

PM_{2.5} source apportionment in the southeastern U.S.: Spatial and seasonal variations during 2001–2005

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[1] The seasonal and spatial variations of source contributions of 112 composite fine particulate matter (PM_{2.5}) samples collected in the Southeastern Aerosol Research and Characterization Study (SEARCH) monitoring network during 2001–2005 using molecular marker-based chemical mass balance (CMB-MM) model were determined. The lowest PM_{2.5} concentration occurs in January with higher values in warm months (maxima in July at four inland sites versus October at the coastal sites). Sulfate shows a similar pattern and plays a primary role in PM_{2.5} seasonality. Carbonaceous material (organic matter plus EC) exhibits less seasonality, but more spatial variations between the inland and coastal sites. Compared with the data at coastal sites, source attributions of diesel exhaust, gasoline exhaust, other organic matter (other OM), secondary sulfate, nitrate, and ammonium in PM_{2.5} mass at inland sites are higher. The difference in source attributions of wood combustion, meat cooking, vegetative detritus, and road dust among the eight sites is not significant. Contributions of eight primary sources to fine OC are wood burning ($17 \pm 19\%$), diesel exhaust ($9 \pm 4\%$), gasoline exhaust ($5 \pm 7\%$), meat cooking ($5 \pm 5\%$), road dust ($2 \pm 3\%$), vegetative detritus ($2 \pm 2\%$), cigarette smoke ($2 \pm 2\%$ at four urban sites), and coke production ($2 \pm 1\%$ only at BHM). Primary and secondary sources explain 82–100% of measured PM_{2.5} mass at the eight sites, including secondary ionic species (SO_4^{2-} , NH_4^+ , and NO_3^- ; $41.4 \pm 5.7\%$), identified OM ($24.9 \pm 11.3\%$), “other OM” (unexplained OM, $23.3 \pm 10.3\%$), and “other mass” ($11.4 \pm 9.6\%$). Vehicle exhaust from both diesel and gasoline contributes the lowest fraction to PM_{2.5} mass in July and higher fractions at BHM and JST than other sites. Wood combustion, in contrast, contributes significantly to a larger fraction in winter than in summer. Road dust shows relatively high levels in July and April across the eight sites, while minor sources such as meat cooking and other sources (e.g., vegetative detritus, coke production, and cigarette smoke) show relatively small seasonal and spatial variations in the SEARCH monitoring network.

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1. Introduction

[2] Airborne fine particles with aerodynamic diameter less than $2.5 \mu\text{m}$ (PM_{2.5}) have been of universal concern due to potential adverse effects on human health [Pope *et al.*, 2002; Seagrave *et al.*, 2006]. In response, the U.S. Environmental Protection Agency (EPA) set the National Ambient Air Quality Standards (NAAQS) for ground-level PM_{2.5} at $15 \mu\text{g m}^{-3}$ for an annual average and $65 \mu\text{g m}^{-3}$ for a daily average in 1997, and further lowered the daily standard to $35 \mu\text{g m}^{-3}$ in 2006. Unlike other criteria air pollutants such as ozone, PM_{2.5} is usually comprised of hundreds of organic and inorganic compounds released from various primary sources or formed by chemical reactions in the atmosphere (secondary sources). Therefore, a detailed understanding of composition and sources of PM_{2.5} is needed to investigate the associations between specific components and health

effects and for policy-makers to develop effective control strategies.

[3] The Southeastern Aerosol Research and Characterization Study (SEARCH) was initiated as a multiyear program (since 1998) at eight monitoring sites in four states (GA, AL, MS, FL), and some of the objectives were to investigate the chemical composition, major sources and variability of PM_{2.5} over an expanded geographical coverage and long-term trends [Hansen *et al.*, 2003]. The major PM_{2.5} components, including elemental carbon (EC), organic carbon (OC), sulfate, nitrate, ammonium, trace metals, gaseous pollutants, and meteorological parameters were simultaneously measured at eight sites [Hansen *et al.*, 2003; Edgerton *et al.*, 2005, 2006]. These valuable data sets from the SEARCH program have been widely used by a variety of studies such as source identification of aerosol [Kim *et al.*, 2003], contribution of primary and secondary sources to organic aerosol and PM_{2.5} [Kleindienst *et al.*, 2010], and chemical mass balance (CMB) analysis using particle-phase organic compounds as tracers to quantitatively identify the primary sources of PM_{2.5} in samples from the SEARCH sites [Zheng *et al.*, 2002, 2006b, 2007]. This molecular marker-based CMB (CMB-MM) receptor model was originally developed and applied in the Los Angeles area and San Joaquin Valley of California [Schauer *et al.*, 1996; Schauer and Cass, 2000], and has been successfully applied in PM_{2.5} source apportionment in several regions around the world [Zheng *et al.*, 2002, 2005, 2006a, 2006b; Fraser *et al.*, 2003; Chow *et al.*, 2007; Yin *et al.*, 2010]. Compared to the inorganic-based CMB approach [Watson, 1984], CMB-MM has the advantage to identify some important sources which emit significant amounts of organic materials such as wood burning, meat cooking, diesel exhaust and gasoline-powered motor vehicle emissions [Schauer *et al.*, 1996].

[4] By combining the CMB model with the SEARCH monitoring data collected from four urban and four rural/suburban sites during April, July, and October 1999 and January 2000, Zheng *et al.* [2002] identified the major contributors to PM_{2.5} organic carbon concentrations in the southeastern United States. Then, Zheng *et al.* [2007] quantified the primary emission source contributions to daily PM_{2.5} mass concentrations for a summer month in 2001 and a winter month in 2002 in Atlanta, GA. The modeling results in these previous studies showed that the main contributors to PM_{2.5} mass included primary sources of wood burning, diesel and gasoline vehicle exhaust, meat cooking, road dust, and secondary sources (SO₄²⁻, NO₃⁻, and NH₄⁺).

[5] With multiyear source apportionment results of PM_{2.5} in this study, we are able to investigate the spatial and temporal variation patterns of PM_{2.5} sources in this important region of the U.S. In previous studies, Zheng *et al.* [2002] observed the distinct seasonality in source contributions, with higher contributions from wood burning in colder months and higher proportions of secondary organic aerosol formation in the summer season. Liu *et al.* [2005] analyzed the spatial differences and correlations with data sets of two pairs of urban-rural sites in the SEARCH network from January 2000 to December 2002. By using a one-year data set from May 2004 to April 2005, Ding *et al.* [2008] investigated the spatial and seasonal trends in biogenic secondary organic aerosol tracers and water-soluble organic carbon at one rural site and three urban sites located in the

southeastern United States. Distinct spatial distributions were observed for all tracers with the highest concentration at the rural site and the lowest level at a coastal site. This work is a follow-up to these previous studies, focusing on the spatial and seasonal variations based on a larger data set from eight sites during 2001 to 2005. To our best knowledge, this is the first study to reveal spatial and seasonal variations of PM_{2.5} sources based on such a multiyear data set and to investigate the trend of sources of PM_{2.5} in this region. The direct comparison of these PM_{2.5} source apportionment results from different sites can be made because samples were collected with the same type of sampler and analyzed by the same protocol and laboratory.

2. Experimental Methods

2.1. Sample Collection

[6] PM_{2.5} samples were collected at the four paired urban-rural or urban-suburban sites in the SEARCH monitoring network: Jefferson Street (JST), Atlanta-Yorkville (YRK) in GA; North Birmingham (BHM)-Centreville (CTR) in AL; Gulfport (GFP)-Oak Grove (OAK) in MS; and Pensacola (PNS)-Outlying Landing Field #8 (OLF) in FL (see Figure S1 in the Supporting Information). Detailed descriptive information about the eight sites was provided previously [Zheng *et al.*, 2002; Hansen *et al.*, 2003]. Briefly, the four urban sites are located in commercial/industrial/residential (JST, BHM), commercial/residential (GFP), and residential areas (PNS). The rural sites (CTR, OAK, and YRK) are located at least 50 km from major cities, while OLF is a suburban site located at about 20 km northwest of downtown Pensacola. All of these sites were selected to minimize the influence of local sources. The terrain in the southeastern area is generally hilly and is covered heavily by subtropical vegetation. All areas experience both controlled and uncontrolled wood combustion in the cooler seasons.

[7] Filter-based 24-h integrated PM_{2.5} samples were collected daily or every third day at the eight sites using a three-channel Particle Composition Monitor (PCM; Atmospheric Research & Analysis, Inc., Plano, TX) sampling system. A 37-mm-diameter quartz filter was used in Channel 3 (PCM3) at a flow rate of 16.7 L min⁻¹ downstream of a carbon denuder. One to three 0.5 cm² punches were cut from each PM_{2.5} filter for analysis of organic and elemental carbon (OC/EC) at the Desert Research Institute (DRI) using a thermal/optical reflectance (TOR) protocol [Chow *et al.*, 1993, 2001]. PM_{2.5} mass was measured with a Federal Reference Method (FRM) sampler (Rupprecht & Patashnick Model 2025 sequential sampler) on 47-mm diameter Teflon filters. Other measurements of PM_{2.5} composition such as ionic species (SO₄²⁻, NO₃⁻, NH₄⁺) were acquired from filter samples collected in other channels of the PCM. Back-up filter samples were applied for the correction of volatilization loss from the front filter, and regular field blank samples were collected to subtract potential contamination [Edgerton *et al.*, 2005].

