et al., 1995, 2002). Both acute and chronic exposures to $PM_{2.5}$ have been associated with increased mortality rates and hospital visits, as well as cardiopulmonary disease, heart attacks, decreased lung function, and asthma (Dockery et al., 1993; Ebelt et al., 2000; Peters et al., 2001; Pope et al., 1995; Vedal, 1997). PM is chemically complex, being comprised of numerous primary and secondary components, including ionic and organic compounds and dozens of trace elements. It is still unknown which specific components of $PM_{2.5}$ are more prone to cause specific health effects, although recent studies have started to address this issue (e.g., Metzger et al., 2004a, b; Peel et al., 2005; Hauck et al., 2004; Heal et al., 2005). However, the association between health outcomes and specific $PM_{2.5}$ components raises several issues: it is not obvious that the major cause for the health outcome is actually measured (it is impractical to measure every single $PM_{2.5}$ species) or is possibly measured inaccurately due to analytical issues; the actual health effects may be due to a combination of pollutants; many species are correlated which limits the ability to isolate species health impacts. A complementary approach is to examine associations between health outcomes and sources contributing to ambient $PM_{2.5}$ (Laden et al., 2000; Manchester-Neesvig et al., 2003; Mar et al., 2000; Tsai et al., 2000). By means of source apportionment (SA), source impacts on the receptor can be quantified, and their health impacts examined. A source-impact oriented approach could help target and regulate the sources that contribute most to adverse health effects. It could also allow for better multi-component epidemiologic modeling, as the number of major source-impact categories is typically far fewer than the number of PM components. Finally, this approach can help identify health effects of unmeasured species present in emissions from specific source categories. For example, preliminary studies have found an association between mortality and combustion-related $PM_{2.5}$ (from motor vehicles, coal combustion, and wood burning), but not soil-related $PM_{2.5}$, in both cohort (Laden et al., 2000) and time-series (Mar et al., 2000) studies. Ito et al. (2004) mention that source-oriented evaluation of PM health effects need to take into consideration the uncertainty associated with spatial representativeness of the species measured at a single monitor.

Here, we compare two conceptually different approaches to $PM_{2.5}$ SA, receptor-based modeling (represented in this study by the chemical mass balance (CMB) model) and emissions-based air-quality models (represented in this study by EPA's Models-3 suite of models). We address issues associated with using these techniques for time-series health studies, with special emphasis on the degree to which these approaches provide source impact estimates that are both spatially and temporally representative. So far, the focus in the SA and health literature has been on the use of various factor analytical (FA) techniques (such as PCA or PMF), to associate health outcomes with factors associated with sources of $PM_{2.5}$ (Laden et al., 2000; Manchester-Neesvig et al., 2003; Mar et al., 2000; Tsai et al., 2000). Here, we address a slightly different approach, using the CMB receptor model, which, especially in its extended form presented here (CMB–Lipschitz global optimizer (LGO)), is more explicit in terms of identifying the sources in question. The main difference between CMB and FA approaches is that CMB uses emission composition data to derive source impacts, whereas FA techniques derive the source compositions from trends in the ambient data during the process of estimating source impacts. In an FA application, the investigator assigns names to the obtained factors (i.e., identifies the factors as sources) based on their chemical composition; the factors are unique for each data set/site analyzed. For example, Ito et al. (2004) illustrate that uncertainties in "naming" the factors in FA applications might cause source impacts to appear less spatially representative than the true case because the derived factor for a given source category will be different at each site. Both approaches have advantages and disadvantages; however, in terms of assessing the spatial and temporal representativeness of receptor and emissions-based models for use in health studies, FA techniques share many of the same characteristics and issues as presented here for CMB (both being based on measured ambient data).

2. Methods

We used both receptor-based (CMB) and emissions-based (EPA's Models-3) air-quality modeling approaches to conduct SA of $PM_{2.5}$ in Atlanta, GA and other sites in the southeastern US. The CMB receptor-based model (US-EPA, 2001) makes use of

speciated ambient $PM_{2.5}$ measurements (major ions, carbon fractions, trace elements) and typical compositions of emissions from various source categories to quantify the source contribution to measured concentrations at the receptor. It is based on the following mass balance equation, which is solved for S_j (a vector of source contributions) (US-EPA, 2001):

$$C_i = \sum_{j=1}^{n} f_{ij} S_j + e_i, \quad (1)$$

where C_i is the ambient concentration of chemical species i ($\mu g\,m^{-3}$ in $PM_{2.5}$), $f_{i,j}$ the fraction of species i in emissions from source j, S_j the contribution (source-strength) of source j ($\mu g\,m^{-3}$ in $PM_{2.5}$), n the total number of sources, and e_i the error term.

Here, we applied both an extended version of CMB, referred to as CMB–LGO (Marmur et al., 2005) and the standard CMB. CMB–LGO uses measured ambient gaseous concentrations (SO_2, CO, and NO_y) to bound acceptable solutions to the mass balance equation above. For each solution obtained, ambient SO_2, CO, and NO_y concentrations are calculated based on the obtained $PM_{2.5}$ source contribution and typical gas-to-particle ratios at the source (e.g., $SO_2/PM_{2.5}$ from the various sources). The calculated value is then compared to the ambient measurement and, in case of significant differences (e.g., under/overprediction of more than a factor 3), the model then searches for the next best fit to the mass balance equation that would also adhere to the gas-phase constraints (Marmur et al., 2005). This approach reduces collinearity between sources, which is one of the major limitations to source identification using CMB (i.e., the inability to distinguish between sources with similar $PM_{2.5}$ emissions compositions). The reduced collinearity is because sources that share fairly similar $PM_{2.5}$ composition may have very different gaseous emissions. For example, gasoline and diesel engines have fairly similar $PM_{2.5}$ compositions (rich in organic and elemental carbon), but differ significantly with regards to CO and NO_x emissions. Likewise, collinearity caused by crustal elements found in both soil-dust and coal-fired power-plant emissions is significantly reduced with the introduction of the SO_2 constraint (SO_2 is abundant in power-plant emissions, but not present in resuspended soil dust). However, use of CMB–LGO has some limitations, some of which might be important in time-series health studies. First, collinearity is not eliminated completely, and part of the daily fluctuations in the amount of mass apportioned between several sources might be due to collinearity, hence introducing an error to the time-series epidemiologic analysis. There are also uncertainties associated with the source profiles used. The composition might not necessarily represent typical local source compositions since locally accurate source profiles are not always available. The composition may vary temporally (e.g., the effect of driving mode on the composition of $PM_{2.5}$ from mobile sources) whereas constant values are used. Finally, the model relies on local (receptor) ambient measurements which might be significantly affected by local sources within 1–2 km and, therefore, might not represent the health study area, and might also contain measurement errors.

For these reasons, we also evaluated the use of emissions-based air-quality models for epidemiologic analyses. Such models have been used for gas-phase simulations and source-impact analysis of ozone for decades, and are typically three-dimensional (3-D) representations of the atmosphere. More recently, they are being applied to $PM_{2.5}$ SA as well, apportioning mass the either sources or regions (Boylan et al., 2002; Held et al., 2005; Odman et al., 2004). The 3-D air-quality model simulates the source impacts by solving the conservation equation expressed as

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (U c_i) = \nabla \rho D_i \nabla \left(\frac{c_i}{\rho}\right) + R_i(c_1, c_2, \ldots, c_n, T, t) + S_i(x, t), \quad i = 1, 2, \ldots, n, \quad (2)$$

where c_i is the concentration of species i, U the wind velocity vector, D_i the molecular diffusivity of species i, R_i the rate of concentration change of species i by chemical reaction, $S_i(x, t)$ the source/sink of i at location x and time t, ρ the air density, and n the number of predicted species.

The conservation equation describes the formation, transport, and fate of air pollutants, including components for processing emissions, meteorology, topography, and atmospheric chemistry (Russell and Dennis, 2000). SA can be performed using direct sensitivity methods, such as direct decoupled method (Dunker, 1981; Hakami et al., 2003), inert tracer methods, or by multiple applications of the model with and without emissions from target sources ("brute force"). Here, we applied brute force to the US-EPA's Models-3 suite of models, including MM5 (Grell et al., 1995) as the

meteorological model to simulate atmospheric physical dynamics; SMOKE (Houyoux et al., 2003) as the emissions processor to calculate spatial and temporal trends in emissions based on the annual emissions inventory; and the community multiscale air-quality (CMAQ) model (Byun and Ching, 1999) to simulate atmospheric reactivity, transport, and deposition of chemical contaminants.

