Source identification of PM$_{2.5}$ in an arid Northwest U.S. City by positive matrix factorization

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Abstract

Spokane, WA is prone to frequent particulate pollution episodes due to dust storms, biomass burning, and periods of stagnant meteorological conditions. Spokane is the location of a long-term study examining the association between health effects and chemical or physical constituents of particulate pollution. Positive matrix factorization (PMF) was used to deduce the sources of PM$_{2.5}$ (particulate matter ≤ 2.5 μm in aerodynamic diameter) at a residential site in Spokane from 1995 through 1997. A total of 16 elements in 945 daily PM$_{2.5}$ samples were measured. The PMF results indicated that seven sources independently contribute to the observed PM$_{2.5}$ mass: vegetative burning (44%), sulfate aerosol (19%), motor vehicle (11%), nitrate aerosol (9%), airborne soil (9%), chlorine-rich source (6%) and metal processing (3%). Conditional probability functions were computed using surface wind data and the PMF deduced mass contributions from each source and were used to identify local point sources. Concurrently measured carbon monoxide and nitrogen oxides were correlated with the PM$_{2.5}$ from both motor vehicles and vegetative burning.

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1. Introduction

Located in a semiarid, eastern Washington valley, Spokane is subject to frequent dust storms and pollutant-trapping temperature inversions. As a result, Spokane is classified as a non-attainment area for PM_{10} (particulate matter ≤ 10 μm in aerodynamic diameter). According to the emission inventories (Washington State Department of Ecology, 1994), the major anthropogenic source of PM_{10} in Spokane is airborne soil and vehicle exhaust. Because of the recurrent exceedances of the air quality standard, a number of studies have been conducted to understand particulate air pollutants in Spokane (Kantamaneni et al., 1996; Norris, 1998; Claiborn et al., 1998; Haller et al., 1999; Villasenor et al., 2001). Currently, Spokane is the location of a long-term, time series study examining the association between health effects and constituents of particulate pollution (Norris et al., 2000).

As part of this study, advanced source apportionment methods for the airborne particulate matter are required. Positive matrix factorization (PMF) (Paatero, 1997) has been shown to be a powerful alternative to traditional receptor modeling of airborne particulate matter (Huang et al., 1999; Willis, 2000; Qin et al., 2002). PMF has been successfully used to assess particle source contributions in the Arctic (Xie et al., 1999), in Hong Kong (Lee et al., 1999), in Thailand (Chueinta et al., 2000), in Phoenix (Ramadan et al., 2000), in Vermont (Polissar et al., 2001), in three northeastern U.S. cities (Song et al., 2001), in Atlanta (Kim et al., in press).

The objectives of this study are to identify particulate matter sources and estimate their contributions to the particle mass concentrations. In the present paper, PMF was applied to a $PM_{2.5}$ (particulate matter ≤ 2.5 μm in aerodynamic diameter) compositional data set of daily samples collected during a 3-year period at a residential monitoring site in Spokane, WA. The resolved $PM_{2.5}$ sources and their seasonal trends are discussed. To help identify the likely locations of the PMF-identified sources, a conditional probability function was calculated. The results of this study will be used for the testing of relationship between $PM_{2.5}$ sources and observed health effects associated with exposure to $PM_{2.5}$.

