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Source apportionment for urban PM_{10} and $\text{PM}_{2.5}$ in the Beijing area

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Airborne particulate matter (PM_{2.5} and PM₁₀) samples were collected at the Beijing Normal University sampling site in the urban area of Beijing, China in dry and wet seasons during 2001 2004. Concentrations of 23 elements and 14 ions in particulate samples were determined by ICP-AES and IC, respectively. Source apportionment results derived from both Positive Matrix Factorization (PMF) and Chemical Mass Balance (CMB) models indicate that the major contributors of PM_{2.5} and PM₁₀ in Beijing are: soil dust, fossil fuel combustion, vehicle exhausts, secondary particulate, biomass burning and some industrial sources. We have identified both regional common sources, such as vehicular emissions, particulate of secondary origin and biomass burning, as well as country-specific problems, such as sand storms and soil dust that should be addressed for effective air quality control.

airborne particulate matter, source apportionment, receptor model

As Beijing will host the 2008 Olympic Games, a series of measures, such as fuel shift from coal to natural gas, moving high-polluting industries out of Beijing, phasing out leaded gasoline, upgrading gasoline quality to meet EURO3 emission standards as well as abatement technologies for sulfur dioxide and particulate removal from point and mobile sources, and greening of unpaved land, have been taken to improve the air quality in Beijing. The SO_2 and NO_x yearly average levels have been improved significantly since 1998. However, in spite of these efforts, the PM₁₀ levels in Beijing remained rather high^[1]. The reason is that Beijing is located in a semiarid area with scarce precipitation. There are consequently natural sources of air particulate, such as the bare land and dried riverbeds, which become sources of air particulate during sunny/windy days. These natural sources are more difficult to be dealt with in a semiarid area. A considerable amount of particulate is transferred from remote arid areas as well. On the other hand, aerosol is also receiving worldwide attention, as it would have potential impacts on the global climate change^[2].

Earlier studies on aerosol characterization in Beijing were focused on TSP (total suspended particulate) and $PM_{10}^{[3 \ 6]}$. Chen et al.^[7] initiated characterization of PM_{2.5} and used CMB (chemical mass balance) model to identify diesel exhaust, coal combustion and re-entrained dust as the major sources in Beijing. Yao et al.^[8,9] studied the composition and formation mechanism of water-soluble ions in PM2.5. Limited numbers of samples and chemical species measured in the above studies made clear characterization difficult. The current routine monitoring program of air pollutants in Beijing run by the Beijing Municipal Environmental Monitoring Center includes only measurements of PM₁₀ mass concentrations but not PM2.5 levels, as there is no air quality standard for PM_{2.5} in China. Recent publications were mainly focused on the identification of organic or min-

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eral compounds in PM2.5 samples^[10 19]. Using simple factor analysis methods, Sun et al.^[20] identified 4 6 sources in PM_{2.5} samples. Our project supported by SIDA and AIT focuses on this gap long-term simultaneous sampling and analyses of PM_{2.5} and PM₁₀, as well as source apportionment. This paper describes some of our findings from our monitoring data during the first phase (2001 2003) at an urban sampling site in Beijing.

1 Sampling and analytical methods

PM_{2.5} and PM₁₀ samples have been collected at a sampling site about 200 300 m from the Third Ring Road, a heavy traffic artillery in the city. Samplers were located on the roof (40 m high) of the Building of Science and Technology, Beijing Normal University. In order to avoid the street canyon effect, this height of the sampling site has been particularly selected to collect those fine particles formed from primary and secondary sources, including the exhausts from traffic vehicles and the sources from long-range transport. Two sets of aerosol samplers with three interchangeable sampling heads (PM_{2.5}, PM₁₀, and TSP) from the Beijing Geological Instrumental Factory (Model TSP, PM₁₀, PM_{2.5}, with the air flow rate fixed at 77 L/min) were used for sampling PM_{2.5}, and PM₁₀, separately. We have also used a dichotomous sampler donated by US EPA and AIT for comparison and QA/QC purposes.

