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# **Biomass burning impact on PM<sub>2.5</sub> over the southeastern US during 2007: integrating chemically speciated FRM filter measurements, MODIS fire counts and PMF analysis**

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## Abstract

Archived Federal Reference Method (FRM) Teflon filters used by state regulatory agencies for measuring PM<sub>2.5</sub> mass were acquired from 15 sites throughout the southeastern US and analyzed for Water-Soluble Organic Carbon (WSOC), water-soluble ions and carbohydrates to investigate biomass burning contributions to fine aerosol mass. Based on over 900 filters that spanned all of 2007, levoglucosan and K<sup>+</sup> were studied in conjunction with MODIS Aqua fire count data to compare their performances as biomass burning tracers. Levoglucosan concentrations exhibited a distinct seasonal variation with large enhancement in winter and spring and a minimum in summer, and were well correlated with fire counts, except in winter when residential wood burning contributions were significant. In contrast, K<sup>+</sup> concentrations had no apparent seasonal trend and poor correlation with fire counts. Levoglucosan and K<sup>+</sup> only correlated well in winter ( $r^2=0.59$ ) when biomass burning emissions were highest, whereas in other seasons they were not correlated due to the presence of other K<sup>+</sup> sources. Levoglucosan also exhibited larger spatial variability than K<sup>+</sup>. Both species were higher in urban than rural sites (mean 44% higher for levoglucosan and 86% for K<sup>+</sup>). Positive Matrix Factorization (PMF) was applied to analyze PM<sub>2.5</sub> sources and four factors were resolved: biomass burning, refractory material, secondary light absorbing WSOC and secondary sulfate/WSOC. The biomass burning source contributed 13% to PM<sub>2.5</sub> mass annually, 27% in winter, and less than 2% in summer, consistent with other source apportionment studies based on levoglucosan, but lower in summer compared to studies based on K<sup>+</sup>.

## 1 Introduction

Biomass burning is a major source of ambient PM<sub>2.5</sub> (particulate matter with aerodynamic diameter less than 2.5 μm) and has significant impacts on human health (Lighty et al., 2000), regional and global air quality (Lelieveld et al., 2001) and climate (Penner

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et al., 1992; Hobbs et al., 1997). Numerous studies have attempted to assess the impact of biomass burning on local and regional  $PM_{2.5}$  concentrations. Typically, either source-oriented or receptor-oriented methods are used to quantify biomass burning contributions. Source-oriented approaches use Chemical Transport Models (CTMs) to simulate the emissions from biomass burning sources, but are limited by large uncertainties in fire emission inputs (Zeng et al., 2008; Tian et al., 2009). Receptor-oriented approaches quantify source contribution through measurements of specific marker species for biomass burning (e.g. Schauer et al., 1996; Rogge et al., 1998; Schauer and Cass, 2000; Zheng et al., 2002) and have been used more widely.

A number of chemical species have been used as particle-phase biomass burning emission tracers. Water-soluble potassium ( $K^+$ ) has been used extensively as an inorganic tracer to apportion biomass burning contributions to ambient aerosol (Ramadan et al., 2000; Kim et al., 2003a, b; Ma et al., 2003; Liu et al., 2005; Lee et al., 2008).  $K^+$  is not an ideal tracer as it has other sources, such as sea salt and soil dust (Duvall et al., 2008; Wang et al., 2005). Attempts have been made to eliminate these sources by calculating non-sea-salt non-dust  $K^+$  (Cachier et al., 1991; Puxbaum et al., 2007; Pio et al., 2008), but this requires knowledge of the source characteristics and an assumption that they are invariant among different locations and seasons. Organic compounds are the largest component produced from fires and there are specific compounds found to be exclusively emitted from biomass burning. The most commonly used organic tracer is levoglucosan, a sugar anhydride produced during the combustion of cellulose (Simoneit et al., 1999; Puxbaum et al., 2007; Zheng et al., 2007; Sullivan et al., 2008;).

$PM_{2.5}$  concentrations are high in the southeastern US (Goldstein et al., 2009) and the impact of biomass burning emissions in this region have been extensively studied (Tanner et al., 2004; Liu et al., 2005; Zheng et al., 2006, 2007; Lee et al., 2008; Zeng et al., 2008; Tian et al., 2009; Yan et al., 2009;). Zeng et al. (2008) suggest that prescribed fire emissions can result in a daily increase of  $PM_{2.5}$  mass up to  $25 \mu g m^{-3}$ , leading to  $PM_{2.5}$  nonattainment in regions affected by the fires. Tian et al. (2009) have attributed 55% and 80% of  $PM_{2.5}$  to prescribed burning in January and March of 2002,

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respectively. Receptor model studies using levoglucosan as a biomass-burning tracer suggest that wood burning is the dominant contributor (9%–51%) to OC and one of the major sources of PM<sub>2.5</sub> at several urban and rural sites in the southeast during September 2003 and January 2004 (Zheng et al., 2006, 2007). Using both PMF and CMB receptor models and K<sup>+</sup> as a tracer, Lee et al. (2008) attribute 5%–20% of the PM<sub>2.5</sub> mass to biomass burning emissions at four sites in Georgia and Alabama from January 2000 to December 2002.

These previous studies indicated that biomass burning was a major PM<sub>2.5</sub> source with contributions that varied with seasons; however, they are based on data for only a few months at a limited number of sites and are further restricted by uncertainties in biomass burning emissions and tracer concentrations. In this study, an extensive data set on PM<sub>2.5</sub> mass concentrations and chemical speciation was generated from archived FRM filters acquired from state regulatory agencies for the year of 2007. This study compares K<sup>+</sup> and levoglucosan as biomass-burning tracers and employs a PMF analysis to quantify average mass contributions from biomass burning and other sources for the southeastern US throughout 2007.

## 2 Methodology

### 2.1 FRM filter sampling

State agencies use an EPA Federal Reference Method (FRM) to determine ambient PM<sub>2.5</sub> mass at sites throughout the country to assess compliance with National Ambient Air Quality Standards (NAAQS). Once mass has been determined gravimetrically, the filters are stored and are eventually discarded. For this study, archived FRM filters that had been in storage (in the dark at a  $T < -20^{\circ}\text{C}$ ) for roughly a year were acquired from state regulatory agencies in Georgia (GA Department of Natural Resources), South Carolina (SC Department of Health and Environmental Control) and Alabama (AL Department of Environmental Management and the Jefferson Co. Department of

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Health). The FRM method for collecting ambient fine particles onto Whatman 47 mm Teflon filter substrates involved 24-h integrated sampling at a nominal flow rate of 16.7 L/min with PM<sub>2.5</sub> sharp cut cyclone size selector or PM<sub>2.5</sub> WINS impactor and with no gas-denuders (Patashnick et al., 2001). Fifteen sampling sites throughout the southeastern US were chosen within the EPA FRM monitoring network, on the basis of geographic location, site type (i.e. urban and rural) and source influences. Among these fifteen sites, eight were urban and seven were rural. Table 1 lists the sites, and their locations are shown in Fig. 1. A subset of all field samples corresponding to a one-in-six-day sampling schedule produced 60 filters for analysis per site and a total of 900 filters. In addition to these filters, a series of field blanks (36 filters) and replicated filters (43 filters) were included for quality control.

## 2.2 Chemical analyses

This suite of filters was extracted in water and a number of chemical components and physical properties were quantified. Each archived FRM Teflon filter was placed in a pre-cleaned 30 ml Nalgene amber HDPE bottle and extracted with 30 ml of 18-M $\Omega$  Milli-Q water via 30 min sonication. The liquid extract was then filtered using a 0.45  $\mu$ m PTFE syringe filter and transferred to a separate pre-cleaned 30 ml Nalgene amber bottle. Various chemical analyses were performed on aliquots from these bottles, which were stored in the refrigerator ( $\sim 4^{\circ}\text{C}$ ) for at most one week until all analyses were completed.

For each species quantified, overall uncertainty was determined based on a sum of squares that included measurement uncertainties (e.g. liquid extraction volumes, etc.), variability in calibrations performed throughout the analyses of all 900 filters, variability in field blanks and water blanks, and precision based on variability of a standard placed at intervals of every tenth sample throughout the sample queues.

Water-Soluble Organic Carbon (WSOC) in the extract was quantified using a Sievers Model 800 Turbo Total Organic Carbon analyzer (GE Water Systems, Boulder, CO; for more details see Sullivan et al., 2004). The instrument was calibrated using a

series of sucrose standards ( $N=5$ , linear regression  $r^2=0.9998$ ; variability in slope based on three separate calibrations throughout the analysis period was  $\sim 3\%$ ). The method Limit Of Detection (LOD) of  $0.33 \mu\text{g Carbon m}^{-3}$  ( $\mu\text{g C m}^{-3}$ ) for WSOC was estimated by three times the standard deviation of field blanks, translated to ambient air concentration assuming, in all cases, a flow rate of  $16.7 \text{ L/min}$  and  $24 \text{ h}$  sampling period. Overall measurement uncertainty was  $9\%$ .