In terms of $PM_{2.5}$ SA, the major difference between a receptor model and an emissions-based air-quality model is the starting point. While a receptor model's starting point is the ambient measurement, from there going backwards to estimate source contributions, the starting point of the air-quality model is the processed emissions inventory, going forward by simulating the transport and transformation of pollutants and ultimate air-quality impact. The emissions used are typically processed from annual, county level emissions, using statistical daily/weekly/seasonal temporal trends (such as peak morning rush hour and lower weekend emissions from mobile sources) and spatial information. Starting from an estimate of expected emissions at a location and time, atmospheric processes taking place during transport from source to receptor are simulated. CMAQ results provide more regionally representative values than those provided by an analysis based on a local measurement. However, results from air-quality models include uncertainties arising from each step of the process (meteorological modeling, emission estimates, air-quality modeling). Studies have shown that the emission inventory is one of the more uncertain, but particularly important, inputs to the air-quality modeling process (NRC, 1991; Seinfeld, 1988). This is especially important in the current application where daily variability in source impacts is sought.

2.1. Model application

SA using CMAQ was performed on a daily basis for the months of July 2001 and January 2002. For actually applying either of these methods to a time-series health study, a much larger data set is desired (typically, several years of data). However, focusing on the reduced time periods allows more detailed examination of the issues involved in using results from either approach. Also, obtaining several years of CMAQ-based SA results using the brute-force method requires significant computational resources. As tracer and other direct SA methods become available in CMAQ, it will be possible to expand this type of analysis by tracking source impacts efficiently, without having to re-apply the model for each source category examined. A longer CMB–LGO analysis has been completed and analyzed (Marmur et al., 2005).

The Models-3 air-quality modeling system was applied over much of the US using a 36 km grid, and a finer 12 km grid was used over northern Georgia (Fig. 1). More detailed model information is presented elsewhere (Park et al., 2005, 2006a, b). We focused on two urban (Atlanta, GA and Birmingham, AL) and two rural (Yorkville, GA and Centerville, AL) southeastern aerosol research and characterization (SEARCH) sites (Fig. 1), which include data on total $PM_{2.5}$ mass (gravimetric measure) and its components (Hansen et al., 2003; Kim et al., 2003). The main objectives of SEARCH include the understanding of composition and sources of PM in the southeast (Hansen et al., 2003; Kim et al., 2003). The SEARCH data are also being used for health studies in Atlanta in which associations of $PM_{2.5}$ with respiratory illnesses and cardiovascular disease have been observed (Metzger et al., 2004a, b; Peel et al., 2005). In the southeastern US, a major part of the total $PM_{2.5}$ is secondary (Hansen et al., 2003; Kim et al., 2003; Marmur et al., 2005), i.e., formed in the atmosphere from

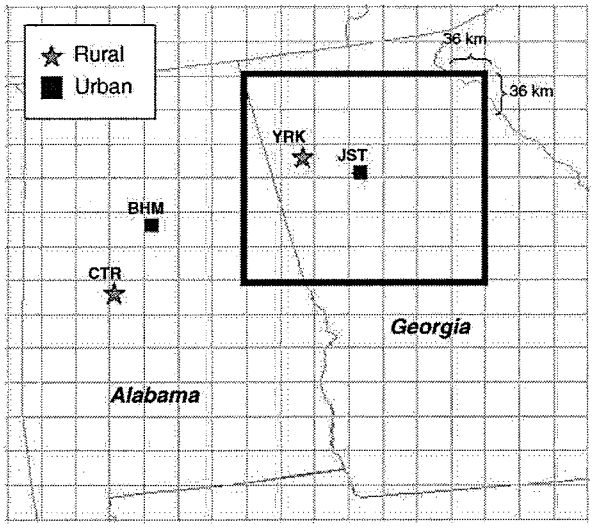


Fig. 1. Locations of SEARCH monitoring sites in Georgia (JST and YK) and Alabama (BHM and CTR), over plotted by the grids (36 km) of the air-quality model (only a part of the 36 km domain is shown here). The rectangle that contains the YRK and JST sites is the sub-domain of the air-quality model with a grid size of 12 km (12 km grids are not shown here).

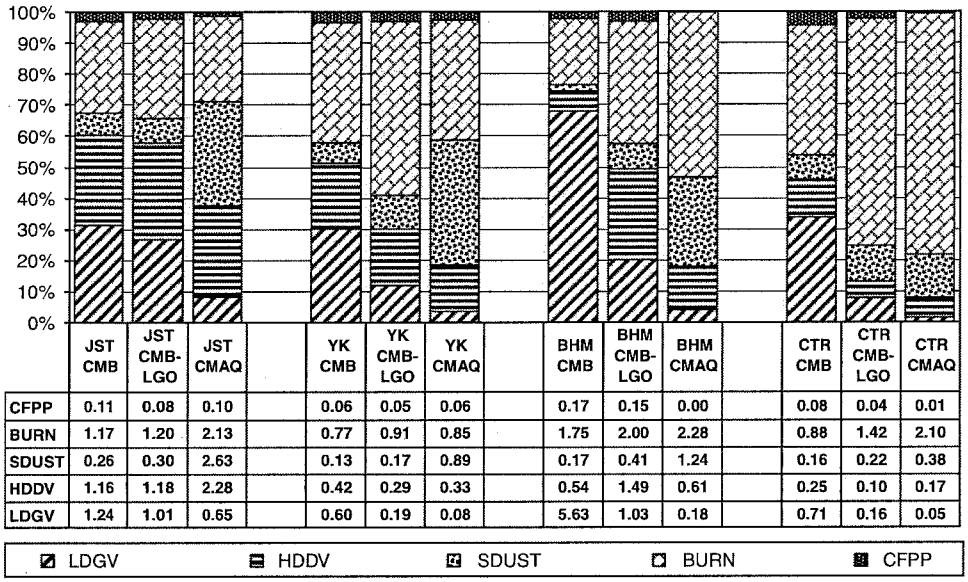
precursor gases. The major secondary $PM_{2.5}$ components are sulfate from the oxidation of SO_2 and secondary OC particles formed from volatile organic compounds emissions. The emphasis in this study is on sources of primary $PM_{2.5}$, i.e., PM emitted directly from emissions sources, due to the fact that receptor models are limited in their ability to link secondary compounds to emission sources (Burnett et al., 1998). We focused on primary $PM_{2.5}$ emissions from five source categories: gasoline vehicles, diesel vehicles, soil dust, vegetative/wood burning, and coal-fired power plants (in the CMB–LGO analysis these were noted as LDGV, HDDV, SDUST, BURN, and CFPP, respectively). These categories were previously identified as affecting the Atlanta airshed (Kim et al., 2004; Marmur et al., 2005; Zheng et al., 2002). Other categories, such as meat cooking, were not quantified due to lack of tracers and collinearity issues (Marmur et al., 2005), and were likely apportioned to the unexplained fraction of OC (which also includes secondary organic aerosol (SOA)). Regarding the major secondary $PM_{2.5}$ compounds, relating ambient sulfate concentrations to power-plant emissions is fairly straight forward since these account for the vast majority of SO_2 emissions (US-EPA, 1999). The sources and chemistry of SOA are still being investigated; biogenic compounds, such as monoterpene emissions from vegetation, are believed to be a major source of SOA (Carreras-Sospedra et al., 2005; Lim and Turpin, 2002). As the chemistry of SOA becomes better understood, air-quality models, such as CMAQ, may be able to provide a useful tool to assess the health outcomes associated with elevated levels of SOA.

For each one of the primary source categories investigated, emissions from SMOKE were tracked separately, and the air-quality model was applied six times: a base-case run including all sources, and five additional runs withholding one source category at a time (domain-wide exclusion) (Park et al., 2005, 2006b). A "source category" in this case is defined as a summation of emissions from numerous source classification codes from the emissions inventory. Specifically, the "gasoline" SMOKE category is a summation of emissions from on-road and off-road gasoline-engine powered vehicles; the "diesel" category is a summation of emissions from on-road and off-road diesel-engine powered vehicles; "dust" emissions are a summation of emissions from both paved and unpaved roads; "wood-burning" emissions are a summation of emissions from industrial and residential wood burning, prescribed burning and wildfires; "power-plant" emissions are a summation of emissions from all coal-fired boilers. Emissions from natural-gas combustion and meat charbroiling were tracked separately as well (Park et al., 2005, 2006b), but are not presented here, as they were not resolved by the receptor model (Marmur et al., 2005). Withholding emissions of primary $PM_{2.5}$ particles may shift secondary formation products to other particles, changing their size and deposition velocities and change their rate of coagulation with particles from other sources. However, a comparison of the results obtained by the brute-force method and a tracer method (where no sources are being withheld) showed very minor differences in the amount of mass apportioned to each source category and near perfect agreement in modeled daily variability of source impacts (Baek et al., 2005). CMB–LGO (Marmur et al., 2005) was also applied for the same time periods, using source profiles representing these five source categories (Chow et al., 2004b; Cooper, 1981; Zielinska et al., 1998). We compared the two models used here in terms of their ability to estimate long-term (monthly) and short-term (daily) source impacts, the degree of correlation between the various source categories, and the factors driving each model. In addition, we also addressed the issue of spatial variability in source impacts using concentrations of major $PM_{2.5}$ tracers from an additional monitoring site in the Atlanta metropolitan area (South-Dekalb (SDK) site).