[8] For organic speciation analysis, the remaining PM_{2.5} filters after taking punches for OC/EC analysis were combined into composite samples by site and by month in each year from July 2001 to January 2005. Four months (January, April, July, and October) were selected to represent the four

seasons. Totally, there are 14 composite samples at each site: 2001 (July), 2002–2004 (January, April, July, and October), and 2005 (January) (Table S1 in auxiliary material Text S1).¹ For the elimination of potential contamination from sampling and handling processes, field blanks from the eight sites were combined into five composite samples and analyzed along with other PM_{2.5} samples. Concentrations of PM_{2.5} mass and components (SO₄²⁻, NO₃⁻, NH₄⁺, Al, Si, OC, EC, and organic tracers) of each composite sample were calculated based on the mass and air volume sampled on the remaining filters.

2.2. Organic Speciation Analysis

[9] The analytical procedure for organic compounds in the composite PM_{2.5} samples was described in detail previously [Zheng *et al.*, 2002, 2006b] and briefly summarized here. Prior to solvent extraction, filter samples were spiked with an internal standard mixture (IS) which contains 16 isotopically labeled compounds, including benzaldehyde-*d*₆, dodecane-*d*₂₆, hexadecane-*d*₃₄, eicosane-*d*₄₂, octacosane-*d*₅₈, hexatriacontane-*d*₇₄, ααα-20*R*-cholestane-*d*₄, decanoic acid-*d*₁₉, heptadecanoic acid-*d*₃₃, phthalic acid-3,4,5,6-*d*₄, acenaphthene-*d*₁₀, chrysene-*d*₁₂, dibenz(ah)anthracene-*d*₁₄, cholesterol-2,2,3,4,4,6-*d*₆, 4,4'-dimethoxybenzophenone-*d*₈, and levoglucosan-¹³C₆. Each sample was extracted with hexane (twice) and a 2:1 mixture of benzene and 2-propanol (three times). The extracts were filtered, combined, and concentrated first by rotary evaporation, and then by ultra pure nitrogen blown down to the volume of the IS spiked. Each concentrated extract was then divided into half. One aliquot was stored in a freezer for future use and the other half was subject to methylation using freshly prepared diazomethane for converting the organic acids to their methyl esters; then the methylated aliquot was silylated with N, O-bis (trimethylsilyl) trifluoroacetamide (BSTFA) plus 1% trimethylchlorosilane (TMCS) (Pierce Biotechnology, Inc.) to convert levoglucosan and cholesterol to their trimethylsilyl derivatives.

[10] The methylated and silylated extracts were analyzed by a Hewlett Packard (HP) 6890 gas chromatography with a HP mass selective detector (GC/MSD) using a HP-5MS fused silica capillary column (30 m × 0.25 mm i.d. × 0.25 μm film thickness). Chromatographic conditions include: carrier gas: ultra pure helium (1.0 mL min⁻¹); injection mode: splitless; injector and GC/MS interface temperature: 300°C; scan range: 50–550 amu; electron ionization mode: 70 eV; oven temperature program: isothermal hold at 65°C for 2 min, temperature ramp of 10°C min⁻¹ to 300°C, and isothermal hold at 300°C for 20 min. The identification and quantification of target organic species in PM_{2.5} were achieved by using a set of primary authentic standards and secondary standards, and the uncertainties for the quantification of most organic compounds were approximately ± 20% [Zheng *et al.*, 2002, 2006b]. The concentrations of organic species were corrected from field blanks.

[11] The QA/QC procedure for GC/MS analysis was performed before analyzing each batch of samples, including solvent blank run, instrumental sensitivity checks, and relative response factor checks. Detailed description of

instrumental QA/QC was given by Zheng *et al.* [2006b]. Two replicates of Standard Reference Material for urban dust (SRM 1649a) purchased from the National Institute of Standards and Technology (NIST) were analyzed to check the recovery and accuracy of the organic compound analysis by comparing to certified values of 12 PAHs provided by NIST. The result indicates that the relative standard deviation (RSD) for most compounds is less than 5%, and the recovery of PAHs with certified values in SRM 1649a ranges from 68% for fluoranthene to 107% for benzo(k) fluoranthene.

2.3. CMB Approach

[12] Eight source emission profiles used in the present study include diesel-powered vehicles [Schauer *et al.*, 1999a; Lough *et al.*, 2007], combined catalyst and non-catalyst-equipped gasoline-powered vehicles [Schauer *et al.*, 2002], wood combustion [Fine *et al.*, 2002], paved road dust [Schauer, 1998], meat cooking [Schauer *et al.*, 1999b; McDonald *et al.*, 2003], vegetative detritus [Rogge *et al.*, 1993], cigarette smoke [Rogge *et al.*, 1994], and coke production emissions [Weitkamp *et al.*, 2005; Robinson *et al.*, 2006]. In our previous studies, source profiles of wood combustion and road dust were modified to be more representative of the southeastern area and applied successfully in source apportionment of PM_{2.5} mass in the SEARCH monitoring network [Zheng *et al.*, 2002, 2006b]. The source profile of meat cooking was also modified by combining the emission rates of total organic carbon and cholesterol from chicken under-char [McDonald *et al.*, 2003] with the emission rates of the other compounds from Schauer *et al.* [1999b]. The modifications and reasons for such modifications are described in [Zheng *et al.*, 2002, 2006b]. These modified source profiles were applied in the present study to ensure consistency with our previous studies. Coke production profile is used in BHM samples because some coke plants are located just about 2 km away from BHM site [Zheng *et al.*, 2006b].

[13] The selection of organic tracers used in the present study is based on the recommendation by Schauer *et al.* [1996]. All these organic tracers have been examined for their chemical stability [Schauer *et al.*, 1996]. However, recent studies suggested that some organic tracers (such as cholesterol, hopanes, steranes, and levoglucosan) could react in the atmosphere, and therefore may lead to possible underestimation of the contributions from meat cooking, gasoline vehicles, and wood burning [Weitkamp *et al.*, 2008a, 2008b; Hennigan *et al.*, 2010]. However, the decay of these markers is examined in a much simpler system than the real atmosphere with complex aerosol composition and varying atmospheric condition such as oxidant type and concentration as well as temperature. Therefore, the decay itself needs to be further examined. In addition, this effect should be minor in this study because the estimate of wood burning source is not based on levoglucosan alone, but on a variety of species such as some PAHs and EC. In our CMB analysis, the fit for different wood burning species is within the acceptable range of the model.

[14] OC and EC results with the TOR protocol in each PM_{2.5} sample were converted to the equivalent TOT (thermal-optical transmittance) values [Birch and Cary, 1996; Chow *et al.*, 2001, 2004], since OC and EC in source

¹Auxiliary materials are available in the HTML. doi:10.1029/2011JD016572.

profile samples were measured with the TOT protocol. The conversion equations were established by 329 PM_{2.5} quartz-filter samples (8 × 10 in), which were collected using high-volume PM_{2.5} samplers at SEARCH sites and simultaneously analyzed by both TOT and TOR methods. The details were presented elsewhere [Cheng *et al.*, 2011], and a brief introduction and converting formulas (Table S3 in auxiliary material Text S1) are included in the Supporting Information.

[15] Source contributions to PM_{2.5} mass were computed by CMB8.2 software distributed by the USEPA. The target for the percent mass explained by the CMB model is 100 ± 20%. Other diagnostics include R² (target 0.8–1.0), χ² (target 0–4.0), t-test (target > 2.0), degree of freedom (DF, target > 5), no clusters, and calculated-to-measured ratio (C/M ratio for fitting species, target 0.5–2.0).

3. Results and Discussion

3.1. PM_{2.5} Mass and Composition

[16] Concentrations of PM_{2.5} mass and major components of monthly composite samples from the eight sites in the southeastern United States are presented in Table S1 in auxiliary material Text S1. These data were calculated from the best estimated (BE) data published on the SEARCH website [Edgerton *et al.*, 2005]. OC and EC data were measured by the TOR method [Chow *et al.*, 1993, 2001]. Organic matter (OM) was calculated by multiplying TOR OC by a factor of 1.7 for urban samples and 2.0 for rural samples [Yu *et al.*, 2005], which are different from a factor of 1.4 commonly used previously [Hansen *et al.*, 2003; Edgerton *et al.*, 2005]. This assumption may lead to an underestimate for the summer season when the OM/OC ratio is usually found higher than in other seasons.

[17] Figure 1 shows the averaged PM_{2.5} mass and species from July 2001 to January 2005 at the eight sites. The average PM_{2.5} mass concentrations at JST (17.4 ± 3.1 μg m⁻³, avg. ± one standard deviation, similarly hereinafter) and BHM (18.3 ± 4.0 μg m⁻³) exceed the annual NAAQS standard (15 μg m⁻³), while PM_{2.5} mass concentrations at other sites could meet the standard. Of the PM_{2.5} components at the eight locations, OM is the largest contributor to mass, ranging from 28 ± 7% (3.35 ± 0.78 μg m⁻³ at GFP) to 43 ± 8% (5.69 ± 1.71 μg m⁻³ at CTR), followed by SO₄²⁻ (28 ± 2%). NH₄⁺ accounts for about 10% at almost all sites. EC is the highest at JST (8 ± 2%) and BHM (10 ± 4%), while it is only about 5% at other sites. NO₃⁻ makes up 3–6% at the eight sites. “Other mass” or unexplained mass varies from 8 ± 7% at JST to 25 ± 8% at GFP, including sea salt components, metal oxides such as silica and alumina, and other species not measured [Edgerton *et al.*, 2005].

