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Source apportionment of PM2.5 in Beijing by positive matrix factorization

Yu Song[a], Yuanhang Zhang[a,*], Shaodong Xie[a], Limin Zeng[b], Mei Zheng[c], Lynn G. Salmon[d], Min Shao[b], Sjaak Slanina[b]

[a]*Department of Environmental Sciences, Peking University, Beijing 100871, China*
[b]*State Key Lab of Air Pollution Control and Simulation, Peking University, Beijing 100871, China*
[c]*School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA 30332-0340, USA*
[d]*Environmental Engineering Science Department, California Institute of Technology, Pasadena, CA 91125, USA*

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Abstract

Air pollution associated with atmospheric fine particulate matter (PM2.5, i.e., particles with an aerodynamic diameter of 2.5 μm or less) is a serious problem in Beijing, China. To provide a better understanding of the sources contributing to PM2.5, 24-h samples were collected at 6-day intervals in January, April, July, and October in 2000 at five locations in the Beijing metropolitan area. Both backward trajectory and elemental analyses identified two dust storm events; the distinctly low value of Ca:Si (<0.2) and high Al:Ca (>1.7) in Beijing PM2.5 appear indicative of contributions from dust storms. Positive matrix factorization (PMF) was used to apportion sources of PM2.5, and eight sources were identified: biomass burning (11%), secondary sulfates (17%), secondary nitrates (14%), coal combustion (19%), industry (6%), motor vehicles (6%), road dust (9%), and yellow dust. The lower organic carbon (OC), elemental carbon (EC), SO_4^{2-}, and Ca values of yellow dust enable it to be distinguished from road dust. The PMF method resolved 82% of PM2.5 mass concentrations and showed excellent agreement with a previous calculation using organic tracers in a chemical mass balance (CMB) model. The present study is the first reported comparison between a PMF source apportionment model and a molecular marker-based CMB in Beijing.

Keywords: China; Fine particle; Dust storm; Source profiles; Backward trajectories

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1. Introduction

Beijing, the capital of China, suffers from air pollution that has risen dramatically since the onset of rapid urbanization in the 1980s. The concentration of PM10 (particles with aerodynamic diameters of less than 10 μm) monitored by the Beijing Environmental Protection Bureau (BJEPB) during 2000–2004 showed particulate matter to be the major problem in Beijing (BJEPB, annual reports for 2000–2004). In these years, the annual PM10 concentrations ranged between 140 and 165 μg m^{-3}, while the PM2.5 exhibited mass concentrations of

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*Corresponding author. Tel.: +86 10 62756592; fax: +86 10 62751927.
E-mail addresses: songyu@pku.edu.cn (Y. Song), yhzhang@pku.edu.cn (Y. Zhang).

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$>100\,\mu g\,m^{-3}$, or about 60% of the PM10 concentrations in Beijing (He et al., 2001). PM2.5 is considered to be the more harmful to human respiration, as well as being responsible for degrading visibility (Bergin et al., 2001), and any efforts to improve the air quality of Beijing will require a clear understanding of the potential PM2.5 sources.

Several studies have identified possible sources of particulate matter in Beijing. He et al. (2001) used the chemical composition of PM2.5 collected from sites at Tsinghua University and Chegongzhuang from July 1999 to September 2000, and suggested sources to be dust storms, motor vehicle emissions, and biomass burning. Duan et al. (2004) demonstrated that summer burning of biomass could contribute 46–70% of the organic carbon (OC) in aerosols at the Ming Tombs, a rural site, and 10–43% of the OC at the Temple of Heaven, an urban site. Dan et al. (2004) regarded biomass burning and traffic and/or industry emissions as providing the major sources of OC and elemental carbon (EC) during summer, with coal consumption being the dominant contributor in winter. More recently, Zheng et al. (2005) used the chemical mass balance receptor model (CMB) to apportion OC in fine particles and the total PM2.5 mass as well. They concluded that the major sources of PM2.5 in Beijing were dust, secondary sulfate, secondary nitrate, coal combustion, mobile sources, secondary ammonium, biomass aerosol, cigarette smoke, and vegetative detritus. However, some of the source profiles used for their CMB model were the ones derived in the United States and may not be directly applicable in Beijing.

Unfortunately, measurements of source profiles have a difficult methodology and are time-consuming. A different but highly effective tool exists in using factor analysis to apportion sources without the chemical profiles, and in China, this option offers a viable alternative given the absence of a local source profile library.

The novel analysis provided by positive matrix factorization (PMF) is a powerful technique for particle apportionment. Cities where it has been applied successfully include Hong Kong (China; Lee et al., 1999), Toronto (Canada; Lee et al., 2003), Atlanta (USA; Kim et al., 2004), and Pittsburg (USA; Zhou et al., 2004). An excellent review of PMF modeling is presented in Hopke (2003).

In this study, we used the PMF method to determine PM2.5 sources in Beijing using the raw data from Zheng et al. (2005). Interpretation of the results was aided by comparing them to those obtained from the CMB model.

2. Methods

2.1. Data description

In January, April, July, and October 2000, PM2.5 samples were collected in Beijing for 24 h at five sites simultaneously at 6-day intervals. The five sampling sites are shown in the Supplemental Materials as Fig. S1, including the Ming Tombs (OT), the airport (NB), Beijing University (BJ), Dong Si EPB (XY), and Yong Le Dian (CH). Details of the sites are given in Zheng et al. (2005). In total, 100 samples were taken over 4 months. Three parallel filters were collected during each 24-h sampling period. For each sample, the 24-h PM2.5 mass concentrations were obtained and the chemical composition was then analyzed for sulfate, nitrate, chloride, and ammonium ions by ion chromatography (IC) and for metals by X-ray fluorescence (XRF) spectroscopy. The OC and EC were determined by NIOSH thermal-optical procedures. The detailed organic speciation obtaining at monthly was ascertained by GC/MS. Details of the particulate sampling procedures and the analytical methods are given in Zheng et al. (2005).

Most of the data presented in this paper are the same as those used by Zheng et al. (2005), which contained monthly data, particularly of organic tracers, averaged over 5 samples from each month. In addition, this paper considers the 24-h data, which include the OC, EC, and inorganic tracers, but the monthly organic speciation is not included.