2. Sample collection and chemical analysis

The $PM_{2.5}$ compositional data used in this study consisted of measurements made between January, 1995 and December, 1997 at a residential monitoring site (Rockwood), located 8 km north of the central business district. In Spokane, the prevailing winds are from the southwest and north. Wind speeds (upper 25%) are from the southwest where grass and cereal fields are mostly located. The location of the Rockwood site is presented in Fig. 1. The Rockwood monitoring site is located near (< 10 m) a four-lane, paved road that has a traffic count of approximately 10,000 vehicles per day (Kantamaneni et al., 1996). Daily $PM_{2.5}$ filter samples were collected using a Versatile Air Pollutant Sampler (VAPS, University Research Glassware). The VAPS allowed for simultaneous sampling of $PM_{2.5}$ on quartz and Teflon filters. The VAPS included an acid gas denuder and an ammonia denuder upstream of the Teflon filter. The quartz filter that was used to collect $PM_{2.5}$ for carbon measurements did not include an upstream denuder and thus generated a small, but
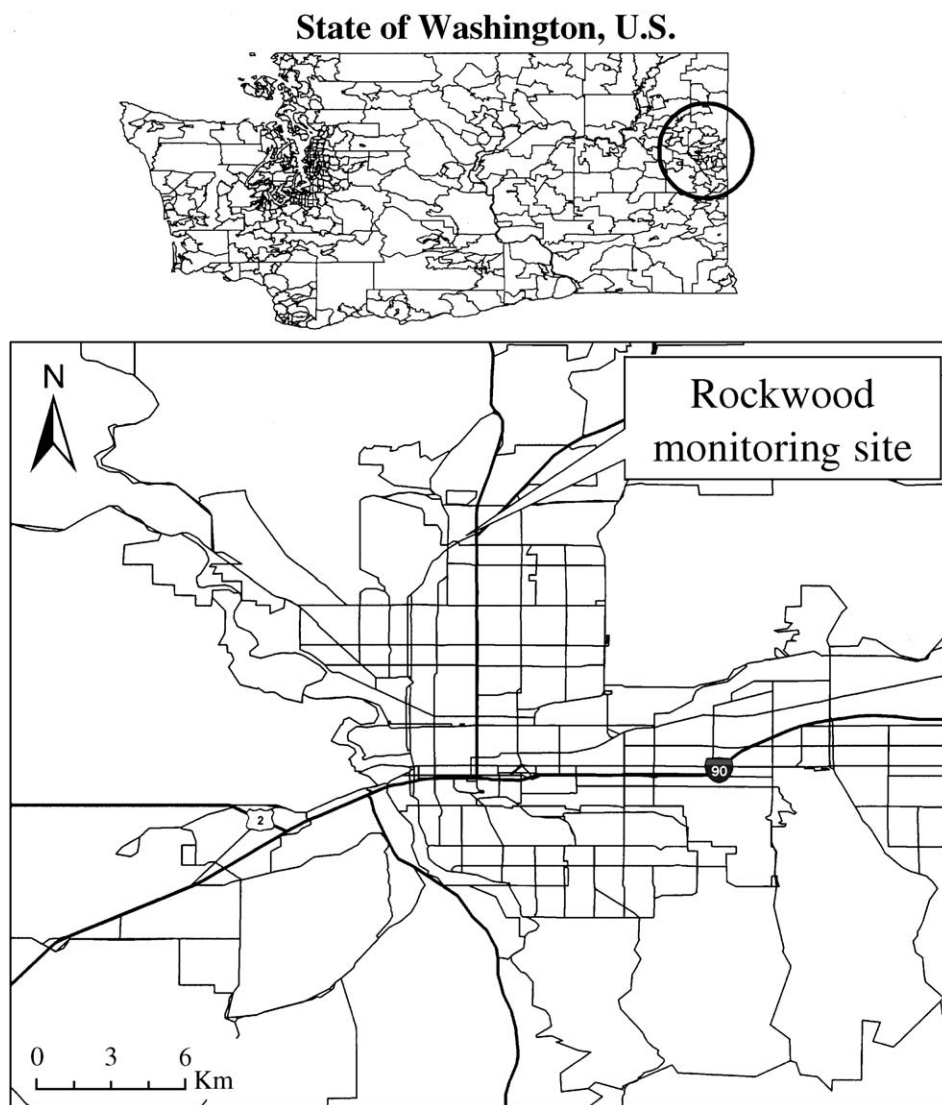
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Fig. 1. Location of the Rockwood monitoring site in Spokane, WA.

unknown, carbon artifact (McDonald et al., 2000). The details of this sampling system are presented elsewhere (Haller et al., 1999; Claiborn et al., 2000).

The Teflon filter samples were analyzed via energy dispersive X-ray fluorescence (XRF) (Dzubay et al., 1988) for chemical elements by the US EPA National Exposure Research Laboratory at RTP, NC. Each quartz filter was equilibrated at a relative humidity of 40–45%, cut in half, and a 0.33 cm^2 punch taken from one half. The punch was analyzed via the Thermal Manganese Oxidation (TMO) method for organic carbon (OC) and total carbon

Table 1
Summary of fine particle mass and 24 species concentrations used for PMF modeling

Species	Concentration (ng/m^3)				Number of BDL values (%)	Number of missing values (%)
	Arithmetic mean	Geometric mean[a]	Minimum	Maximum		
PM$_{2.5}$	12,069.9	10,198.7	2065.2	45,849.7	0 (0.0)	0 (0.0)
SO$_4^{2-}$	887.5	731.7	0.966	3960.5	18 (1.9)	129 (13.7)
NH$_4^+$	318.9	206.5	0.027	1888.6	333 (35.2)	14 (1.5)
NO$_3^+$	667.2	172.4	1.29	4952.4	250 (26.5)	77 (8.1)
Cl	17.1	7.94	0.1	248.0	524 (55.4)	12 (1.3)
C	3800.5	2855.5	22.7	19,527.4	298 (31.5)	0 (0.0)
As	0.994	0.58	0.100	4.90	923 (97.7)	10 (1.1)
Br	1.71	1.20	0.100	13.2	661 (69.9)	9 (1.0)
Cu	23.0	12.5	0.400	500.9	71 (7.5)	11 (1.2)
Mn	3.33	1.92	0.100	24.5	420 (44.4)	10 (1.1)
Pb	5.11	3.19	0.100	45.2	555 (58.7)	10 (1.1)
Zn	11.1	8.05	0.400	58.2	78 (8.3)	9 (1.0)
Al	143.7	81.4	1.30	1272.8	466 (49.3)	9 (1.0)
Si	315.9	177.6	1.70	3553.7	69 (7.3)	9 (1.0)
K	85.6	67.2	0.300	447.9	3 (0.3)	11 (1.2)
Ca	59.0	39.5	0.600	466.1	57 (6.0)	9 (1.0)
Fe	136.1	96.0	2.80	1113.5	1 (0.1)	9 (1.0)

[a] Data below the limit of detection were replaced by half of the reported detection limit values for the geometric mean calculations.

