SOURCE APPORTIONMENT OF FINE ORGANIC AEROSOL USING CMAQ TRACERS

Jaemeen Baek*, Sun-Kyoung Park, Yongtao Hu, Armistead G. Russell
School of Civil and Environmental Engineering
Georgia Institute of Technology, Atlanta GA 30332
Corresponding author e-mail: gtg700i@mail.gatech.edu

ABSTRACT

Source apportionment of fine aerosol is essential to identify effective strategies to lower the particulate matter levels via efficient emission control strategies. For years, the chemical mass balance (CMB) receptor model has been used for source apportionment of fine organic aerosol, but many factors, such as uncertainties in measurement and incompleteness of source profile, limit accuracy of CMB. To complement these limitations, a 3-D photochemical model, CMAQ was used as an alternative approach to source apportionment.

One approach to conducting source apportionment using CMAQ is to use a Brute Force approach (CMAQ-BF), but it is computationally expensive as this method requires multiple runs. To overcome this drawback, tracers of organic and inorganic aerosols which account for the contributions from important sources were added to CMAQ. This method (CMAQ-TR) is computationally efficient because multiple source contributions can be calculated in one run of SMOKE and CMAQ. To evaluate usefulness of CMAQ-TR method, two results from CMAQ-TR and CMAQ-BF were compared and they agree well. Further usefulness of CMAQ-TR method was sought by quantifying ratios of local emissions to transported emissions in Atlanta, Georgia. Within the Atlanta region, transported emissions accounted for from 5% (meat cooking) to 99% (primary metal process) of the primary organic aerosol concentrations depending on source categories.

1. INTRODUCTION

There is increasing evidence that fine particles affect human health (Dockery and Pope 1994; Peel et al. 2002; Metzger et al. 2004). Effective control of particle levels requires identifying the relative importance of emission sources that contribute to particle concentrations. There are two major methods to address this problem; one is a receptor model and the other is an emissions-based air quality model.

Chemical mass balance (CMB) model with or without organic tracers is one of the more commonly applied receptor models. Initially, CMB relied on elemental concentrations as tracers. Recently, organic molecular markers have been used as tracer species (Schauer et al. 1996; Zheng et al. 2002; Schauer 2003; Zheng et al. 2005).

However, the accuracy of CMB results are limited by the accuracy of input data such as measured concentrations, source profiles, and completeness of sources treated. As an alternative approach, the Community Multiscale Air Quality model using a Brute Force method (CMAQ-BF) (Marmur et al. 2005; Park et al. 2005; Park et al. 2005) was used to achieve the same goal.

The drawback of CMAQ-BF is that it is computationally expensive as this method requires multiple runs: A Brute Force method is executed by repeated runs of CMAQ with different emission inventories and an equal number of emissions inventories to a number of source categories is needed.

Instead of a Brute Force method, using tracer species which can be used as fingerprints of specific sources is useful to decrease the computational burden. One example of using tracers is the CIT photochemical airshed model with tracers and has been applied to the source apportionment of fine aerosols and visibility impairment (Kleeman and Cass 2001; Mysliwiec and Kleeman 2002; Held et al. 2004; Ying et al. 2004; Held et al. 2005).

In this study, tracers for primary organic aerosols were added to CMAQ to apportion sources of fine organic aerosols in the United States. The CMAQ-TR method is computationally economical because multiple source contributions can be calculated in one run of CMAQ.

In addition to domain-wide source apportionment, quantifying contributions of specific

* *Corresponding author:* Jaemeen Baek. School of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta GA 30332. Corresponding author e-mail: gtg700i@mail.gatech.edu

local emissions to ambient pollutants is useful to estimate the effectiveness of control strategies because different control strategies are often applied by regions. Sensitivity analysis using a decoupled direct method (DDM) gives comprehensive aspects about regional impacts to air quality (Cohan 2004). But if we want to look at many source categories, sensitivity analysis becomes demanding because it needs as many emission inventories as source categories. Thus if a number of target regions are small, CMAQ-TR would be a handy way. To test usefulness of CMAQ-TR method for analysis of regional impacts, it was used to quantify the impact of Atlanta area to air quality within itself and outside of it.