2.2. Model description

The PMF model was developed by Paatero and Tapper (1994) and Paatero (2004). A conventional factor analysis model can be written as,

$$X = GF + E, \tag{1}$$

where X is the $n \times m$ matrix of ambient element concentrations, G is the $n \times p$ matrix of source contributions, F is the $p \times m$ matrix of source profiles, and E is the matrix of residuals not fitted by the model and is defined as

$$e_{ij} = x_{ij} - \sum_{k=1}^{p} f_{ik} g_{kj}, \tag{2}$$

where $i = 1, \ldots, n$ samples; $j = 1, \ldots, m$ elements; $k = 1, \ldots, p$ sources. To decrease the rotational freedom, PMF applies non-negativity constraints to the parameters. The objective function, $Q(E)$, based on the uncertainties inherent in each observation that are to be minimized, is defined as

$$Q(E) = \sum_{i=1}^{m} \sum_{j=1}^{n} \left[\frac{e_{ij}}{s_{ij}} \right]^2, \tag{3}$$

where s_{ij} is an uncertainty estimate in the jth element measured in the ith sample. The standard deviations for each data point, x_{ij}, are calculated at each iteration step according to

$$s_{ij} = c_1 + c_2 \max\left(|x_{ij}|, \sum_{k=1}^{p} |g_{ik} f_{kj}| \right), \tag{4}$$

where c_1 is the uncertainty for each data point, and c_2 is adjusted to 0.15 in this study, since $Q(E)$ is close to the number of individual elements of X.

Excluding species that were frequently present at concentrations below the detection limit (BDL), only 24 species were selected for PMF analysis: OC, EC, NO_3^-, SO_4^{2-}, NH_4^+, Na, Al, Si, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Pb, and Mg. Those few BDL data present in this data set were replaced by values corresponding to one-half the appropriate analytical detection limit (Lee et al., 2003).

2.3. Two dust storms in April 2000

Dust storms (DS in this paper) are defined as the dust transported to Beijing from great distances, such as from Inner Mongolia or Mongolia. They occur occasionally in northern China during spring, but are not frequent. During a dust storm, severe air pollution can occur over with thousands of kilometers. The ambient air contains of a mixture of external dust transported over long distances, and internal or local dust. It is difficult to distinguish between these two dust types because their chemical compositions are similar, but we were able to accomplish this by PMF analysis.

Samples from two DS were captured during the five sampling periods in April 2000. Fig. 1 shows the variations in Al, Si, Ca, and total mass concentrations at the five sites during the entire 20-day interval. The total mass concentration on 25 April was high at 216 $\mu g\,m^{-3}$. The crystal elements Al, Si, and Ca were much higher during 7 and 25 April than in the other sampling intervals. In contrast, on 19 April, PM2.5 had the highest measured anthropogenic pollution of 246 $\mu g\,m^{-3}$, but its crystal element concentrations were low. Both DS could be identified from the backward trajectories, i.e., the "successive" locations, back in time, of a particle arriving at a given height above ground at a given location and time, as well as by the hourly SO_2 and PM10 concentrations monitored on 7 and 25 April.

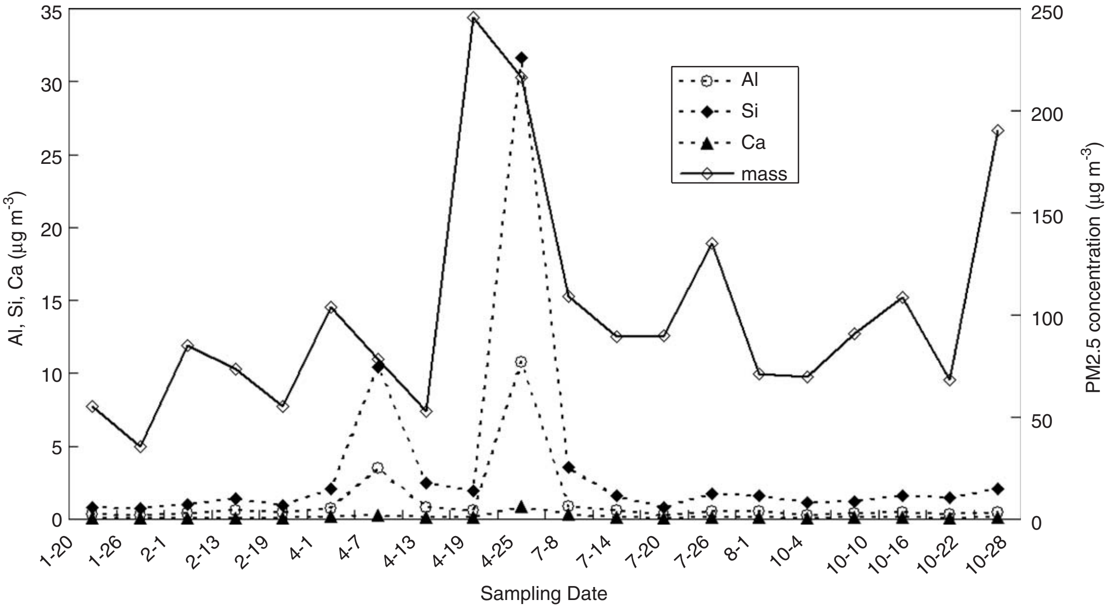

Fig. 1. Trends in Al, Si, Ca, and PM2.5 concentrations. Values were averaged over five sites.

The PM10 and SO_2 data were collected from the routine monitoring network maintained by BJEPB. The concentrations are averaged from measurements at five stations distributed within Beijing: the Ming Tombs, Qianmeng, Gucheng, Nongzhanguan, and the Olympic Center, determined using tapered element oscillating microbalance (TEOM) particulate monitors. Air flow was generated by a pump and controlled by a programmable timer that automatically turned on and off at a preset time. The flow rate was 16.7 L min^{-1} and regularly checked to assure it remained constant. SO_2, an indicator of gaseous pollution, was regarded as an indicator of local contributions, given the low level of SO_2 emissions in Inner Mongolia (TRACE-P data; http://atmos.cgrer.uiowa.edu/EMISSION_DATA/) from where the main mass of the dust probable originated (Xuan, 2005). PM10 concentrations rose on 6 April at 11:00 LST to drop back on 7 April at 04:00 LST (Fig. 2a) and again rose on 25 April at 07:00 LST and fell back on 26 April at 06:00 LST (Fig. 2b). Fig. 2a shows the PM10 concentrations as becoming saturated at 1500 μg m^{-3} because the "analog output" in the TEOM was selected for online data acquisition and the full scale set as 1500 μg m^{-3}; the true concentrations may well have been higher. During both intervals when high PM10 concentrations were observed, SO_2 concentrations were low. The dust was presumably transported from long distances by strong advection. The duration of the DS on 7 April was only 4 h and much shorter than the 18 h on 25 April. In other words, only the dust tail was captured on 7 April, which is why the total mass concentration of PM2.5 on 7 April was much less than on 25 April.