3. Results

SA results were analyzed for average source contributions, daily variability, and factors/species driving the apportionment process. Source impacts at four sites were studied using CMB–LGO and CMAQ: Jefferson Street (JST), an urban site in Atlanta, GA; Yorkville (YK), a rural site in northwestern GA; Birmingham (BHM), urban site in Birmingham, AL; and Centerville (CTR), a rural site in AL (Fig. 1). We report the calculated source contributions using CMB–LGO and CMAQ, as well as those using regular CMB (without incorporating the gaseous data as in CMB–LGO) for reference.

When analyzing these results for use in a time-series health study, it is important to consider two aspects: temporal (daily) variation and spatial representativeness. Inaccuracies in either introduce



	JST CMB	JST CMB-LGO	JST CMAQ		YK CMB	YK CMB-LGO	YK CMAQ		BHM CMB	BHM CMB-LGO	BHM CMAQ		CTR CMB	CTR CMB-LGO	CTR CMAQ
CFPP	0.11	0.08	0.10		0.06	0.05	0.06		0.17	0.15	0.00		0.08	0.04	0.01
BURN	1.17	1.20	2.13		0.77	0.91	0.85		1.75	2.00	2.28		0.88	1.42	2.10
SDUST	0.26	0.30	2.63		0.13	0.17	0.89		0.17	0.41	1.24		0.16	0.22	0.38
HDDV	1.16	1.18	2.28		0.42	0.29	0.33		0.54	1.49	0.61		0.25	0.10	0.17
LDGV	1.24	1.01	0.65		0.60	0.19	0.08		5.63	1.03	0.18		0.71	0.16	0.05

Fig. 2. Average source contributions ($\mu g\,m^{-3}$) to primary $PM_{2.5}$ over a 2-month period (July 2001 and January 2002) of gasoline vehicles (LDGV), diesel vehicles (HDDV), soil dust (SDUST), vegetative/wood burning (BURN), and coal-fired power plants (CFPP) at four SEARCH sites in Georgia and Alabama.

errors in the epidemiologic analysis. The temporal (daily, in this case) variation in source impacts is the major factor driving a time-series health study, as short-term health effects are sought (other trends in the data, such as seasonal effects, are controlled for). For other purposes, such as air-quality management and control strategy development, longer-term results (e.g., seasonal) can be used. Obtaining such results involves less uncertainty; as demonstrated above (Fig. 2), the two approaches agree reasonably well on a monthly average basis, but differences in the daily source impacts are evident (Figs. 3–6). Spatial representativeness of air-quality data is an important issue when such data are used in health and exposure studies. This is especially true in this type of application because the spatial heterogeneity of source impacts is likely higher than that of individual species. Though there are not many data available to fully investigate this issue, consider the extreme example of two different but constant sources, both emitting significant amounts of the same pollutant (e.g., OC from wood burning and meat charbroiling), each located next to a monitoring site. Both sites will show an OC impact and likely follow similar meteorological trends, resulting in high correlations between OC levels at the two sites. However, the dominant source contributing to the OC is completely different. This need for increased temporal and spatial accuracy places greater burdens on the SA approach than might be demanded for air-quality management that focuses on reducing annual averaged levels.

3.1. Average source impacts

Analyzing the average source contributions at the four sites examined (Fig. 2), biomass burning appears to be a major source of primary $PM_{2.5}$ in the region, with contributions ranging between 27% and 77% (higher fractions in the rural sites). Average wood-burning contributions obtained using the three techniques were fairly similar at YRK and BHM, while CMAQ values were significantly higher at CTR and JST. Calculated average source contributions of primary $PM_{2.5}$ from coal-fired power plants are small, $<4\%$ for all sites using all techniques, and the average values obtained using the various techniques are in good agreement. Calculated impacts from diesel vehicles were quite different at JST (CMAQ value higher) and BHM (CMB–LGO value higher). The average contribution ranged between 5% and 31% of the primary $PM_{2.5}$, with higher fractions at the urban sites. Major differences were observed for the gasoline-vehicle primary $PM_{2.5}$ contributions, with consistently higher values obtained by CMB compared to CMB–LGO. This is likely caused by some

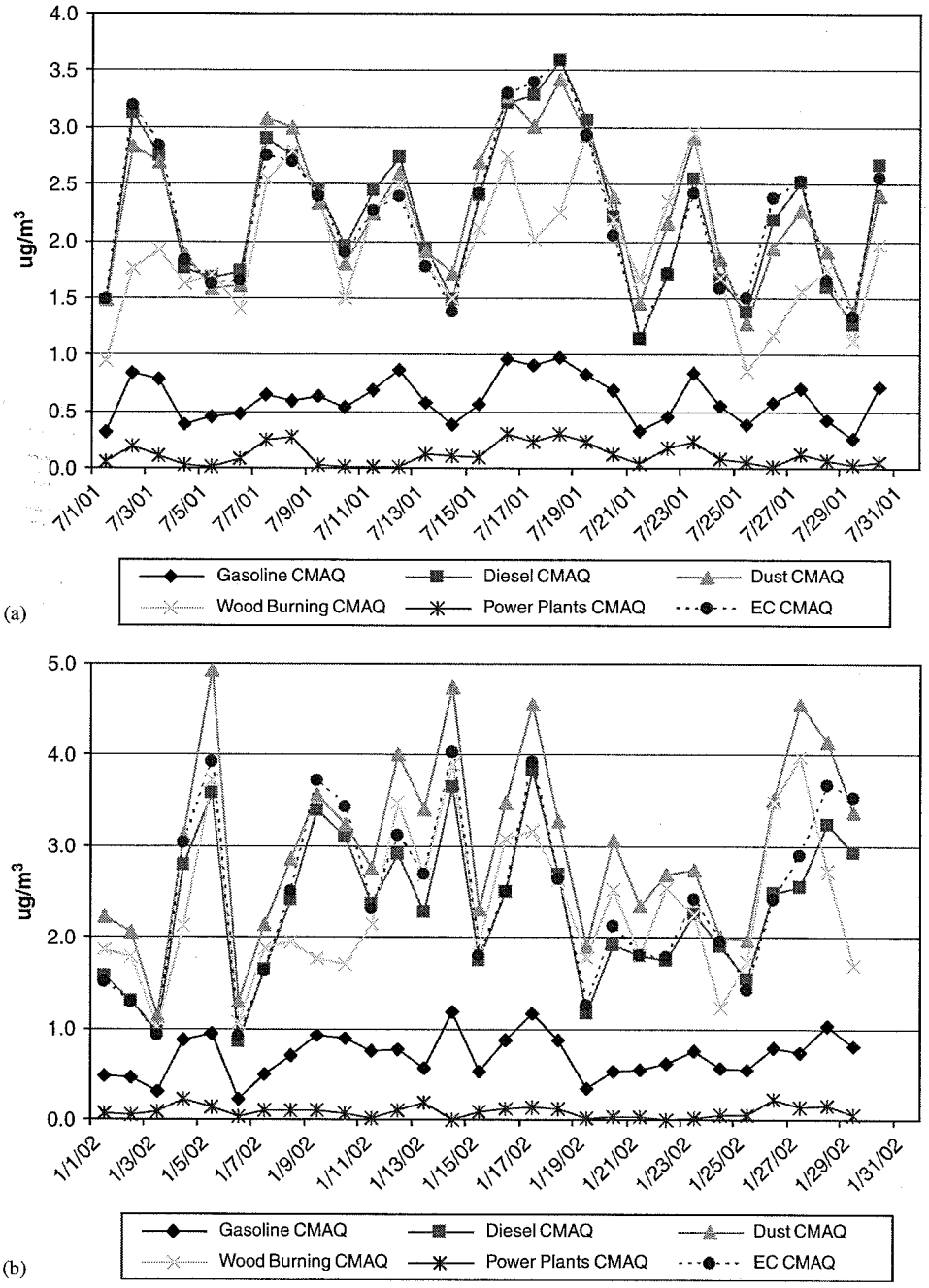


Fig. 3. Daily source-specific contributions ($\mu g\,m^{-3}$) to primary $PM_{2.5}$ at JST from gasoline vehicles, diesel vehicles, soil dust, vegetative/wood burning, and coal-fired power plants for the periods July 2001 (a) and January 2002 (b) using CMAQ. Also plotted are modeled EC levels at JST.

