



PM_{2.5} water-soluble elements in the southeastern United States

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PM_{2.5} water-soluble elements in the southeastern United States: automated analytical method development, spatiotemporal distributions, source apportionment, and implications for health studies

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Abstract

Water-soluble redox-active metals are potentially toxic due to the ability to catalytically generate reactive oxygen species (ROS) in vivo, leading to oxidative stress. As part of the Southeastern Center for Air Pollution and Epidemiology (SCAPE), we developed a method to quantify water-soluble elements, including redox-active metals, from a large number of filter samples ($N = 530$) in support of the Center's health studies. $PM_{2.5}$ samples were collected during 2012–2013 at various sites (three urban, two rural, a near-road, and a road-side site) in the southeastern US, using high-volume samplers. Water-soluble elements (S, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Se, Br, Sr, Ba, and Pb) were determined by extracting filters in deionized water and re-aerosolized for analyses by X-ray fluorescence (XRF) using an online aerosol element analyzer (Xact, Cooper Environmental). Concentrations ranged from detection limits (nominally 0.1 to 30 ng m^{-3}) to $1.2 \text{ } \mu\text{g m}^{-3}$, with S as the most abundant element, followed by Ca, K, Fe, Cu, Zn, and Ba. Positive Matrix Factorization (PMF) identified four factors that were associated with specific sources based on relative loadings of various tracers. These include: brake/tire wear (with tracers Ba and Cu); biomass burning (K); secondary formation (S, Se, and WSOC); and mineral dust (Ca). Of the four potentially toxic and relatively abundant metals (redox active Cu, Mn, Fe, and redox-inactive Zn), 51 % of Cu, 32 % of Fe, 17 % of Mn, and 45 % of Zn, were associated with the brake/tire factor. Mn was mostly associated with the mineral dust factor (45 %). These two factors were higher in warm (dryer) periods that favored particle re-suspension. Zn was found in a mixture of factors, with 26 % associated with mineral dust, 14 % biomass burning, and 13 % secondary formation. Roughly 50 % of Fe and 40 % of Cu was apportioned to the secondary formation factor, likely through increased solubility by sulfur-driven aerosol acidity. Linkages between sulfate and water-soluble Fe and Cu may account for some of the past observed associations between sulfate/sulfur oxide and health outcomes. For Cu, Mn, Fe, and Zn, only Fe was correlated with $PM_{2.5}$ mass ($r = 0.73\text{--}0.80$). Overall, mobile source emissions generated through mechanical processes (re-entrained road

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cially Fe and Cu, can act as a catalyst for the reactions, e.g., Fenton reaction that convert hydrogen peroxide to the more toxic hydroxyl radicals (Liochev and Fridovich, 2002; Stohs and Bagchi, 1995). Metal-mediated formation of free radicals may lead to DNA modifications, enhanced lipid peroxidation, and altered calcium and sulfhydryl homeostasis (Valko et al., 2005). Water-soluble transition metals have been identified as the potential contributors to reactive oxygen species (ROS) production by different ROS probes such as the DTT (dithiothreitol) (Charrier and Anastasio, 2012), AA (Ascorbate Acid) (Strak et al., 2012; Fang et al., 2015b), and macrophage (Saffari et al., 2014; Verma et al., 2010) assays.

Since most metals in PM have low solubilities (e.g., Zn ~ 50 %; Cu and Mn 10–40 %; Fe < 10 %) (Birmili et al., 2006; Espinosa et al., 2002), total element concentrations cannot be used to assess the roles of redox-active metals' potential effects on human health. The objective of this work, within the framework of the Southeastern Center for Air Pollution & Epidemiology (SCAPE) study, was to provide a reliable measurement of PM_{2.5} water-soluble element concentrations from filter samples collected at seven sites in the Southeastern US that represent different degrees of anthropogenic and traffic influence. A cost effective and automated method was required since over 500 filters were available for analyses. The resulting unique large and comprehensive dataset allowed for robust statistical analyses and has informed studies on particle ROS generating activities based on the DTT (Verma et al., 2014, 2015) and AA assays (Fang et al., 2015b). The data have provided new insights into the health effects of particulate water-soluble metals. This work focuses on a description of the measurement techniques, discussion on the spatio-temporal distribution, and source apportionment of water-soluble elements, with a specific focus on four important health-related water-soluble metals (Fe, Cu, Mn, and Zn).

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4. Georgia Tech, GA (GT), a rooftop site on Georgia Tech campus, roughly 30 m a.g.l. (above ground level), 840 m from the RS site, providing an intermediate location between RS and the central urban site (JST);

5. Birmingham, AL (BHM), an urban site within a few kilometers of significant transportation and industrial sources;

6. Centerville, AL (CTR), the rural pair of BHM, surrounded by forests and a lightly traveled rural road;

7. East St. Louis, IL (Sauvain et al., 2008), an urban residential/light commercial area approximately 3 km east of the central business district of St. Louis, MO.

JST, YRK, BHM, and CTR are all part of the Southeastern Aerosol Research and Characterization Study (SEARCH) network sites (Hansen et al., 2003). The sampling approach involved paired simultaneous measurements: one high-volume sampler always at JST and the other sampler moved among RS, GT and YRK on a monthly basis, during different seasons. Paired sampling at BHM and CTR was also undertaken for a month and coincided with the Southern Oxidant and Aerosol Study (SOAS). Detailed sampling schedule and map can be found in Table S1 and Fig. S1 (in the Supplement). Samples were collected from June 2012 to September 2013. In November 2012, two Hi-Vol samplers were co-located at JST for side-by-side comparisons. A total of 530 filters were collected as part of the study. In all cases, collected filter samples were immediately wrapped in prebaked aluminum foil and stored at -18°C until analyzed.