The NOAA hybrid single particle Lagrangian integrated trajectories (HYSPLIT) model (Draxler and Rolph, 2003) was used with FNL (Final GDAS run) meteorological data to calculate the 24-h back-trajectories, starting at altitudes of 100, 500, and 1500 m in Beijing (39.8°N, 116.47°E), and from 7 April at 02:00 h (6 April, 18:00 UTC) and 25 April at 11:00 h (25 April, 03:00 UTC). The trajectories of both events passed over northern China, specifically Mongolia and Inner Mongolia, where the main source (Gobi Desert) occurs. Almost the same trajectories were found at all three altitudes (Fig. 3), indicating that the dust was driven by strong synoptic winds (winds associated with large-scale events such as warm and cold fronts), as is typical for DS.

Fig. 4 shows PM2.5 ratios of Al:Si, Mg:Si, Mg:Ca, Mg:Al, Ca:Si, and Al:Ca determined over the observational period. The Ca:Si fell and Al:Ca rose dramatically on 7 and 25 April. The average value of the Ca:Si ratio was 0.52 on non-DS days and 0.20 on DS days, whereas Al:Ca was 0.67 on non-DS days and 1.70 on DS days. He et al. (2001) recorded a distinct reduction in Ca:Si in their weekly PM2.5 data during a DS on 21–26 April 2000. Unfortunately, no data are given in their data sets for the 7–12 April period. Either a significantly low value of Ca:Si or a significantly high value of Al:Ca in PM2.5 can be taken as a good indicator of a dust storm in Beijing.

For 7 and 25 April, Al:Si, Ca:Si, Mg:Si, Al:Ca, Mg:Al, and Mg:Ca were 0.34 and 0.34, 0.20 and 0.20, 0.06 and 0.06, 1.71 and 1.76, 0.16 and 0.17, and 0.28 and 0.29, respectively. The values of the ratios on both days agreed closely. As a consequence, only one chemical profile of the external dust was used for PMF analysis. The values can be derived by mixing or averaging external yellow dust with internal road dust; the Ca:Si ratio should be less than 0.2 and that of Al:Ca should be larger than 1.7 in PM2.5 derived from yellow dust alone.

The above values differed somewhat from the chemical composition of mineral dust particles found in Chinese loess areas, where Ca:Si is 0.37 and Al:Ca is 1.00 (Kruger et al., 2004, 2005). However, as Xuan (2005) noted, the Gobi Desert is the dust source, not the China loess. Furthermore, the Al:Ca values were close to ratios calculated from the elemental contents of surface soils in China (Zheng, 1994), whereas they are 1.6–1.7 in Mongolia and Inner Mongolia (Xuan, 2005). Cheng et al. (2005) recently found Al:Ca to be 2.05 in PM2.5 sampled in a tall tower in Hunshan Dake Sandland, a desert considered as one possible source of Beijing's dust.

3. Results and discussion

Rotational ambiguity is always a problem in factor analysis methodology. The parameters *FPEAK* and the matrices *GKEY* and *FKEY* in PMF can be used to control the rotation and assist in finding a more reasonable solution (Paatero, 2004). By setting a positive/negative value of *FPEAK*, the routine is forced to add/subtract *G*-factor vectors to/from each other and subtract/add the corresponding *F*-factors from/to each other, thereby yielding more physically realistic solutions.