High-Performance Anion-Exchange Chromatography with Pulsed Amperometric Detection (HPAEC-PAD) was utilized to quantify various carbohydrate compounds, including levoglucosan. This technique involved a Dionex DX-500 series ion chromatograph coupled with Dionex ED 50 electrochemical detector with a gold working electrode operating in integrating amperometric mode, and a Dionex CarboPac PA-1 anion-exchange column with gradient elution of  $200 \text{ mM NaOH}$  at a flow rate of  $0.5 \text{ mL min}^{-1}$ . The elution profile was as follows:  $0\text{--}8 \text{ min}$ , isocratic elution with  $10 \text{ mM NaOH}$ ;  $8\text{--}25 \text{ min}$ , linear gradient elution from  $10 \text{ mM}$  to  $60 \text{ mM NaOH}$ ;  $25\text{--}38 \text{ min}$ , column cleaning step with  $180 \text{ mM NaOH}$ ; and  $38\text{--}56 \text{ min}$ , column re-equilibration step with  $10 \text{ mM NaOH}$ . Detailed descriptions of eluent preparation, peak detection and calibrations of HPAEC-PAD have been presented elsewhere (Engling et al., 2006; Sullivan et al., 2008). Calibrations were based on serial dilutions from a stock solution made by dissolving individual compounds in solid form (purchased from Sigma-Aldrich) with Milli-Q water. The LOD for the various carbohydrates was estimated at  $6 \text{ ng/m}^3$  and overall measurement uncertainty for levoglucosan was  $21\%$ . Previous studies have found that the peaks of levoglucosan and arabitol, a sugar alcohol from fungal spores, cannot be fully separated by a CarboPac PA-1 column (Caseiro et al., 2007; Sullivan et al., 2008), as a result the integrated peak area of levoglucosan was corrected for this interference by arabitol. Ambient mannitol level is found to be  $1.5$  times higher than that of arabitol (Bauer et al., 2008), and since mannitol can be quantitatively measured with the CarboPac PA-1 column, the arabitol concentration was determined by dividing mannitol by  $1.5$ . The peak area of arabitol was then calculated and subtracted from the levoglucosan peak area. This correction adds uncertainty to the levoglucosan concentration,

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but typically it is minor since mannitol concentrations are generally low, especially in winter when levoglucosan is most important (Fig. 2).

To validate the HPAEC-PAD method, levoglucosan concentrations were determined from a series of quartz fiber filters ( $n=33$ ) via an independent GC-MS measurements (Zheng et al., 2006). A portion of the same filters were extracted in water and levoglucosan was measured by HPAEC-PAD following the method described above. The two datasets showed good agreement, with a linear regression  $r^2$  of 0.92, and a zero intercept regression slope of  $1.09 \pm 0.04$  ( $\pm 1$  STD).

Water-soluble anions (chloride, nitrate, sulfate and oxalate) and cations (sodium, ammonium, potassium, magnesium and calcium) were quantified on a dual channel Dionex DX-500 Ion Chromatograph with suppression and conductivity detection, employing a Dionex AS11-HC anion column and Dionex CS12A cation column. Calibrations were based on NIST traceable liquid standards. Uncertainties were 40% for  $\text{Na}^+$ , 28% for  $\text{NH}_4^+$ , 63% for  $\text{Mg}^{2+}$ , 17% for  $\text{K}^+$ , 7% for  $\text{SO}_4^{2-}$  and 36% for oxalate.  $\text{Ca}^{2+}$  uncertainty was 755%, which was subsequently found to be due to a contaminated DI water supply. Thus,  $\text{Ca}^{2+}$  results are not reported. Instead,  $\text{Mg}^{2+}$  is used as a mineral dust indicator in the following analysis. LODs for the ions were in the range of 0.01 ( $\text{K}^+$ ) to 0.1 ( $\text{SO}_4^{2-}$ )  $\mu\text{g}/\text{m}^3$ .

The UV-VIS light absorption spectra over wavelength range of 250 to 800 nm for the liquid extracts were also determined with a Liquid Waveguide Capillary Cell (LWCC) to investigate the link between Brown Carbon and biomass burning. The absorbance is determined using the ratio of transmitted to incident light intensity following Beer's Law, such that the absorbance is linear with the absorbers concentration and molar absorptivity, and LWCC path length ( $\sim 1$  m). Here, we use the product of all three since in our samples molar absorptivities are not known. The results of this aspect of the study are discussed in detail by Hecobian et al. (2010).

$\text{PM}_{2.5}$  mass concentrations were determined gravimetrically by each of the three state regulatory agencies following their protocols. It is important to note that FRM Teflon filters are not designed for  $\text{PM}_{2.5}$  composition measurements. Although

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Teflon filters are known to be relatively inert to gas absorption, the un-denuded sampling method may contribute to positive artifacts. Furthermore, this sampling method and the year-long storage of the Teflon filters may also lead to loss of semi-volatile components (Watson et al., 2009). Effort has been made to estimate these semi-volatile components retained on FRM Teflon filters (Frank, 2006). In this paper, known semi-volatile species such as nitrate are not discussed, and the reported WSOC and other components (including FRM PM<sub>2.5</sub> mass) should be viewed as a measure of the more non-volatile species associated with ambient PM<sub>2.5</sub>.

To assess the quality of the FRM filter data, the results were compared to a number of components, i.e. PM<sub>2.5</sub> mass, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, and SO<sub>4</sub><sup>2-</sup>, from seven co-located EPA Speciation sites (South Dekalb, Rome, Macon, AUG-BRS, COL-CRS, ATNS-UGA and N'BHM; see Table 1), where the chemical analyses were done through Research Triangle Institute on quartz fiber filters and were completely independent of the FRM data. The results are shown in Fig. 3. All components show good agreement between the two types of filters, with linear regression slopes from 0.88 to 1.15. It's noteworthy that the Speciation NH<sub>4</sub><sup>+</sup> were on average 15% higher than the FRM NH<sub>4</sub><sup>+</sup> (a slope of 1.15), reflecting possible loss of semi-volatile NH<sub>4</sub><sup>+</sup> associated with NO<sub>3</sub><sup>-</sup> from the FRM filters.

### 2.3 MODIS fire counts

Fire counts detected by remote sensing have been used for examining seasonal biomass burning emissions (Eva and Lambin, 1998; Duncan et al., 2003; Zeng et al., 2008). In this work, MODIS fire count data from the NASA Aqua satellite (Giglio et al., 2003) were used as a measure of outdoor biomass burning over the southeast in 2007. The data set had a horizontal resolution of 1 km×1 km, and a time resolution of 1 day. The sum of fire counts in each month was used to investigate monthly and seasonal variations. Fire counts for seven states were used, which included the three states in which our sampling sites were located and four other surrounding states to ensure that all possible biomass burning source regions were considered. An analysis of fires over continental United States indicated no evidence for long-range transport of smoke



to the sites during 2007; however, episodic influence from longer-range transport of smoke cannot be ruled out.

## 2.4 Positive Matrix Factorization (PMF) source apportionment

A PMF analysis on the chemical and physical data was used to quantify contributions from biomass burning and other sources to the FRM PM<sub>2.5</sub> mass concentrations. Detailed information on EPA-PMF (v3.0) can be found on US EPA website (<http://www.epa.gov/heasd/products/pmf/pmf.html>). Two datasets, (i.e. a concentration/physical property dataset and an uncertainty dataset) are required for PMF input. PMF is able to identify underlying covariance among chemical or physical parameters.

While most previous PMF source apportionment studies use concentrations of chemical species as objects for analysis, in this work, along with chemical species (WSOC, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, oxalate, xylose, and levoglucosan), the UV-VIS light absorption data at 365 nm (abs) from the FRM filter water-extract were also incorporated. Light absorption is an important aerosol property dependent on source and composition of the aerosols (Andreae and Gelencser, 2006). Data from the 15 sampling sites on each sampling date (including concentrations of chemical species and light absorption) were merged to form the concentration dataset. Considering potentially different source types for urban and rural sites, PMF analysis was also performed separately using two datasets from urban and rural sites, and the results, i.e. source composition profiles and contributions, showed little difference from those discussed in Sect. 3.3. Missing values for each component were replaced with the mean concentration of this component, and values below the LOD were replaced with half of the detection limit of corresponding component (Polissar et al., 1998), ensuring that all values were positive. The uncertainty for each component was determined based on the methods discussed in Sect. 2.2. For missing data points and values below the LOD, the uncertainties were assigned as 4 and 5/6 times the concentration values, respectively (Polissar et al., 1998). Numerous PMF runs were performed with 3–7 factors and various combinations of the concentration and absorption data set. Based on Q values (the objective

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function to be minimized) and physical interpretation of the solution, four factors appeared to be the optimal solution. The model output files include factor profiles, relative factor contributions and residuals (unexplained fractions).