(TC); the remaining half was extracted and analyzed by colorimetry for ammonium (NH$_4^+$) and by ion chromatography for filterable sulfate (SO$_4^{2-}$) and nitrate (NO$_3^-$) (Chow, 1995).

In addition, the local air pollution control agency (Spokane County Air Pollution Control Authority, SCAPCA) provided hourly measurements of carbon monoxide (CO) as well as nitrogen oxides (NO$_X$). It is worth noting that the TMO analysis that was performed on all the Spokane filters for determining the carbon content does not correct for pyrolytically formed char, and therefore overestimates the ratio of EC to OC (Chow et al., 1993) when compared to other methods that make this correction. For this reason, only total carbon (C), the sum of OC and EC, was used in the data analysis. A total of 945 daily samples collected between January 1995 and December 1997 and 16 species were used for the PMF analysis. Summaries of PM$_{2.5}$ species and measurements used in this study are shown in Tables 1 and 2, respectively.

Table 2
Summary of measurements used for conditional probability function and correlation analysis

	Number of values	Mean	Minimum	Maximum
NO$_X$ (ppb)	114	35.8	3.50	166.2
SO$_2$ (ppb)	868	2.57	0.03	26.9
CO (ppm)	502	0.48	0.004	3.71
Wind speed (mile/h)	22,680	44.4	0.10	61.0
Wind direction (°)	22,680	NA[a]	NA	NA

[a] Not available.

3. Data analysis

The general receptor modeling problem can be stated in terms of the contribution from p independent sources to all chemical species in a given sample (Miller et al., 1972; Hopke, 1985, 1991) as follows

$$X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \qquad (1)$$

where x_{ij} is the jth species concentration measured in the ith sample, g_{ik} is the particulate mass concentration from the kth source contributing to the ith sample, f_{kj} is the jth species mass fraction from the kth source, e_{ij} is residual associated with the jth species concentration measured in the ith sample, and p is the total number of independent sources. The corresponding matrix equation is

$$\mathbf{X} = \mathbf{GF} + \mathbf{E} \qquad (2)$$

where $\mathbf{X}$ is a $n \times m$ data matrix with n measurements and m number of elements. $\mathbf{E}$ is a $n \times m$ matrix of residuals. $\mathbf{G}$ is a $n \times p$ source contribution matrix with p sources, and $\mathbf{F}$ is a $p \times m$ source profile matrix. As noted by Henry (1987), there are a potentially infinite number of possible solutions to this bilinear factor analysis problem (rotations of $\mathbf{G}$ matrix and $\mathbf{F}$ matrix). To decrease rotational freedom, PMF uses non-negativity constraints on the factors. PMF provides a solution that minimizes an object function, $Q(E)$, based upon uncertainties for each observation (Paatero, 1997, 2000). This function is defined as

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{m} \left[\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right]^2 \qquad (3)$$

where u_{ij} is an uncertainty estimate in the jth element measured in the ith sample. The receptor modeling problem is then to minimize $Q(E)$ with respect to $\mathbf{G}$ and $\mathbf{F}$ with the constraint that each of the elements of $\mathbf{G}$ and $\mathbf{F}$ is to be non-negative. This problem is solved iteratively as a weighted linear least squares problem (Paatero and Tapper, 1993). One of the matrices, $\mathbf{G}$ or $\mathbf{F}$, is taken as known and the chi-squared is minimized with respect to the other matrix. Then the role of $\mathbf{G}$ and $\mathbf{F}$ are reversed so that the matrix that has just been calculated is fixed and the other is calculated by minimizing $Q(E)$. This process then continues until convergence. The iteration speed was improved by Paatero (1997) adopting a global optimization scheme in which the elements of $\mathbf{G}$ and $\mathbf{F}$ vary simultaneously in each iterative step.

The parameter, FPEAK, and the matrix, FKEY, are used to control the rotations (Lee et al., 1999; Paatero, 2000; Paatero et al., 2002). By setting a non-zero value of FPEAK, the routine is forced to add one $\mathbf{G}$ vector to another and subtract the corresponding $\mathbf{F}$ factors from each other and thereby yield more physically realistic solutions. The external information can be imposed on the solution to control the rotations. If specific species in the source profiles are known to be zero, then it is possible to pull down those values toward

lower concentration through appropriate settings of FKEY. The details of setting up the FPEAK and FKEY in the input file are presented elsewhere (Paatero, 2000).