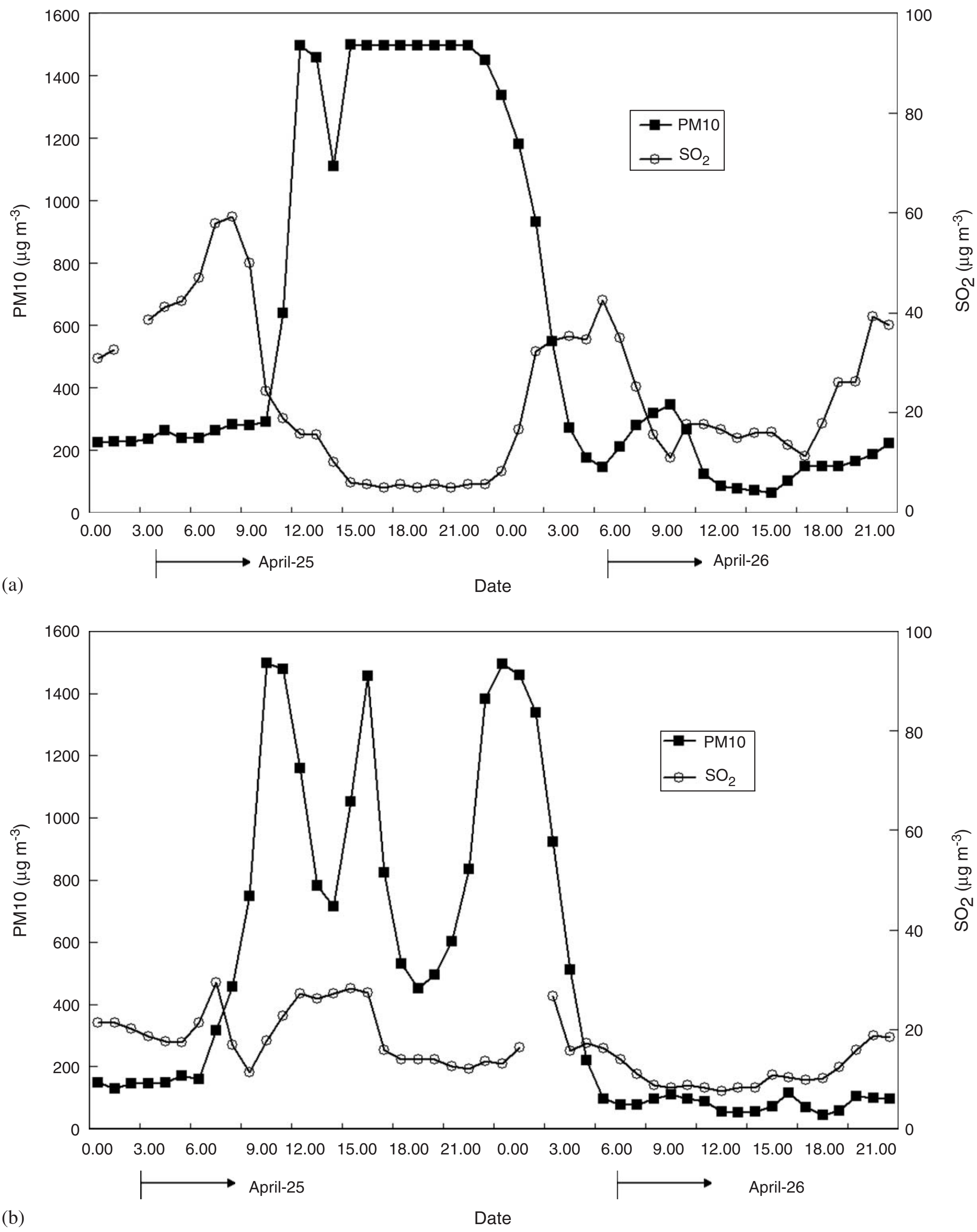

Fig. 2. The 1-h SO_2 and PM10 concentrations during the two dust events: (a) 6–7 April and (b) 25–26 April. The PM10 concentrations appear to be saturated at $1500\,\mu g\,m^{-3}$ but the actual number may have been much higher because the "analog output" used in TEOM online data acquisition was set to full scale at $1500\,\mu g\,m^{-3}$.

Sometimes an entry in matrices G (source contributions) or F (species concentrations) is in conflict with a priori information, so that the value is large when a priori information indicates that the value should be small or zero. Such an entry can be reduced or even forced to zero by setting the corresponding control variable in *GKEY* or *FKEY* to a positive value. Several entries can be suppressed in the same way, if needed.

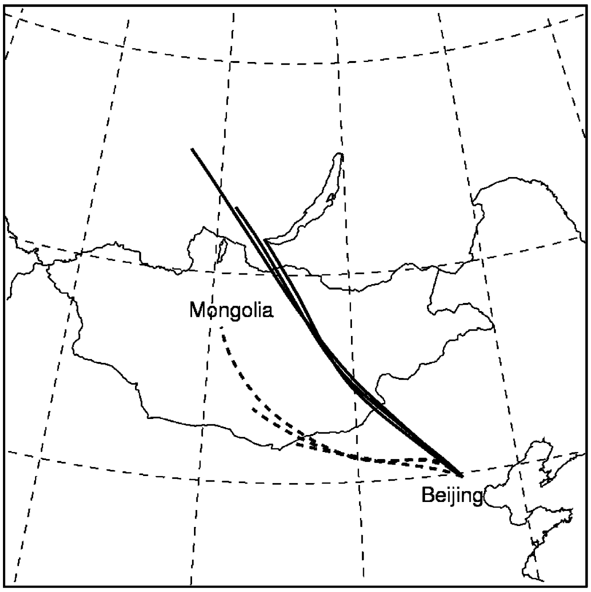

Fig. 3. The 24-h backward trajectories commencing on 7 April at 03:00 h (solid lines) and on 25 April at 11:00 h (dash lines) 100, 500, and 1500 m above ground level in at Beijing (39.8°N, 116.47°E).

As outlined above, only two DS occurred during the 20 sampling days, i.e., the contributions by DS should approach zero in the other 18 days when DS failed to occur. However, we found that the contributions were not zero on non-DS days after we ran the PMF analysis in the standard way, possibly due to the difficulty of distinguishing DS dust from local sources. We therefore used *GKEY* to reduce these values toward zero, and after several trials, set the *GKEY* values to 11 on non-DS days in order to achieve this. In the two DS days, the values for *GKEY* were maintained a value of zero. After numerous runs, no rotation was selected ($FPEAK = 0$) and eight possible particulate sources were identified. The eight probable particulate sources were determined as biomass burning, secondary sulfates, secondary nitrates, coal combustion, industry, motor vehicles, road dust, and yellow dust. However, it was found that SO_4^{2-} was unreasonably high in the chemical profiles of