2.1.2 Filter preparation

Four punches of the Hi-Vol filter (5.07 cm^2 each) were extracted in 15 mL of deionized (DI) water ($> 18\text{ M}\Omega\text{ cm}^{-1}$) in a sterile polypropylene centrifuge tube (VWR International LLC, Suwanee, GA, USA) by sonication (Ultrasonic Cleanser, VWR International LLC, West Chester, PA, USA) for half an hour. Extracts were then filtered using PTFE

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liquid sample (at least 6 mL) was loaded by a DIONEX automated sampler (AS40, DIONEX Corporation, Sunnyvale, CA, USA) through a SelectPro two-position fluid processor valve (Alltech, Deerfield, IL, USA) to a 5 mL PEEK sample loop (Upchurch Scientific, Inc., Oak Harbor, WA). After 2.5 min, at which point the sample loop had been completely filled with extract liquid from the Dionex autosampler, the valve was switched to the “Inject” position and all 5 mL of sample injected to an ultrasonic nebulizer (CETAC U5000 AT+, CETAC Technologies, Omaha, NE, USA) via a carrier DI flow of 0.42 mL min⁻¹ (DI₁, Fig. 1) propelled by a peristaltic pump (Ismatec, Cole-Parmer Instrument Company, Vernon Hills, IL, USA). In the continuous flow ultrasonic nebulizer, liquid sample was converted to a fine aerosol spray and directed by filtered carrier room air of 1.62 L min⁻¹ through an evaporator at 136 °C followed by a condenser at 3 °C. The dry aerosolized sample was neutralized by a Kr-85 (Model 3077A, TSI) ion source and then mixed with clean filtered (Pall HEPA Capsule) make-up air drawn into the Xact and through the filter tape by the Xact’s flow control system. After 14 min, at which point all sample in the sample loop had been transferred to the Xact filter tape, the system switched to “Load” position again, and DI water with HNO₃ (final concentration = 2 %) was loaded onto the sample loop (2.5 min), after which the system switched to “Inject” position again, and the 2 % HNO₃ was directed to the nebulizer and the Xact. This cycle was performed to wash off any metal residuals in the liquid system. After 11 min, the filter tape was automatically advanced to a position where total mass for each element from the sample was measured by XRF analysis. At the same time, the system repeated the process for measuring the next sample. To further ensure no carry-over between samples, a faster DI flow (DI₂, 2.09 mL min⁻¹, Fig. 1) was used for flushing while the SelectPro valve was at the “Load” position. Inserting 2 % HNO₃ between samples was found to be an effective method to eliminate carry-over between samples (Fig. S2).

The final ambient concentration of each element was calculated as follows:

$$C_a = \frac{(C_{\text{sample}}V_{\text{sample}} - C_{\text{blank}}V_{\text{blank}})}{m} \times \frac{15 \text{ mL} \frac{A_{\text{filter}}}{A_{\text{punches}}}}{5 \text{ mL} \times Qt} \quad (1)$$

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where C_a is the specific element ambient concentration (ng m^{-3}); C_{sample} (C_{blank}) and V_{sample} (V_{blank}) are the concentration of element (ng m^{-3}) and volume of air (m^3) drawn through the filter tape for sample (blank), respectively, both reported by the XRF in the 30 min sampling time. 15 mL is the volume of DI water used for the filter extraction and 5 mL is the sample liquid volume loaded to the Xact. A_{filter} is the total particle collection area of the Hi-Vol filter (m^2) and A_{punches} is the area used for this analysis (m^2). Q is the Hi-Vol sampling flow rate ($\sim 1.13 \text{ m}^3 \text{ min}^{-1}$) and t is the sampling duration (min). m is the calibration factor determined by multiple external element standard solutions, discussed below (Sect. 2.3).

A similar automated system with the DIONEX sampler, SelectPro valve, and a peristaltic pump was also used to measure water-soluble organic carbon (WSOC) on the extracts from the same Hi-vol filters. Filter extracts ($\sim 6 \text{ mL}$) that had been loaded into a 5 mL sample loop, were first passed through a 1 m Liquid Wave-guide Capillary Cell (LWCC-M-100; World Precision Instruments, Inc., FL, USA), where absorbance at 365 nm wavelength (BrC) was measured (not included in this work). The extracts then entered a TOC analyzer (Sievers Model 900, GE Analytical Instruments, Boulder, CO, USA) for determining WSOC concentration (Sullivan et al., 2006).

2.3 Calibration

Multiple element-ion standard stock solutions [$(\text{NH}_4)_2\text{SO}_4$, CaCl_2 , $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2$, CuSO_4 , ZnCl_2 , and MnCl_2] were prepared by dissolving powders in DI water with HNO_3 (2% final concentration) and stored in a refrigerator ($T = 4^\circ\text{C}$). CuSO_4 and $(\text{NH}_4)_2\text{Fe}(\text{SO}_4)_2$ were obtained from Sigma-Aldrich; CaCl_2 , MnCl_2 , and ZnCl_2 were obtained from Alfa Aesar; $(\text{NH}_4)_2\text{SO}_4$ was from Fisher Scientific. Final standard solutions were diluted from stock (20–200 times dilution), 2% HNO_3 added, transferred to DIONEX vials, and ran through the same system as described above.

Figure 2 shows the system calibration using serial dilutions of multiple element-ion standards. Linear regression yielded r^2 larger than 0.98 for all cases and similar slopes

(0.47–0.60), indicating that the nebulizer efficiency and other losses in the system were not dependent on the specific element. Slopes from measured mass versus calculated mass of element-ion standard solutions for all standards were averaged and used as the calibration factor ($m = 0.53 \pm 0.05$ in Eq. 1) to interpret all elements in samples.