An ICP-AES instrument (Model: ULTIMA, JOBIN-YVON Company, France) was used for the analyses of 23 elements (Al, Fe, Mn, Mg, Ti, Sc, Na, Eu, Ce, Sr, Ca, Co, Cr, Ni, Cu, Pb, Zn, Cd, V, S, As, Se and Sb) in PM_{2.5} and PM₁₀ samples collected on Whatman 41 filters. A Dionex Model 600 Ionic Chromatograph was employed for the analysis of important anions and cations. We have developed a new IC method with gradient elution for the satisfactory separation and simultaneous determination of organic anions (acetate, formate and oxalate), methanesulfonic acid (MSA, a marker of biogenic sulfur from marine and soil microorganisms) and inorganic anions (fluoride, chloride, nitrite, nitrate, sulfate and phosphate) within 14 min with a sample loop volume of 25 i $L^{[21]}$. The method has been applied successfully to the determination of anions in aerosol/rain samples in China. Besides, cations such as NH_4^+ , K^+ , Na^+ , Ca^{2+} , and Mg^{2+} have also been determined by ion chromatography.

Data quality assurance including inter-laboratory comparison of SRM measurements has been taken into account for the above-mentioned analytical methods and the sampler performance as well.

2 Input data processing for modeling

Both PMF and CMB8 were employed in the study of source apportionment. PMF (positive matrix factorization) is a new variant of factor analysis. The 2-way model in the PMF package was applied to our Beijing datasets by the courtesy of the author of PMF in the University of Helsinki^[22]. The PMF approach differs from earlier factor analysis and principal component analysis (PCA), because error estimates of the data matrix are utilized and the non-negativity of the resulting factors is intergraded in computation^[23 25]. The CMB8 model^[26] recommended by US EPA was also used to estimate the contribution of sources to the aerosol mass.

For PMF modeling, a simplistic rule was applied to determining the standard-deviation value: std-dev (S_{ii}) . It was adopted from our observed value X_{ii} . The sum of the analytical uncertainty (three times of the standard deviation of parallel runs on the same day) and the sampling uncertainty (8% of the observed value X_{ij}) was assigned to each measured value as the overall uncertainty. The missing observations in the data matrix (X_{ii}) were replaced by the mean value of the variable. The code EM controls how the S_{ij} is computed. As the option EM= -14 is recommended for environmental work, we used this option to perform our source analysis of atmospheric pollutants. Based on the Q value, it is possible to judge the quality of the fit. The theoretical value of Qshould be approximately equal to the number of data points in the data set. This can help us to determine the optimal number of factors.

It is also necessary to process the input data before computation. All aerosol samples (both fine and coarse particles) collected at the mixed site of BNU have been normalized for 24 h. The total numbers of samples and chemical species employed for PMF analysis are shown in Table 1.

The chemical mass balance (CMB) model requires grouping of similar contributing sources into source types, and the development of appropriate composition profiles for each source type. Furthermore, the results derived from this model should be compared with those from other modeling techniques. To date, there have been no established source profiles in China based on the local aerosol data, so we used two kinds of source profiles for computation, one is from the "speciate" software developed by US EPA^[27], and the other is the profiles developed for the Beijing aerosol by Dr. Su Ge from the Oregon Graduate Institute of Science and Technology^[28]. A summary of the source profiles we used is listed in Table 2.

Since the aerosol loading in the Beijing area (a semiarid area) is generally quite high, sampling periods were constrained to 12 or 8 h and were not consistent. All raw data have been normalized to the 24-h basis for receptor modeling. Both PM_{10} and $PM_{2.5}$ data were computed. Tables 3 and 4 give the ranges and uncertainties of annual average elemental composition of PM_{10} and $PM_{2.5}$ samples, respectively. The uncertainties listed in these two tables were used as input data for model calculation.

In CMB8 model, both yearly and seasonal average values were inputted as the fitting sample data. The chemical composition of PM_{10} includes 22 species: S, Na, Mg, K, Al, Ca, V, Co, Cr, Cd, Mn, Fe, Ni, Cu, Ti, Zn, Pb, Ce, As, Se, $SO_4^{2^-}$ and Cl⁻. Organic carbon (OC) and elemental carbon (EC) concentrations were included in $PM_{2.5}$ data.

Table 1 The total numbers of samples and chemical species selected in PMF modeling

Particle size	Total number of samples	Chemical species
PM_{10}	77	9 ions: NH ₄ ⁺ , K ⁻ , F ⁻ , MSA, CH ₃ COO ⁻ , HCOO ⁻ , Cl ⁻ , NO ₃ ⁻ , SO ₄ ²⁻ ;
PM _{2.5}	72	23 elements: As, Se, Cr, Sb, Zn, Sr, Pb, Ni, Co, Cd, Fe, Mn, Mg, V, Ca, Cu, Ti, Sc, Eu, Al, Ce,
		Na, and S