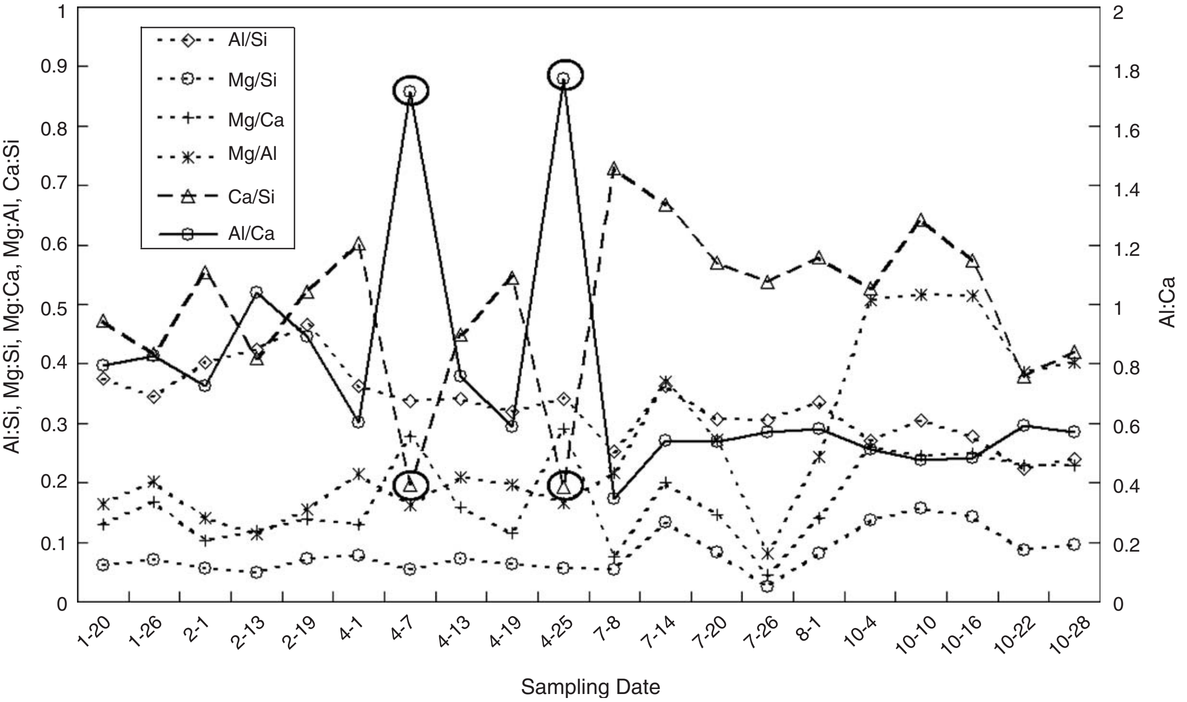

Fig. 4. Ratios of Al:Si, Mg:Si, Mg:Ca, Mg:Al, Ca:Si, and Al:Ca. The distinctively low Ca:Si and high Al:Ca on 7 April and 25 April are denoted by the larger circles.

biomass burning, secondary nitrate, and motor vehicle emissions. Consequently, *FKEY* values were set to 5, 3, and 5 to reduce these sulfate values mildly (Lee et al., 1999).

Finally, the solution provided by PMF analysis is reasonable. The source profiles (matrix *F*) are displayed in Fig. 5. The seasonal source contributions to PM2.5 mass concentrations at the five sites are shown in Fig. 6. The average percentages of each contributory source during the four months, compared to the results of Zheng et al. (2005), are listed in Table 1.

Biomass burning is characterized by high OC, EC, and K (Watson and Chow, 2001; Liu et al., 2003). Agrarian cultivation in Beijing suburbs is still extensive. The farmers burn the crop remnants each spring and fall to fertilize the soil. The incineration of fallen leaves throughout the city in the fall also contributes to biomass burning numbers (Duan et al., 2004), with the contribution from this source in October of 15.2 $\mu g\,m^{-3}$ being higher than in other seasons (Fig. 6). The annual calculated percentage of biomass burning is 11%, larger than the 6% of Zheng et al. (2005; Table 1).

The presence of secondary sulfates is identified by a high sulfate concentration. It is formed by photochemical reactions especially in the summer when solar radiation and the ambient temperature are high (Seinfeld and Pandis, 1998). A high relative humidity (RH) would accelerate the dry conversion processes, with Yao et al. (2003) finding the sulfate concentration to be higher in summer than spring, as a result of in-cloud processing in Beijing. Clearly the 31.5 $\mu g\,m^{-3}$ contribution of secondary sulfate in July (Fig. 6) was much higher than in other months, perhaps owing to the high conversion rates of SO_2 from local contributions.

Secondary nitrate is characterized by high nitrate and ammonium concentrations. The precursor gas of nitrate, NO_x, is emitted by traffic and stationary sources such as electricity generating plants. Ammonium nitrate is semi-volatile at high temperatures (Seinfeld and Pandis, 1998) and hence NH_4NO_3 would not be expected to be present in aerosol during the day because of the low RH in Beijing, although this was reported by Yao et al. (2003). The secondary nitrate contributions in January (9.1 $\mu g\,m^{-3}$), April (22.0 $\mu g\,m^{-3}$), and October (24.3 $\mu g\,m^{-3}$) were higher than in July (7.3 $\mu g\,m^{-3}$), probably because NH_4NO_3 was present.

The percentages of secondary sulfates and secondary nitrates were 17% and 14%, respectively. A contribution from ammonium is included in both sources. The sum of 31% in Table 1 is slightly higher than the 29% sum of secondary sulfates (17%), secondary nitrates (6%), and ammonium (6%) found by CMB (Zheng et al., 2005). This difference could have arisen from the CMB model only including secondary sulfates, secondary nitrates, and ammonium, whereas the secondary profiles derived by PMF contained other components, especially secondary OC.

Coal combustion is represented by high OC, EC, and Cl, in accordance with source profiles determined in the laboratory (Zheng et al., 2005). Coal is still a major energy source in Beijing. In 2003, nearly 2.4×10^7 t of coal provided 51.1% of the total energy consumption of 4.7×10^7 t. Coal burning is very common way of providing heat with the season lasting nearly half a year. As expected, the contribution was much higher in January (22.7 $\mu g\,m^{-3}$), April (20.4 $\mu g\,m^{-3}$), and October (19.9 $\mu g\,m^{-3}$), and lowest in July (0.7 $\mu g\,m^{-3}$). The contribution of coal combustion to PM2.5 was 19% (Table 1), considerably higher than the 7% reported by Zheng et al. (2005). It should be noted that the chemical profile provided by PMF analysis included secondary products of coal combustion, such as sulfates, nitrates, and OC. These were not included in the organic tracer CMB model of Datong coal (Zheng et al., 2005), where the contribution of coal combustion to PM2.5 was estimated from results of OC source apportionment. In addition, the CMB model did not include particulate matter from high-efficiency coal combustion that contained very little organics (Zheng et al., 2005), but the results of PMF analyses likely included these emissions.

Metal elements such as Cr, Fe, Ni, and Mn are present at high concentrations and attributed to industrial sources. Some metal manufacturing plants are located in Beijing, and the Capital Steel Corporation Limited, located in the Shijingshan industry area, produces six million tons of steel each year (http://www.bjepb.gov.cn/). A relative small contribution of 6% was found to have come from this source, which was not apportioned by Zheng et al. (2005).