The intercepts were not included in the final ambient concentration calculation (Eq. 1) since the intercept of sample and blank cancel out after blank subtraction. The standards were made in the range of typical sample concentrations and the intercepts were negligible (< 2 % of typical ambient levels). One standard ion solution was measured for every five filter samples and the coefficients of variation [CV, calculated as standard deviation (σ)/mean, %] were less than 10 % throughout the analyses of all filters, indicating that the system was capable of stable and reproducible operation.

2.4 Blanks, LOD and uncertainties

The limits of detection (LOD) based on blank levels obtained from Hi-Vol filter blanks ($N > 40$), and various uncertainties, as well as overall uncertainties for all elements are given in Table 1. The concentration of blanks was calculated using Eq. (1) assuming 23 h sampling at $1.13 \text{ m}^3 \text{ min}^{-1}$ and LOD was determined by three times the standard deviations of blanks. Values below LOD for S, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Se, Br, Sr, Ba, and Pb were assigned as half of LOD values in all the statistical analyses below. Other elements detected by the Xact, such as Sc (Scandium), V (Vanadium), Cr (Chromium), Co (Cobalt), Ni (Nickel), Ge (Germanium), Rb (Rubidium), Ag (Silver), Cd (Cadmium), and Hg (Mercury), for which > 50 % of aerosol samples were below the detection limits, are not included in the table or subsequent discussion. Calculation of uncertainties are discussed in Sect. 2.5. For most elements, overall uncertainties are less than 20 %. Ti, Sr, and Cu have the highest uncertainties of 28, 25, and 25 %, respectively.

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2.5 Source apportionment

A source apportionment analysis was performed with Positive Matrix Factorization (PMF) (Paatero and Tapper, 1994) using EPA PMF 5.0 software. PMF analysis was applied on the combined data from JST (summer, fall, winter 2012, and spring 2013), GT (fall, winter 2012, and fall 2013), and RS (winter 2013 and fall 2013) (total $N = 299$). Although the road-side site generally has higher levels of metals than the urban site (JST) and near-road site (GT) (discussed in Sect. 3.2.2), merging RS data with JST and GT in the analysis did not alter the PMF solutions substantially (factor profiles and source contributions), but the larger number of input data resulted in a more robust PMF solution. 14 elements (S, K, Ca, Ti, Mn, Fe, Cu, Zn, As, Se, Br, Sr, Ba, and Pb) and WSOC were run in the model with Ti and As categorized as weak species (low S/N signals). The concentrations, together with the uncertainties, were used as the input for PMF runs. Missing data were replaced by species median with 400% uncertainty and values below LOD were assigned as half of LOD values with uncertainties of 5/6 the concentration (Polissar et al., 1998). For other data, uncertainties for each species were determined by multiplying the concentration by overall uncertainties (%). Overall uncertainty was calculated from the sum of square of various uncertainties including filter sampling (5%), extraction (5%), blanks (1σ of multiple blanks, 2–15% depending on species), calibration (1σ of slope, 10%), and analytical uncertainty. The analytical uncertainty for elements was obtained by analyzing the same sample, a composite of extracts from 11 selected road-side samples 20 times and calculating the coefficient of variation (CV, %). The measured uncertainty for each element, based on two side-by-side Hi-Vol samplers at JST, was also included in the overall uncertainty. This was done because the calculated uncertainties for some elements (e.g., Cu and Sr) are much smaller than the measured uncertainties from collocated measurements (Table 1). By combining two uncertainties, the uncertainties for all elements are slightly overestimating. Uncertainty from collocated measurements was calculated as the relative uncertainty of the slope ($1\sigma/\text{slope}$), which was based on an orthogonal

regression (discussed below, see Fig. 4). Both uncertainties and the combined overall uncertainties for each element are given in Table 1. The PMF model was executed with 3–8 factors. Based on minimized Q values and physical interpretation of the solutions, a 4-factor solution was found to be optimal. Details on determining the optimal factor and bootstrapping results can be found in the Supplement.

3 Results and discussions

3.1 Comparison with ion chromatography (IC)

As a further test of the system, sulfate concentrations were determined on a subset of the Hi-Vol filter samples by ion chromatography and compared to the elemental analysis of sulfur. 200 filters that included samples from JST and GT were extracted in DI water (same procedure discussed in Sect. 2.1, but without adding HNO_3 in the extracts) and inorganic sulfate (IC-sulfate) was measured by an ion chromatography (IC, DX500 with UTAC-ULP1 concentrator column, AG11 guard column, and AS11 anion column, DIONEX, CA). IC-sulfate was then divided by 3 to convert to sulfur mass (molar mass of sulfate and sulfur are 96 and 32 g mol^{-1} , respectively) and directly compared to XRF-sulfur. Orthogonal regression shows good quantitative agreement and correlation coefficient (r^2) of 0.96 (Fig. 3). The discrepancy between sulfur measured by XRF and IC (slope of IC-sulfur vs XRF-sulfur = 0.79 ± 0.01) may be attributed to roughly 20% contributions from additional sulfur species, such as organosulfates that are not detected by the IC. Lower sulfur measured by IC versus XRF has also been observed in other studies (Shakya and Peltier, 2015; Tolocka and Turpin, 2012; He et al., 2001). However, Hidy et al. (2014) found no statistical evidence for organosulfates in the Southeast by this difference method.