[18] Figure 2 presents seasonal averages of PM_{2.5} mass and composition at the eight sites. These averaged mass concentrations show a distinct seasonal variation with the lowest values in wintertime (January) and higher in warm months at all sites. PM_{2.5} mass peaks in summer (July) at four inland locations (JST, BHM, YRK, and CTR), while at four coastal sites (GFP, PNS, OAK, and OLF) mass concentrations reach maxima in October.

[19] The seasonal variability of major species in PM_{2.5} can be also seen in Figure 2. These variations were similar to our previous findings [Edgerton *et al.*, 2005]. Briefly, SO₄²⁻

shows the same seasonal and spatial trends with PM_{2.5} mass, which is lower in the cool season (January) but higher in warm seasons. In fact, a significant correlation (R² = 0.67) was found between SO₄²⁻ and PM_{2.5} concentrations. The higher SO₄²⁻ concentrations in warm months are linked to increased photochemical activity and/or increased SO₂ emissions in summer as a result of peak electricity generation [Edgerton *et al.*, 2005]. However, NO₃⁻ concentrations show an opposite trend, with the highest values in January and lower in warm months at all of the sites, and the ratios of NO₃⁻ concentrations in January to those in July range from 3.1 (OLF) to 6.3 (CTR). This trend is consistent with the thermodynamics of gas/particle partitioning of NH₄NO₃ [Edgerton *et al.*, 2005; Bassett and Seinfeld, 1983]. In contrast, carbonaceous material (OM plus EC) shows less seasonal variation, although the highest percentage in PM_{2.5} is observed in January (Figure 2). For the spatial variation, higher concentrations and contributions of carbonaceous material to PM_{2.5} are found at the inland sites (6.11–8.80 μg m⁻³, and 44–50%) than at the coastal sites (3.90–5.33 μg m⁻³, and 33–43%).

3.2. Source Apportionment of PM_{2.5} Mass

3.2.1. Concentrations of Tracers and OC Source Apportionment

[20] Of more than 100 organic compounds quantified in the composite PM_{2.5} samples, 28 organic species as well as Al, Si, and EC were selected as the tracer input for CMB-MM modeling. These organic tracers include 9 *n*-alkanes, 3 branched alkanes, 7 hopanes and steranes (H + S), 6 polycyclic aromatic hydrocarbons (PAHs), levoglucosan, cholesterol, and nonanal. The concentrations of these tracers in the samples at the eight sites are listed in Table S2 in auxiliary material Text S1.

[21] Based on these tracers, the contributions of up to eight emission sources to OC concentrations were evaluated in the southeastern area (Table 1). The CMB results are statistically significant with R² (average 0.84 ± 0.05), χ² (average 3.43 ± 1.12), and DF (average 15 ± 4). The average OC concentration for the 112 composite samples in the present study is 3.22 ± 1.37 μg m⁻³. Model results revealed that the primary sources contributing to OC are wood burning (17 ± 19%) (percentages refer to % OC), diesel exhaust (9 ± 4%), gasoline-powered motor vehicle exhaust (5 ± 7%), meat cooking (5 ± 5%), road dust (2 ± 3%), and vegetative detritus (2 ± 2%), as well as cigarette smoke only identified at the four urban sites (2 ± 2%), and coke production only detected at BHM (2 ± 1%). On average, the explained OC accounted for 40 ± 21% of “measured OC” for the composite samples at the eight sites (from 32 ± 11% at BHM site to 47 ± 21% at PNS). Therefore, the unexplained OC (i.e., “other OC”) is quite significant at these monitoring sites and may include other unidentified primary sources, secondary OC formed by atmospheric reactions, as well as potential biases introduced by the representativeness of source profiles.

3.2.2. PM_{2.5} Mass Source Apportionment

[22] According to the source apportionment of fine OC, the contributions of the eight primary emission sources to PM_{2.5} mass concentrations were calculated using the ratios of fine OC to total PM_{2.5} from source profiles. The secondary fraction of inorganic ions (SO₄²⁻, NO₃⁻, NH₄⁺) was

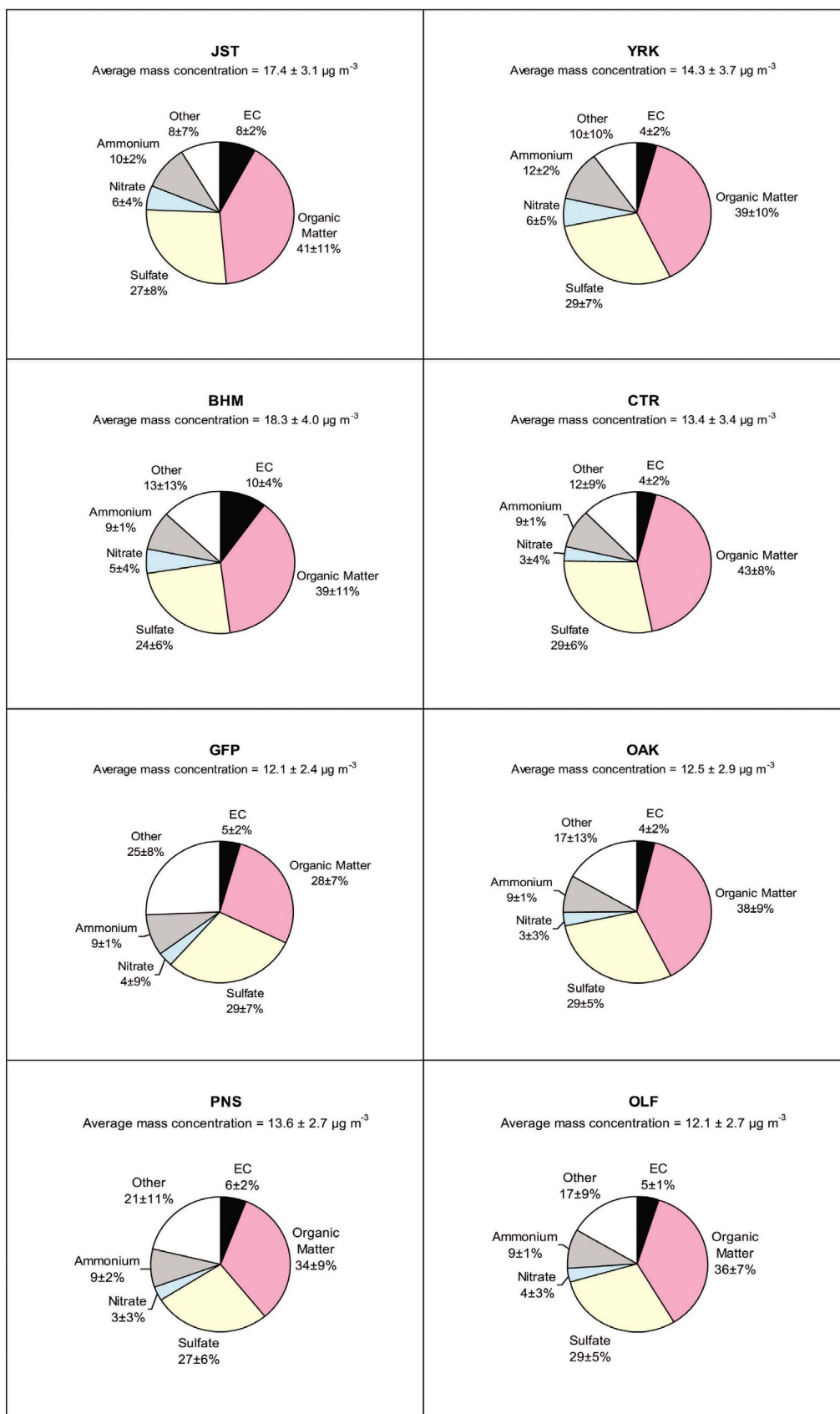


Figure 1. Averaged mass concentrations of PM_{2.5} and composition for the eight SEARCH sites from July 2001 to January 2005.