collinearity between the gasoline-vehicle source profile (OC fraction of 0.55) and the secondary/other OC profile (OC fraction of 1.0), included to account for secondary OC (and any OC not apportioned to one of the primary sources included in the analysis). Without bounding acceptable solutions based on the gaseous species, the optimal solution obtained by CMB contains a high gasoline-vehicle contribution, likely including some secondary OC formation. Such a high gasoline-vehicle contribution would suggest much higher ambient CO concentrations should be present than are

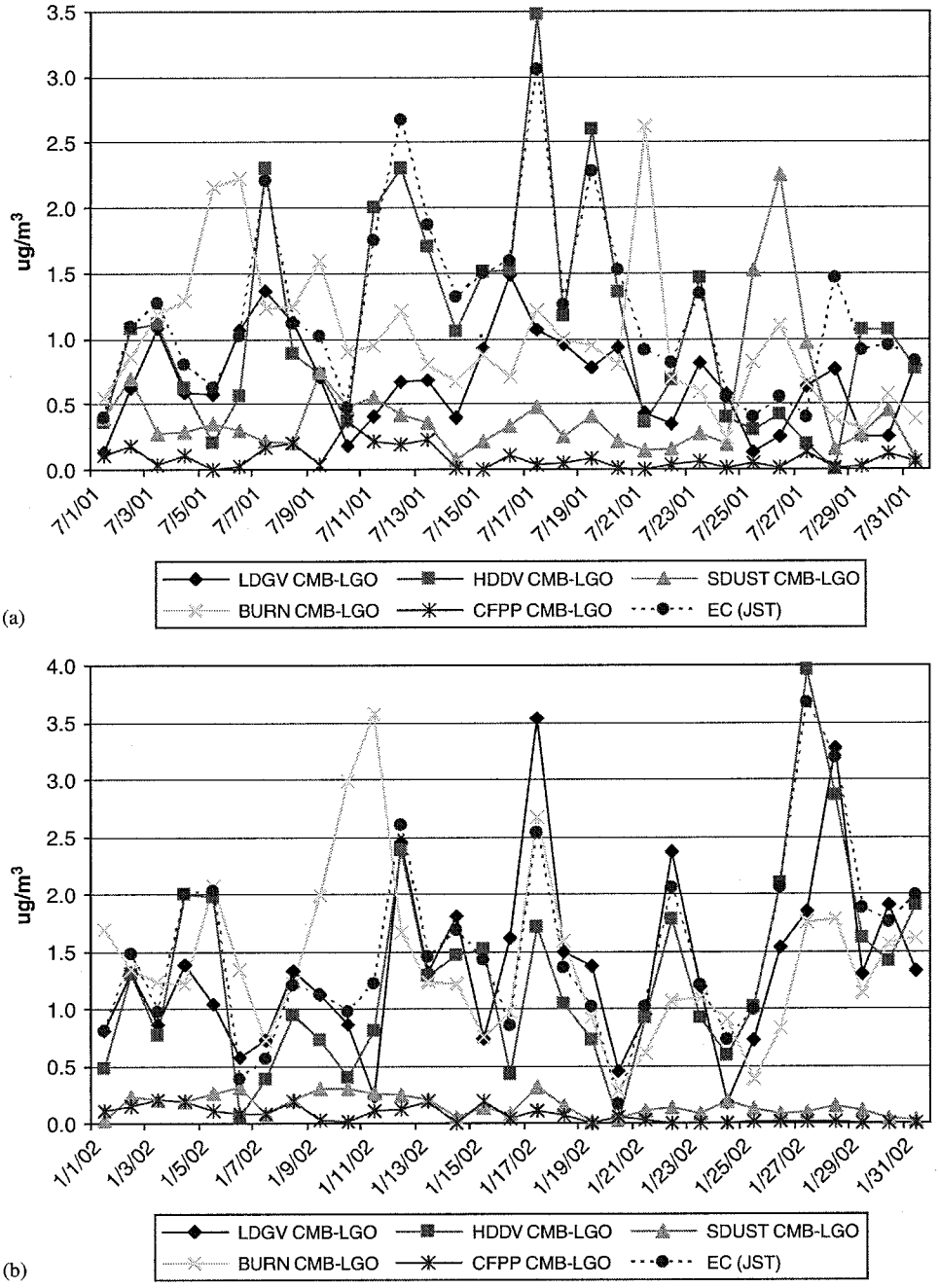


Fig. 4. Daily source-specific contributions ($\mu g\,m^{-3}$) to primary $PM_{2.5}$ at JST from gasoline vehicles (LDGV), diesel vehicles (HDDV), soil dust (SDUST), vegetative/wood burning (BURN), and coal-fired power plants (CFPP) for the periods July 2001 (a) and January 2002 (b) using CMB–LGO. Also plotted are measured EC levels at JST.

measured. When the acceptable solutions are bounded by CO, as done in CMB–LGO, lower gasoline-vehicle contributions and higher secondary/other OC contributions are obtained, 3.1 $\mu g\,m^{-3}$ on average, compared to 1.2 $\mu g\,m^{-3}$ using regular CMB. The solutions obtained by CMB indicate that even at the rural sites (YK and CTR), the contributions from mobile sources comprise approximately 50% of the primary $PM_{2.5}$, and that the gasoline-vehicle contribution at BHM is nearly 70% of the primary $PM_{2.5}$, with an extremely high gasoline-to-diesel ratio of 10.4. On the other hand,

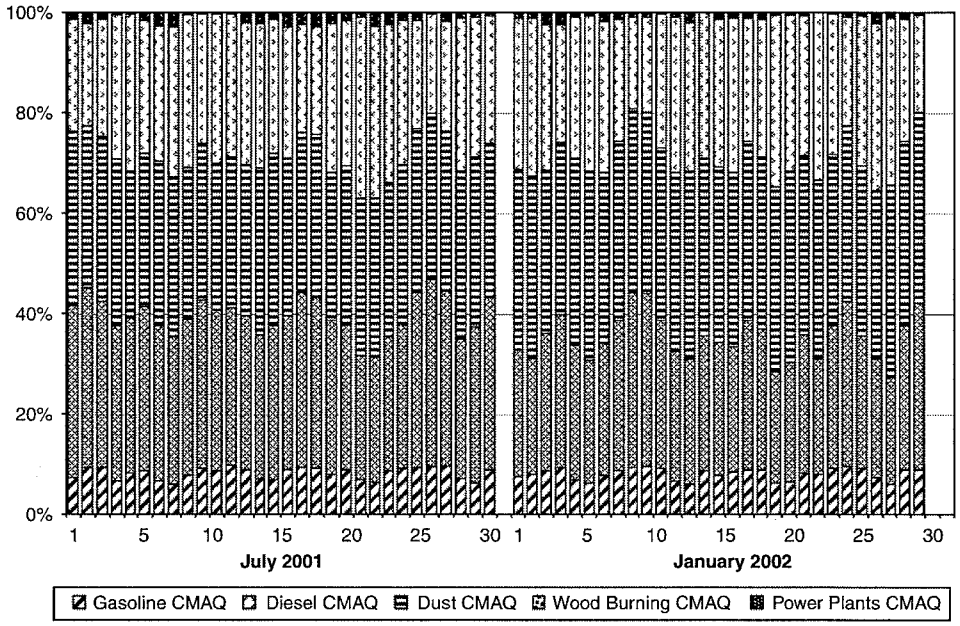


Fig. 5. Daily fractional source contribution (relative to overall contribution from the five categories examined) at JST using CMAQ.

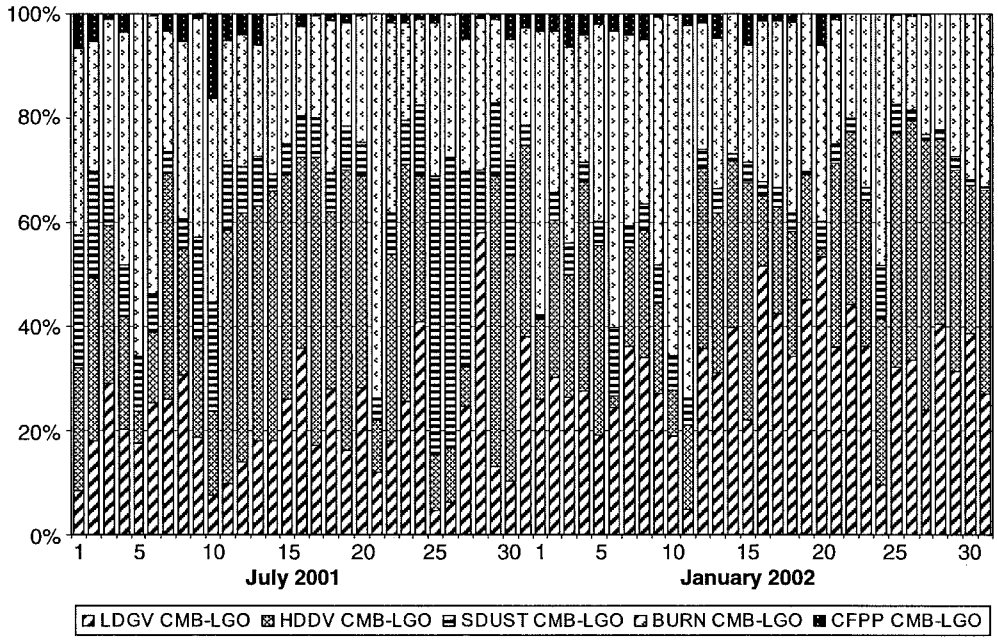


Fig. 6. Daily fractional source contribution (relative to overall contribution from the five categories examined) at JST using CMB–LGO.