Thus the objectives of this study are 1) to evaluate reliance on CMAQ-TR method in source apportionment of fine organic aerosol, 2) to apply CMAQ-TR method for simulating impacts of regional emissions on ambient organic aerosol concentration.

2. METHODS

Emissions inventories based on EPA national emission inventory (NEI) 1999 were processed with the Sparse Matrix Operator Kernel Emissions (SMOKE) version 1.5 (US EPA 2004), and meteorological fields were prepared with the NCAR's 5th generation Mesoscale Model (MM5) version 3.5.3 (PSU/NCAR 2003). For air quality modeling, CMAQ version 4.3 (Byun and Ching 1999; CMAS 2005) was used for both CMAQ-TR and CMAQ-BF methods.

Periods of simulation were July 1st to 31st, 2001 and January 1st to 31st, 2002 and domain was the continental United States and parts of Mexico and Canada with a 36km grid. The projection used is the unified Regional Planning Organization (RPO) national grid. More information of the air quality modeling system and the model evaluation results are available in elsewhere (Park, et al. 2005).

For a CMAQ-BF method, five source categories which are diesel exhaust, wood burning, road dust, meat cooking, and natural gas combustions were selected. Six emission inventories were created for five sources; one was the base case emissions inventory which includes every source and others were source specific emission inventories. In source specific inventories, emissions from the target source category was removed based on EPA source classification codes (SCC). SMOKE and CMAQ were run for six times with each of six emission inventories. The source contribution was defined as a difference between fine organic aerosol concentrations simulated in CMAQ with the base case inventory and those with the source specific inventories.

As a CMAQ-TR method, tracers were added in SMOKE and CMAQ in following ways. First, two sets of tracers were defined; one set included six source categories which are same categories as in CMAQ-BF except "other" category. The other group included thirty-two tracers which represent detailed source categories such as natural gas combustion in external boilers, natural gas combustion in internal engines, wild land fires, prescribed forest fires, and industrial processes. Six source categories were used for comparison with CMAQ-BF and thirty two categories were used for simulation of regional impacts of emissions from Atlanta.

Secondly, speciation process of fine particular matter (PM2.5) in SMOKE was modified. Different from CMAQ-BF, only one emissions inventory was used as a SMOKE input file and tracer information was added into two of SMOKE speciation files; a speciation profile and a speciation reference. Emissions of each of tracers were calculated by speciating PM2.5 total emissions into defined tracers in one run of SMOKE. A SMOKE output file contained separated emissions from each of source categories. Emissions of tracers are set to be one thousands of the original emissions to avoid possible interaction between tracers and other species.

In CMAQ, the tracers were treated as non-reactive species with same deposition properties as those of anthropogenic primary organic aerosol which were pre-defined in CMAQ.

To simulate contributions of emissions from the Atlanta region (as shown in Figure 4a) to ambient particle levels, emissions in corresponding cells to the Atlanta region were removed in the SMOKE output files and CMAQ was run twice; with emissions within the Atlanta region and without the Atlanta region. Impact of the Atlanta region was calculated by subtracting concentrations.

3. RESULTS

3.1. EVALUATION OF CMAQ-TR

The consistency of CMAQ-TR in estimating the impacts of sources was first assessed based on the results from CMAQ-BF for July 2001 and for January 2002. Daily organic aerosol

concentrations from CMAQ-TR were compared with those from CMAQ-BF at the Jefferson street

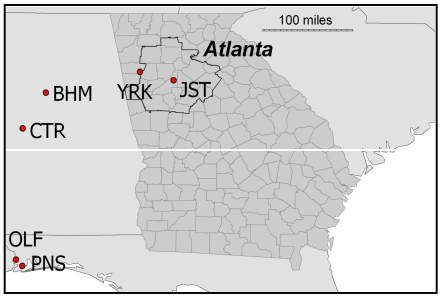

Figure 1. Map of SEARCH monitoring sites

(JST) station (Figure 1).