The application of PMF depends on the estimated uncertainties for each of the data values. The uncertainty estimation provides a useful tool to decrease the weight of missing and below detection limit data in the solution as well as to account for the variability in the source profiles. The procedure of Polissar et al. (1998) was used to assign measured data and the associated uncertainties as the input data to PMF. The concentration values were used for the measured data, and the sum of the analytical uncertainty and 1/3 of the detection limit value was used as the overall uncertainty assigned to each measured value. Values below the detection limit were replaced by half of the detection limit values and their overall uncertainties were set at 5/6 of the detection limit values. Missing values were replaced by the geometric mean of the measured values and their accompanying uncertainties were set at four time this geometric mean value. Uncertainty estimates for the measured data were usually less than 50% of corresponding measured values and relative uncertainty estimates from 100% to 250% were used for below detection limits values. Relative error estimates for missing values were equal to 400% of replaced geometric mean values. Therefore, half of the detection limit values that were used for the below detection limit values and geometric mean values that were used for missing values had lower weights in comparison to actual measured values. In addition, the estimated uncertainties of species that have scaled residuals larger than ± 2 need to be increased to reduce their weights and consequently reduce their residuals (Polissar et al., 1998; Paatero, 2000). This individual data point weighting permits the influence of the values to be related to the level of confidence the analyst has in the data.

Another important aspect of weighting of individual data points is the handling of extreme values. Environmental data typically shows a positively skewed distribution and often with a heavy tail due to extreme values as well as outliers. Such high values would distort the solution. To reduce their influence on the solution, PMF offers a robust mode that is a technique of iterative re-weighing of the individual data values (Paatero, 2000). In PMF routine, a data point was classified as an extreme value if the residual of the model fit exceeds four times the error estimate. The error estimate values of those extreme values were then increased so that the influences of them were reduced.

The results of PMF were then normalized by a scaling constant, s_k so that the quantitative source contributions as well as profiles for each source were obtained. Specifically,

$$x_{ij} = \sum_{k=1}^{p} (s_k g_{ik}) \left(\frac{f_{kj}}{s_k} \right) \qquad (4)$$

where s_k is determined by regressing total PM$_{2.5}$ mass concentrations in the ith sample, m_i, against estimated source contribution values.

$$m_i = \sum_{k=1}^{p} s_k g_{ik} \qquad (5)$$

This regression provides useful indicators of the quality of the solution. If the regression produces a negative s_k, it suggests that too many sources have been used. If a scaled

source profile exceeds unity, then it suggests that too few sources may have been chosen.

4. Conditional probability function

To identify likely locations of local point sources, a conditional probability function (CPF) (Ashbaugh et al., 1985) was calculated using source contribution estimates from PMF coupled with wind direction values measured at the site. To minimize the effect of atmospheric dilution, daily fractional mass contributions from each source relative to the total of all sources were used rather than using the absolute source contributions. The same daily fractional contribution was assigned to each hour of a given day to match to the hourly wind data. Specifically, the CPF is defined as

$$\text{CPF} = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \qquad (6)$$

where $m_{\Delta\theta}$ is the number of occurrences from wind sector $\Delta\theta$ that are upper 25 percentile of the fractional contributions, $n_{\Delta\theta}$ is the total number of observations from the same wind sector. In this study, $\Delta\theta$ was set to be $11.3°$. Calm winds (< 1 m/s) were excluded from this analysis. The sources are likely to be located in the directions that have high conditional probability values.
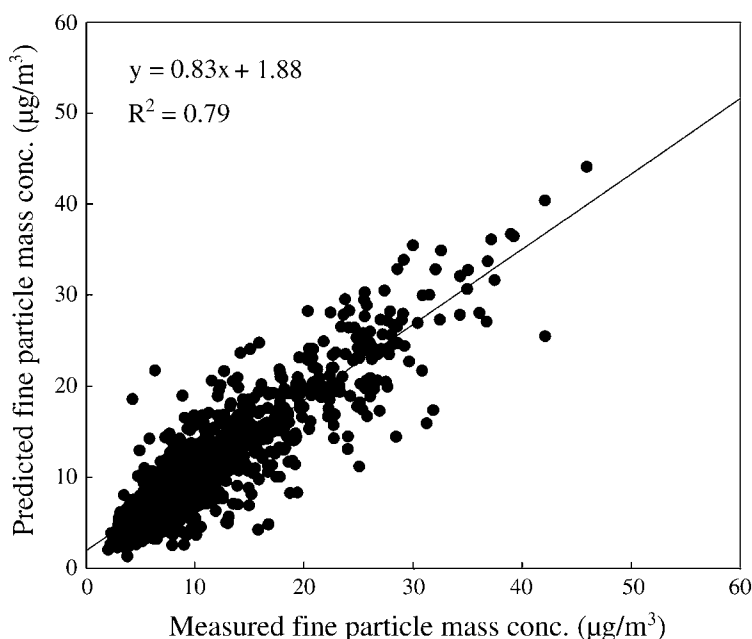

Fig. 2. Measured versus predicted PM$_{2.5}$ concentrations.