solutions obtained by CMB–LGO and CMAQ indicate that the gasoline-to-diesel ratio is <1 and that wood burning is the major source of primary $PM_{2.5}$ at the rural sites. These results demonstrate the collinearity problem often encountered with regular CMB SA. Significant differences in the soil-dust contribution are observed when CMAQ estimates are compared to the CMB and CMB–LGO results. CMAQ soil-dust impacts seem to be extremely overpredicted, up to 10 times compared to the receptor-based results (the ambient data shows much lower levels of crustal elements

than is modeled using CMAQ). This is a well-known issue in CMAQ, where resuspended dust is assumed to be uniformly vertically mixed in the bottom layer of the model grid, while in practice, much of the resuspended dust is removed locally by impaction to surfaces (vehicles, leaves, etc.). Often a 75% removal factor (DRI, 2000) is applied for soil dust (as in this case); however, soil-dust impacts are still overpredicted.

3.2. Conceptual differences between CMB and CMAQ

To illustrate the conceptual differences between SA from a receptor model and a 3-D air-quality model, we will focus on the results from CMB–LGO and CMAQ for the Atlanta urban site (JST). Different source impacts using CMAQ follow a similar day-to-day trend (Fig. 3), driven mainly by meteorology (mixing height and ventilation). This trend is also similar to the temporal trend in modeled EC levels (also shown in Fig. 3), which further indicates the strong effect of meteorology on the temporal variation. CMB–LGO results, on the other hand, exhibit less correlated source-category trends (Fig. 4). In the case of CMB–LGO, the trends in the diesel source-category and measured EC levels (also shown in Fig. 4) are similar, but these trends are different than the trends of other source categories. These differences between the daily trends in CMAQ and CMB–LGO are more clearly shown when the fraction of each source category's contribution to the total is plotted as a time series (Figs. 5 and 6). CMAQ SA results show little variation in relative source impact while CMB–LGO results show substantial variation.

Another useful way of interpreting these results is by means of a correlation matrix (R-values in Table 1). Relatively high correlations between the CMAQ source categories and low correlations between the CMB–LGO categories are observed. As an example, in terms of the health study, the high correlation (0.94) between the gasoline and diesel categories using CMAQ might limit the ability to distinguish between the health outcomes of these two categories (even if these were a true reflection of the source impacts), while the results from CMB–LGO (significantly lower correlation, $R = 0.54$) will allow such differentiation (even if these variations were mainly a reflection of collinearity). These correlations (Table 1) also indicate the level of agreement between the two techniques.

Table 1
Correlations (R) between CMB–LGO and CMAQ-based source contributions to $PM_{2.5}$ at JST

	LDGV (CMB–LGO)	HDDV (CMB–LGO)	SDUST (CMB–LGO)	BURN (CMB–LGO)	CFPP (CMB–LGO)	Gasoline (CMAQ)	Diesel (CMAQ)	Dust (CMAQ)	Wood burning (CMAQ)	Power Plants (CMAQ)
LDGV (CMB–LGO)	1.00									
HDDV (CMB–LGO)	0.53	1.00								
SDUST (CMB–LGO)	−0.32	−0.16	1.00							
BURN (CMB–LGO)	0.31	0.10	−0.04	1.00						
CFPP (CMB–LGO)	−0.13	0.06	0.10	−0.06	1.00					
Gasoline (CMAQ)	0.58	0.52	−0.05	0.28	−0.01	1.00				
Diesel (CMAQ)	0.49	0.52	0.03	0.27	0.05	0.94	1.00			
Dust (CMAQ)	0.70	0.59	−0.24	0.30	−0.06	0.85	0.85	1.00		
Wood burning (CMAQ)	0.62	0.61	−0.33	0.18	−0.02	0.67	0.63	0.86	1.00	
Power Plants (CMAQ)	0.25	0.42	−0.10	−0.10	0.16	0.40	0.49	0.39	0.38	1.00

The sources of the discrepancy include errors of both CMAQ and CMB results. However, these have different magnitudes with respect to different sources. Correlations between the corresponding source impacts using the two techniques are poor for soil dust (−0.24), wood burning (0.18), and power plants (0.16), and more reasonable for the mobile sources (0.58 for gasoline vehicles, 0.52 for diesel vehicles, and 0.59 for the sum of these two categories).

One of the major sources of possible error in results from CMAQ comes from the uncertainty in emissions estimates. Emissions used as input to CMAQ have little daily variation (Fig. 7). Soil-dust and wood-burning $PM_{2.5}$ daily emissions are assumed to be constant (except for power plants on the 4 July and New Year's Day holidays). Mobile-source and power-plant emissions are given a weekly trend, with emissions modified on the 4 July and New Year's Day holidays. In reality, soil-dust emissions would depend on wind speed, humidity and recent rain fall, all having a significant effect on the amount of soil resuspended into the air (Hien et al., 2002). Stronger winds will lead to increased resuspension of dust (though this also leads to increased mixing of the atmosphere, lowering concentrations of pollutants). On the other hand, relatively small amounts of dust will be resuspended following rain events. Such effects are not captured by SMOKE, which can explain the low correlation with the CMB–LGO results (−0.24). Also, wood burning is a source with varying activity, including prescribed and agricultural burning, residential wood combustion, and industrial use of wood bark as fuel, that is not captured by SMOKE (unless specific information about wild fires and prescribed burning is incorporated). The constant emission rate used might explain the low

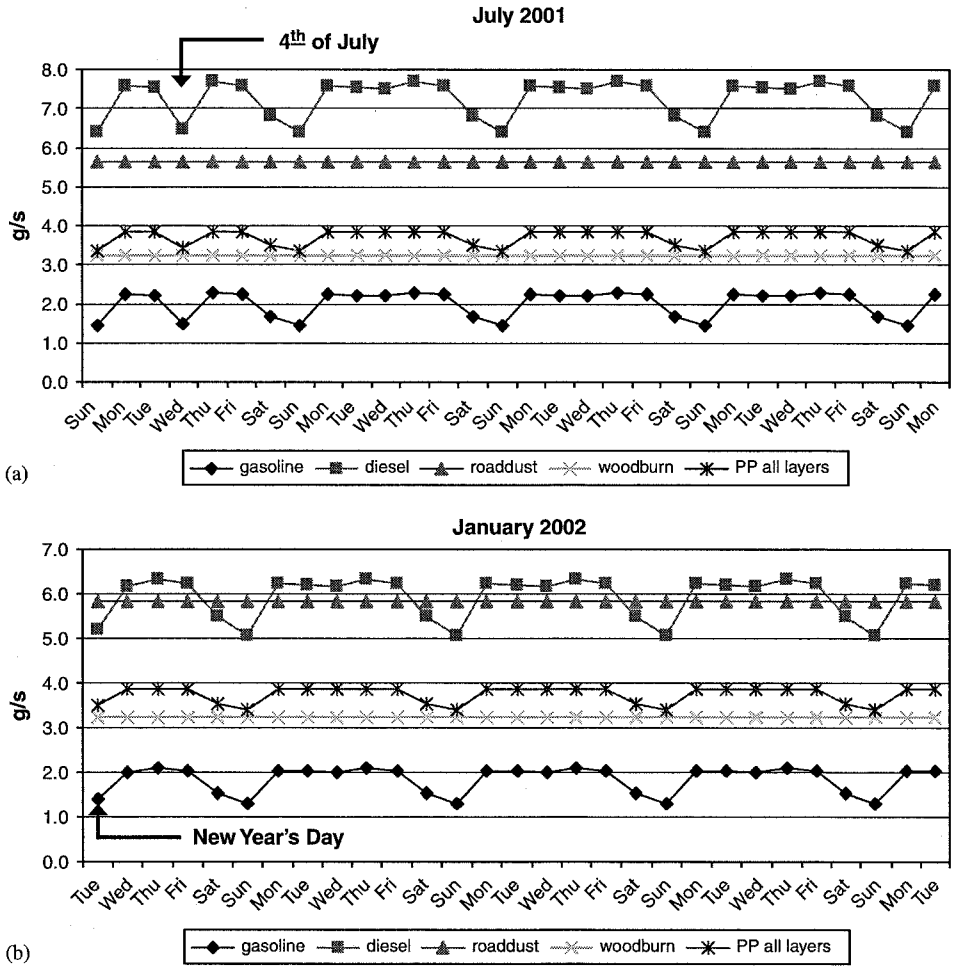


Fig. 7. Average daily modeled emissions in the model cell where the JST site is located.

correlation with the receptor model results (0.18). The differences between the variations in power-plant contributions may partially be due to a relatively coarse plume characterization using a 12 km grid (and a 36 km grid for the Alabama sites), though uncertainties in the Se measurement and source profile fraction may contribute to the discrepancy as well. The temporal pattern of mobile-source emissions has been studied in detail (Sawyer et al., 2000). Therefore, actual mobile-source emissions are probably more like the typical trends used in the model than the emissions from other sources. So it is not surprising that the correlations for the mobile sources are significantly higher (0.59 for the sum of the mobile sources). Further, mobile-source emissions are more ubiquitous, so a specific local source will have a smaller effect. Results from the other three sites studied, BHM, CTR, and YK, show similar trends in differences between CMAQ and CMB–LGO results, as shown by correlation matrices (Tables 2–4).