Motor vehicle emissions are characterized by high Pb, Zn, and EC. Pb was used in gasoline in China as an antidetonator. The government has recently implemented the Europe-3 standard in China with unleaded petrol (ULP) starting to replace (http://www.bjepb.gov.cn/) the present lead-bearing fuel. Zn is emitted from lubricant oil, brake linings, and

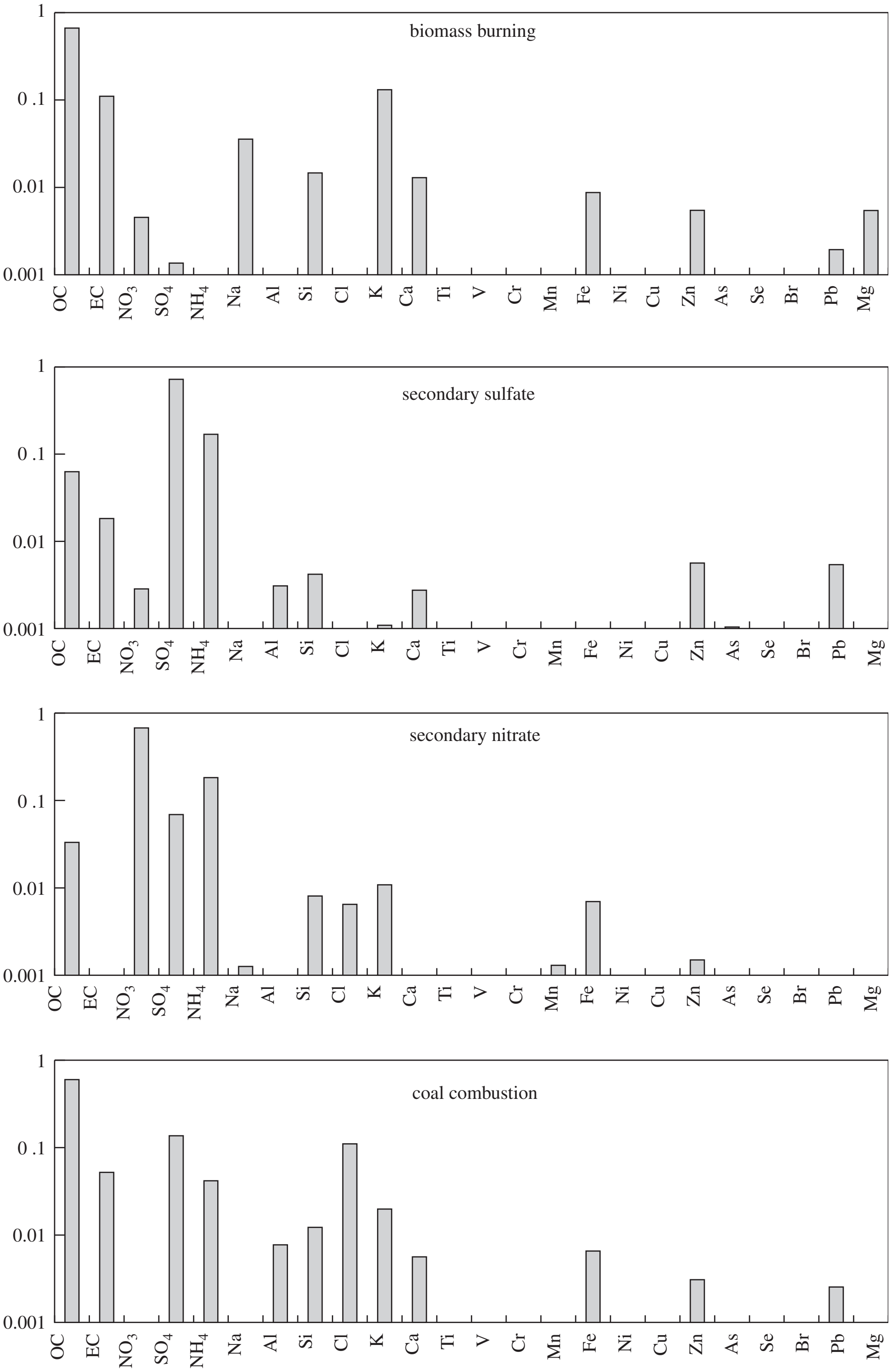

Fig. 5. Sources profiles resolved from PM2.5 samples for biomass burning, secondary sulfates, secondary nitrates, coal combustion, industry, motor vehicles, yellow dust, and road dust.