Mass concentrations of organic aerosol calculated using the two methods match well with each other (Table 1). Figure 2 shows that daily average concentration from each method and results from CMAQ-TR and those from CMAQ-BF agree well. Spatial distributions for the entire model domain match with each other as well (Figure 3). Contributions to ambient primary organic aerosol of wood burning, diesel exhaust, meat cooking were 45%, 10% and 14% in sequence.

Table 1. Monthly average contributions of each of source categories ($\mu g/m^3$). BF: Brute force method, TR: Tracer method

Source	2001. July		2002. January	
categories	BF	TR	BF	TR
Wood burning	0.92	0.91	1.87	1.85
Meat cooking	0.37	0.35	0.58	0.58
Road dust	0.22	0.21	0.39	0.39
Natural gas comb.	0.29	0.29	0.44	0.45
Diesel exhaust	0.38	0.38	0.40	0.41

Mean fractional error for 5 categories between CMAQ-BF and CMAQ-TR was 4.5% (July 2001) / 3.4% (January 2002) and overall mean fractional bias was 0.21% (July 2001) / 0.5% (January 2002) (Table 21). Slight differences between two results were mainly due to differences in emissions; while speciating tracer emissions in SMOKE, cut off of sixth decimal places occurred for some of speciation profiles.

3.2. IMPACT OF LOCAL EMISSIONS BY THE ATLANTA REGION

To discern how far emissions from the Atlanta region impact organic aerosol concentrations, we calculated source contributions at six SEARCH sites. Only Jefferson Street and Yorkville had significant impacts from Atlanta-based sources on primary PM levels, and other sites such as Birmingham (BHM) and Centerville (CTR) had less than a 2% influence of the Atlanta region. Table 3 shows monthly average impacts of organic particulate matter at JST and YRK.

Changes in total primary anthropogenic organic aerosol are shown in Figures 4b and 5a. The maximum decrease occurred at center of the Atlanta region and it spread over to nearby area. However, changes in separate source categories had different trends both in spatial distribution and quantities (Table 3).

Contributions of point sources in Atlanta were 19% at JST and 5% at YRK. Distillate oil combustion for electricity generation had a maximum impact at outside of Atlanta (Figures 4c and 5b). Industrial Asphalt roofing processes (Figures 4d and 5c) had a broader impact than the other sources.

Regional impacts of prescribed forest fires and wild fires at both sites were 50%, which meant that 50% of organic aerosol in the atmosphere came from outside of Atlanta. In July 2001 and January 2002, Florida and Alabama had high PM2.5 impacts from those two categories. Different from point sources and forest fires, most of the impact of PM2.5 emissions from mobile sources and area sources were local (Figures 4e, 4f, 5d and 5e). For these categories, the maximum contribution is located within Atlanta and it was almost 90%.

4 CONCLUSIONS

Comparison of CMAQ-BF and CMAQ-TR demonstrated consistency. Source apportionment results suggest that wood burning is the largest contributor to primary organic aerosol over Atlanta area and 50% of it was originated outside of Atlanta. Other important contributors to primary organic aerosol include meat cooking, diesel engines, which were local emissions.