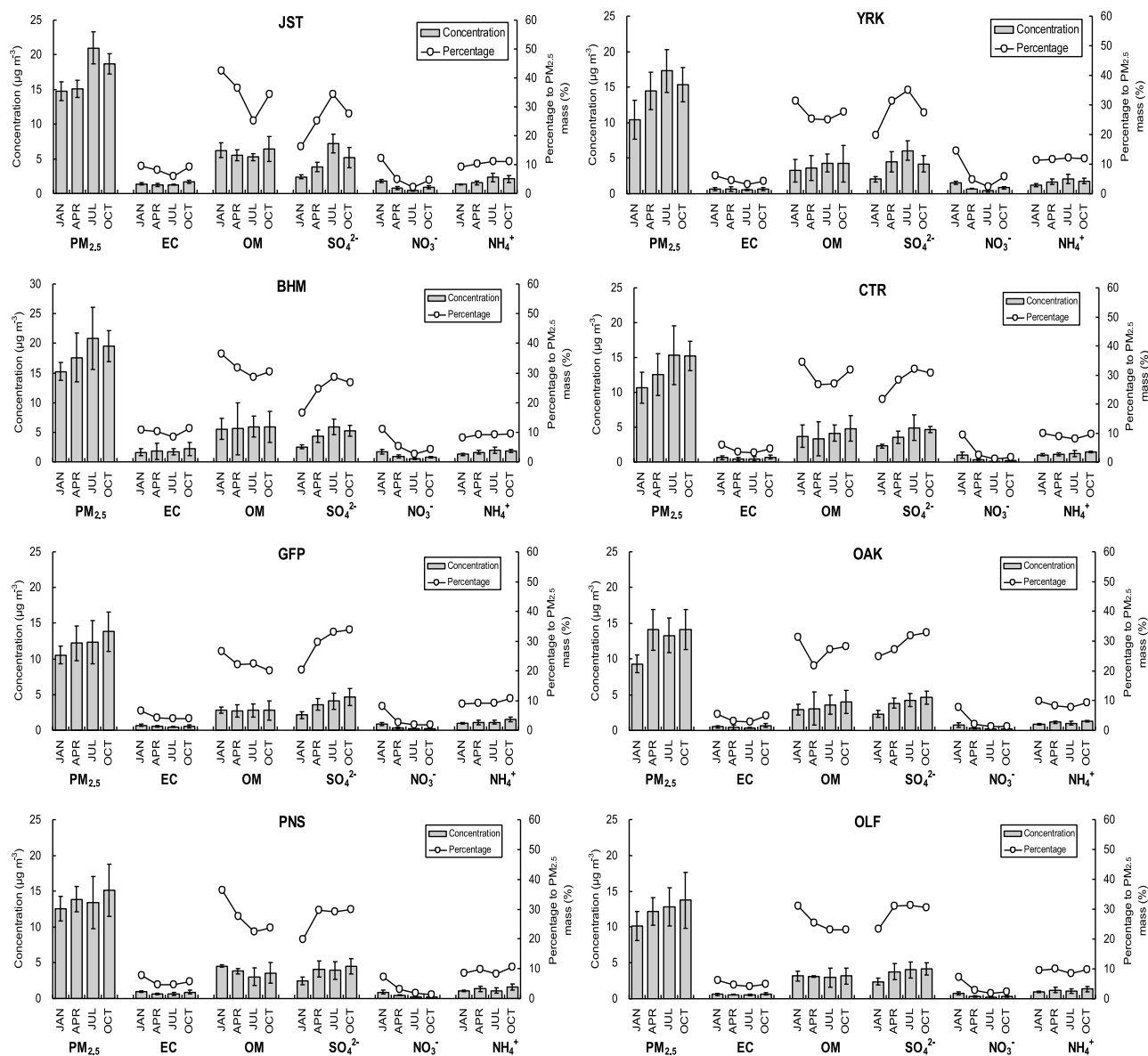


Figure 2. Seasonal averaged concentrations of PM_{2.5} mass and major components, and percentage contributions of components to PM_{2.5} mass at the eight SEARCH sites during 2001–2005.

calculated by the difference between the measured concentrations and the amount estimated in the primary source emissions [Zheng *et al.*, 2002]. In fact, the primary sulfate, nitrate and ammonium only contribute very small amounts to the total measured concentrations of ionic species (about $0.6 \pm 0.4\%$, $2.0 \pm 1.3\%$, and $0.8 \pm 0.5\%$ respectively). These primary and secondary contributions are used to reconstruct PM_{2.5} mass of each sample in the present study. “Other OM” was calculated from “other OC” multiplied by a factor of 1.7 for urban samples and 2.0 for rural samples [Yu *et al.*, 2005]. As can be seen in Table 2, the identified primary and secondary sources contribute from $82 \pm 8\%$ at GFP to $100 \pm 7\%$ at JST of measured PM_{2.5} mass in the southeastern monitoring network.

[23] On average of all the 112 composite samples in the present study, the PM_{2.5} mass concentration is $14.2 \pm 3.8 \mu\text{g m}^{-3}$. The largest contributor is secondary ionic

species (SO_4^{2-} , NO_3^- , and NH_4^+), and their total concentration is $5.90 \pm 1.85 \mu\text{g m}^{-3}$ and the contribution to the total mass is $41.4 \pm 5.7\%$. About half of OM is identified by the eight primary sources (the total concentration is $3.42 \pm 1.63 \mu\text{g m}^{-3}$ and the contribution to PM_{2.5} is $24.9 \pm 11.3\%$), and “other OM” contributes about $23.3 \pm 10.3\%$ of PM_{2.5} mass. The unexplained fraction of PM_{2.5} mass, which accounts for $11.4 \pm 9.6\%$ of total PM_{2.5} mass in the present study and may be linked to a combination of species not measured (such as metal oxides), particle-bound water, and analytical error (including OC conversions between TOR and TOT methods) etc. [Edgerton *et al.*, 2005]. However, this unexplained mass would be less if a higher ratio of OM to OC is used (e.g., 2.2–2.6 by Turpin *et al.* [2000]).

[24] Figure 3 illustrates the averaged source apportionment of PM_{2.5} mass at the eight sites. Among all source contributors, secondary sulfate accounts for the largest fraction

Table 1. Source Contributions ($\mu\text{g m}^{-3}$) to Fine OC at the Eight SEARCH Sites During 2001–2005^a

Sampling Site		Diesel Exhaust	Gasoline Exhaust	Wood Combustion	Meat Cooking	Road Dust	Vegetative Detritus	Cigarette Smoke	Coke Production	Other OC	Measured OC	R ²	χ^2	DF
JST	Mean	0.39	0.40	0.87	0.18	0.04	0.06	0.13		2.74	4.81	0.86	3.22	19
	Stdev	0.12	0.25	1.05	0.12	0.05	0.09	0.05		1.21	0.81	0.02	0.46	3
	Min	0.17	0.07	0.09	0.01	0.00	0.01	0.04		0.11	3.74	0.82	2.56	12
	Max	0.59	0.93	3.39	0.43	0.19	0.33	0.21		4.52	6.79	0.90	4.38	23
YRK	Mean	0.28	0.22	0.38	0.14	0.05	0.07			1.89	2.86	0.84	3.34	13
	Stdev	0.14	0.16	0.33	0.10	0.04	0.07			0.97	0.91	0.06	1.18	4
	Min	0.13	0.03	0.06	0.00	0.01	0.01			0.42	1.56	0.70	1.90	9
	Max	0.64	0.46	0.99	0.35	0.11	0.25			3.54	4.11	0.92	5.32	23
BHM	Mean	0.49	0.21	0.54	0.13	0.07	0.03	0.08	0.10	3.45	5.06	0.85	3.39	19
	Stdev	0.35	0.13	0.51	0.14	0.07	0.01	0.04	0.09	1.27	1.69	0.04	0.91	2
	Min	0.08	0.06	0.07	0.03	0.02	0.02	0.02	0.01	0.92	1.70	0.78	1.76	15
	Max	1.33	0.51	1.48	0.46	0.27	0.06	0.13	0.30	5.48	7.85	0.92	5.37	21
CTR	Mean	0.24	0.05	0.54	0.11	0.04	0.02			2.04	2.96	0.85	3.16	13
	Stdev	0.12	0.03	0.59	0.06	0.04	0.02			1.02	0.92	0.06	1.49	5
	Min	0.07	0.02	0.06	0.03	0.01	0.00			0.33	1.15	0.73	1.24	5
	Max	0.44	0.08	2.07	0.19	0.13	0.05			3.59	4.52	0.94	6.01	22
GFP	Mean	0.14	0.11	0.36	0.25	0.06	0.03	0.03		1.26	2.23	0.85	3.20	16
	Stdev	0.06	0.12	0.52	0.12	0.08	0.03	0.02		0.59	0.52	0.04	1.04	4
	Min	0.03	0.00	0.02	0.06	0.01	0.01	0.01		0.06	1.57	0.79	1.69	10
	Max	0.22	0.47	1.70	0.42	0.27	0.10	0.08		2.36	3.21	0.91	4.93	22
OAK	Mean	0.21	0.33	0.51	0.14	0.08	0.07			1.45	2.50	0.85	3.13	13
	Stdev	0.10	0.48	0.57	0.14	0.10	0.06			0.97	0.80	0.06	1.05	4
	Min	0.04	0.06	0.07	0.04	0.00	0.01			0.00	1.06	0.75	0.98	7
	Max	0.37	1.37	1.94	0.49	0.29	0.20			2.60	3.68	0.95	4.92	20
PNS	Mean	0.21	0.15	0.82	0.10	0.07	0.05	0.08		1.58	3.02	0.82	4.29	16
	Stdev	0.10	0.11	0.87	0.07	0.07	0.04	0.05		0.72	0.75	0.03	0.83	3
	Min	0.09	0.03	0.05	0.03	0.01	0.01	0.02		0.41	1.38	0.77	2.78	9
	Max	0.42	0.38	2.89	0.27	0.25	0.12	0.20		2.79	3.95	0.89	5.50	22
OLF	Mean	0.27	0.22	0.38	0.11	0.05	0.05			1.38	2.29	0.83	3.72	12
	Stdev	0.11	0.17	0.56	0.08	0.08	0.06			0.63	0.45	0.06	1.41	4
	Min	0.10	0.03	0.03	0.00	0.00	0.00			0.06	1.45	0.72	2.08	6
	Max	0.47	0.46	2.07	0.26	0.30	0.22			2.30	3.17	0.93	6.85	19

^aMean: arithmetic mean; Stdev: standard deviation; Min: minimum; Max: maximum; DF: degree of freedom.

of PM_{2.5} mass ($27.2 \pm 6.3\%$), and shows no significant variation across the widely distributed eight sites (from $3.51 \pm 1.12 \mu\text{g m}^{-3}$ at OLF to $4.64 \pm 2.16 \mu\text{g m}^{-3}$ at JST, average $3.93 \pm 1.53 \mu\text{g m}^{-3}$). It should be noted that the BHM site was located just about 2 km away from some coke plants, but the sulfate concentration at BHM is not the highest (also can be seen in Figures 1 and 2). Secondary ammonium ($1.36 \pm 0.47 \mu\text{g m}^{-3}$) and nitrate ($0.61 \pm 0.47 \mu\text{g m}^{-3}$) contribute $9.5 \pm 1.7\%$ and $4.7 \pm 4.0\%$ of PM_{2.5} mass, respectively, and also show small variation across the eight sites (Table 2).