5. Results

To determine the number of sources, it is reasonable practice to test different numbers of sources and use the one with both adequate fit to the data and the most physically meaningful results. Also, since rotational ambiguity exists in factor analysis modeling (Paatero et al., 2002), PMF was run several times with different FPEAK values to determine the range within which the objective function $Q(E)$ value in Eq. (3) remains relatively constant. The optimal solution should lie in this FPEAK range. In this way, subjective bias was reduced an extent. The final PMF solutions were determined
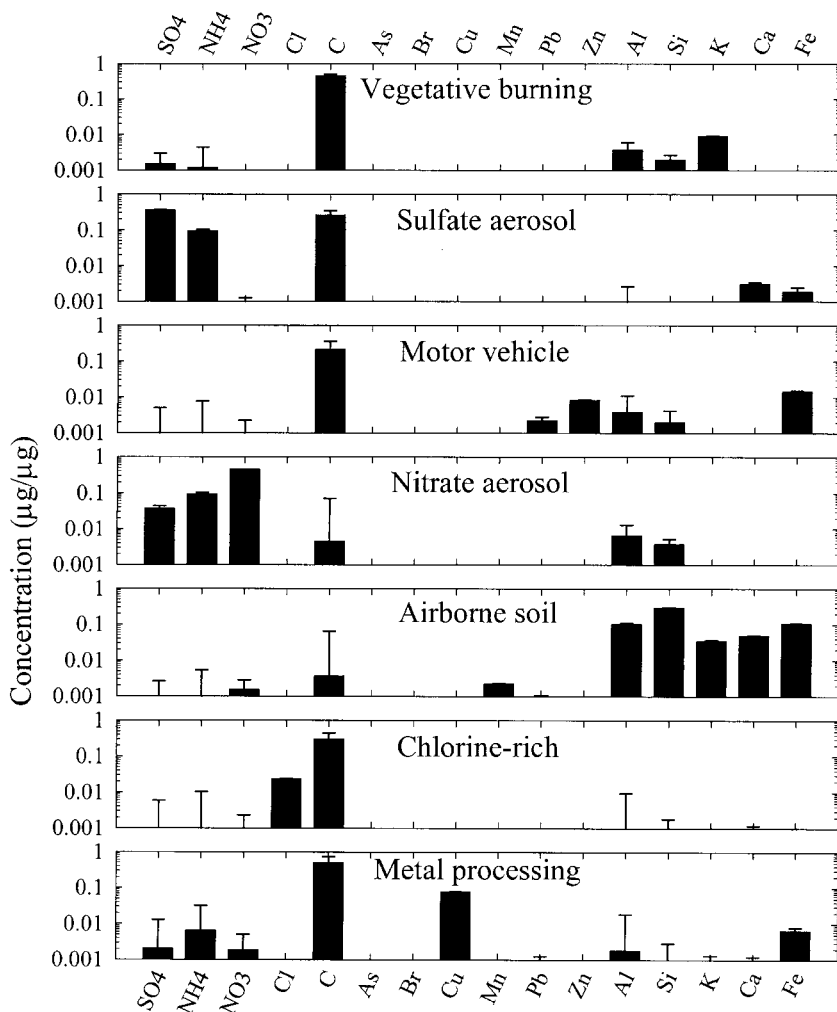

Fig. 3. Source profiles resolved from PM$_{2.5}$ samples (prediction ± standard deviation).

by trial and error with different number of sources, different FPEAK values, and different FKEY matrices with the final choice based on the evaluation of the resulting source profiles.