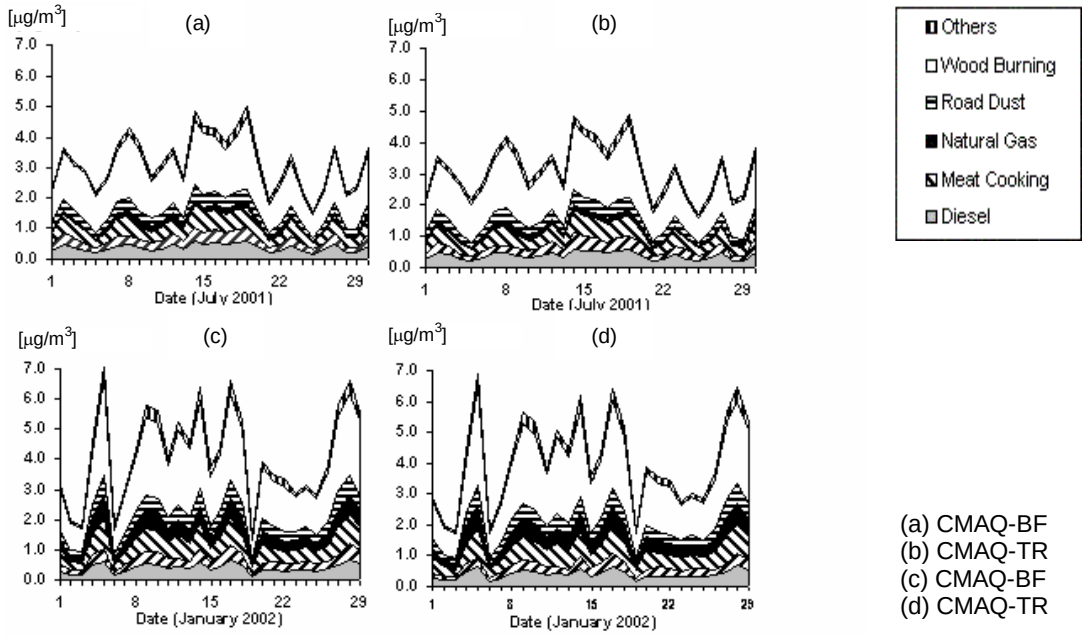

Figure 2. Daily average mass concentrations of primary organic aerosols simulated by CMAQ-BF and CMAQ-TR

Table 2. Comparison of simulated contributions between CMAQ-BF and CMAQ-TR by each source category. R^2 is a correlation coefficient between both methods. Statistics are calculated at Jefferson street site. Negative MFB means results from CMAQ-TR were lower than those of CMAQ-BF.

Date	STAT	Wood burning	Meat Cooking	Natural gas	Diesel	Road dust
2001, Jul	R^2	1.00	1.00	0.99	1.00	1.00
	MFB(%)	-0.63	-2.41	-1.70	0.44	-3.83
	MFE(%)	1.45	2.73	5.22	1.71	4.15
2002, Jan	R^2	1.00	1.00	1.00	1.00	1.00
	MFB(%)	-0.02	0.26	1.84	2.17	-1.69
	MFE(%)	2.68	2.92	3.95	2.68	3.96

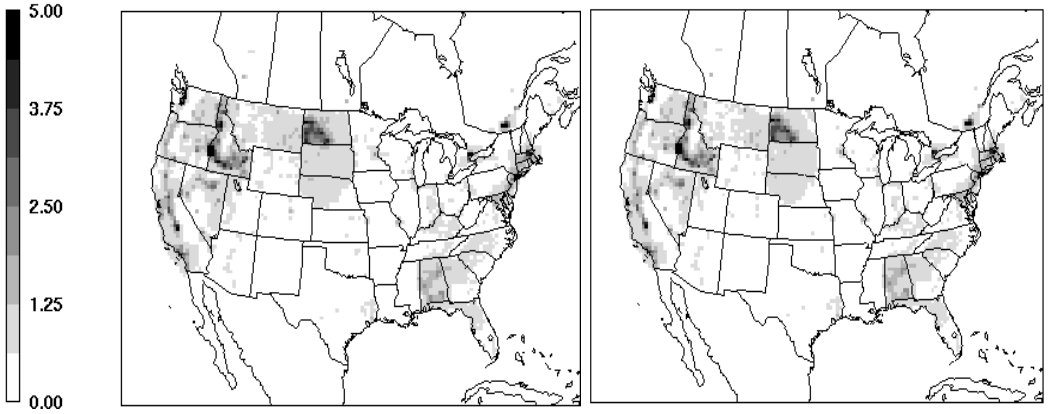

Figure 3. Daily mass contributions to ambient primary organic aerosols calculated using CMAQ-BF(left-hand side) and CMAQ-TR. January 2002. (μg/m^3)