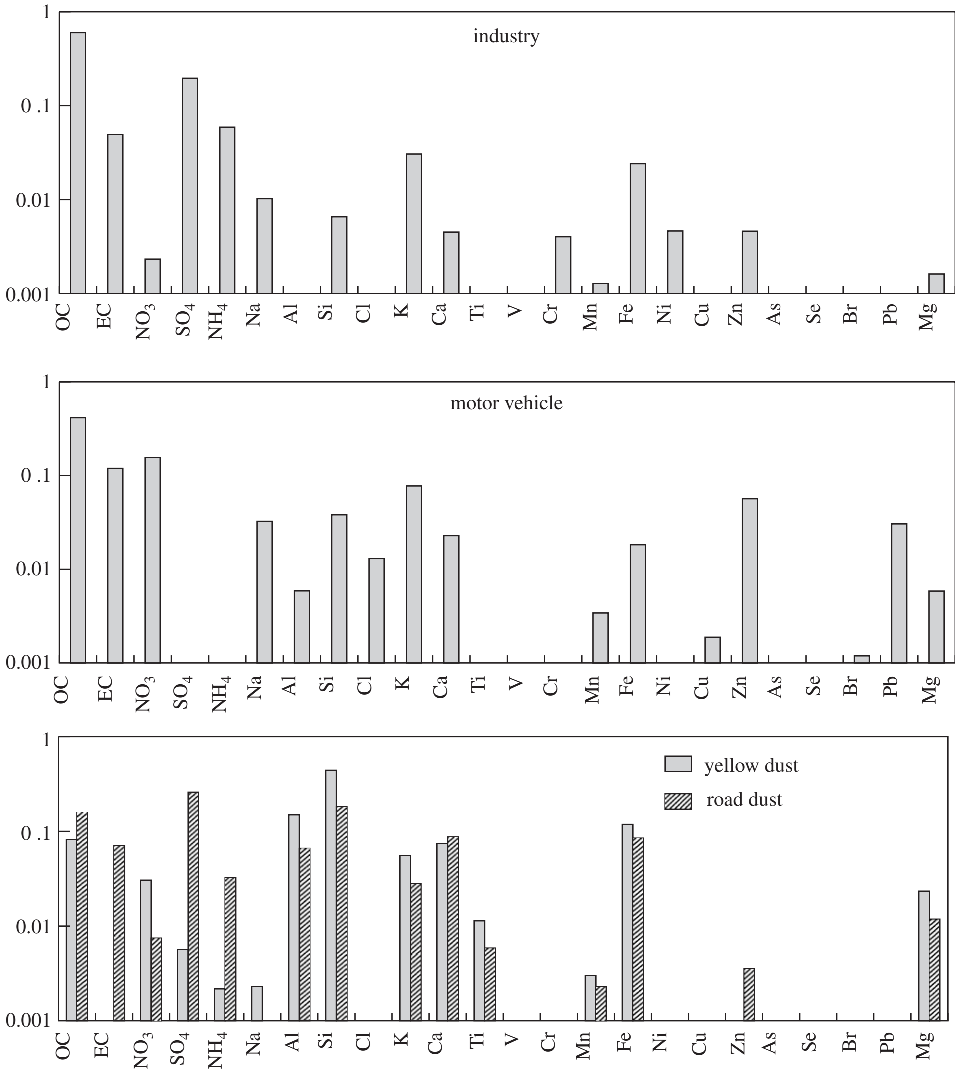

Fig. 5. (*Continued*)

tires (Zhou et al., 2004). Both Pb and Zn have also been used as indicators of gasoline combustion in Hong Kong particle apportionment (Lee et al., 1999). EC is often emitted from engines, and the mass contribution of motor vehicle exhaust was 6% (Fig. 6 and Table 1), very close to the 7% apportioned by Zheng et al. (2005).

Both yellow dust and road dust are high in the crystal elements Si, Al, Ca, Mg, Fe, and Ti (Watson and Chow, 2001; Kim et al., 2004; Zhao and Hopke, 2004). The values in yellow dust differ from those in road dust. OC, EC, SO_4^{2-}, and Ca in road dust is higher than in yellow dust (Fig. 6), presumably because road dust is more exposed to human activities, with the higher OC and EC values coming from rotted vegetation and rubbish, and combustion sources, the higher SO_4^{2-} values from coal combustion, and Ca largely from construction activities. Si, Al, and Mg are richer in yellow dust; Ca:Si was 0.17 and 0.48, and Al:Ca was 2.0 and 0.76 in yellow dust and road dust, respectively. Yellow dust contributed $38\,\mu g\,m^{-3}$ (48%) to the total mass of PM2.5 on 7 April and $118\,\mu g\,m^{-3}$ (55%) on 25 April. In contrast, road dust contributed only $5\,\mu g\,m^{-3}$ (7%) on 7 April and $18\,\mu g\,m^{-3}$ (8%) on 25 April. Yellow dust only appeared in April and was apportioned only for that month. Since DS seldom occur in Beijing, the annual percentages of source contributions were averaged to exclude yellow dust. Road dust was 9%, much less than

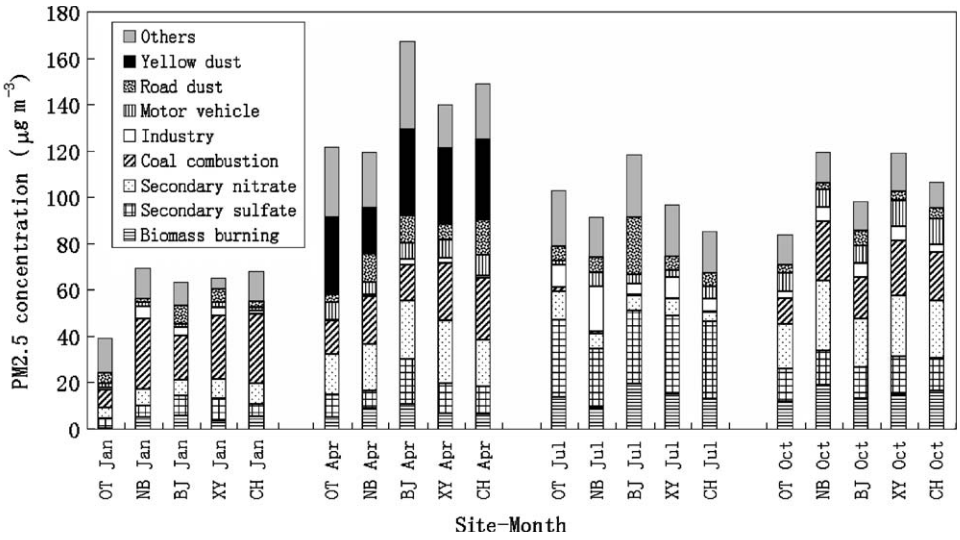

Fig. 6. Source contributions to mass concentrations in January, April, July and October.

Table 1
The comparison of average source contributions (%) to PM2.5 mass concentrations between PMF method and CMB approach

PMF		CMB	
Identified sources	Average source contribution (%)	Identified sources	Average source contribution (%)
Biomass burning	11	Biomass aerosols	6
Secondary sulfates	17	Secondary sulfates	17
Secondary nitrates	14	Secondary nitrates	10
		Secondary ammonium	6
Coal combustion	19	Coal combustion	7
Motor vehicles	6	Diesel and gasoline exhaust	7
Road dust	9	Dust	20
Industry	6		
		Cigarette smoke	1
		Vegetative detritus	1
		Other organic matter	13
Others	18	Others	12

the 20% of Zheng et al. (2005), in large part because yellow dust was not distinguished from road dust in their study; although they in fact used different dust profiles from those used for other seasons to estimate dust in April. If their April data is excluded, the contribution of road dust in the other three months is about 10%, which is closer to the PMF results (Dr. Zheng Mei, personal communication). It can also be noted that Zhao and Hopke (2004) used PMF analysis to separate local soils from Asian dusts. In summary, PMF analysis accounted for 82% of PM2.5, compared to 88% in Zheng et al.'s (2005) study.

4. Conclusions

The PMF model was used successfully to apportion PM2.5 from different sources in Beijing. The data used had a 24-h resolution. The interpretation of the PMF analytical results was improved by comparison to earlier CMB results, which used averaged weekly data. The contribution from coal consumption found in this study was 19%, much higher than the 7% in Zheng et al. (2005). The present result seems more reasonable given that coal combustion provides Beijing with over 50% of its energy. PMF analysis enabled yellow dust to be separated from road dust during the April sampling, which had not been resolved in the study by Zheng et al. (2005). The contribution from the road dust was 9%, much less than the 20% from CMB results.