In order to quantify the contributions to  $PM_{2.5}$  mass from each factor-related source, Multivariate Linear Regression (MLR) analysis was performed to scale relative factor contributions to the measured total  $PM_{2.5}$  mass. Light absorption data were not included in the regression analysis since it was not a mass concentration measurement.

### 3 Results and discussion

#### 3.1 Spatiotemporal variations of levoglucosan and water-soluble potassium ( $K^+$ )

The 12 months of 2007 were separated into four seasons as: January, February and December (winter); March, April and May (spring); June, July and August (summer); September, October and November (fall). Table 2 presents the annual and seasonal mean concentrations of levoglucosan and  $K^+$ , together with those of  $PM_{2.5}$  mass, Water-Soluble Organic Carbon (WSOC), ammonium ( $NH_4^+$ ), sulfate ( $SO_4^{2-}$ ), oxalate and xylose. Figure 4 shows the seasonal mean concentrations of levoglucosan and  $K^+$  at each site.

##### 3.1.1 Levoglucosan

In general, levoglucosan concentrations obtained in this study are comparable to those in previous studies in the same region (Zheng et al., 2007; Ding et al., 2008), but are considerably higher than those at other locations in the US, such as Pittsburgh, PA (annual mean at  $\sim 21 \text{ ng/m}^3$ ) (Robinson et al., 2006), suggesting much larger impact from biomass burning in the southeast. The annual mean concentration of levoglucosan

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for all sampling sites was  $107.5 \text{ ng/m}^3$  (Table 2). Clear seasonal variations of levoglucosan concentrations were observed, with significantly higher concentrations in spring and winter and minimum levels in summer. Mean concentrations for spring and winter were  $180 \text{ ng/m}^3$  and  $170 \text{ ng/m}^3$ , respectively; whereas the mean summer concentration was only  $19 \text{ ng/m}^3$ , which is within the range of levels (5 to  $52 \text{ ng/m}^3$ ) reported by Puxbaum et al. (2007) at six background stations in Europe. Significantly enhanced levoglucosan concentrations at several sites were detected for several days in spring. For instance,  $2950 \text{ ng/m}^3$  of levoglucosan was recorded at the Macon site on 12 May, almost 2 orders of magnitude higher than the level observed six days before ( $31 \text{ ng/m}^3$ ). Similarly, on 30 May, levoglucosan concentrations at COL-CRS, N'BHM, Wylam, Ashland and Providence all exceeded  $1,000 \text{ ng/m}^3$ , much higher than those at other sites on the same day ( $3.93 \text{ ng/m}^3$ – $346 \text{ ng/m}^3$ ). Such unusual levoglucosan concentrations at these sites were due to significant but sporadic impacts from unique wildfire events in spring of 2007. Extensive wildfires started in the Okefenokee Swamp in southern Georgia/northern Florida in April and spread across Georgia and adjacent states during the following two months (Yan et al., 2010).

Figure 4a also shows the spatial distributions of levoglucosan for different seasons. In general, there are significant site-to-site variations, suggesting varied impacts from biomass burning emissions among the sampling sites. Winter and spring show the largest variations in terms of absolute concentrations. In spring, variability is largely due to the sporadic impacts from wildfire events at different sites. In winter, biomass burning is likely mainly in the form of residential wood burning (discussed in more detail below), thus spatial variability at these time periods would reflect population densities and frequency of burning in different regions. Levoglucosan concentrations in urban sites are generally higher than those in rural sites, except during summer when rural sites tend to have slightly higher levoglucosan concentrations, implying that residential wood burning in urban areas is an important source of levoglucosan when other forms of biomass burning, such as wildfires, are absent. High spatial variability was observed for levoglucosan during all seasons, as indicated by the large Relative Standard

Deviations (RSD, standard deviations of concentrations in each season divided by the corresponding seasonal mean concentration) (51%–79%).

### 3.1.2 Water-soluble Potassium ( $K^+$ )

Compared to levoglucosan,  $K^+$  concentrations exhibited different spatiotemporal variability. First,  $K^+$  had an opposite seasonal pattern to levoglucosan with much less seasonal variability (from 45 ng/m<sup>3</sup> in winter to 71 ng/m<sup>3</sup> in summer, Table 2). Smaller site-to-site concentration differences suggested by the lower RSD values (25%–75%) for all four seasons indicate more uniform  $K^+$  concentrations compared to levoglucosan. The significantly higher levels of  $K^+$  at South Dekalb and AUG-BRS sites during summer (Fig. 4b) are from  $K^+$  spikes detected at these two sites on 5 July, and attributed to pyrotechnique (fireworks) emissions. Excluding these data, RSD value in summer (75%) would be much smaller, and  $K^+$  concentrations would be even more uniform (The 5 July sampling day was excluded from all further analysis). Similar to levoglucosan,  $K^+$  tended to have slightly higher concentrations at urban sites (Fig. 4b).

A comparison of 24-h averaged levoglucosan and  $K^+$  shows different degrees of correlation in winter and summer (Fig. 5). In winter, when biomass burning is expected to be more prevalent, levoglucosan and  $K^+$  are reasonably correlated ( $r^2=0.59$ ). In summer, however, there is almost no correlation between levoglucosan and  $K^+$  ( $r^2=0.02$ ), reflecting distinctly different behaviours of these two tracers when biomass-burning emissions are expected to be significantly lower.

### 3.2 Relationships between $K^+$ , levoglucosan and MODIS fire counts

MODIS fire count data can be used as a measure of wildfire and prescribed burning events, two of the major forms of biomass burning over the southeast (Zeng et al., 2008). Other forms of biomass burning such as residential wood burning cannot be detected by remote sensing and thus are not reflected in the fire counts. Since our sampling dates are evenly distributed in each month, the monthly fire counts over the

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region encompassing our sampling sites should reflect the outdoor biomass burning impacts on our measured PM<sub>2.5</sub> mass and composition.

In 2007 there was a distinct seasonal variation of fire counts, with maximum in spring (counts ranging from 2156 to 4822) and minimum in summer (646–1214) (Fig. 6), indicating extensive outdoor biomass burning events in spring, and fewer such events in summer. The highly concentrated fire points in May along Georgia-Florida boarder (Fig. 6) are the Okefenokee Swamp fires discussed above.

The relationships between monthly fire counts and monthly mean concentrations of levoglucosan and K<sup>+</sup> are shown in Figs. 7 and 8. Fire counts track levoglucosan concentrations well, except in January and December (Fig. 7), consistent with residential wood burning during these months (Tian et al., 2009). For example, fire counts dropped from a March peak of 4822 by a factor of 8 to 646 in July, corresponding to levoglucosan concentrations changing from 294 ng/m<sup>3</sup> to 15 ng/m<sup>3</sup>, a factor of 9 decrease. Levoglucosan and K<sup>+</sup> concentrations are also shown as a function of fire counts in Fig. 8. For the levoglucosan and fire counts correlation,  $r^2$  is 0.54; excluding January and December,  $r^2$  is 0.86. K<sup>+</sup> concentrations show poor correlation with fire counts throughout the year ( $r^2=0.16$ , and 0 excluding January and December in Fig. 8). Because residential burning is minimal in summer, and fire count data show a large decrease in outdoor burning during summer, it is expected that biomass burning emissions in summer would be significantly less than the cooler seasons. Levoglucosan had a similar trend to fire counts, whereas K<sup>+</sup> did not. Scatter at the lower concentrations in summer months may be due to some fraction of levoglucosan being lost through oxidation (Hennigan et al., 2010). The difference in levoglucosan concentrations in January and December when fire counts are low (Fig. 8) suggests residential wood burning throughout the southeastern US, on average, contributed roughly 135 ng/m<sup>3</sup> to 145 ng/m<sup>3</sup> of levoglucosan during these two months. This corresponds to approximately 2.6 μg/m<sup>3</sup> of PM<sub>2.5</sub> mass (based on an average emission factor determined in Sect. 3.3.3).

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### 3.3 Source apportionment of PM<sub>2.5</sub> using PMF

#### 3.3.1 Source profiles and relative contributions

Composition profiles for the 4 factors resolved by PMF are shown in Fig. 9 (left panel). Values in the composition profiles represent average fractions (in percent) of those species distributed amongst the four factors. Relative contributions of the four factors obtained from the PMF output were averaged among all sampling sites for each sampling date, shown on the right panel, to illustrate their temporal variation throughout the year.