Table 3. Contributions of the Atlanta region to ambient primary organic aerosol

		2001, July				2002, January			unit: μg/m^3
	Source types	Base case	Without Atlanta	difference (μg/m^3)	Contribution (%)	Base case	Without Atlanta	difference (μg/m^3)	Contribution (%)
JST	Point	0.18	0.15	0.03	19.0	0.22	0.18	0.04	18.4
	mobile	0.62	0.07	0.55	88.9	0.55	0.04	0.51	92.0
	forest fire	0.32	0.15	0.17	52.5	0.42	0.21	0.21	49.7
	Others	2.34	0.29	2.05	87.5	2.86	0.35	2.51	87.6
YRK	Point	0.19	0.18	0.01	5.8	0.23	0.22	0.01	4.8
	mobile	0.14	0.06	0.08	59.2	0.12	0.05	0.07	58.8
	forest fire	0.37	0.17	0.20	55.2	0.44	0.23	0.21	47.7
	Others	0.84	0.31	0.52	62.7	0.97	0.39	0.58	59.4

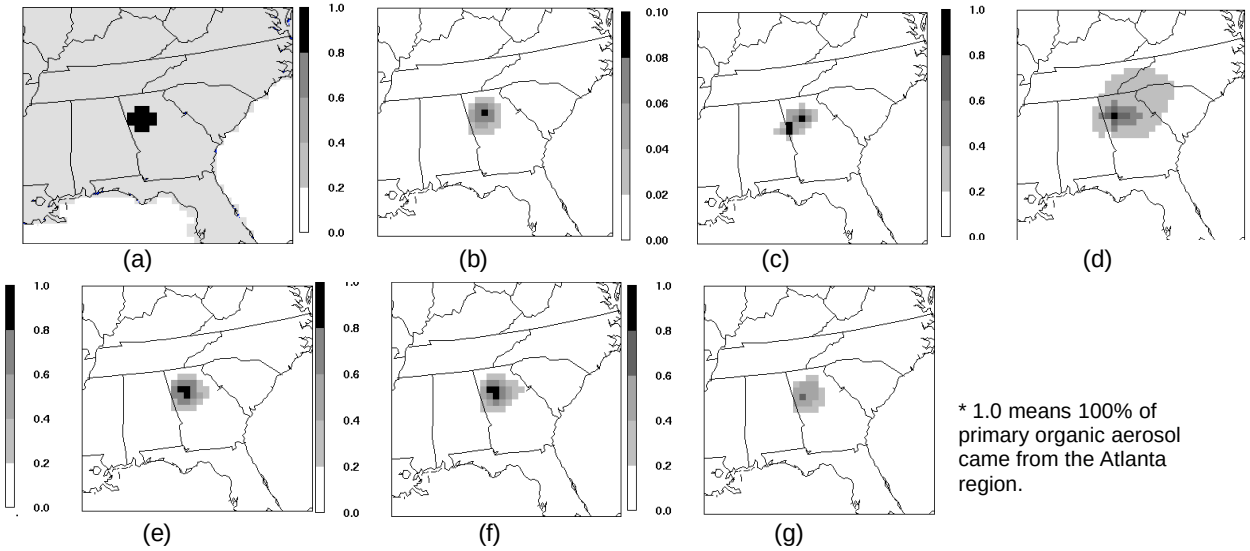

Figure 4. July 2001 monthly average contributions of emissions from the Atlanta region to ambient organic aerosol by each source category. (a) Black area indicates the Atlanta region, b) total primary anthropogenic organic aerosol c) distillate oil combustion in electricity generation, d) industrial process – asphalt roofing, e) diesel exhaust, f) meat cooking, g) wild land fire