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3.2 Water-soluble elements

3.2.1 Inter-comparisons of two collocated Hi-Vols

Inter-comparisons of elements from two collocated Hi-Vol samplers ($N = 11$) at JST in November 2012 are shown in Fig. 4, and results included in Table 1. Orthogonal regressions resulted in strong correlations for most elements ($r^2 = 0.73$ – 0.99) with moderate correlations for Cu ($r^2 = 0.57$) and Sr ($r^2 = 0.61$). The slopes show percentage differences (1-slope) are 1–10% for Cu, K, and Ti, 11–20% for S, Zn, Ca, Br, As, and Sr, and 20–36% for Pb, Mn, Se, Fe, and Ba. Overall, the two Hi-Vols show good agreement for measuring water-soluble elements, considering the uncertainties from sampling, filter preparations, and extractions. The uncertainties in slope (1σ) were used in calculating the overall uncertainties (Sect. 2.3). The intercepts were relatively small and thus ignored.

3.2.2 Spatial and temporal trends

Monthly average concentrations of water-soluble elements at various sampling sites are given in Fig. 5. Three seasons (summer, fall, and winter) were grouped based on the temperature profiles in 2012 and 2013, consistent with our previous work (Fang et al., 2015a; Verma et al., 2014). As seen from Fig. 5, mass concentrations of water-soluble elements span a wide range, from 0.1 ng m^{-3} to $1.2 \mu\text{g m}^{-3}$. S is the most abundant water-soluble element of the group measured, comprising $71 \pm 14\%$ of the total measured element mass. Ca, K, Fe, Cu, Zn, and Ba follow with average ($\pm 1\sigma$) fractions of 16 (± 12)%, 8 (± 5)%, 1.8 (± 1.3)%, 1.4 (± 2.1)%, 1.4 (± 1.4)%, and 0.5 (± 0.6)%.

Seasonal variability of all elements can be examined from the Atlanta urban sites (JST, GT, and RS). Besides S, commonly found to have higher concentration in summer due to the higher SO_2 oxidation rates in warm seasons (Hidy et al., 2014), most of the water-soluble elements also had higher concentrations in summer/fall, such as Ca, Fe, Cu, Mn, Sr, and Se. The seasonal variability of these elements may be explained by

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cific elements identified as various source tracers. Note, metals as source tracers are typically based on total metals, whereas here we are using the measured water-soluble concentrations. Factor profiles and time series plots (from the JST site) are shown in Fig. 7a and b, respectively. The percentage contribution of the various factors (sources) to the four important health-related metals (Cu, Fe, Zn, and Mn) is shown in Fig. 8. Breakdown of sources for the other water-soluble elements and WSOC can be found in Fig. S3.

The factor with high loadings for Ti, Fe, Cu, Zn, and Ba is identified as a brake/tire wear source as it includes products from brake pads or linings, such as Cu (Adachi and Tainosho, 2004; Sternbeck et al., 2002; Garg et al., 2000), Fe (Adachi and Tainosho, 2004; Garg et al., 2000; Hopke et al., 1980), Ti (Adachi and Tainosho, 2004), and Zn (Adachi and Tainosho, 2004; Sternbeck et al., 2002), and tracers of tire wear, e.g., Zn (Harrison et al., 2012). A biomass burning factor is identified by high concentrations of K, Br, As, and Pb. While K is a typical component in biomass burning aerosols, Br (Turn et al., 1997) and Pb (Richard et al., 2011) have also been found in wood combustion. The time series plot (Fig. 7b) showed a higher contribution of this factor in winter than in summer ($winter_{avg}/summer_{avg} \approx 1.35$), consistent with the observed winter enhancement of biomass burning emissions in Atlanta (Zhang et al., 2010). Some water-soluble metals (e.g., Se, Fe, Br, Pb, As, Mn, Ba, and Zn) were apportioned to the biomass burning factor (see Figs. 8 and S3). The total form of some of these metals (Fe, Mn, Zn, and Cu) (Chang-Graham et al., 2011) and water-soluble Fe have been seen in biomass burning in other studies (Oakes et al., 2012). The third factor, referred to as secondary formation, is characterized by high S, WSOC, and Se, and some Fe, with higher contributions in summer than winter ($summer_{avg}/winter_{avg} \approx 2.37$). For the last source, a mineral dust origin is suggested by high loadings of Ca, Mn, and Sr, all indicators of crustal material. The results have implications for health studies.

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important ROS sources (Bates et al., 2015). In contrast, these redox active metals are not found at significant levels in the biomass burning factor (Fig. 7), which has been found to be a contributor to the PM_{2.5} DTT activity in the SCAPE study (Verma et al., 2014, 2015; Bates et al., 2015), indicating that redox active organic species dominate in that case.

The major identified source for Mn is mineral dust (45 %) with other sources making relatively similar contributions: secondary formation (18 %), biomass burning (16 %), and brake/tire wear (17 %). At YRK, the rural site least affected by traffic, Mn correlates best with Ca ($r = 0.91$) and Sr ($r = 0.82$, Table S3) in summer and K ($r = 0.86$), Ca ($r = 0.86$), Zn ($r = 0.89$), and Sr ($r = 0.79$) in winter, all indicative of regional mineral dust contributions, consistent with the regional characteristic of Mn discussed in Sect. 3.2.2.

For Fe, besides the brake/tire factor, a large fraction (47 %) is apportioned to the secondary formation factor, and Fe correlates well with S at JST ($r = 0.71$), YRK ($r = 0.76$), and GT ($r = 0.73$) in summer, and at JST ($r = 0.76$) in fall 2012 and RS ($r = 0.74$) in fall 2013. Fe and S are moderately correlated at RS in fall 2012 ($r = 0.62$), and GT ($r = 0.55$) in fall 2013. Highest correlations occur when secondary atmospheric processing (oxidation) is strong. These results are consistent with a previous study involving single particle chemical analysis on PM_{2.5} particles in Atlanta, which showed that sulfate is an important proxy for Fe solubility (Oakes et al., 2012) by affecting aerosol pH, or as an indicator of iron sulfates, which are soluble and possibly formed at some point earlier in the particles lifespan under acidic conditions. The correlation between Fe and S might explain past associations found in other studies between sulfate/sulfur oxide and health endpoints (Atkinson et al., 2010; Sarnat et al., 2008; Pope et al., 2002; Gwynn et al., 2000; Dockery et al., 1996; Raizenne et al., 1996).