[25] Among the primary sources, diesel exhaust ($1.41 \pm 0.96 \mu\text{g m}^{-3}$) is the largest contributor ($9.9 \pm 5.5\%$) to PM_{2.5} mass in the southeastern monitoring network. It accounts for the greatest fraction ($2.49 \pm 1.79 \mu\text{g m}^{-3}$) at the BHM site but the lowest at GFP ($0.69 \pm 0.29 \mu\text{g m}^{-3}$). The second most important primary source is wood combustion, with the average concentration of $0.74 \pm 0.89 \mu\text{g m}^{-3}$ (from $0.48 \pm 0.69 \mu\text{g m}^{-3}$ at GFP to $1.17 \pm 1.41 \mu\text{g m}^{-3}$ at JST) and average contribution of $5.8 \pm 7.4\%$ to PM_{2.5} mass at the eight sites. As mentioned above, wood burning is the largest contributor to OC followed by diesel exhaust. However, due to a lower OC to PM_{2.5} ratio in the diesel exhaust source profile

than the ratio in the wood burning source profile, diesel exhaust becomes the most important contributor to PM_{2.5} mass when converting OC source apportionment results to PM_{2.5} mass source apportionment results. The other major primary contributors to PM_{2.5} mass include meat cooking (average $0.45 \pm 0.34 \mu\text{g m}^{-3}$ and $3.3 \pm 2.8\%$), road dust (average $0.45 \pm 0.52 \mu\text{g m}^{-3}$ and $3.2 \pm 4.4\%$), and gasoline-powered vehicle emission (average $0.41 \pm 0.42 \mu\text{g m}^{-3}$ and $3.0 \pm 3.6\%$). Coke production emissions are taken into account at the BHM site, and it contributes $0.24 \pm 0.21 \mu\text{g m}^{-3}$ and $1.3 \pm 1.0\%$ to the PM_{2.5} mass at this site. Other minor but statistically significant primary sources include cigarette smoke (average $0.18 \pm 0.12 \mu\text{g m}^{-3}$ and $1.2 \pm 0.8\%$) at urban sites and vegetative detritus (average $0.15 \pm 0.16 \mu\text{g m}^{-3}$ and $1.1 \pm 1.3\%$).

[26] The unexplained organic matter (other OM), which was calculated by the unidentified OC (other OC), contributes a significant fraction ($\sim 1/4$) of total PM_{2.5} mass. This component has been considered to be possibly associated with secondary organic aerosol (SOA) formed by chemical reactions in the atmosphere [Zheng *et al.*, 2002; Yin *et al.*, 2010]. In this study, a positive correlation ($R^2 = 0.41$) was observed between “other OM” and the sum

Table 2. Source Contributions ($\mu\text{g m}^{-3}$) to PM_{2.5} Mass at the Eight SEARCH Sites During 2001–2005^a

Sampling Site	Diesel Exhaust	Gasoline Exhaust	Wood Combustion	Meat Cooking	Road Dust	Vegetative Detritus	Cigarette Smoke	Coke Production	Other OM	Secondary Sulfate	Secondary Nitrate	Secondary Ammonium	Identified Mass	PM _{2.5} Mass	Percent Mass Explained
JST	Mean	0.74	1.17	0.54	0.34	0.18	0.29		4.66	4.64	0.96	1.81	17.3	17.4	100
	Stdev	0.62	0.46	0.36	0.36	0.27	0.12		2.06	2.16	0.58	2.4	2.4	3.1	7
	Min	0.85	0.13	0.02	0.02	0.02	0.08		0.19	2.03	0.29	1.26	12.8	13.3	88
	Max	3.00	1.72	1.27	1.42	1.02	0.47		7.68	8.39	2.14	3.04	20.4	22.5	110
YRK	Mean	1.44	0.41	0.42	0.35	0.22			3.52	4.17	0.88	1.68	13.1	14.3	92
	Stdev	0.71	0.29	0.31	0.29	0.21			1.94	1.86	0.47	0.54	3.6	3.7	10
	Min	0.65	0.06	0.01	0.04	0.03			0.72	1.62	0.27	0.94	7.68	8.87	75
	Max	3.27	0.85	1.03	0.88	0.78			7.08	7.25	1.82	2.90	19.1	19.1	109
BHM	Mean	2.49	0.39	0.37	0.57	0.09	0.17	0.24	5.87	4.43	0.98	1.63	17.8	18.3	98
	Stdev	1.79	0.24	0.41	0.50	0.04	0.08	0.21	2.16	1.64	0.52	0.42	4.6	4.0	14
	Min	0.41	0.12	0.08	0.12	0.05	0.04	0.04	1.57	2.09	0.34	1.02	11.1	13.3	73
	Max	6.76	0.95	1.37	2.08	0.17	0.29	0.75	9.32	6.81	2.07	2.33	25.9	25.9	117
CTR	Mean	1.20	0.10	0.33	0.31	0.08			3.77	3.81	0.44	1.19	11.7	13.4	87
	Stdev	0.60	0.05	0.18	0.28	0.05			1.96	1.49	0.43	0.28	3.2	3.4	8
	Min	0.35	0.04	0.08	0.04	0.00			0.56	2.03	0.13	0.75	7.22	8.20	67
	Max	2.26	0.15	0.57	1.01	0.16			6.87	6.96	1.48	1.67	16.7	19.6	99
GFP	Mean	0.69	0.21	0.73	0.48	0.11	0.07		2.14	3.54	0.42	1.14	9.89	12.1	82
	Stdev	0.29	0.23	0.35	0.64	0.09	0.05		1.01	1.29	0.30	0.31	2.0	2.4	8
	Min	0.15	0.01	0.18	0.06	0.02	0.01		0.10	1.53	0.19	0.75	7.36	9.23	69
	Max	1.12	0.87	1.26	2.05	0.32	0.18		4.02	5.76	1.03	1.71	12.9	16.7	94
OAK	Mean	1.05	0.60	0.41	0.60	0.20			3.13	3.65	0.35	1.07	10.8	12.5	87
	Stdev	0.53	0.89	0.42	0.77	0.19			1.59	1.15	0.29	0.22	2.5	2.9	12
	Min	0.22	0.10	0.11	0.03	0.02			0.84	1.81	0.12	0.68	6.87	7.42	50
	Max	1.86	2.55	1.46	2.24	0.61			5.09	5.61	1.14	1.43	14.9	16.6	101
PNS	Mean	1.07	0.29	0.31	0.53	0.16	0.18		2.68	3.65	0.45	1.25	11.5	13.6	85
	Stdev	0.50	0.21	0.20	0.54	0.11	0.05		1.23	1.20	0.33	0.36	2.1	2.7	9
	Min	0.45	0.05	0.09	0.09	0.02	0.05		0.70	1.92	0.00	0.73	8.06	9.92	70
	Max	2.12	0.70	0.79	1.95	0.36	0.43		4.75	5.38	1.29	1.84	15.4	19.2	101
OLF	Mean	1.40	0.42	0.33	0.41	0.15			2.54	3.51	0.42	1.13	10.4	12.1	86
	Stdev	0.54	0.31	0.25	0.63	0.18			1.18	1.12	0.23	0.30	2.1	2.7	7
	Min	0.52	0.06	0.01	0.02	0.00			0.11	1.87	0.20	0.70	7.67	7.98	73
	Max	2.40	0.86	0.77	2.27	0.68			4.54	5.18	0.97	1.71	14.6	18.1	99

^aMean: arithmetic mean; Stdev: standard deviation; Min: minimum; Max: maximum.