 Table 2
 A list of codes used in "NAME" for computation

"NAME" cod	e Source Type	Description	Elemental species	Ionic species	Carbon species	Reference
EPAMO1	motor vehicle	light-duty vehicles with unleaded gasoline	18 elements	none	OC, EC	Speciate (US EPA)
MOPB1	motor vehicle	light-duty vehicles with leaded gasoline	18 elements	none	OC, EC	Speciate (US EPA)
RESICO	coal combustion	residential space heating	20 elements	none	OC, EC	Speciate (US EPA)
CRUST	soil dust	earth's crust	21 elements	none	EC	Speciate (US EPA)
IRON1	industrial	cast iron induction furnace	19 elements	none	none	Speciate (US EPA)
STEEL	industrial	steel production-average	16 elements	SO4 ²⁻ NO3 ⁻	OC	Speciate (US EPA)
FIELD	biomass burning	field burning composite	16 elements	SO_4^{2-} NO_3^{-}	OC, EC	Speciate (US EPA)
BIOMASS	biomass burning	biomass burning	10 elements	none	none	AIT
SECON	secondary particulate	e secondary sulfate	10 elements	\mathbf{SO}_4^{2-}	none	Ge Su (China)
(NH4)2SO4	secondary particulate	e ammonium sulfate	1 element	SO_4^{2-}	none	AIT
Constuction	construction	construction particles-ca	11 elements	none	none	AIT
Traffic	traffic	traffic dust	none	none	EC	AIT

Table 3The uncertainties of the annual average elemental composition of PM_{10} samples as input data for model computation (i g/m³)

Species	Range of the average concentration Range of the uncertainty		e uncertainty	Species	Range of the average concentration		Range of the uncertainty		
Mass	200.952	237.274	17.4862	20.3919	V	0.01201	0.04068	0.01086	0.01315
S	4.27965	11.7268	0.34717	0.9429	Ca	7.25048	12.288	0.596	0.9992
As	0.02834	0.06752	0.01757	0.0207	Cu	0.04006	0.09153	0.01370	0.0178
Se	0.00667	0.02374	0.00263	0.0217	Ti	0.26339	0.32163	0.02707	0.0317
Cr	0.01774	0.02940	0.0104	0.01135	Al	4.13160	5.77789	0.33533	0.46703
Zn	0.32760	0.86630	0.03101	0.07410	Na	1.60425	2.08274	0.24834	0.28662
Sr	0.04627	0.07873	0.01330	0.01590	Cl	2.51434	6.51632	0.26805	0.58821
Pb	0.11387	0.38897	0.02711	0.04912	NO ₃	10.20249	24.7304	1.14320	2.30543
Ni	0.03991	0.09147	0.00919	0.01332	SO_4	12.08780	37.6187	1.67622	3.71871
Cd	0.00316	0.01182	0.01015	0.01085	PO_4	0.12088	0.60379	0.11257	0.15120
Fe	3.01252	5.27205	0.25060	0.43136	Na^+	0.40149	0.86747	0.13322	0.17050
Mn	0.07388	0.20907	0.01671	0.02753	NH_4	5.56866	15.3823	0.59369	1.37879
Mg	1.28860	1.55350	0.10999	0.13118	\mathbf{K}^+	1.0206	2.69884	0.23165	0.36591
Ce	0.001433	0.1355	0.00087	0.00206	C_2O_4	1.01	1.14	0.027	0.1487
HCOO	0.0107	0.6682	0.00219	0.0022	CH ₃ COO	0.0026	0.2066	0.0018	0.1095

Table 4 The uncertainties of the annual average elemental composition of PM_{2.5} samples as input data for model computation (i g/m³)

Species	Range of the average concent	tration Range of the	uncertainty	Species	Range of the aver	rage concentration	Range of the	uncertainty
Mass	115.77 159.27	10.672	14.1516	Ca	1.5060	4.0208	0.1367	0.3379
S	3.3686 8.0495	0.2743	0.6488	Cu	0.0347	0.0728	0.0133	0.0163
As	0.0310 0.0434	0.0178	0.0188	Ti	0.0516	0.1478	0.0101	0.0178
Se	0.0066 0.0151	0.0063	0.0107	Al	0.6569	2.6441	0.0574	0.2163
Cr	0.0226 0.0346	0.0108	0.0118	Na	0.7150	1.4273	0.1772	0.2342
Sb	0.0065 0.0160	0.0038	0.0091	Cl	1.9629	4.7199	0.2239	0.4445
Zn	0.3132 0.6088	0.0299	0.0535	NO ₃	7.0217	16.9709	0.8887	1.6847
Sr	0.0143 0.0260	0.0107	0.0117	SO_4	9.2855	25.8340	1.4520	2.7759
Pb	0.0960 0.3227	0.0257	0.0438	PO_4	0.1180	.5664	0.1123	0.1482
Ni	0.0360 0.0763	0.0089	0.0121	Na^+	0.3448	1.2626	0.1287	0.2021
Cd	0.0027 0.0066	0.0011	0.0014	NH_4	5.0072	24.3308	0.5488	2.0947
Fe	1.1255 1.4587	0.0996	0.1263	K^+	0.8313	2.7530	0.2165	0.3702
Mn	0.0399 0.0857	0.0140	0.0177	OC	18.4331	44.0717	1.8469	4.4108
Mg	0.2335 0.7712	0.0256	0.0686	EC	7.4016	22.4210	0.7438	2.2457
Ce	0.0085 0.0164	0.00013	0.00148	C_2O_4	0.052	1.039	0.01236	0.1574
V	0.0089 0.0441	0.0036	0.0134	HCOO	0.00383	0.3282	0.00057	0.05865