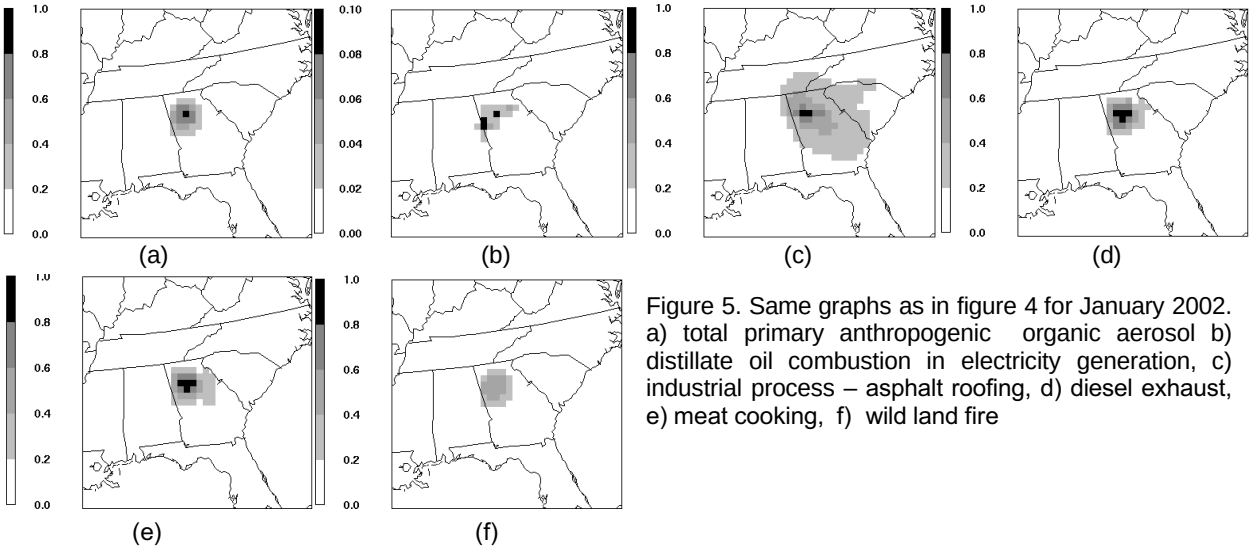

Figure 5. Same graphs as in figure 4 for January 2002. a) total primary anthropogenic organic aerosol b) distillate oil combustion in electricity generation, c) industrial process – asphalt roofing, d) diesel exhaust, e) meat cooking, f) wild land fire

REFERENCES

Byun, D. W., and J. K. Ching, 1999: *Science algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) modeling system.* Atmospheric modeling division, National Exposure Research Laboratory, U.S. Environmental Protection Agency.

CMAS, 2005. *Community modeling & analysis system*. http://www.cmascenter.org.

Cohan D. S., 2005: *Photochemical formation and cost-efficient abatement of ozone: high order sensitivity analysis.* PhD thesis. Georgia Institute of Technology.

Dockery, D. W., and C. A. Pope, 1994: Acute respiratory effects of particulate air-pollution. *Annual Review of Public Health,* **15**, 107-132.

Held, T., Q. Ying, A. Kaduwela, and M. Kleeman, 2004: Modeling particulate matter in the San Joaquin Valley with a source-oriented externally mixed three-dimensional photochemical grid model. *Atmospheric Environment,* **38**, 3689-3711.