When comparing the results it is important to recognize some of the limitations and issues arising from the use of CMB–LGO, contributing to the discrepancy between the two sets of results. First, as previously mentioned, there is some collinearity between the source categories, introducing more fluctuations in source impacts than is likely true. In addition, CMB–LGO is limited by the availability of ambient data. One such example is the zero contribution of diesel vehicles on 28 July 2001 estimated using CMB–LGO (see Fig. 4), which is unlikely. This "error" is the result of there being no EC measurement on that day, so EC concentration was estimated as the monthly average. The high uncertainty associated with that value resulted in a low EC weighting in the error function; thus, EC levels were very poorly reconstructed for that day (17% of the filled-in value) and nearly no mass was apportioned to the HDDV category. There are also issues pertaining to the use of temporally constant source profiles in CMB applications. For instance, $PM_{2.5}$ composition from mobile sources depends on driving mode (Shah et al., 2004), and the composition of $PM_{2.5}$ from wood burning depends on the type of wood and the burning practices (Chow et al., 2004b). Also, any receptor-based analysis is driven by point measurements, with very local influences. Finally, measurement error of $PM_{2.5}$ components introduces noise to the apportionment process.

Table 2
Correlations (R) between CMB–LGO and CMAQ-based source contributions to $PM_{2.5}$ at YK

	LDGV (CMB–LGO)	HDDV (CMB–LGO)	SDUST (CMB–LGO)	BURN (CMB–LGO)	CFPP (CMB–LGO)	Gasoline (CMAQ)	Diesel (CMAQ)	Dust (CMAQ)	Wood burning (CMAQ)	Power Plants (CMAQ)
LDGV (CMB–LGO)	1.00									
HDDV (CMB–LGO)	0.14	1.00								
SDUST (CMB–LGO)	−0.35	−0.18	1.00							
BURN (CMB–LGO)	0.10	−0.09	0.02	1.00						
CFPP (CMB–LGO)	0.33	0.11	−0.26	−0.07	1.00					
Gasoline (CMAQ)	0.47	0.64	−0.29	0.15	0.11	1.00				
Diesel (CMAQ)	0.42	0.57	−0.29	0.24	0.15	0.97	1.00			
Dust (CMAQ)	0.57	0.41	−0.31	0.29	0.15	0.83	0.88	1.00		
Wood burning (CMAQ)	0.33	0.29	−0.26	0.26	0.03	0.66	0.72	0.77	1.00	
Power Plants (CMAQ)	0.13	0.45	−0.17	0.10	0.00	0.70	0.70	0.56	0.28	1.00

Table 3
Correlations (R) between CMB–LGO and CMAQ-based source contributions to $PM_{2.5}$ at BHM

	LDGV (CMB–LGO)	HDDV (CMB–LGO)	SDUST (CMB–LGO)	BURN (CMB–LGO)	CFPP (CMB–LGO)	Gasoline (CMAQ)	Diesel (CMAQ)	Dust (CMAQ)	Wood burning (CMAQ)	Power Plants (CMAQ)
LDGV (CMB–LGO)	1.00									
HDDV (CMB–LGO)	0.65	1.00								
SDUST (CMB–LGO)	−0.03	0.08	1.00							
BURN (CMB–LGO)	0.20	0.16	0.11	1.00						
CFPP (CMB–LGO)	0.58	0.71	0.00	0.12	1.00					
Gasoline (CMAQ)	0.42	0.52	−0.07	0.31	0.37	1.00				
Diesel (CMAQ)	0.39	0.48	−0.05	0.29	0.35	0.97	1.00			
Dust (CMAQ)	0.50	0.50	−0.21	0.22	0.41	0.86	0.88	1.00		
Wood burning (CMAQ)	0.38	0.47	−0.02	0.28	0.39	0.76	0.80	0.79	1.00	
Power Plants (CMAQ)	—	—	—	—	—	—	—	—	—	—

Table 4
Correlations (R) between CMB–LGO and CMAQ-based source contributions to $PM_{2.5}$ at CTR

	LDGV (CMB–LGO)	HDDV (CMB–LGO)	SDUST (CMB–LGO)	BURN (CMB–LGO)	CFPP (CMB–LGO)	Gasoline (CMAQ)	Diesel (CMAQ)	Dust (CMAQ)	Wood burning (CMAQ)	Power Plants (CMAQ)
LDGV (CMB–LGO)	1.00									
HDDV (CMB–LGO)	0.36	1.00								
SDUST (CMB–LGO)	−0.27	−0.19	1.00							
BURN (CMB–LGO)	−0.12	−0.16	−0.02	1.00						
CFPP (CMB–LGO)	0.72	0.34	−0.20	−0.12	1.00					
Gasoline (CMAQ)	0.36	0.30	−0.23	0.25	0.47	1.00				
Diesel (CMAQ)	0.35	0.34	−0.23	0.28	0.47	0.94	1.00			
Dust (CMAQ)	0.57	0.44	−0.36	0.19	0.56	0.81	0.85	1.00		
Wood burning (CMAQ)	0.20	0.39	−0.22	0.29	0.20	0.65	0.64	0.74	1.00	
Power Plants (CMAQ)	0.18	−0.07	−0.09	0.01	0.28	0.48	0.46	0.25	0.10	1.00

Table 5
Correlations (R) between source contributions and ambient species concentrations measured at JST

	CMB–LGO					CMAQ				
	LDGV	HDDV	SDUST	BURN	CFPP	Gasoline	Diesel	Dust	Wood Burning	Power Plants
EC	0.67	0.94	−0.22	0.25	0.00	0.56	0.55	0.64	0.64	0.36
OC	0.69	0.76	−0.25	0.34	0.13	0.48	0.45	0.58	0.57	0.30
Al	−0.23	−0.12	0.96	−0.02	0.06	−0.03	0.03	−0.18	−0.25	−0.07
As	0.21	0.23	−0.01	0.15	0.34	0.15	0.14	0.19	0.15	−0.06
Br	0.59	0.40	−0.24	0.46	−0.02	0.41	0.40	0.46	0.43	0.29
Ca	−0.08	0.17	0.63	0.19	0.53	0.18	0.28	0.00	−0.05	0.00
Fe	0.37	0.42	0.60	0.29	0.18	0.42	0.44	0.30	0.16	0.15
K	0.46	0.24	0.09	0.71	−0.01	0.25	0.24	0.29	0.23	−0.04
Mn	0.19	0.29	0.43	0.14	0.24	0.42	0.41	0.20	0.15	0.24
Pb	−0.04	0.03	0.03	−0.03	0.51	−0.02	−0.02	−0.07	−0.06	−0.07
Se	−0.08	0.15	0.13	0.08	0.58	0.07	0.09	−0.05	−0.03	0.01
Si	−0.19	−0.10	0.98	0.03	0.19	0.02	0.10	−0.14	−0.26	−0.06
Ti	−0.12	0.00	0.90	0.09	0.15	0.07	0.12	−0.10	−0.23	−0.06
Zn	0.82	0.44	−0.32	0.41	−0.11	0.54	0.42	0.58	0.53	0.17
SO_2	0.41	0.24	−0.15	0.34	0.58	0.32	0.28	0.40	0.31	0.20
CO	0.81	0.67	−0.24	0.34	−0.06	0.58	0.53	0.77	0.67	0.31
NO_y	0.90	0.63	−0.23	0.33	−0.09	0.57	0.49	0.73	0.64	0.17

3.3. Temporal variation in source impacts

To assess the degree to which results from CMB–LGO and CMAQ track the temporal variation in expected source tracer species, we calculated the correlations between these modeled daily source impacts and the daily ambient species concentrations at JST (Table 5). CMB–LGO source impacts are correlated with the expected tracers: EC, OC, Zn, CO, and NO_y for gasoline and diesel vehicles; crustal elements (e.g., Si and Al) for the soil-dust contribution; potassium (K) for wood burning; selenium (Se) and SO_2 for power plants. CMAQ SA is not based on the ambient data, but a similar test gives additional insight to the factors driving the CMAQ SA results. All of the source impacts are most correlated with EC. EC is the most abundant component of $PM_{2.5}$ that is entirely primary and, therefore, is in part an indicator of atmospheric stability. (Other major $PM_{2.5}$ components, such as sulfate or OC, are either nearly entirely secondary in nature, or contain a large portion that is secondary.) Variations in CMAQ-based source impacts are mainly due to variations in meteorology, and more specifically to the results of meteorological modeling; hence, these impacts are mostly correlated with measured EC (R-values ranging between 0.36 and 0.64). Hogrefe et al. (2001) showed that meteorological models, and hence air-quality simulations based on those results, do not capture fine-scale temporal and spatial variations.