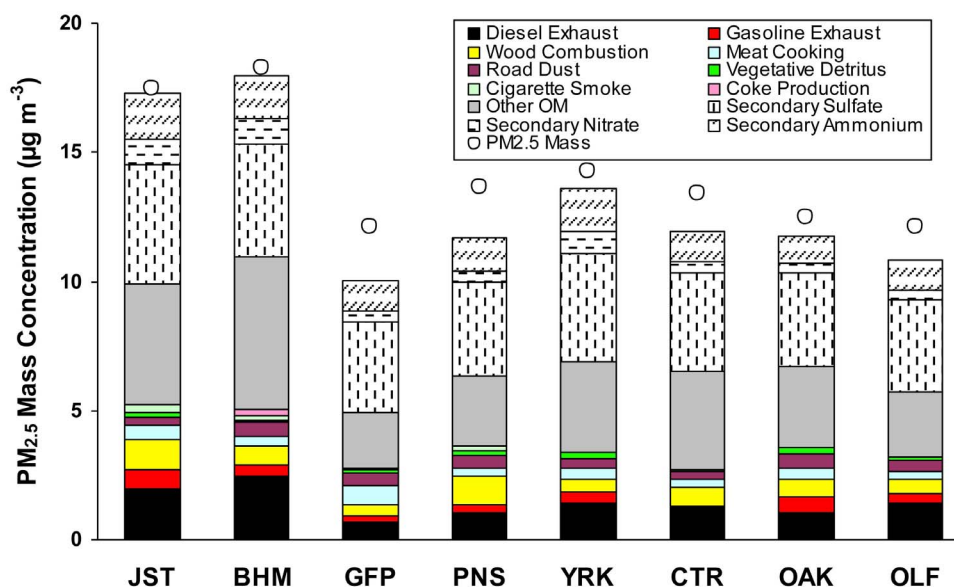


Figure 3. Averaged contributions of apportioned sources to PM_{2.5} mass at the eight SEARCH sites from July 2001 to January 2005.

of secondary SO_4^{2-} , NO_3^- , and NH_4^+ ion concentrations for all 112 composite samples, while a stronger correlation ($R^2 = 0.57$) exists for samples collected in summer (July) when photochemical reactions are favored.

3.3. Seasonal and Spatial Variability of Source Contributors

[27] Figure 4 presents the averaged concentrations of apportioned sources in PM_{2.5} mass by month and by site in the southeastern area. Data in four months of each year during 2001–2005 were averaged to represent four seasons. As can be seen in Figure 4, different sources show very different seasonal patterns and spatial characteristics across the sites. In general, compared with the data at coastal sites, source concentrations of diesel exhaust, gasoline exhaust, other OM, and secondary sulfate, nitrate, and ammonium in PM_{2.5} mass at inland sites are higher. Source concentrations of diesel exhaust, other OM, as well as some secondary ions at inland rural sites are even higher than those at coastal urban sites. Differences in source concentrations of wood combustion, meat cooking, vegetative detritus, and road dust among the eight sites are not significant.

[28] The seasonal variation of the contribution of diesel exhaust is not significant compared with other sources, as shown in Figure 4. On average, diesel exhaust contributes the least mass in July (from $0.51 \pm 0.11 \mu\text{g m}^{-3}$ at GFP to $1.45 \pm 0.26 \mu\text{g m}^{-3}$ at JST, average $1.00 \pm 0.32 \mu\text{g m}^{-3}$). The greatest seasonal variation can be seen at the BHM site ($1.41 \pm 0.74 \mu\text{g m}^{-3}$ in July versus $3.86 \pm 2.58 \mu\text{g m}^{-3}$ in October), followed by JST ($1.45 \pm 0.26 \mu\text{g m}^{-3}$ in July and $2.62 \pm 0.38 \mu\text{g m}^{-3}$ in April).

[29] The contribution from gasoline-powered motor vehicles is larger in wintertime ($0.76 \pm 0.39 \mu\text{g m}^{-3}$) except at the CTR and OLF sites, while it is relatively small in July ($0.22 \pm 0.12 \mu\text{g m}^{-3}$). The highest contribution occurs in January at the JST ($1.24 \pm 0.39 \mu\text{g m}^{-3}$) and OAK ($1.18 \pm 1.24 \mu\text{g m}^{-3}$) sites. The concentrations of main tracers for

gasoline exhaust (hopanes and steranes, H + S) also show the highest values in winter samples at JST. Another study also reported that the contribution of gasoline exhaust in JST was $2.42 \mu\text{g m}^{-3}$ in January 2002 versus $0.38 \mu\text{g m}^{-3}$ in July 2001 [Zheng *et al.*, 2007]. At the OAK site, the high average contribution of gasoline exhaust in winter is due to a very high level in January 2005 ($2.55 \mu\text{g m}^{-3}$). Three high molecular weight PAHs (indeno[123-cd]pyrene, benzo(ghi)perylene, and coronene) have much higher concentrations in this January 2005 composite sample (a total of 0.50 ng m^{-3}) than the other three seasons in 2005 (averaged 0.04 ng m^{-3}). These three PAHs were thought to be useful tracers for gasoline exhaust, especially for separation from diesel exhaust [Zielinska *et al.*, 2004; Chow *et al.*, 2007].

[30] Wood combustion in winter contributes $1.64 \pm 0.71 \mu\text{g m}^{-3}$ to PM_{2.5}, but much less in summer ($0.19 \pm 0.08 \mu\text{g m}^{-3}$) (Figure 4). This seasonal variation is consistent with the levoglucosan concentrations in the samples (average $114.1 \pm 79.6 \text{ ng m}^{-3}$ in winter versus $17.1 \pm 10.9 \text{ ng m}^{-3}$ in summer). Tian *et al.* [2009] described the fact that wood burning and prescribed fires are performed heavily in cooler seasons in Georgia, Southeastern United States. Levoglucosan is a known marker for wood burning [Simoneit *et al.*, 1999] and has been used widely in CMB-MM modeling study.

[31] Road dust contributes the highest level in July (average $0.89 \pm 0.27 \mu\text{g m}^{-3}$), followed by April ($0.34 \pm 0.17 \mu\text{g m}^{-3}$), January ($0.11 \pm 0.05 \mu\text{g m}^{-3}$), and October ($0.17 \pm 0.09 \mu\text{g m}^{-3}$). The unexplained mass also shows a similar seasonal variation as road dust [Edgerton *et al.*, 2005]. Road dust consists of soil dust particles as well as resuspended vehicle exhaust particles, tire dust, and plant fragments [Cass, 1998]. Liu *et al.* [2005] reported in a positive matrix factorization (PMF) study that a soil factor peaked during April 2001, July 2001, and July 2002 in the four inland sites, and attributed the April 2001 event to unusual Asian dust transportation [Szykman *et al.*, 2003] and the two July

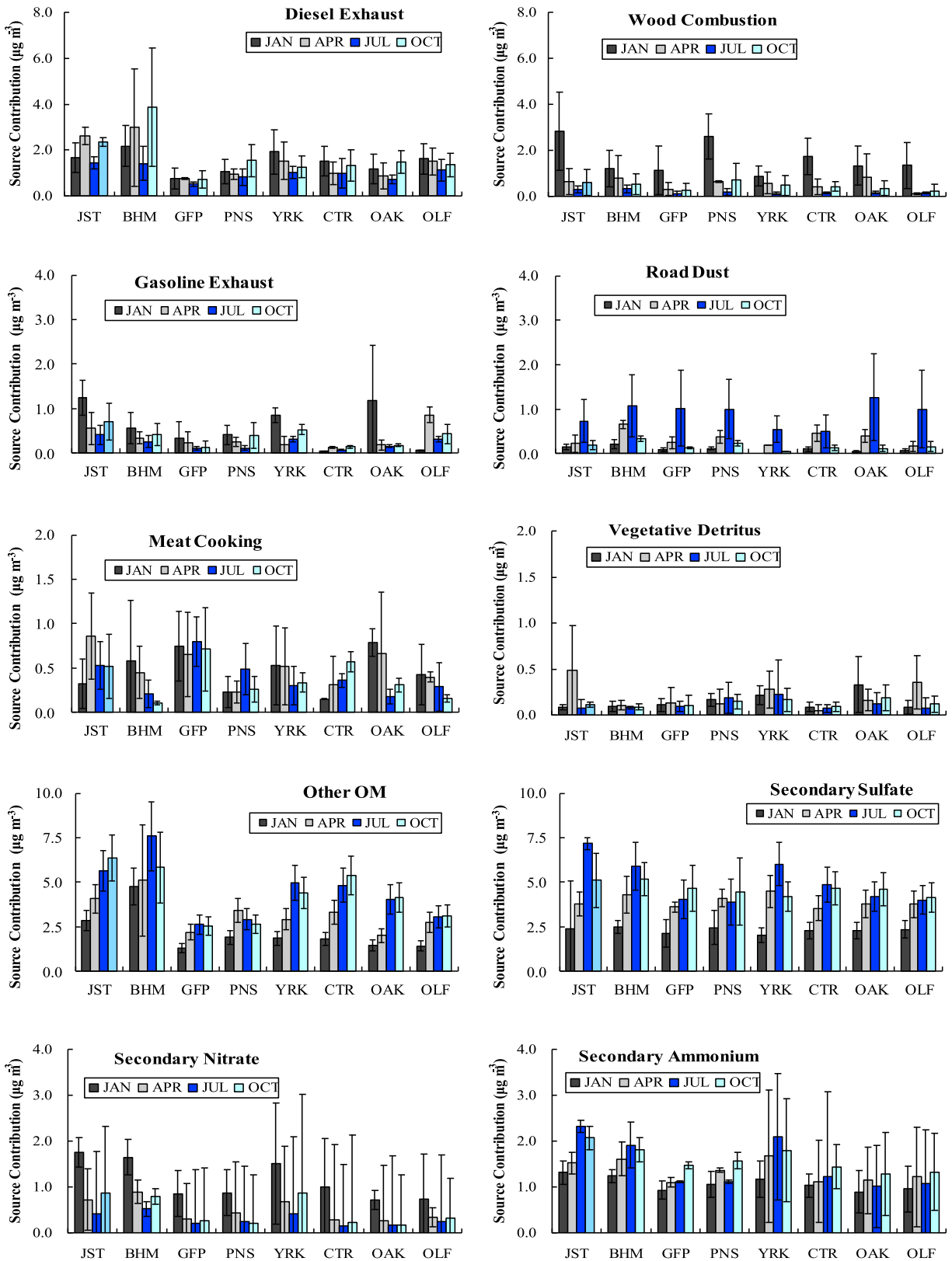


Figure 4. Seasonal average of source concentrations in PM_{2.5} mass at the eight SEARCH sites during 2001–2005.

episodes to Saharan dust impacts. *Prospero* [2001] showed that African dust can be carried by the summer trade winds into the southeastern United States. Other emission sources, such as vegetative detritus, coke production, and cigarette smoke are found to be minor contributors to PM_{2.5} and also have smaller seasonal variations.