In this study, the eight source model produced a negative s_k (Eq. (5)) that indicated too many sources had been used. The motor vehicle source and chlorine-rich source were combined and shown as one source in the six-source model. In the seven source model, a value of FPEAK = 0.1, and a FKEY matrix (a value of 3 for NH_4^+ in both motor vehicle and chlorine-rich source profiles) provided the most physically reasonable
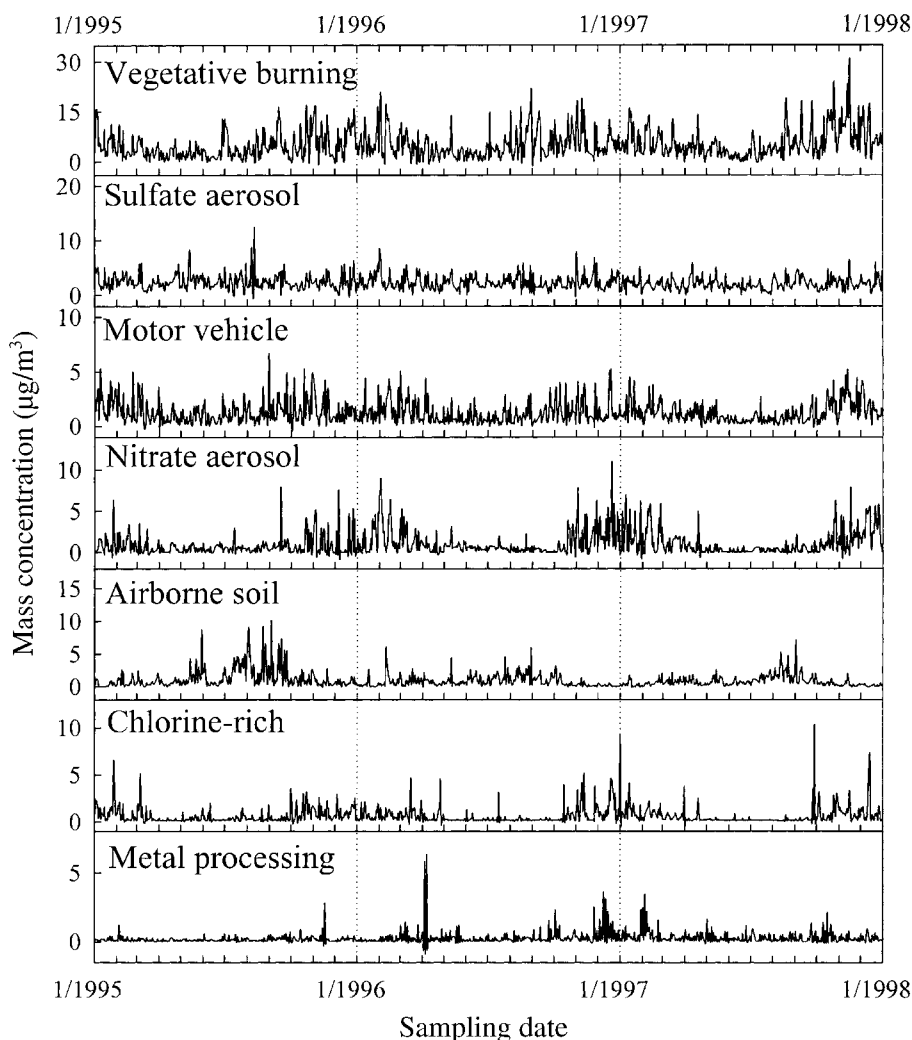

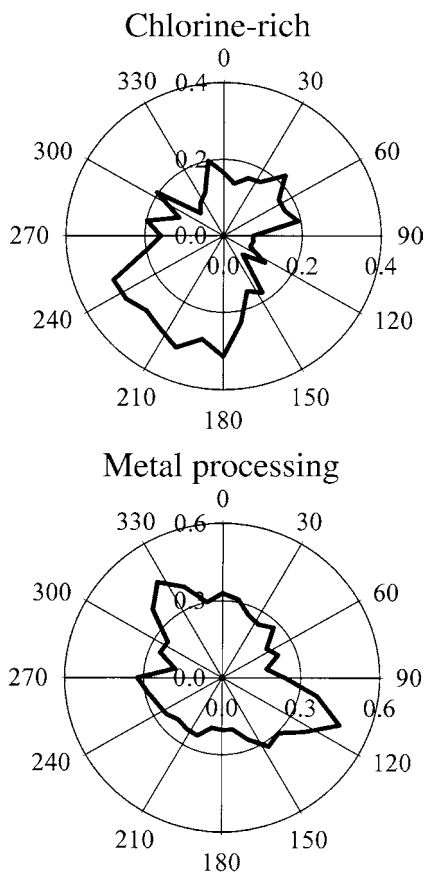Fig. 4. Time series plot of source contributions.

Fig. 5. Hourly CPF plots for the highest 25% of the mass contribution from point sources.

source profiles. Large uncertainties were assigned to several elements so that their scaled residuals were smaller than ± 2: NH_4^+ (three times its accompanying uncertainty), total carbon (two times), Br (two times), Pb (four times), and Al (three times).