Factor 1 is characterized by high levels of levoglucosan (i.e. 77% of levoglucosan is in factor 1), xylose (50%), and UV-VIS light absorption (55%). Also, associated with this factor are K<sup>+</sup> (16%), WSOC (14%), oxalate (11%), and NH<sub>4</sub><sup>+</sup> (7%). Factor 1 composition profile is consistent with biomass burning emissions. Levoglucosan is the dominant component and almost exclusively associated with this factor, consistent with it being a unique biomass burning tracer (Simoneit et al., 1999; Schkolnik and Rudich, 2006). Xylose is also a carbohydrate emitted in biomass burning (Simoneit, 2002). As the above analysis has shown, K<sup>+</sup> is not as unique a biomass-burning tracer, and it is associated with other factors, mostly factor 2. Biomass burning emissions also produce significant levels of Brown Carbon (Andreae and Gelencser, 2006; Hecobian et al., 2010), which accounts for the light absorption factor. Oxalate has also been found in biomass burning smoke (Kundu et al., 2009). The appreciable amount of WSOC associated with this factor is consistent with previous studies suggesting biomass burning and Secondary Organic Aerosol (SOA) formation are the two major sources of WSOC (Fuzzi et al., 2006; Sullivan et al., 2006; Weber et al., 2007; Saarikoski et al., 2008). Factor 1 seasonal trend shows that averaged over the southeast, biomass burning is most prevalent in the cold months, with events in the springtime due to wildfires in South Georgia. These data show little biomass burning impacts during the summer.

Factor 2 is characterized by refractory material; Na<sup>+</sup> (51%), K<sup>+</sup> (43%), and Mg<sup>2+</sup> (51%), indicating PM<sub>2.5</sub> from mineral dust (e.g. Lee et al., 1999). These refractory

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materials are likely related to coarse-mode particles, windy conditions and possibly long-range transport. The factor 2 time series has no clear seasonal pattern.

Factors 3 and 4 are thought to be linked to secondary aerosol formation. Factor 3 has the highest percentages of WSOC (56%) and oxalate (56%), along with light absorbing species (e.g. Brown Carbon) (34%), and  $K^+$  (15%). Factor 4 is distinguished by  $NH_4^+$  (86%) and  $SO_4^{2-}$  (90%), as well as a considerable amount of WSOC (22%), but little oxalate and little light absorbing species. In this factor, the molar ratio of  $NH_4^+$  to  $SO_4^{2-}$  was 1.6, indicating that for much of the southeast sulfate is not fully neutralized, as noted in other studies (Lee et al., 2008) (any  $NH_4^+$  associated with nitrate is not considered). Both factors are correlated with temperature ( $r^2=0.62$  for F3 and  $r^2=0.81$  for F4) and the high levels of WSOC in both factors and sulfate in factor 4 are attributed to secondary aerosol formation processes that are known to be prevalent during summer (Lim and Turpin, 2002; Kondo et al., 2007; Weber et al., 2007; Miyazaki et al., 2009). The differences between factors 3 and 4 and the linking of WSOC, oxalate and light absorbing organics (Brown Carbon) suggest insights into secondary organic aerosol formation, which is investigated further elsewhere (Hecobian et al., 2010; Zhang et al., 2010).

### 3.3.2 Contributions of factors to yearly $PM_{2.5}$ mass, levoglucosan, $K^+$ and WSOC

Using Multivariate Linear Regression (MLR), the four factors isolated by PMF, along with the residual, can be used to estimate source contributions (percent of total mass and concentrations in  $\mu g/m^3$ ) to overall  $PM_{2.5}$  mass and various  $PM_{2.5}$  components. Results are summarized in Table 3 and Figs. 10 and 11.

The four factors resolved by PMF explained 82% of the total  $PM_{2.5}$  mass (Fig. 10), with the largest proportion due to factor 4 (secondary sulfate/WSOC) at 38% for the yearly average, factor 3 (secondary light absorbing WSOC) at 25%, and factor 1 (biomass burning) at 13%. The 18% associated with the residual is likely from sources

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related to unmeasured species contributing to PM<sub>2.5</sub> mass, such as Elemental Carbon (EC) and water-insoluble organic species, which can be associated with primary emissions from both biomass burning and fossil fuel combustion. At the seven co-located EPA Speciation sites (see Sect. 2.2, all located at urban areas), measurements were made of organic and elemental carbon by Thermal Optical Reflectance (TOR) and Transmittance (TOT) and several elements by Energy Dispersive X-Ray Fluorescence (EDXRF). Incorporating OC, EC, Ca, Cu and Fe data along with the FRM chemical species/light absorption data that were measured at these seven sites, a PMF analysis on this dataset resolved 5 factors. A mobile source emission factor characterized by abundant EC (70%) and Cu (57%) was identified, contributing 8% of the PM<sub>2.5</sub> mass on an annual basis. Meanwhile, the residual fraction dropped from 18% to 7%. The other 4 factors were very similar to the PMF analysis based on just the FRM data shown in Figs. 9, 10 and 11 (Details are provided in the supplementary material, Figs. 1S and 2S: <http://www.atmos-chem-phys-discuss.net/10/7037/2010/acpd-10-7037-2010-supplement.pdf>).

Relative contributions of various sources (factors) to WSOC, levoglucosan, and K<sup>+</sup> are also shown in Fig. 10. On an annual basis, most of the WSOC is associated with secondary light absorbing WSOC source (56%), followed by secondary sulfate/WSOC (22%) and biomass burning (14%). For levoglucosan, the dominant source is the biomass-burning factor (77%). In contrast, K<sup>+</sup> has several sources, with refractory material (43%) being the largest, and each of the three other sources contributing from 7 to 16%.

Table 3 shows the separated fractional contributions of the factors to PM<sub>2.5</sub> mass by season. Although biomass burning only contributed 13% on an annual basis, it accounted for 27% of the PM<sub>2.5</sub> during winter and only 2% in summer, consistent with the PMF results that included Speciation data, in which case biomass burning contributed 29% and 1% in winter and summer, respectively (Table 1S: <http://www.atmos-chem-phys-discuss.net/10/7037/2010/acpd-10-7037-2010-supplement.pdf>). Factors involving secondary aerosol formation

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processes (F3 and F4) show the opposite trend, contributing 46% in winter and 77% in summer. This rather large relative increase in summertime secondary sources corresponds to an average WSOC increase of roughly  $1 \mu\text{gC}/\text{m}^3$  from winter to summer (Table 2).

Figure 11 is similar to the time series plot for each factor (Fig. 9, right panel), but in this case the contributions in terms of estimated mass of each factor (source) throughout the year are given. While refractory material (F2) contributions remained consistently low (<7%), secondary sulfate/WSOC was the largest  $\text{PM}_{2.5}$  contributor throughout the year (30%–46%), a result partially due to the relatively large and uniform distribution of sulfate in this region (Kim et al., 2003a, b; Liu et al., 2005). Biomass-burning sources (F1) dominate in winter and on average make little contribution to  $\text{PM}_{2.5}$  mass in summer. The secondary light absorbing WSOC factor (F3) also shows enhancement in summer, but was higher relative to the other secondary factor (F4) in March during a period of unusually warm weather. The source of this factor will be addressed in detail in Zhang et al. (2010).

### 3.3.3 Comparisons to other studies

Studies using a variety of techniques have investigated contributions of biomass burning to air quality in the southeast. These results are compared to the PMF analysis using the FRM filter data presented here. First,  $\text{PM}_{2.5}$  mass to levoglucosan ratios from emission studies have been found to vary from  $9.7 \pm 2.4$  g/g for hardwoods to  $24.4 \pm 4.3$  g/g for softwoods (Fine et al., 2002), and 11.5 g/g for a prescribed burning episode in Georgia during April 2004 (Lee et al., 2005). PMF factor 1 characterizes the biomass-burning source for the southeast in 2007. From factor 1, the annual  $\text{PM}_{2.5}$  mass to levoglucosan ratio was  $18.3 \pm 5.4$  ( $\pm$  STD) g/g, roughly midway between hard and softwood emissions, but higher than emissions from prescribed burning, consistent with contributions from residential burning which favor hardwoods. Emission studies have also characterized ratios of  $\text{K}^+$  to levoglucosan, with median ratios in the range of (0.03–0.16) (Fine et al., 2001, 2002, 2004a, b; Lee et al., 2005), depending on the

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type of material burnt. For factor 1 the  $K^+$ /levoglucosan ratio was 0.10, similar to the emission profiles. In contrast, for the FRM dataset the ratio is 0.50 in terms of annual mean and 0.26 for winter. These values are not in the range found in the studies mentioned above, likely due to other sources for  $K^+$ .