We also performed a sensitivity analysis for the CMB–LGO solution, changing the ambient concentrations of one $PM_{2.5}$ component by one ($\pm$) standard deviation of the log-normalized values per analysis. Results from this analysis (Table 6) indicate that the gasoline-vehicle contribution is most sensitive to Zn, CO, and NO_y concentrations. (Zn is present in the lubricating oil of both gasoline and diesel vehicles; however, its fraction in emissions from gasoline vehicles is higher.) The diesel vehicle category is most sensitive to EC. Soil dust is most sensitive to Si concentrations (Al was not included in the model error function, Marmur et al., 2005 and, therefore, was not included in the sensitivity analysis). Vegetative burning is most sensitive to K levels. The power-plant contribution is most sensitive to SO_2 and Ca. The sensitivity of the secondary/other OC category to OC levels, and the lack of such sensitivity in the other categories, indicates that OC is not a driver of the SA of primary $PM_{2.5}$ (OC is first apportioned to the sources of primary $PM_{2.5}$, and only thereafter any OC unaccounted for is apportioned to the other/secondary OC category; hence, any change in OC levels would affect the other/secondary OC category first).

Table 6
Change in average species concentrations and average source attributions corresponding to a decrease/increase of one standard deviation of the log-normalized species concentrations

Average conc. relative to base case ($-\sigma_{log}/+\sigma_{log}$)		Average source-attribution relative to base case ($-\sigma_{log}/+\sigma_{log}$)					
		LDGV	HDDV	SDUST	BURN	CFPP	Other OC
EC	0.55/1.81	1.04/0.89	0.34/2.13	0.99/1.02	0.98/1.00	1.03/0.98	1.06//0.91
OC	0.57/1.77	0.97/1.01	1.02/1.00	1.00/1.00	0.95/1.01	1.01/1.00	0.30/2.27
As	0.45/2.22	1.00/1.00	1.00/0.99	1.00/1.00	0.98/1.04	1.00/1.00	1.01/0.99
Br	0.48/2.07	1.00/1.00	1.01/0.99	1.01/0.99	0.94/1.06	1.00/1.00	1.02/0.98
Ca	0.57/1.76	0.98/1.02	1.01/0.98	1.01/0.97	0.98/1.02	0.72/1.44	1.01/0.99
Cu	0.31/3.20	0.99/1.03	1.00/0.99	1.00/1.00	1.00/1.01	1.00/1.00	1.00/0.99
Fe	0.60/1.67	0.98/1.00	1.01/1.00	0.91/1.00	1.01/1.00	0.99/1.00	1.00/1.00
K	0.63/1.59	1.01/1.00	1.06/0.90	1.01/0.98	0.69/1.47	1.03/0.96	1.09/0.86
Mn	0.46/2.18	1.00/1.00	1.00/1.00	0.95/1.07	1.00/1.00	1.00/1.00	1.00/1.00
Pb	0.34/2.93	0.99/1.04	1.00/0.99	1.00/1.00	1.00/1.01	1.00/0.99	1.00/0.99
Se	0.48/2.10	1.00/0.98	1.00/1.00	1.00/0.97	1.03/1.00	1.03/1.19	1.00/1.00
Si	0.41/2.46	1.01/0.97	0.99/1.02	0.40/2.07	1.02/0.96	1.04/0.84	0.99/1.01
Ti	0.64/1.55	1.01/0.99	1.00/1.00	0.94/1.08	1.00/1.00	0.99/1.01	1.00/1.00
Zn	0.53/1.88	0.62/1.23	1.12/0.93	1.05/0.96	1.00/1.01	1.22/0.95	1.07/0.95
SO_2	0.34/2.95	0.89/0.97	0.99/1.02	1.10/0.82	1.66/1.05	0.37/2.23	1.00/0.99
CO	0.58/1.72	0.72/1.08	1.14/0.97	1.03/0.99	0.99/1.00	1.19/0.96	1.05/0.98
NO_y	0.53/1.88	0.71/1.06	0.93/1.03	1.05/0.99	1.09/0.99	1.06/0.98	1.04/0.99

Table 7
Average and standard deviation of the ratio of ambient concentration (Conc.) to minimum detection limit (MDL)

Species	$(Conc./MDL)_{avg}$
As	1.82 ± 2.01
Ba	0.58 ± 0.25
Br	5.55 ± 3.26
Ca	4.15 ± 2.59
Fe	161 ± 89.1
K	8.68 ± 4.58
Mn	2.01 ± 1.43
Pb	4.02 ± 11.63
Se	2.11 ± 1.86
Si	9.91 ± 11.5
Ti	0.77 ± 0.56
Zn	14.5 ± 8.81

The higher the ratio, the more accurate the measurement.

It is important to analyze the measurement accuracy of the driving species, especially in the case of trace level metals, to ensure that they are significantly above the detection limit. Average ratios of ambient concentrations to the minimum detection limits for various trace metals (Table 7) indicate that Zn concentrations are typically well above the detection limit, adding confidence in the gasoline-vehicle source attribution; so are the concentrations of Si, K, and Ca, which were identified as key species in the attribution to the soil-dust, vegetative-burning, and power-plant categories, respectively. One interesting result of the sensitivity analysis was that the CFPP contribution was not very sensitive to Se levels, even though Se is a unique marker for coal combustion. However, the data in Table 7 indicate that the accuracy of the Se measurement is low compared to Ca, and in some cases Se concentrations may be near or lower than the detection limit. This explains why lowering the Se concentrations did not cause a reduction in the CFPP contribution (as the concentration approaches the detection limit, the weight in the error function is reduced), and why increasing Se concentration did cause an increase in this contribution (as Se levels increase above the detection limit, so does the weight in the error function).

In terms of the number of species influencing each category, if each category was driven by only one species, one might consider doing the health analysis using ambient concentrations of that species, without the apportionment into categories. However, most of these key species are not unique indicators of a single source category; they are present in emissions from several categories (e.g., Si and Ca in both soil-dust and power-plant emissions) and do not represent one specific source category. Some of the categories are driven by more than one species, such as Zn, CO, and NO_y for LDGV and SO_2 and Ca for CFPP, and others have "secondary"

driving species, such as Fe and Mn for SDUST and Br for BURN. Finally, SA results can be used to reveal which are the species most associated with various source categories, if one were to interpret an association with a species as an indication of a source-related health outcome.

3.4. Spatial representativeness

A major issue regarding the use of receptor-based SA results in health studies is the spatial representativeness of the site. For example, Ito et al. (2004) report that the temporal correlation of source impacts across three monitors in New York city, 2–6 miles apart, varied significantly for sources of primary PM. To address this issue for the Atlanta study, we also examined speciated $PM_{2.5}$ data from the EPA-STN (Speciation Trends Network) monitor at SDK, located 15.3 km south east of the JST site. The SDK site is located near the I-285 interstate ("perimeter") that encircles much of the Atlanta metropolitan area, while the JST site is located more towards the center of Atlanta, approximately 2 km east of the I-75/85 interstate (Fig. 8). As the STN and SEARCH networks differ with respect to the carbon analyses method, thermal optical transmittance (TOT) and thermal optical reflectance (TOR), respectively (Chow et al., 2004a), it is not possible to conduct a CMB analysis of the SDK data using the same source profiles used for the JST case (these were based on TOR carbon measurements). Also, SO_2 and CO were not monitored at the SDK site, so CMB–LGO cannot be used. Instead, we compared levels and variations in major tracers for the various categories at the two sites. Data from March 2001 to December 2002 were used (samples at SDK were collected every third day, so 220 samples were available for comparison). We looked at K as an indicator for wood burning, Si and Fe as indicators for soil dust, and Se for coal-fired power plants (even though the CFPP contributions were more sensitive to Ca concentrations, Ca is by no means a unique tracer for CFPP, and is often correlated with elements, such as Si and Fe, indicative of soil dust). There is no unique $PM_{2.5}$ marker to separate gasoline and diesel vehicles contributions. To evaluate mobile sources as a whole, EC seems to be the most suitable (OC is partially secondary), but the comparison between EC at JST and SDK will include some noise due to the two different techniques used (TOR and TOT). EC is also emitted from wood-burning and other processes, so it cannot be view entirely as mobile-source related. We also examine Zn data, as it was correlated with gasoline vehicles impacts at the other sites examined.