Table 3
Pairwise correlation coefficients between source contributions and independent gas measurements

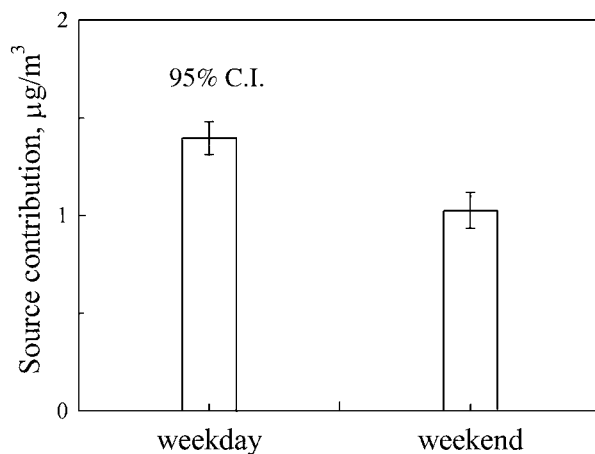| Source	CO	SO_2	NO_X
Vegetative burning	0.66	0.38	0.54
Sulfate aerosol	0.09	0.28	0.15
Motor vehicle	0.71	0.38	0.75
Nitrate aerosol	0.27	0.18	0.25
Airborne soil	− 0.06	0.08	0.25
Chlorine-rich	0.37	0.11	0.23
Metal processing	0.20	0.07	0.33

Fig. 6. The comparisons of PMF resolved motor vehicle contributions between weekday and weekend.

A comparison of the daily reconstructed $PM_{2.5}$ mass contributions from all sources with measured $PM_{2.5}$ mass concentrations is shown in Fig. 2. When the uncertainties associated with this data set are considered, the squared correlation coefficient of 0.79 indicates that the resolved sources effectively account for most of the variation in the $PM_{2.5}$ mass concentration. Fig. 3 presents the identified source profiles and Fig. 4 shows time series plots of estimated daily contributions to $PM_{2.5}$ mass concentrations from each source. Fig. 5 shows conditional probabilities of source locations for local point sources. The pairwise correlations over time of the source contributions against independently measured gaseous concentrations are presented in Table 3.

In Fig. 6, the motor vehicle contributions are compared between weekdays and weekends. The average source contributions of each source to the $PM_{2.5}$ mass concentration are summarized in Table 4. The seasonal average contributions are also presented in Fig. 7 (summer: April–September; winter: October–March). Vegetative burning has the highest average source contribution to the $PM_{2.5}$ mass concentration (44%). The second highest contributor is sulfate aerosol accounting for 19% of the $PM_{2.5}$ mass concentration. The average contributions of motor vehicle, nitrate aerosol, and airborne soil are 11%, 9%, and

Table 4
Average source contributions to fine particle mass concentration

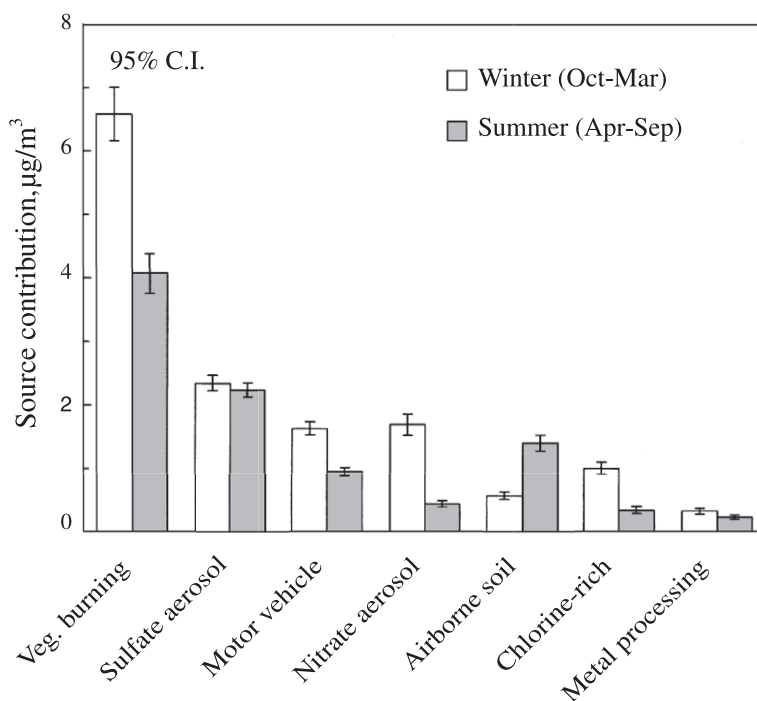|	Average source contribution (standard error)	
	Mass contribution ($\mu g/m^3$)	% Contribution
Vegetative burning	5.28 (0.14)	44.3 (1.2)
Sulfate aerosol	2.30 (0.04)	19.3 (0.4)
Motor vehicle	1.29 (0.03)	10.8 (0.3)
Nitrate aerosol	1.05 (0.05)	8.9 (0.4)
Airborne soil	1.01 (0.04)	8.5 (0.3)
Chlorine-rich	0.68 (0.03)	5.7 (0.3)
Metal processing	0.29 (0.01)	2.5 (0.1)

Fig. 7. The seasonal comparison of source contributions to PM$_{2.5}$ mass concentrations (mean ± 95% confidence interval).