PM_{2.5} mass is regulated and has been associated with adverse health endpoints in many studies (Laden et al., 2000; Pope et al., 2002, 2004; Metzger et al., 2004; Sarnat et al., 2008). Overall, water-soluble iron was highly correlated with PM_{2.5} mass ($r = 0.73$ – 0.80 , Table S4), due to a correlation with sulfate (e.g., role of sulfate on aerosol pH). Interestingly, Fe was correlated with PM_{2.5} mass even in some cases

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when S did not co-vary with PM_{2.5} concentration. These cases are all in winter when the *r* values between PM_{2.5} mass and S were 0.52, 0.35, 0.26, and 0.23, while those with Fe were 0.80, 0.75, 0.76, and 0.74 at JST (Dec), JST (March), GT (March), and RS (Feb), respectively. The exceptions were the two rural sites (YRK and CTR), where water-soluble Fe was moderately or not correlated with PM_{2.5} at all (*r* = 0.69 at YRK *r* = 0.3 at CTR) while S and PM_{2.5} still had high correlations (*r* = 0.78 and 0.75 at YRK and CTR, respectively).

Mn and Zn show some correlations with PM_{2.5} mass as well, but only during two periods (for Mn, *r* = 0.84 at GT and 0.76 at RS, both in winter, for Zn, *r* = 0.71 and 0.76 at JST in December and March, respectively). No significant correlations were ever found between PM_{2.5} and Cu. It has been demonstrated that water-soluble metals (Zn, Cu, and Fe) can have adverse effects on the respiratory system and our observations suggest that some water-soluble metals, especially Fe, are correlated with PM_{2.5} mass. However, some epidemiological studies point to organic carbon (Peel et al., 2005; Metzger et al., 2004) and PM_{2.5} mass (Sarnat et al., 2008; Pope et al., 2002, 2004; Metzger et al., 2004; Laden et al., 2000) but not metals. Li et al. (2009) suggests that redox-active organic chemicals could play major roles in PM toxicity and metals may synergize with organic PM components to further escalate oxidative stress. Thus, these water-soluble metals could play both dominant and important secondary roles in driving observed associations between fine particles and adverse health.

4 Conclusions

Over 500 PM_{2.5} filter samples (23 h integration time) were collected during 2012–2013 at multiple sites (three urban, two rural, one near-road, and one road-side site) in the southeastern US, using paired (simultaneous measurements at two different sites) high-volume samplers, as part of the Southeastern Center for Air Pollution & Epidemiology (SCAPE) project. A focus of SCAPE was assessing the role of PM_{2.5} associated reactive oxygen species (ROS) on health effects. Because water-

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Table 1. Limits of detection (LOD), blanks ($N > 40$) and uncertainties for all water-soluble elements.

Element	LOD, ng m ⁻³	Blank, ng m ⁻³	Analytical uncertainty, %	Sum of square of various uncertainties, %	Uncertainty from collocated measurements, %	Overall uncertainty, %
Sulfur (S)	6.19	6.64	2.19	7.77	2.38	8.13
Potassium (K)	16.45	11.80	2.47	9.89	6.06	11.63
Calcium (Ca)	29.64	32.23	2.38	10.46	11.76	15.87
Titanium (Ti)	0.09	0.06	15.79	21.08	18.68	28.22
Manganese (Mn)	0.11	0.06	3.57	8.54	6.49	10.74
Iron (Fe)	3.11	1.54	12.59	13.94	14.06	19.83
Copper (Cu)	0.91	0.70	2.37	8.57	23.85	25.38
Zinc (Zn)	1.87	1.89	4.56	9.90	8.24	12.90
Selenium (Se)	0.02	0.01	8.94	12.17	7.79	14.46
Bromine (Br)	0.03	0.02	3.93	8.50	4.49	9.62
Strontium (Sr)	0.11	0.08	3.57	9.63	23.53	25.46
Barium (Ba)	1.13	0.37	3.29	10.59	14.08	17.73
Arsenic (As)	–	0	6.97	10.17	3.53	10.77
Lead (Pb)	0.14	0.12	7.45	11.77	1.32	11.85

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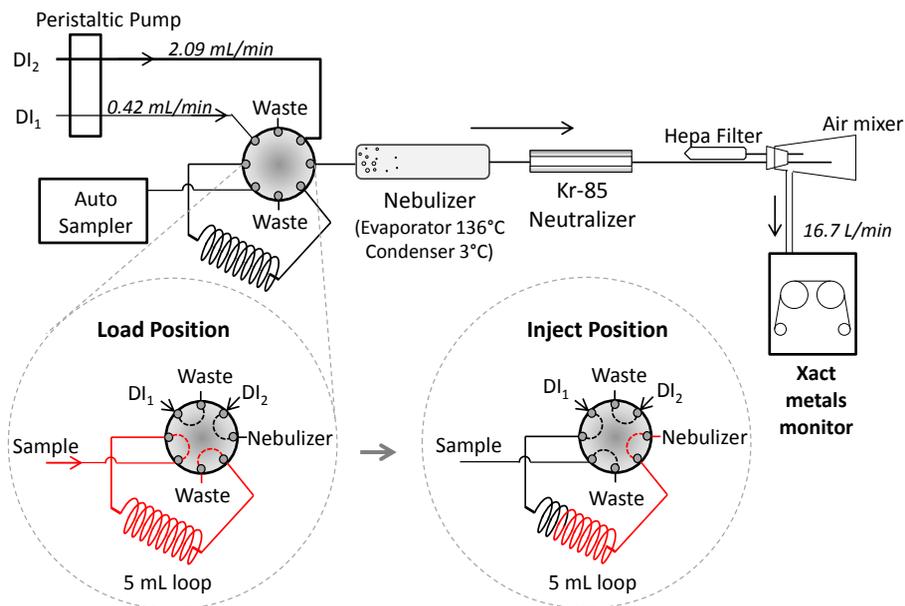


Figure 1. Schematic of automated system developed to measure elements in the water-soluble aerosol extracts using an online XRF element analyzer (XactTM 625).