3 Results and discussion

We performed testing runs with different numbers of factors to find an optimal one with the most physically meaningful results. The results given by 8 factors were the most reasonable and easy for interpretation. It was decided to adopt 8 factors for all the runs. The output files of G and F factors are the computed sample and compound factor matrices, respectively. The F factor matrix refers to the source profile values in ng/µg, while the G factor matrix includes source contribution values to samples in μ g/m³.

Another output file is the explained variation (EV), which is dimensionless and summarizes how important each factor is in explaining the observed matrix. The value of EV ranges from 0.1 to 1.0, from no explanation to complete explanation. The definition considers that all of the data X_{ij} are explained by P factors and the residual, as if the residual would be a (P+1) factor. In short, P+1 factors explain 100% of X_{ij} .

In the results of the BNU site, 8+1 factors were obtained for the PM₁₀ computation. About 83% of the observed data X_{ij} can be explained by the 8 factors, while for PM_{2.5} data, about 78% was explained. The EV values of the 8 *F* factors for PM₁₀ and PM_{2.5} data are given in Tables 5 and 6, respectively.

With the observed *F*-explained variation, we found that some sources are common for PM_{10} and $PM_{2.5}$ at the BNU site during the three-year period, and all the sources have their own characteristic compounds.

Among the 8 factors resolved from BNU data, 6 sources types are common to both PM_{10} and $PM_{2.5}$ particles:

• Source 1 represents the "soil-related" sources with typical crustal elements, i.e. Al, Fe, Se, Ti, Ca, and Sr, including air-borne road dust and fugitive dust. This is an important source of Beijing aerosol.

• Source 2 is rich in K⁺ and Cl⁻, representing the source of biomass-burning emissions. It includes most likely household combustion of agricultural residues and firewood, plus litter incineration. As we explained earlier, the EC/OC data are rare and samples were not taken simultaneously as the Whatman filter samples. For this reason, we did not include EC/OC data.

• Source 3 is dominated by SO_4^{2-} , NO_3^{-} and NH_4^{+} , representing the secondary particles found in both coarse and fine particles.

• Source 4 represents sources related to the fossil fuel combustion, including oil and coal combustion sources. They are all characterized by the pollution elements, such as As, Pb, Cd, Sb and Se.

• Source 5 is dominated by CH_3CHOO^- and $HCOO^-$, which often were derived from automobile exhausts. The motor vehicle source in some cases cannot be identified clearly due to the absence of OC and EC data.

• Source 6 is a minor industrial emission source, often characterized by Cu, Ni, Zn, etc., probably from non-ferrous smelters.

Both PM_{10} and $PM_{2.5}$ particles have unknown sources characterized by Ce and Cr, respectively.