9%, respectively. The chlorine-rich source and metal processing contributions are 6% and 3%, respectively.

6. Discussion

The vegetative burning source is characterized by C and K, and its estimated contributions are correlated with both CO and NO$_X$. This source has a seasonal trend of winter high, a minimum in summer, and short-term peaks in late summer and early fall. The winter peaks attributed to vegetative burning are consistent with emissions from residential fireplaces and wood stoves. Wood burning is a ubiquitous ground level source in this area whose emissions are frequently trapped by nighttime surface inversions. The summer peaks from this source are consistent with the timing of forest fire and grass burning. Cereal field stubble is burned in both eastern Washington and Idaho. Most of this activity takes place in the fall in areas well to the south and east of Spokane.

The sulfate aerosol has a high concentration of SO$_4^{2-}$. It also includes some carbon that typically becomes associated with the secondary sulfate aerosol. This carbon association is consistent with previous Phoenix (Ramadan et al., 2000) and northeastern U.S. (Song et al., 2001) aerosol studies. These previous studies show that sulfate aerosol has a strong

seasonal trend of summer high when the photochemical activity is highest. In contrast, a winter sulfate aerosol peak has been reported in the Salt Lake City aerosol study (Mangelson et al., 1997). In Spokane, the sulfate aerosol does not show strong seasonal variation as shown in Figs. 4 and 7.

The motor vehicle source is represented by high C and there is a winter-high seasonal pattern in this source. The estimated mass contributions from this source show associations with both CO and NO_X. The clear weekday-high pattern of this source shown in Fig. 6 is consistent with the commuting patterns. This source is a mixture of diesel emissions and gasoline vehicle particles. In this study, it was not possible to separate diesel emissions from gasoline vehicle sources with only total carbon data.

The nitrate aerosol has high concentration of NO_3^-. It has strong seasonal variation with maxima in the wintertime. These peaks in winter indicate that low temperature and high relative humidity help the formation of nitrate aerosols in Spokane. This is consistent with a previous study of northeastern U.S. sites (Song et al., 2001).

The airborne soil source profile shows the characteristic crustal elements: Si, Al, Fe, Ca and K. As expected, the highest contributions are in the dry, summer season. The increased levels of airborne soil particles in the summer shown in Fig. 4 are consistent with the lower relative humidity during this season. Unpaved roads, construction sites, and wind-blown soil dust could also produce particles of these crustal elements. Previous studies (Kantamaneni et al., 1996; Haller et al., 1999; Claiborn et al., 2000) have shown similar elevated dust impacts during this time for $PM_{2.5}$ as well as PM_{10}.

The sixth source has relatively high mass fractions of C and Cl. The leading candidates for this chlorine dominated source in Spokane are medical centers located 6 km southwest and 10 km south of the monitoring site because these centers generally incinerate contaminated medical refuse. As shown in the CPF plot in Fig. 5, high values come from the direction of these sources. However, this source could be a mixture of unknown chlorine-rich sources and carbon-rich sources if those are co-located and daily emission patterns are similar. The time series of daily contributions from this source are consistent with impacts at a fixed location from an elevated point source, specifically the appearance of large peaks interspersed with very low values. The examination of the relationship between Cl measured by XRF and Na measured by instrumental neutron activation analysis showed no correlation between these two species in either the fine or coarse particles in Spokane (Hoffman et al., 2001). Therefore, this source cannot be due to a marine source.

The seventh source is characterized by high mass fraction of Cu and high mass fraction of C. The origin of this source is likely to be two metal processors located 4 km northeast and 13 km southeast of the Rockwood monitoring site (US EPA, 2000). There are indications of contributions from the direction of the metal processing plants in Fig. 5. In addition, Cu particles could be generated from high-volume air sampling pumps (Lee et al., 1999) such as those located on the roof of Rockwood monitoring site.

7. Conclusion

Daily integrated $PM_{2.5}$ compositional data measured at a monitoring site in Spokane, WA were analyzed through a bilinear source apportionment method, PMF. The PMF

effectively resolved seven sources of PM$_{2.5}$ in Spokane: vegetative burning, sulfate aerosol, motor vehicle, nitrate aerosol, airborne soil, chlorine-rich source, and metal processing. Vegetative burning is the largest PM$_{2.5}$ source in Spokane, accounting for 44% of the mass during the study period. The airborne soil impacts are higher during periods of low relative humidity, and elevated dust impacts occur in the summer. Motor vehicles, chlorine-rich aerosol, and metal processing contributed more to the PM$_{2.5}$ mass concentrations in the winter. This seasonal trend is likely due to the more frequent temperature inversions in winter that trap and concentrate pollutants in the surface-mixing layer. The impacts from the point sources are more clearly seen by using PMF results combined with the CPF plots. Those plots show the direction of chlorine-rich sources. This approach also located potential sources of copper particles from the metal processing facilities.

Acknowledgements

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