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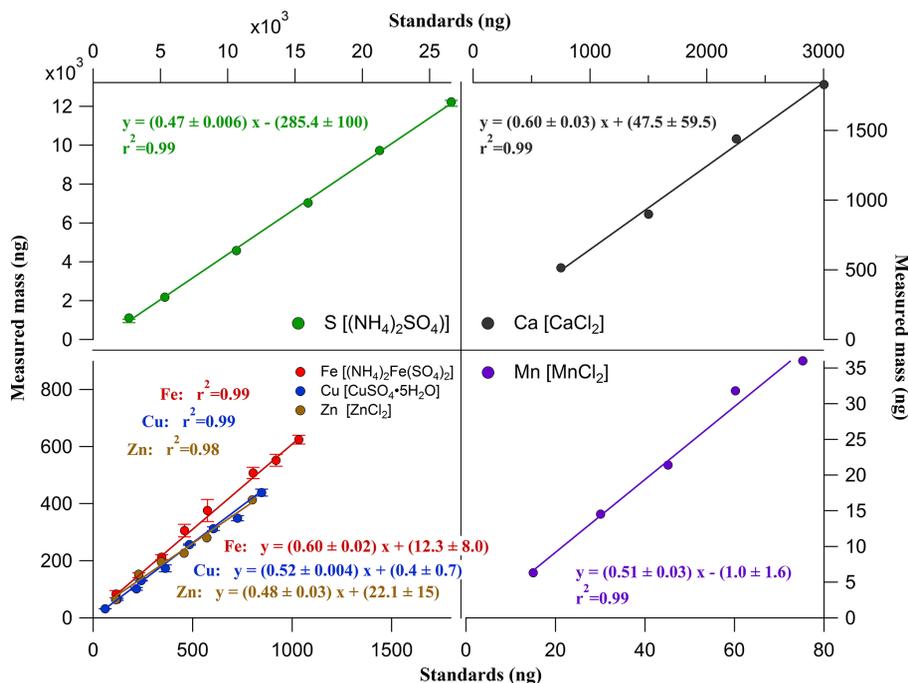


Figure 2. System calibration based on multiple element-ion standard solutions. Error bars represent the standard deviation of three replicates. Slopes and intercepts are based on orthogonal regression with errors as one standard deviation.

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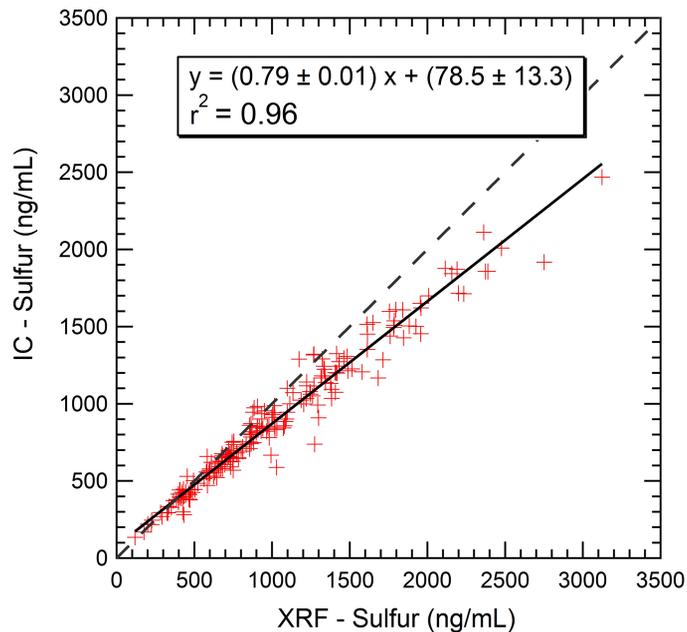


Figure 3. Sulfur measured by XRF and 1/3 of sulfate measured by IC (results of orthogonal regression are shown, along with 1 : 1 ratio by dotted line).