5 Our PMF-predicted annual mean contribution of 13% for biomass burning is comparable with previous PMF results using  $K^+$  as the tracer. For example, Kim et al. (2003a, b) estimated 14% in Atlanta, and Liu et al. (2005) 13% for the southeastern US. However, different seasonal patterns were found in our study and those PMF studies using  $K^+$ . Our PMF results suggest significant biomass burning contribution in winter (27%),  
10 and a small impact (2%) in summer, consistent with a Chemical Mass Balance analysis based on Molecular Markers (CMB-MM) using levoglucosan as the biomass-burning tracer (0.4% in summer) (Zheng et al., 2007). In contrast, previous PMF studies using  $K^+$  predicted similar biomass-burning contributions for all the seasons (e.g. Kim et al., 2003a; Liu et al., 2005). The discrepancy depends on the choice of biomass burning  
15 tracer used. During periods of significant biomass burning emissions, such as the colder months, levoglucosan and  $K^+$  tend to be correlated (Fig. 5) and the predicted biomass burning impacts are similar, whether using levoglucosan or  $K^+$  as the tracer. During these periods the biomass-burning source for  $K^+$  appears to dominate other sources. However, due to the relatively higher contributions of non-biomass burning  
20  $K^+$  sources observed in summer, it is expected that a source apportionment analysis using  $K^+$  as a tracer would overestimate biomass burning emission during those periods.

Alternatively, levoglucosan may be substantially depleted in the summer due to photochemical oxidation reactions leading to a large underestimation of summertime biomass-burning contribution (Hennigan et al., 2010; Hoffmann et al., 2010). This, however, is not consistent with the low fire counts in summer. Considering such uncertainties and that the results from previous studies might overestimate biomass burning contribution in summer (>10%) due to non-biomass burning  $K^+$  sources (e.g. Kim et al., 2003a; Liu et al., 2005), we conclude that summer biomass burning contributions

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are likely in the range of 2%–10%. In winter, levoglucosan is relatively stable and likely provides a reasonable estimate of biomass burning contributions to PM<sub>2.5</sub> levels. Based on a number of studies, including this work, typical winter contribution of biomass burning to PM<sub>2.5</sub> in the southeast (on a mass basis) is estimated to be near 25%, which in December and January appears to be mostly from residential wood-burning.

## 4 Conclusions

We investigate the biomass burning impact on PM<sub>2.5</sub> in the southeastern US in 2007 through analysis of chemical and physical properties of over 900 24-h integrated FRM Teflon filters collected by state regulatory agencies.

Two commonly used biomass burning tracers, i.e. levoglucosan and K<sup>+</sup>, were compared in conjunction with MODIS Aqua fire counts. Levoglucosan concentrations showed large seasonal variations and correlated well with fire counts, except in winter (January and December), when residential wood burning not detected by satellites led to increased levoglucosan. During these months, residential burning was estimated to contribute 2.6 μg/m<sup>3</sup>, on average, to the ambient PM<sub>2.5</sub> mass throughout the region. K<sup>+</sup> concentrations exhibited no apparent seasonal trends and poor correlation with fire counts. Levoglucosan and K<sup>+</sup> correlated well with each other ( $r^2=0.59$ ) in winter, suggesting their common origin from biomass burning when emissions were relatively high. In other seasons, K<sup>+</sup> poorly correlated with levoglucosan apparently due to additional K<sup>+</sup> sources. Both levoglucosan and K<sup>+</sup> concentrations were higher at urban sites than at rural sites, with levoglucosan showing larger spatial variations than K<sup>+</sup>. Comparison of K<sup>+</sup> and levoglucosan measurements in conjunction with fire count data suggested that K<sup>+</sup> was not a good tracer for biomass burning emissions due to multiple sources in addition to biomass burning, whereas levoglucosan was a reasonable indicator of biomass burning emissions, including emissions from wildfires and prescribed

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burnings, as well as emissions from wood combustion for residential heating, despite chamber study results that show levoglucosan loss by reaction with hydroxyl radical (Hennigan et al., 2010).

Positive Matrix Factorization (PMF) was applied to analyze PM<sub>2.5</sub> sources from the FRM data. Four factors were resolved, including a biomass burning factor characterized by high levels of levoglucosan and light absorbing compounds (Brown Carbon), a refractory component characterized by K<sup>+</sup>, Na<sup>+</sup> and Mg<sup>2+</sup>, and two secondary aerosol components, one characterized by high WSOC, organic acids (oxalate) and light absorbing compounds, and the other by high SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, and WSOC. Secondary sulfate/WSOC component was the largest source of PM<sub>2.5</sub> in all seasons and combined, the two secondary sources dominated in all seasons. The biomass burning source contributed 13% to the PM<sub>2.5</sub> mass annually, 27% in winter, and only approximately 2% in summer. The refractory component contributed the least and was consistent throughout the year. Overall, the results show that K<sup>+</sup> is a poor biomass burning tracer especially in summer and can lead to large over-prediction of biomass burning contributions by source apportionment analyses. Extensive photochemical degradation of levoglucosan may lead to under-prediction of biomass burning in summer, however, in this study there was reasonable consistency between levoglucosan concentrations and summertime outdoor burning quantified by remotely sensed fire count data.

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**Table 1.** Locations and types of the FRM sampling sites in this study.

No.	State	City	Site name	Latitude	Longitude	Type
1	GA	Decatur	South Dekalb*	33.6881	−84.2902	Urban
2	GA	Rome	Rome*	34.2611	−85.3230	Urban
3	GA	Augusta	AUG-BRS*	33.4339	−82.0224	Urban
4	GA	Macon	Macon*	32.7773	−83.6412	Urban
5	GA	Yorkville	Yorkville	33.9285	−85.0453	Rural
6	GA	Sandersville	Sandersville	32.9747	−82.8089	Rural
7	GA	Columbus	COL-CRS*	32.4309	−84.9318	Urban
8	GA	Athens	ATNS-UGA*	33.9179	−83.3446	Urban
9	AL	Birmingham	N'BHM*	33.5530	−86.8149	Urban
10	AL	Wylam	Wylam	33.4997	−86.9241	Urban
11	AL	Providence	Providence	33.4596	−87.3055	Rural
12	AL	Ashland	Ashland	33.2849	−85.8036	Rural
13	AL	Crossville	Crossville	34.2886	−85.9699	Rural
14	SC	Long Creek	Long Creek	34.8053	−83.2377	Rural
15	SC	Trenton	Trenton	33.7400	−81.8536	Rural

\* Sampling stations with co-located EPA Speciation sites.

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**Table 2.** Annual and seasonal mean concentrations ( $\pm$  STD) of measured PM<sub>2.5</sub> components from FRM filters. Winter (J, F, D), Spring (M, A, M), Summer (J, J, A) and Fall (S, O, N).

Component		Annual	Winter	Spring	Summer	Fall
PM <sub>2.5</sub>	$\mu\text{g m}^{-3}$	15.56 $\pm$ 8.67	11.68 $\pm$ 6.25	17.33 $\pm$ 11.19	19.17 $\pm$ 7.87	13.61 $\pm$ 5.74
WSOC	$\mu\text{gC m}^{-3}$	2.07 $\pm$ 1.53	1.45 $\pm$ 1.01	2.47 $\pm$ 2.22	2.58 $\pm$ 1.21	1.70 $\pm$ 0.83
NH <sub>4</sub> <sup>+</sup>	$\mu\text{g m}^{-3}$	1.40 $\pm$ 0.71	1.06 $\pm$ 0.61	1.39 $\pm$ 0.63	1.78 $\pm$ 0.79	1.36 $\pm$ 0.63
K <sup>+</sup>	$\text{ng m}^{-3}$	53.7 $\pm$ 104.1	45.0 $\pm$ 32.1	55.6 $\pm$ 51.8	70.8 $\pm$ 194.8	42.9 $\pm$ 46.2
SO <sub>4</sub> <sup>2-</sup>	$\mu\text{g m}^{-3}$	4.43 $\pm$ 2.99	2.60 $\pm$ 1.90	4.24 $\pm$ 2.40	6.41 $\pm$ 3.68	4.38 $\pm$ 2.46
Oxalate	$\text{ng m}^{-3}$	136.0 $\pm$ 91.3	82.6 $\pm$ 57.2	172.4 $\pm$ 110.5	173.2 $\pm$ 85.6	108.3 $\pm$ 59.5
Levogluconan	$\text{ng m}^{-3}$	107.5 $\pm$ 221.9	169.9 $\pm$ 180.3	180.3 $\pm$ 339.2	18.7 $\pm$ 44.7	55.7 $\pm$ 134.7
Xylose	$\text{ng m}^{-3}$	1.23 $\pm$ 2.44	1.71 $\pm$ 2.12	1.99 $\pm$ 3.84	0.52 $\pm$ 0.78	0.68 $\pm$ 1.31