4. Conclusions

[32] Compositions and sources of 112 composite PM_{2.5} samples collected from the eight SEARCH monitoring sites during 2001–2005 are studied, and the seasonal and spatial variations of major sources to PM_{2.5} mass are investigated in the present study. On average, the PM_{2.5} mass concentration is $14.2 \pm 3.8 \mu\text{g m}^{-3}$ at the eight monitoring sites, though the PM_{2.5} concentrations at JST and BHM exceed the annual NAAQS standard ($15 \mu\text{g m}^{-3}$). Organic material (OM) contributes the largest measured fraction (28–43%) of total PM_{2.5} mass, followed by SO₄²⁻ (~28%), NH₄⁺ (~10%), EC (5–10%) and NO₃⁻ (3–6%). PM_{2.5} mass shows distinct seasonal and spatial trends, with the lowest concentrations in January and higher values in warm months. PM_{2.5} mass peaks in July samples at the four inland sites versus in October samples at the coastal sites. SO₄²⁻ shows similar seasonal and spatial trends with PM_{2.5} and drives PM_{2.5} seasonality. Carbonaceous material shows less seasonality, but it exhibits spatial variations in both concentration and contribution to PM_{2.5} between the inland and coastal sites.

[33] Eight primary contributors to fine OC are wood burning ($17 \pm 19\%$), diesel exhaust ($9 \pm 4\%$), gasoline exhaust ($5 \pm 7\%$), meat cooking ($5 \pm 5\%$), road dust ($2 \pm 3\%$), vegetative detritus ($2 \pm 2\%$), cigarette smoke ($2 \pm 2\%$ at four urban sites), and coke production ($2 \pm 1\%$ only at BHM). The explained OC accounts for $40 \pm 21\%$ of “measured OC” across the eight sites. The unexplained OC is therefore a quite significant fraction and may be dominated by secondary OC formed by atmospheric reactions.

[34] Eighty to 100% of measured PM_{2.5} mass at the eight sites can be explained by the identified sources, in which secondary ionic species (SO₄²⁻, NH₄⁺, and NO₃⁻) contribute $41.4 \pm 5.7\%$ in total, followed by identified OM from the eight primary sources ($24.9 \pm 11.3\%$), unexplained OM ($23.3 \pm 10.3\%$), and the other mass ($11.4 \pm 9.6\%$).

[35] Secondary sulfate accounts for the largest fraction of PM_{2.5} mass ($27.2 \pm 6.3\%$) among all source contributors, and shows the same seasonal trend as PM_{2.5} and measured sulfate, but has very weak variation across the eight sites. Secondary ammonium ($9.5 \pm 1.7\%$) and nitrate ($4.7 \pm 4.0\%$) also show small spatial variations. Among the primary emission sources, diesel exhaust (contributing $9.9 \pm 5.5\%$ to PM_{2.5} mass) and gasoline exhaust ($3.0 \pm 3.6\%$) show strong seasonal and spatial variations, with the lowest contribution in July and obviously higher levels at BHM and JST than other sites. Wood combustion ($5.8 \pm 7.4\%$) contributes a significantly larger fraction in winter than in summer, and shows a very strong correlation ($R^2 = 0.85$) with levoglucosan concentrations. Road dust ($3.2 \pm 4.4\%$) shows higher levels in July and April across the eight sites, which may be due to the effects of African and Asian dust. Meat cooking ($3.3 \pm 2.8\%$) and other primary sources, such as vegetative detritus, coke production, and cigarette smoke,

are minor contributors to PM_{2.5}, and show insignificant seasonal variations.

[36] The averaged other OM accounted for about 40% of OM in January at JST (Figures 2 and 4), which is due to high other OM in the January 2005 sample. This sample may be impacted by some primary sources, which were not identified in this study. In other years, about 90% of OM was identified in the January samples.

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References

- Bassett, M. E., and J. H. Seinfeld (1983), Atmospheric equilibrium model of sulfate and nitrate aerosol, *Atmos. Environ.*, **17**, 2237–2252, doi:10.1016/0004-6981(83)90221-4.
- Birch, M. E., and R. A. Cary (1996), Elemental carbon-based methods for monitoring occupational exposures to particulate diesel exhaust, *Aerosol Sci. Technol.*, **25**, 221–241, doi:10.1080/02786829608965393.
- Cass, G. R. (1998), Organic molecular tracers for particulate air pollution sources, *Trends Anal. Chem.*, **17**, 356–366, doi:10.1016/S0165-9936(98)00040-5.
- Cheng, et al. (2011), Comparison of two thermal-optical methods for the determination of organic carbon and elemental carbon: Results from the southeastern United States, *Atmos. Environ.*, **45**, 1913–1918.
- Chow, J. C., J. G. Watson, L. C. Pritchett, W. R. Pierson, C. A. Frazier, and R. G. Purcell (1993), The DRI thermal/optical reflectance carbon analysis system: Description, evaluation and applications in U.S. Air quality studies, *Atmos. Environ., Part A*, **27**, 1185–1201.
- Chow, J. C., J. G. Watson, D. Crow, D. H. Lowenthal, and T. Merrifield (2001), Comparison of IMPROVE and NIOSH carbon measurements, *Aerosol Sci. Technol.*, **34**, 23–34.
- Chow, J. C., J. G. Watson, L.-W. A. Chen, W. P. Arnott, H. Moosmuller, and K. Fung (2004), Equivalence of elemental carbon by thermal/optical reflectance and transmittance with different temperature protocols, *Environ. Sci. Technol.*, **38**, 4414–4422, doi:10.1021/es034936u.
- Chow, J. C., J. G. Watson, D. H. Lowenthal, L. W. A. Chen, B. Zielinska, L. R. Mazzoleni, and K. L. Magliano (2007), Evaluation of organic markers for chemical mass balance source apportionment at the Fresno Super-site, *Atmos. Chem. Phys.*, **7**, 1741–1754, doi:10.5194/acp-7-1741-2007.
- Ding, X., M. Zheng, L. P. Yu, X. L. Zhang, R. J. Weber, B. Yan, A. G. Russell, E. S. Edgerton, and X. M. Wang (2008), Spatial and seasonal trends in biogenic secondary organic aerosol tracers and water-soluble organic carbon in the southeastern United States, *Environ. Sci. Technol.*, **42**, 5171–5176, doi:10.1021/es7032636.
- Edgerton, E. S., B. E. Hartsell, R. D. Saylor, J. J. Jansen, D. A. Hansen, and G. M. Hidy (2005), The Southeastern Aerosol Research and Characterization Study: Part II. Filter-based measurements of fine and coarse particulate matter mass and composition, *J. Air Waste Manage. Assoc.*, **55**, 1527–1542.
- Edgerton, E. S., B. E. Hartsell, R. D. Saylor, J. J. Jansen, D. A. Hansen, and G. M. Hidy (2006), The Southeastern Aerosol Research and Characterization Study: Part 3: Continuous measurements of fine particulate matter mass and composition, *J. Air Waste Manage. Assoc.*, **56**, 1325–1341.
- Fine, P. M., G. R. Cass, and B. R. T. Simoneit (2002), Organic compounds in biomass smoke from residential wood combustion: Emissions characterization at a continental scale, *J. Geophys. Res.*, **107**(D21), 8349, doi:10.1029/2001JD000661.
- Fraser, M. P., Z. W. Yue, and B. Buzcu (2003), Source apportionment of fine particulate matter in Houston, TX, using organic molecular markers, *Atmos. Environ.*, **37**, 2117–2123, doi:10.1016/S1352-2310(03)00075-X.
- Hansen, D. A., E. S. Edgerton, B. E. Hartsell, J. J. Jansen, N. Kandasamy, G. M. Hidy, and C. L. Blanchard (2003), The Southeastern Aerosol Research and Characterization Study: Part 1. Overview, *J. Air Waste Manage. Assoc.*, **53**, 1460–1471.
- Hennigan, C. J., A. P. Sullivan, J. L. Collett, and A. L. Robinson (2010), Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, *Geophys. Res. Lett.*, **37**, L09806, doi:10.1029/2010GL043088.
- Kim, E., P. K. Hopke, and E. S. Edgerton (2003), Source identification of Atlanta aerosol by positive matrix factorization, *J. Air Waste Manage. Assoc.*, **53**, 731–739.
- Kleindienst, T. E., M. Lewandowski, J. H. Offenberg, E. O. Edney, M. Jaoui, M. Zheng, X. Ding, and E. S. Edgerton (2010), Contribution