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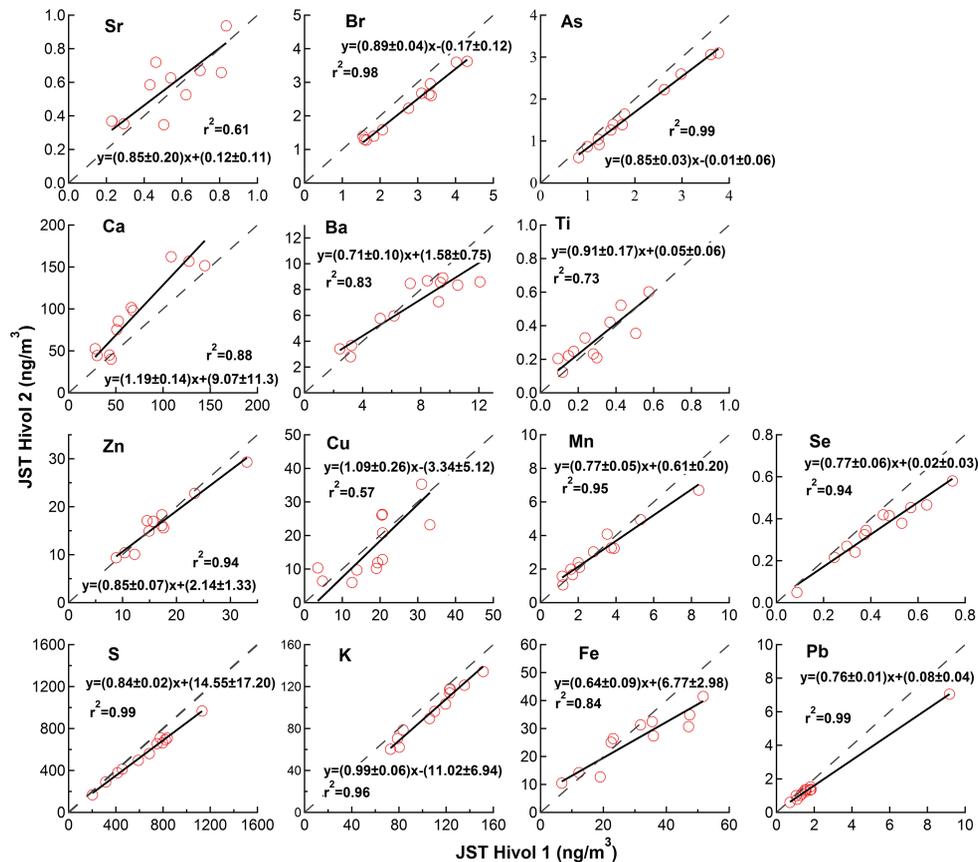


Figure 4. Precision from collocated measurements assessed by filter samples ($N = 11$) collected simultaneously using two Hi-Vol samplers deployed at JST during November 2012. (Analysis was done by orthogonal regression. The dotted line is 1 : 1.)

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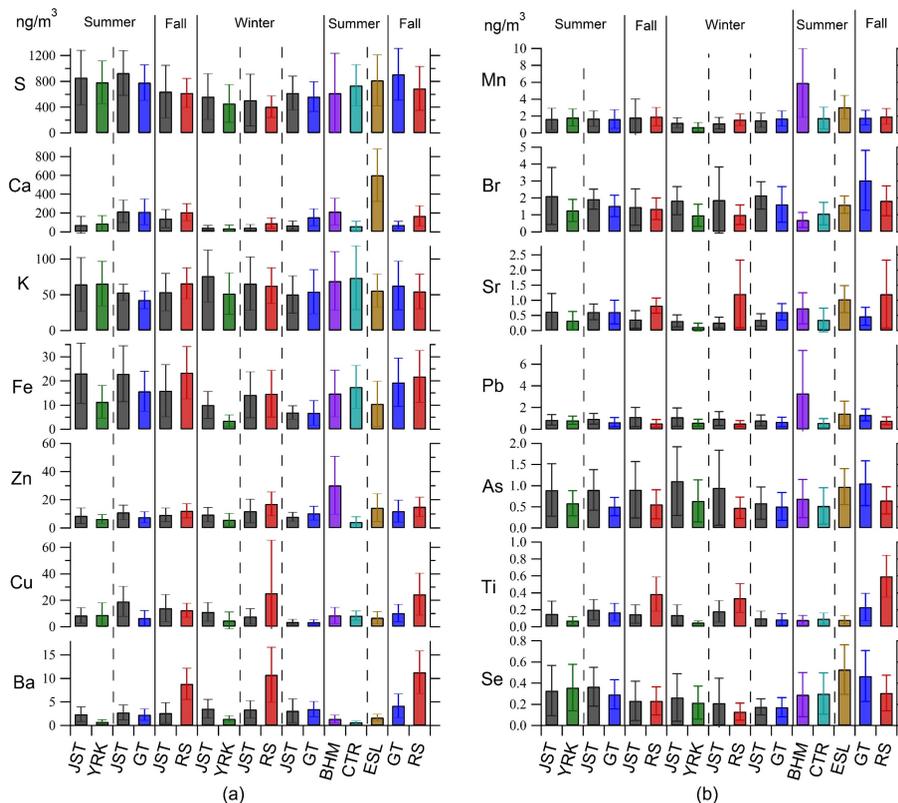


Figure 5. Monthly mean (\pm standard deviation) of water-soluble elements ambient concentration (ng m^{-3}) at various sampling sites. Seasons are separated by solid lines and simultaneous sampling at paired sites are separated by dashed lines (urban – JST, BHM, ESL; rural – YRK, CTR; near-road – GT; road-side – RS).