The contributions from secondary sulfates and nitrates, and motor vehicles were comparable. However, contributions from other organic matter (13%), cigarette smoke (1%), and vegetative debris (1%) were recognized in the CMB results but not apportioned in the PMF analysis. The other organic matter was considered to have been derived from fossil fuel combustion (Zheng et al., 2005). The total percentage of mass concentrations determined by the CMB model was 6% higher than in our study. The most important reason for this can be attributed to the different principles on which the receptor models operate. CMB uses chemical source profiles in a non-negatively constrained regression model in order to obtain the contributions, whereas PMF obtains both the source profiles and the contributions from the observed concentrations using bilinear assumptions.

To control PM2.5 pollution in Beijing, some immediate measures must be undertaken by the government. The total contribution from secondary sulfates and secondary nitrates was the most substantial and accounted for more than 30% annually. These contributions could be addressed by changing the energy source in Beijing from coal to natural gas, enabling control not only of primary PM2.5 emission arising from ashes, but also atmospheric SO_2, which is transformed into sulfates. Improving the quality of gasoline could have profound effects on the concentrations of NO_x and OC, which are also transformed into secondary particulate matter. In addition, watering and road washing can be sued to prevent resuspended dust from entering the air.

We acknowledge that five sampling days are too few to encompass an entire season. An ongoing monitoring system is essential to obtain sufficient data, along with measurements of source chemical profiles in China, to provide base information on chemical tracers.

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Appendix A. Electronic Supplementary Material

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2005.10.039.

References

Bergin, M.H., Cass, G.R., Xu, J., Fang, C., Zeng, L.M., Yu, T., Salmon, L.G., Kiang, C.S., Tang, X.Y., Zhang, Y.H., Chameides, W.L., 2001. Aerosol radiative, physical, and chemical properties in Beijing during June 1999. Journal of Geophysical Research 106, 17,969–17,980.

Cheng, T., Lu, D., Wang, G., Xu, Y., 2005. Chemical characteristics of Asian dust aerosol from Hunshan Dake Sandland in Northern China. Atmospheric Environment 39, 2903–2911.

Dan, M., Zhuang, G., Li, X., Tao, H., Zhuang, Y., 2004. The characteristics of carbonaceous species and their sources in PM2.5 in Beijing. Atmospheric Environment 38, 3343–3352.

Draxler, R.R., Rolph, G.D., 2003. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website (http://www.arl.noaa.gov/ready/hysplit4.html). NOAA Air Resources Laboratory, Silver Spring, MD, USA.

Duan, F., Liu, X., Yu, T., Cachier, H., 2004. Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing. Atmospheric Environment 38, 1275–1282.

He, K., Yang, F., Ma, Y., Zhang, Q., Yao, X., Chan, C.K., Cadle, S., Chan, T., Mulawa, P., 2001. The characteristics of PM2.5 in Beijing, China. Atmospheric Environment 38, 4959–4970.

Hopke, P.K., 2003. Recent developments in receptor modeling. Journal of Chemometrics 17, 255–265.

Kim, E., Hopke, P.K., Edgerton, E.S., 2004. Improving source identification of Atlanta aerosol using temperature resolve carbon factions in positive matrix factorization. Atmospheric Environment 38, 3349–3362.

Kruger, B.J., Grassian, V.H., Cowin, J.P., Laskin, A., 2004. Heterogeneous chemistry of individual mineral dust particles from different dust source regions: the importance of particle mineralogy. Atmospheric Environment 38, 6253–6261.

Kruger, B.J., Grassian, V.H., Cowin, J.P., Laskin, A., 2005. Erratum to "Heterogeneous chemistry of individual mineral dust particles from different dust source regions: the importance of particle mineralogy". Atmospheric Environment 39, 395.

Lee, E., Chan., C.K., Paatero, P., 1999. Application of positive matrix factorization in source apportionment of particulate pollutants in Hong Kong. Atmospheric Environment 33, 3201–3212.

Lee, P.K., Brook, J.R., Dabek-Zlotorzynska, E., Mabury, S., 2003. Identification of the major sources contributing to PM2.5 observed in Toronto. Environmental Science and Technology 37, 4831–4840.

Liu, W., Hopke, P.K., Han, Y., Yi, S., Holsen, T.M., Cybart, S., Kozlowski, K., Milligan, M., 2003. Application of receptor modeling to atmospheric constituents at Potsdam and Stockton, NY. Atmospheric Environment 37, 4997–5007.

Paatero, P., 2004. User's guide for positive matrix factorization programs PMF2 and PMF3, Part 1: tutorial.

Paatero, P., Tapper, U., 1994. Positive matrix factorization: a non-negative factor model with optimal utilization of error estimates of data values. Environmetrics 5, 111–126.

Seinfeld, J.H., Pandis, S.N., 1998. Atmospheric Chemistry and Physics from Air Pollution to Climate Change, Wiley, New York, USA, pp. 649–655.

Watson, J.G., Chow, J.C., 2001. Source characterization of major emission sources in the Imperial and Mexicali valleys along the US/Mexico border. Sciences of the Total Environment 276, 33–47.

Xuan, J., 2005. Emission inventory of eight elements, Fe, Al, K, Mg, Mn, Na, Ca and Ti, in dust source region of East Asia. Atmospheric Environment 39, 813–821.

Yao, X., Lau, A.P.S., Fang, M., Chan, C.K., Hu, M., 2003. Size distributions and formation of ionic species in atmospheric particulate pollutants in Beijing, China: 1—inorganic ions. Atmospheric Environment 37, 2991–3000.

Zhao, W., Hopke, P.K., 2004. Source apportionment for ambient particles in the San Gorgonio wilderness. Atmospheric Environment 38, 5901–5910.

Zheng, C. (Ed.), 1994. Atlas of Soil Environmental Background Value in the People's Republic of China. China Environmental Science Press, Beijing, China.

Zheng, M., Salmon, L.G., Schauer, J.J., Zeng, L., Kiang, C.S., Zhang, Y., Cass, G.R., 2005. Seasonal trends in PM2.5 source contributions in Beijing, China. Atmospheric Environment 39, 3967–3976.

Zhou, L., Hopke, P.K., Paatero, P., Ondov, J.M., Pancras, J.P., Pekney, N.J., Davidson, C.I., 2004. Advanced factor analysis for multiple time resolution aerosol composition data. Atmospheric Environment 38, 4909–4920.