- of primary and secondary sources to organic aerosol and PM_{2.5} at SEARCH network sites, *J. Air Waste Manage. Assoc.*, **60**, 1388–1399, doi:10.3155/1047-3289.60.11.1388.
- Liu, W., Y. Wang, A. Russell, and E. S. Edgerton (2005), Atmospheric aerosol over two urban-rural pairs in the southeastern United States: Chemical composition and possible sources, *Atmos. Environ.*, **39**, 4453–4470, doi:10.1016/j.atmosenv.2005.03.048.
- Lough, G. C., C. G. Christensen, J. J. Schauer, J. Tortorelli, E. Mani, D. R. Lawson, N. N. Clark, and P. A. Gabele (2007), Development of molecular marker source profiles for emissions from on-road gasoline and diesel vehicle fleets, *J. Air Waste Manage. Assoc.*, **57**, 1190–1199, doi:10.3155/1047-3289.57.10.1190.
- McDonald, J. D., B. Zielinska, E. M. Fujita, J. C. Sagebiel, J. C. Chow, and J. G. Watson (2003), Emissions from charbroiling and grilling of chicken and beef, *J. Air Waste Manage. Assoc.*, **53**, 185–194.
- Pope, C. A., III, R. T. Burnett, M. J. Thun, E. E. Calle, D. Krewski, K. Ito, and G. D. Thurston (2002), Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution, *JAMA*, **287**, 1132–1141, doi:10.1001/jama.287.9.1132.
- Prospero, J. M. (2001), African dust in America, *Geotimes*, November, 24–27.
- Robinson, A. L., R. Subramanian, N. M. Donahue, A. Bernardo-Bricker, and W. F. Rogge (2006), Source apportionment of molecular markers and organic aerosol-1. Polycyclic aromatic hydrocarbons and methodology for data visualization, *Environ. Sci. Technol.*, **40**, 7803–7810, doi:10.1021/es0510414.
- Rogge, W. F., L. M. Hildemann, M. A. Mazurek, G. R. Cass, and B. R. T. Simoneit (1993), Sources of fine organic aerosol. 4. Particulate abrasion products from leaf surfaces of urban plants, *Environ. Sci. Technol.*, **27**, 2700–2711, doi:10.1021/es00049a008.
- Rogge, W. F., L. M. Hildemann, M. A. Mazurek, and G. R. Cass (1994), Sources of fine organic aerosol. 6. Cigarette smoke in the urban atmosphere, *Environ. Sci. Technol.*, **28**, 1375–1388, doi:10.1021/es00056a030.
- Schauer, J. J. (1998), Source contributions to atmospheric organic compound concentrations: Emissions measurement and model predictions, PhD dissertation, Calif. Inst. of Technol., Pasadena, Calif.
- Schauer, J. J., and G. R. Cass (2000), Source apportionment of wintertime gas-phase and particle-phase air pollutants using organic compounds as tracers, *Environ. Sci. Technol.*, **34**, 1821–1832, doi:10.1021/es981312t.
- 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, *Atmos. Environ.*, **30**, 3837–3855, doi:10.1016/1352-2310(96)00085-4.
- Schauer, J. J., M. J. Kleeman, G. R. Cass, and B. R. T. Simoneit (1999a), Measurement of emissions from air pollution sources. 2. C-1 through C-29 organic compounds from medium duty diesel trucks, *Environ. Sci. Technol.*, **33**, 1578–1587, doi:10.1021/es980081n.
- Schauer, J. J., M. J. Kleeman, G. R. Cass, and B. R. T. Simoneit (1999b), Measurement of emissions from air pollution sources. 1. C-1 through C-32 organic compounds from meat charbroiling, *Environ. Sci. Technol.*, **33**, 1566–1577, doi:10.1021/es980076j.
- Schauer, J. J., M. J. Kleeman, G. R. Cass, and B. R. T. Simoneit (2002), Measurement of emissions from air pollution sources. 5. C₁-C₃₂ organic compounds from gasoline-powered motor vehicles, *Environ. Sci. Technol.*, **36**, 1169–1180, doi:10.1021/es0108077.
- Seagrave, J., et al. (2006), Lung toxicity of ambient particulate matter from Southeastern U.S. sites with different contributing sources: Relationships between composition and effects, *Environ. Health Perspect.*, **114**, 1387–1393, doi:10.1289/ehp.9234.
- Simoneit, B. R. T., J. J. Schauer, C. G. Nolte, D. R. Oros, V. O. Elias, M. P. Fraser, W. F. Rogge, and G. R. Cass (1999), Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles, *Atmos. Environ.*, **33**, 173–182, doi:10.1016/S1352-2310(98)00145-9.
- Szykman, J., D. Mintz, J. Creilson, and M. Wayland (2003), Impact of April 2001 Asian dust event on particulate matter concentrations in the United States, in *National Air Quality and Emission Trends Report 2003 Special Studies Edition*, Rep. EPA-454/R-03-008, pp. S1–S12, U.S. EPA, Research Triangle Park, N. C.
- Tian, D., Y. T. Hu, Y. H. Wang, J. W. Boylan, and A. G. Russell (2009), Assessment of biomass burning emissions and their impacts on urban and regional PM_{2.5}: A Georgia case study, *Environ. Sci. Technol.*, **43**, 299–305, doi:10.1021/es801827s.
- Turpin, B. J., P. Saxena, and E. Andrews (2000), Measuring and simulating particulate organics in the atmosphere: Problems and prospects, *Atmos. Environ.*, **34**, 2983–3013, doi:10.1016/S1352-2310(99)00501-4.
- Watson, J. G. (1984), Overview of receptor model principles, *J. Air Pollut. Control Assoc.*, **34**, 619–623.
- Weitkamp, E. A., E. M. Lipsky, P. Pancreas, J. Ondov, A. Polidori, B. J. Turpin, and A. L. Robinson (2005), Fine particle emission profile for a large coke production facility based on highly time-resolved fence line measurements, *Atmos. Environ.*, **39**, 6719–6733, doi:10.1016/j.atmosenv.2005.06.028.
- Weitkamp, E. A., K. E. H. Hartz, A. M. Sage, N. M. Donahue, and A. L. Robinson (2008a), Laboratory measurements of the heterogeneous oxidation of condensed-phase organic molecular markers for meat cooking emissions, *Environ. Sci. Technol.*, **42**, 5177–5182, doi:10.1021/es800181b.
- Weitkamp, E. A., A. T. Lambe, N. M. Donahue, and A. L. Robinson (2008b), Laboratory measurements of the heterogeneous oxidation of condensed-phase organic molecular markers for motor vehicle exhaust, *Environ. Sci. Technol.*, **42**, 7950–7956, doi:10.1021/es800745x.
- Yin, J. X., R. M. Harrison, Q. Chen, A. Rutter, and J. J. Schauer (2010), Source apportionment of fine particles at urban background and rural sites in the UK atmosphere, *Atmos. Environ.*, **44**, 841–851, doi:10.1016/j.atmosenv.2009.11.026.
- Yu, L. E., M. L. Shulman, R. Kopperud, and L. M. Hildemann (2005), Fine organic aerosols collected in a humid, rural location (Great Smoky Mountains, Tennessee, USA): Chemical and temporal characteristics, *Atmos. Environ.*, **39**, 6037–6050, doi:10.1016/j.atmosenv.2005.06.043.
- Zheng, M., G. R. Cass, J. J. Schauer, and E. S. Edgerton (2002), Source apportionment of PM_{2.5} in the Southeastern United States using solvent-extractable organic compounds as tracers, *Environ. Sci. Technol.*, **36**, 2361–2371, doi:10.1021/es011275x.
- Zheng, M., L. G. Salmon, J. J. Schauer, L. Zeng, C. S. Kiang, Y. Zhang, and G. R. Cass (2005), Seasonal trends in PM_{2.5} source contributions in Beijing, China, *Atmos. Environ.*, **39**, 3967–3976, doi:10.1016/j.atmosenv.2005.03.036.
- Zheng, M., G. S. W. Hagler, L. Ke, M. H. Bergin, F. Wang, P. K. K. Louie, L. Salmon, D. W. M. Sin, J. Yu, and J. J. Schauer (2006a), Composition and sources of carbonaceous aerosols at three contrasting sites in Hong Kong, *J. Geophys. Res.*, **111**, D20313, doi:10.1029/2006JD007074.
- Zheng, M., L. Ke, E. S. Edgerton, J. J. Schauer, M. Dong, and A. G. Russell (2006b), Spatial distribution of carbonaceous aerosol in the southeastern United States using molecular markers and carbon isotope data, *J. Geophys. Res.*, **111**, D10S06, doi:10.1029/2005JD006777.
- Zheng, M., L. Ke, F. Wang, G. R. Cass, J. J. Schauer, E. S. Edgerton, and A. G. Russell (2007), Source apportionment of daily PM_{2.5} at Jefferson Street, Atlanta, GA during summer and winter, *J. Air Waste Manage. Assoc.*, **57**, 228–242.
- Zielinska, B., J. Sagebiel, W. P. Arnott, C. F. Rogers, K. E. Kelly, D. A. Wagner, J. S. Lighty, A. F. Sarofim, and G. Palmer (2004), Phase and size distribution of polycyclic aromatic hydrocarbons in diesel and gasoline vehicle emissions, *Environ. Sci. Technol.*, **38**, 2557–2567, doi:10.1021/es030518d.
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