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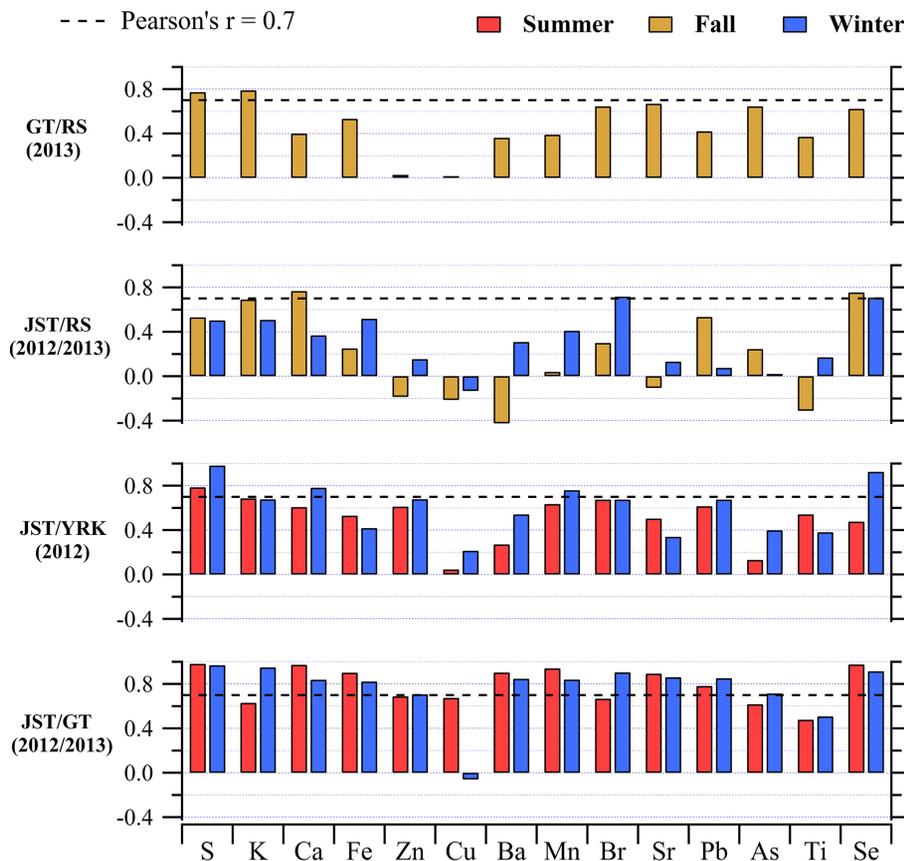


Figure 6. Correlations (Pearson's r) between paired sites for various water-soluble elements in Georgia (JST, RS, GT, and YRK).

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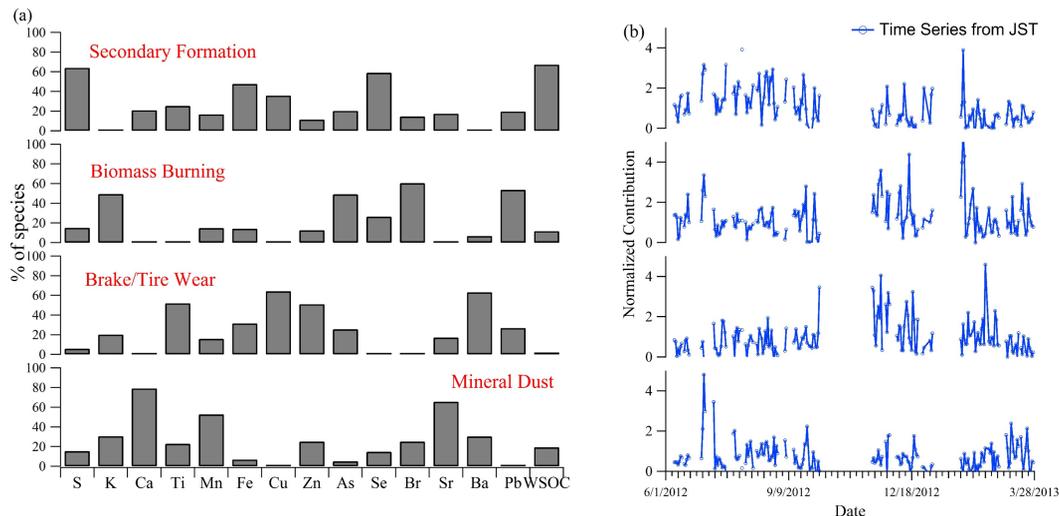


Figure 7. Loading of measured water-soluble elements into various PMF resolved factors for all Atlanta sites (a) and factor time series of source contributions resolved from the Jefferson Street Site (JST, urban Atlanta) (b).

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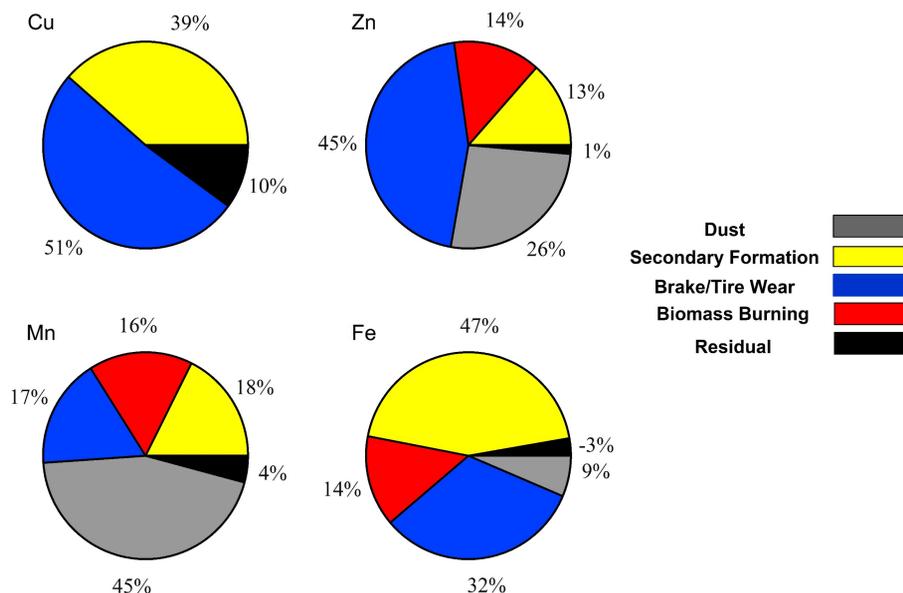


Figure 8. Factor contributions for water-soluble Cu, Zn, Mn, and Fe in PM_{2.5} based on the PMF analyses.

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