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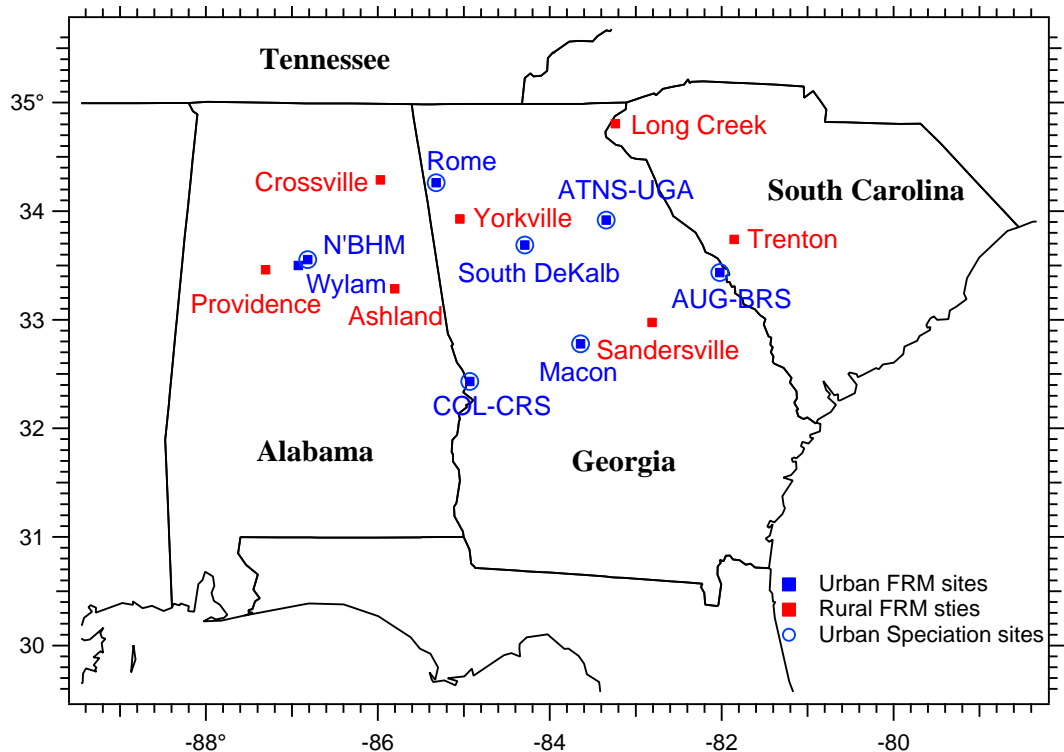
**Table 3.** Seasonal and annual source contributions of each PMF factor to PM<sub>2.5</sub> mass (% of PM<sub>2.5</sub> mass). Winter (J, F, D), Spring (M, A, M), Summer (J, J, A) and Fall (S, O, N).

	Winter	Spring	Summer	Fall	Annual
Biomass Burning (F1)	27	15	2	7	13
Refractory (F2)	7	6	6	6	6
Secondary Light Abs WSOC (F3)	16	27	31	26	25
Secondary Sulfate/WSOC (F4)	30	32	46	44	38
Residual	20	20	16	16	18

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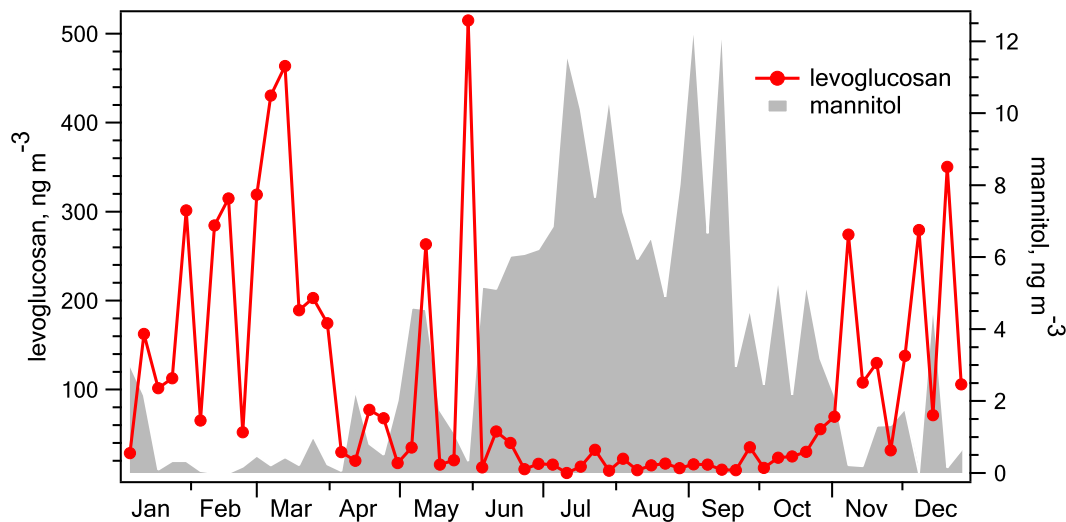


**Fig. 1.** Map of the EPA FRM and co-located Speciation sampling sites used in this study. Site names and locations are given in Table 1.

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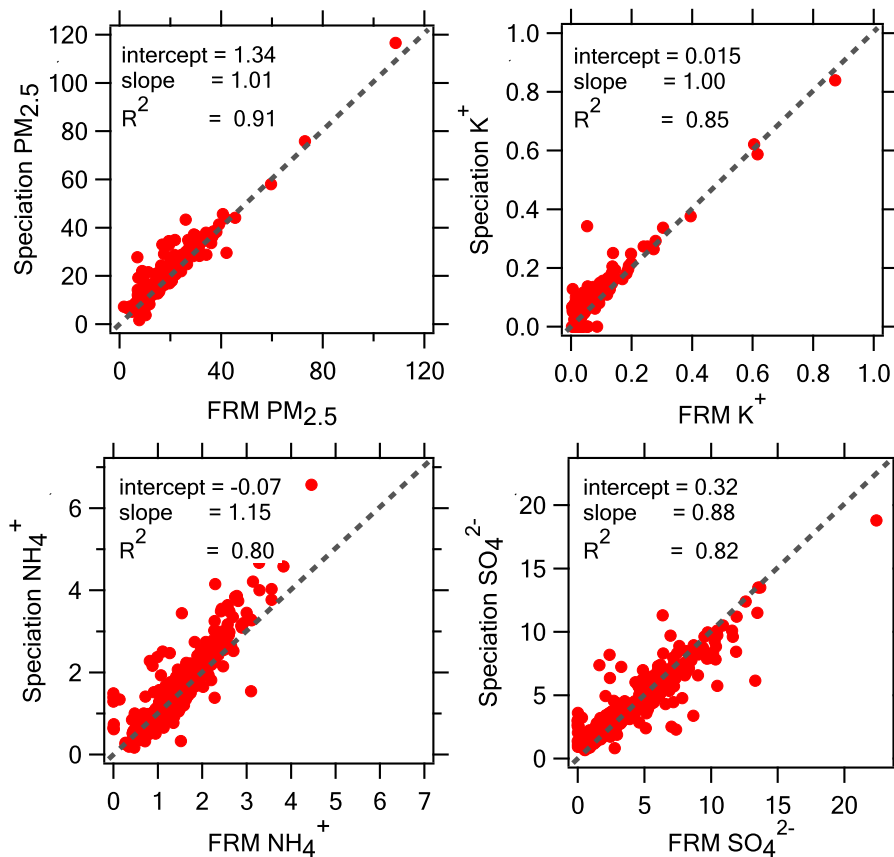
**Fig. 2.** Mean concentrations of levoglucosan and mannitol for all the sites in 2007.

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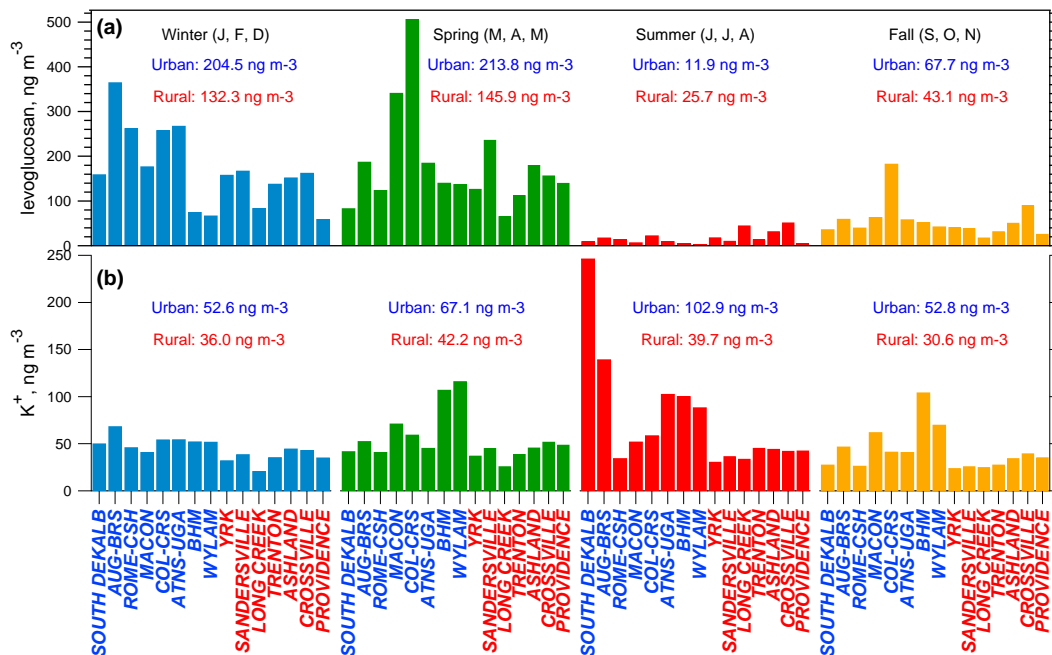
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**Fig. 3.** Intercomparison of  $\text{PM}_{2.5}$  mass,  $\text{K}^+$ ,  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$  concentrations (in  $\mu\text{g}/\text{m}^3$ ) between seven co-located EPA FRM and Speciation sites. FRM  $\text{K}^+$ ,  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$  are from the methods used in the analysis of all sites presented in this study. In all cases  $N=420$ .