Held, T., Q. Ying, M. J. Kleeman, J. J. Schauer, and M. P. Fraser, 2005: A comparison of the UCD/CIT air quality model and the CMB source-receptor model for primary airborne particulate matter. *Atmos. Environ. Accepted for publication.*

Kleeman, M. J., and G. R. Cass, 2001: A 3D Eulerian source-oriented model for an externally mixed aerosol. *Environmental Science & Technology,* **35**, 4834-4848.

Marmur, A., S-K. Park, J. Mulholland, and A. G. Russell, 2005: PM2.5 source apportionment using receptor and source-oriented models: conceptual differences and implications for health studies. *Proceedings of an AAAR international specialty conference -- Particulate matter supersites program and related studies, Atlanta, Georgia*.

Metzger, K. B., P. E. Tolbert, M. Klein, J. L. Peel, W. D. Flanders, K. Todd, J. A. Mulholland, P. B. Ryan, and H. Frumkin, 2004: Ambient air pollution and cardiovascular emergency department visits. *Epidemiology,* **15**, 46-56.

Mysliwiec, M. J., and M. J. Kleeman, 2002: Source apportionment of secondary airborne particulate matter in a polluted atmosphere. *Environmental Science & Technology,* **36**, 5376-5384.

Park, S.-K., C. E. Cobb, K. Wade, J. Mulholland, Y. Hu, and A. G. Russell, 2005a: Uncertainty in air quality model evaluation from spatial variation. *Submitted to Atmos. Environ.*

Park, S.-K., L., B. Ke, A. G. Russell and M. Zheng, 2005b: Source apportionment of PM2.5 using a three-dimensional air quality model and a receptor model. *Proceedings of an AAAR international specialty conference -- Particulate matter supersites program and related studies, Atlanta, Georgia*.

Park, S.-K., A. Marmur, L. Ke, B. Yan, M. Zheng, and A. G. Russell, 2005c: Comparison between chemical mass balance receptor and CMAQ model PM2.5 source apportionment. *Submitted to Environmental Science and Technology*.

Peel, J., P. Tolbert, M. Klein, K. Metzger, W. D. Flanders, K. Todd, J. Mulholland, P. B. Ryan, and H. Frumkin, 2002: Ambient air pollution and respiratory emergency department visits in Atlanta, August 1998 - August 2000 (ARIES/ SOPHIA). *Epidemiology,* **13**, S124-S124.

PSU/NCAR, 2003: *PSU/NCAR Mesoscale modeling system tutorial class notes and user's guide: MM5 modeling system version 3.* Mesoscale and microscale meteorology division, national center for atmospheric research.

Schauer, J. J., 2003: Evaluation of elemental carbon as a marker for diesel particulate matter. *Journal of Exposure Analysis and Environmental Epidemiology,* **13**, 443-453.

Schauer, J. J., W. F. Rogge, L. M. Hildemann, M. A. Mazurek, and G. R. Cass, 1996: Source apportionment of airborne particulate matter using organic compounds as tracers. *Atmospheric Environment,* **30**, 3837-3855.

US-EPA, 2004: *SMOKE User's manual.* http://cf.unc.edu/cep/empd/products/smoke.

Ying, Q., M. Mysliwiec, and M. J. Kleeman, 2004: Source apportionment of visibility impairment using a three-dimensional source-oriented air quality model. *Environmental Science & Technology,* **38**, 1089-1101.

Zheng, M., G. R. Cass, J. J. Schauer, and E. S. Edgerton, 2002: Source apportionment of PM2.5 in the southeastern United States using solvent-extractable organic compounds as tracers. *Environmental Science & Technology,* **36**, 2361-2371.

Zheng, M., L. Ke, F. Wang, G. R. Cass, J. J. Schauer, E. S. Edgerton, and A. G. Russell, 2005: Source apportionment of PM2.5 and their daily variations at Jefferson Street, Atlanta GA during Summer and Winter. *Journal of Air and Waste Association. Submitted for publication*.