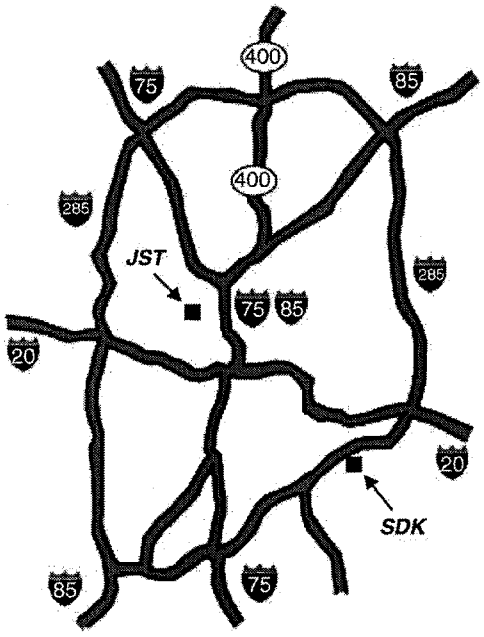


Fig. 8. Location of JST and SDK monitoring sites with respect to major interstates in Atlanta.

Potassium levels at JST and SDK are highly correlated (Fig. 9). This likely indicates a spatially homogeneous source (residential/industrial wood combustion) or distant plume sources (prescribed agricultural burning) hitting the two monitors similarly. Crustal elements (Si, Fe) are also highly correlated, indicating regional/global dust events, and/or soil moisture resuspension effects, assuming that rain events occur similarly at the two sites, and that soil moisture and wind speed have a strong influence on the resuspension of local dust. However, when high Si events are excluded, the correlation is lower, indicating local effects (though Fe correlations still remain high). Se is poorly correlated between the two sites, likely representing the directionality of impacts from power-plant plumes. This is also demonstrated in Wade et al. (2004), where SO_2 concentrations (as a power-plant marker) were the least spatially homogeneous of the primary gaseous pollutants. EC correlations are surprisingly high, considering the differences in measurement (TOR vs. TOT) and location in proximity to major highways. One possible explanation for the high correlation can be the role of atmospheric stability in daily variations of EC (and

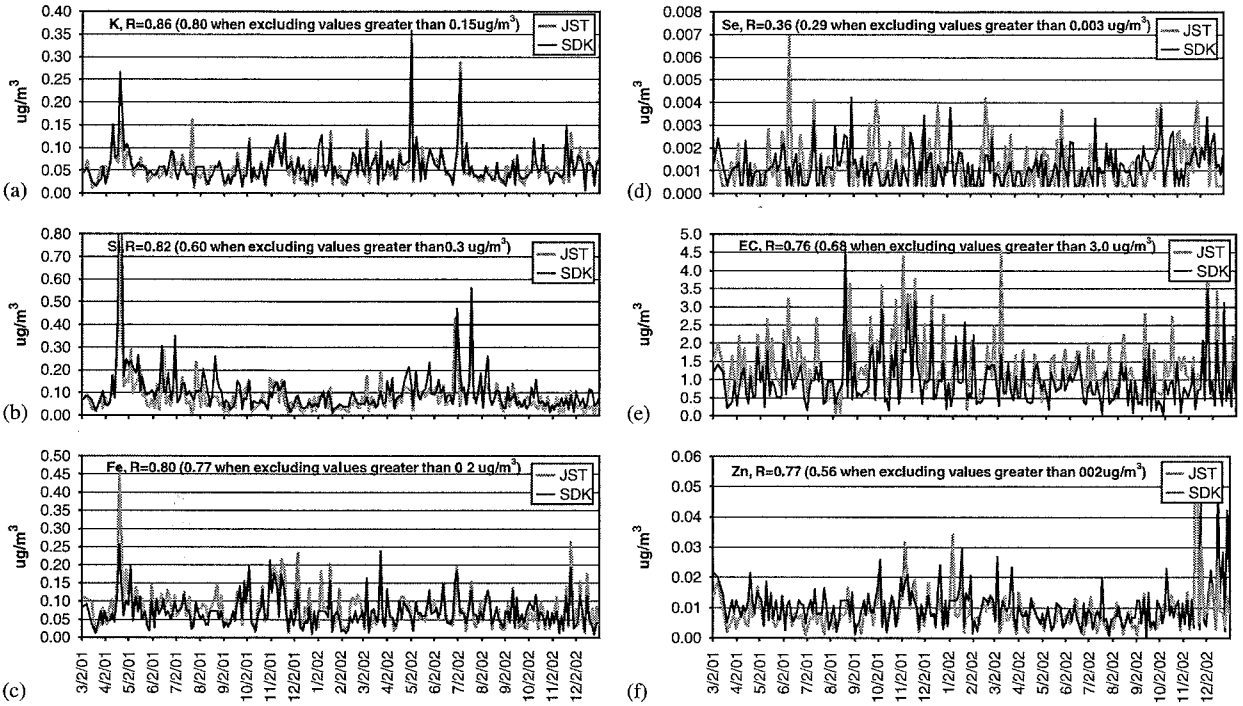


Fig. 9. Time-series comparison between ambient concentrations at two Atlanta monitoring sites located 15.3 km apart (SDK and JST) for K (a), Si (b), Fe (c), Se (d), EC (e), and Zn (f).

other pollutants) concentrations. Zn correlations are also relatively high, but drop when outliers are excluded. These findings are in overall agreement with data from Wade et al. (2004) in which CO (as a mobile-source tracer) measurements at three sites in Atlanta, 11.5–16.8 km apart, were correlated at levels of 0.65–0.76. Such results provide information about spatial variability of source impacts and site representativeness in the Atlanta area. Wood-burning and soil-dust contributions found at either site seem to be relatively spatially representative of the Atlanta urban area, as indicated by the correlations of potassium, silicon, and iron. Power-plant impacts seem to be local, based on the low inter-site selenium correlations. It is difficult to draw conclusions regarding the spatial representativeness of mobile-source impacts due to the lack of a unique marker and CO data. However, weighing both the EC and zinc correlations, it seems that mobile sources impacts are "intermediately" representative, i.e., likely more spatially representative than power-plant impacts, but less than wood-burning and soil-dust impacts. Note that these results are based on a preliminary analysis of two sites in Atlanta, and may represent a local phenomenon.

4. Discussion

Associating health outcomes with sources, rather than pollutants, may have several advantages relating to both the epidemiologic modeling process and the regulatory process. For such analyses, source impacts that capture both the temporal and spatial variability need to be generated. Receptor models, such as CMB–LGO, capture more of the temporal variation in source impacts at a specific receptor site, compared to emissions-based models, though this variation might be overestimated due to collinearity between sources. Being driven by data at a single site, receptor models may have some significant shortcomings with respect to spatial representativeness and exposure issues. SA results from emission-based models, such as CMAQ, may be more spatially representative, as they represent an average grid-cell value. However, limitations in the ability to model fine-scale meteorological fluctuations and daily fluctuations in emissions lead to results being driven mainly by regional meteorological trends (atmospheric stability), likely underestimating the true daily variations in source impacts.

The impact of a lack of spatial representativeness of estimated source impacts, anticipated in receptor modeling output, would likely introduce a bias to

the null in epidemiologic models (i.e., an attenuation of the observed association). The degree of spatial representativeness varies by source, and results for those sources with poor spatial representativeness (such as power plants) will have a greater degree of bias to the null. By characterizing the degree of spatial representativeness, investigators can take measures to handle this issue, such as reducing the study area included in analyses for the less representative sources, or possibly using data from several monitoring sites, if available. The impact of relatively limited capture of true day-to-day variation in the source impacts, anticipated to be more of an issue with emissions-based models than receptor models (though these may overestimate the temporal variation), is also likely to be a bias to the null in the epidemiologic models. In the case of emissions-based models, the fact that the day-to-day variation in the SA estimates is in large part a result of the meteorological conditions, and also influenced to a lesser extent by fixed day-of-week and seasonal patterns in the estimated emissions input, could lead to model instability, intractable confounding by meteorological conditions and temporal factors, such as day-of-week and season, as well as limited usefulness in discerning the relative impacts of the sources on health outcomes.

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