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**Fig. 4.** Seasonal (Winter, Spring, Summer, Fall) variations of levoglucosan **(a)** and K<sup>+</sup> **(b)** concentrations at each site. Site names in blue and red are urban and rural sites, respectively. The mean concentrations of levoglucosan and K<sup>+</sup> by urban/rural segregation for each season are given in the plots.

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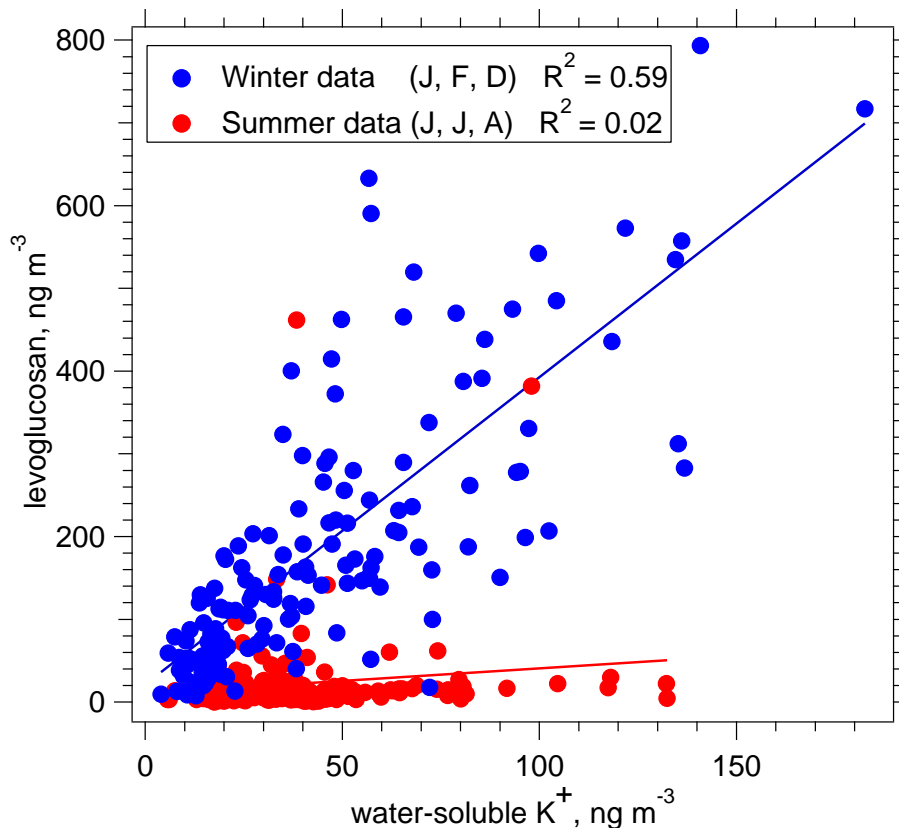
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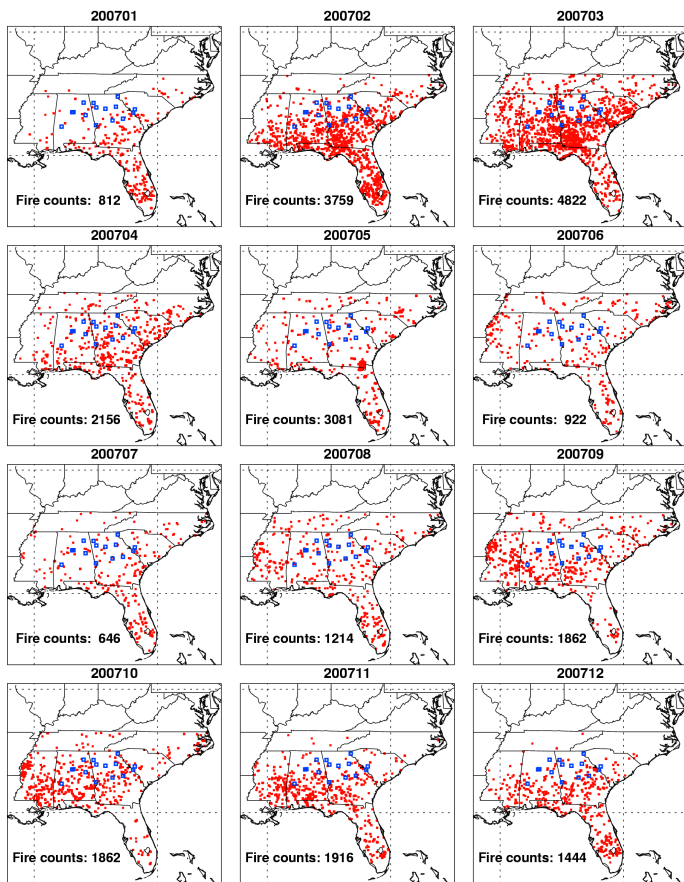
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**Fig. 5.** Relationship between levoglucosan and water-soluble K<sup>+</sup> concentrations in winter (January, February and December) and summer (June, July and August). Data on 5 July 2007 were not plotted and excluded from the regression calculation due to abnormally elevated levels of K<sup>+</sup> associated with fourth of July fireworks.

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### Aqua MODIS fire counts in 2007 over 7 states



**Fig. 6.** Monthly MODIS Aqua fire counts (red points) in 2007 over seven states in the southeastern US. FRM sampling sites are shown as blue square symbols.

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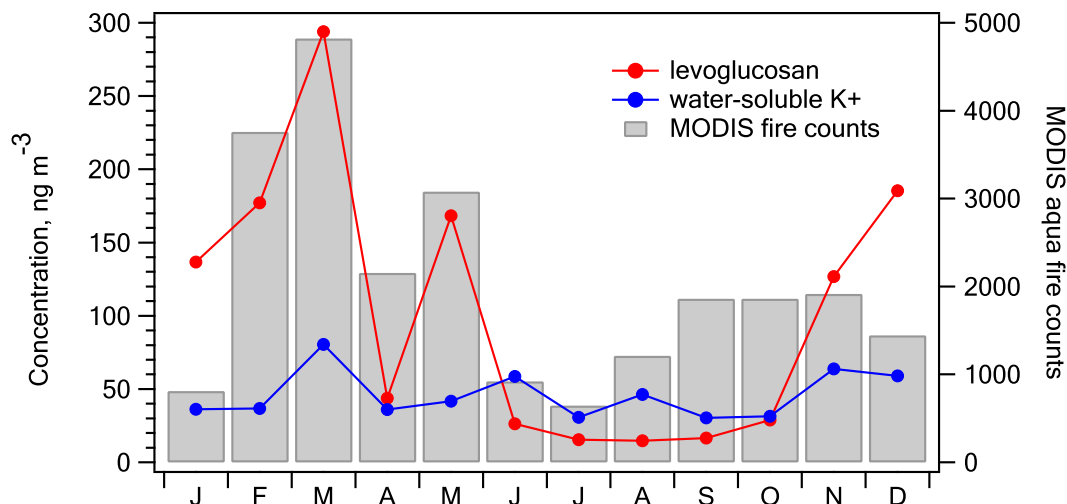
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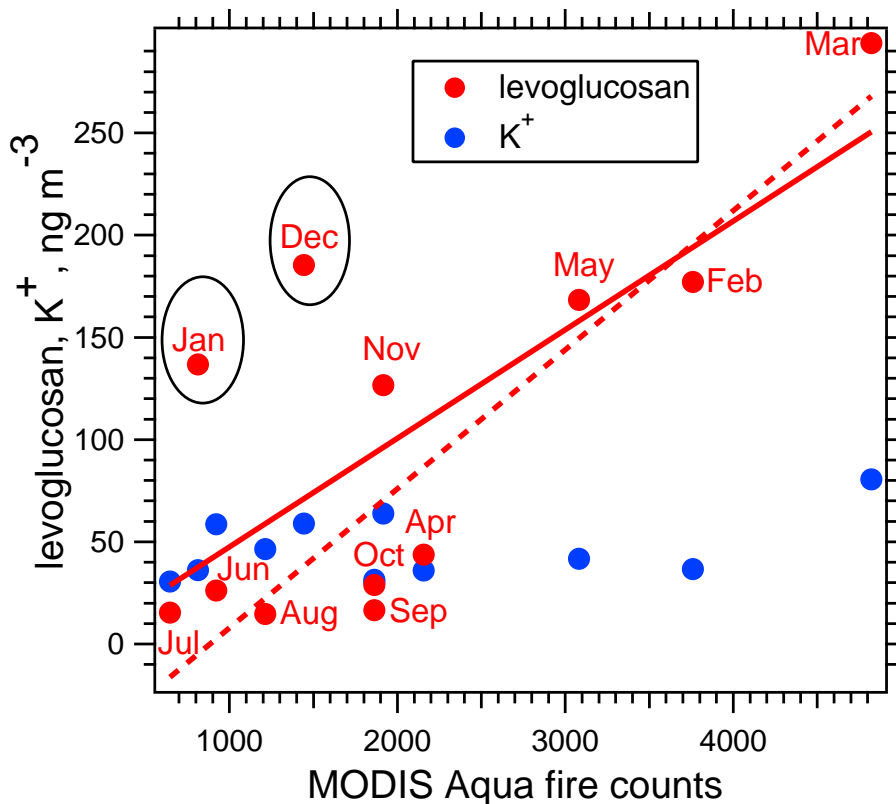


**Fig. 7.** Monthly mean levoglucosan and water-soluble potassium concentrations from all sites compared with monthly mean MODIS Aqua fire counts from seven states in the southeastern US.

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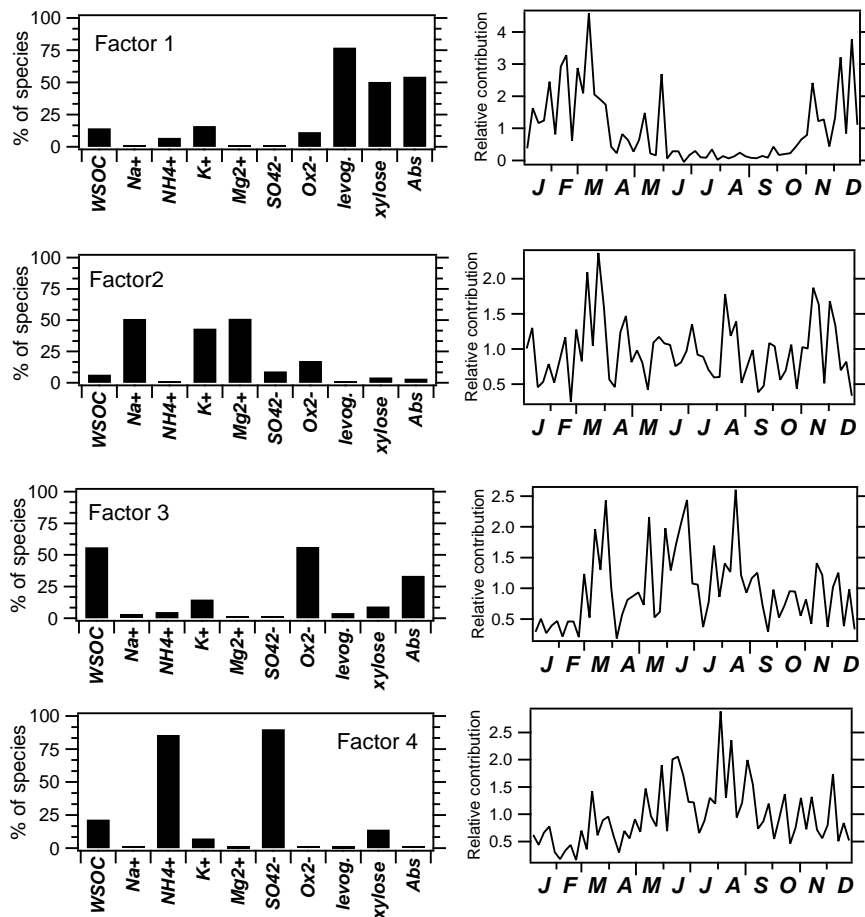


**Fig. 8.** Correlations of monthly mean concentrations of levoglucosan and K<sup>+</sup> versus fire counts. The red solid line is the fit of levoglucosan-fire counts correlation ( $r^2=0.54$ ). The red dashed line is the fit without January and December ( $r^2=0.86$ ).

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**Fig. 9.** Composition profiles (% of total of each species) for the four factors resolved by PMF based on data from the whole year (2007) at all sites (left panel), and the time series of relative contribution of each factor (right panel).

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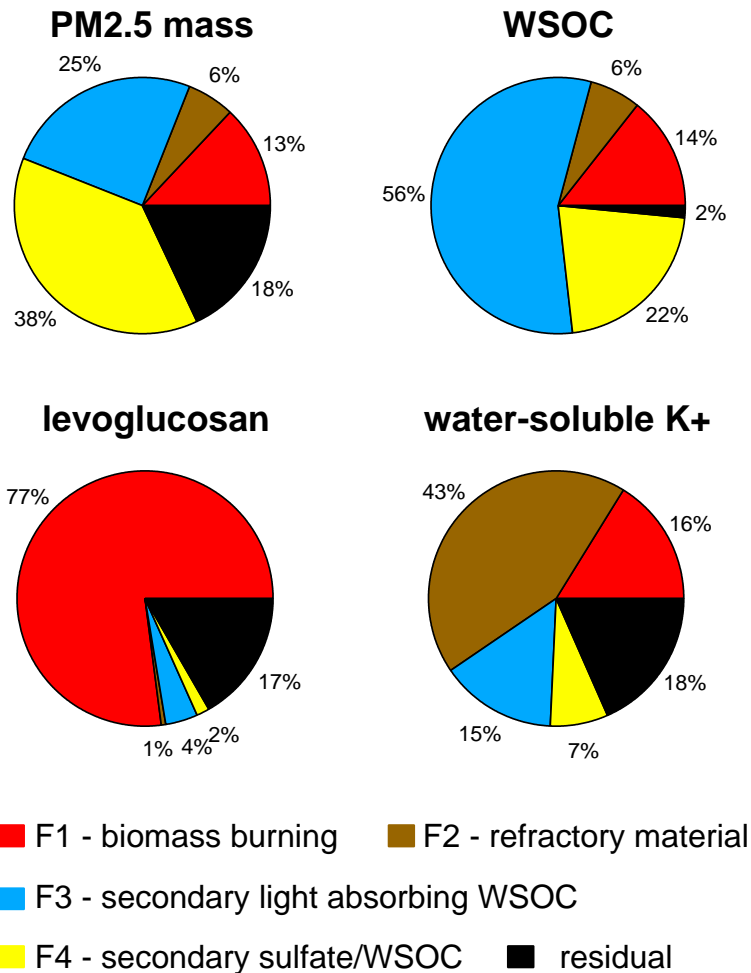
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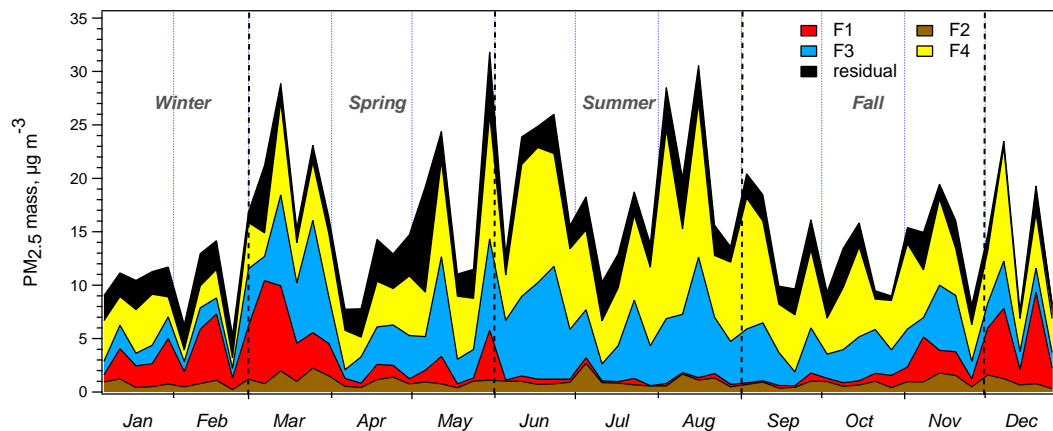
**Fig. 10.** PMF results showing annually averaged source contributions to PM<sub>2.5</sub> mass, WSOC, levoglucosan, and water-soluble K<sup>+</sup>.

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**Fig. 11.** Time series of PM<sub>2.5</sub> composition by four factors resolved by PMF and masses predicted by Multivariant Linear Regression (MLR).

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