Factor	3	1	4	2	6	7	5	8
Ca	0.445	0.09	0.014	0.003	0.074	0.008	0.242	0.004
Al	0.399	0.07	0.031	0.09	0.121	0.034	0.137	0.028
Ti	0.376	0.065	0.004	0.077	0.077	0.068	0.156	0.089
Mg	0.372	0.052	0.053	0.091	0.096	0.002	0.207	0.004
Fe	0.372	0.094	0.033	0.014	0.091	0.081	0.204	0.028
K	0.002	0.47	0.152	0.107	0.007	0.079	0.072	0.005
NO_3	0.099	0.004	0.629	0.002	0.065	0.007	0.04	0.154
NH_4	0.077	0.042	0.592	0.001	0.023	0.117	0.048	0.099
SO_4	0.001	0.06	0.592	0.052	0.013	0.047	0.118	0.116
CH ₃ COO	0.002	0.003	0.004	0.744	0.008	0.004	0.002	0.004
C_2O_4	0.02	0.008	0.031	0.043	0.532	0.004	0.152	0.002
HCOO	0.003	0.316	0.002	0.001	0.339	0.004	0.003	0.129
As	0.073	0.008	0.004	0.109	0.013	0.528	0.003	0.004
Pb	0.078	0.021	0.003	0.001	0.011	0.517	0.001	0.082
Cd	0.035	0.006	0.012	0.004	0.031	0.43	0.002	0.177
Ni	0.037	0.065	0.042	0.013	0.007	0.328	0.34	0.168
Cr	0.009	0.003	0.001	0.054	0.065	0.204	0.311	0.352
Cu	0.131	0.024	0.137	0.005	0.019	0.227	0.301	0.155
Ce	0.011	0.004	0.005	0.004	0.148	0.014	0.013	<u>0.536</u>

Table 5 The EV values for PM_{10} data at the BNU site

Table 6The EV values for PM2.5 data at the BNU site

Factor	3	1	2	8	6	7	4	5
Al	0.495	0.099	0.005	0.009	0.058	0.009	0.149	0.005
Ti	0.483	0.059	0.08	0.015	0.079	0.013	0.095	0.065
Ca	0.436	0.001	0.088	0.004	0.002	0.091	0.004	0.149
Mg	0.435	0.125	0.01	0.061	0.051	0.001	0.132	0.008
Fe	0.404	0.075	0.084	0.124	0.007	0.105	0.064	0.011
Cl	0.026	<u>0.516</u>	0.143	0.145	0.035	0.001	0.001	0.133
К	0.042	<u>0.304</u>	0.258	0.252	0.001	0.001	0.004	0.137
\mathbf{NH}_4	0.001	0.167	0.449	0.141	0.008	0.12	0.025	0.089
SO_4	0.037	0.158	0.449	0.188	0.069	0.083	0.004	0.012
S	0.061	0.153	0.45	0.161	0.007	0.139	0.021	0.008
NO_3	0.039	0.001	0.027	0.436	0.003	0.002	0.348	0.001
HCOO	0.001	0.002	0.106	0.251	0.513	0.002	0.122	0.004
C_2O_4	0.035	0.007	0.026	0.238	0.441	0.007	0.19	0.057
As	0.03	0.046	0.072	0.274	0.019	0.463	0.002	0.093
Sb	0.013	0.004	0.115	0.266	0.005	0.452	0.039	0.105
Pb	0.004	0.114	0.091	0.251	0.031	0.381	0.127	0.001
Ni	0.15	0.208	0.005	0.34	0.002	0.093	0.345	0.009
Zn	0.001	0.241	0.005	0.315	0.017	0.097	<u>0.331</u>	0.037
Cu	0.19	0.176	0.016	0.301	0.029	0.055	0.322	0.007
Cr	0.006	0.003	0.007	0.002	0.159	0.065	0.002	0.514

For the quantitative estimation of mass contributions of the eight resolved factors, the observed PM mass concentrations were regressed against the factor scores using MLR (multi-linear regression). This regression process also provided an additional test for the PMF model and the appropriate number of factors that have been chosen for the analysis.

For the 8-factor solution at the BNU site, the obtained

regression coefficients are all positive. The regression results for the reconstructed (predicted) mass concentrations versus the observed mass concentrations are shown in Figure 1. The *R*-square at the BNU site is 0.817 and 0.878 for the PM_{10} and $PM_{2.5}$ fraction, respectively, indicating that the observed PM mass concentrations are mostly represented by the factors.



Figure 1 Results of regression analysis for the PM_{10} and $PM_{2.5}$ datasets at the BNU site.

The mass concentrations of each factor contributing to the total mass were also derived from this regression process (Table 7). The annual arithmetic means of the total mass contribution apportionment for different types of sources at this site is shown in Figure 2. The source apportionment in Figure 2 is calculated from the average source contribution divided by the regressed mass.

Table 7 PM_{10} and $PM_{2.5}$ annual average source contributions (µg/m³) (PMF)

		PM_{10}	PM _{2.5}
	soil dust	60.7	30.6
	biomass	23.3	17.9
	secondary	29.4	29.4
Source contribution	automobile	39.5	43.7
	fossil	40.9	21.2
	industrial	3.3	7.3
	unknown	-	3.4
Regressed mass	-	197.1	154.3
Measured mass	_	204.2	141.9



Figure 2 Source apportionment by PMF model at the BNU site.

The BNU results in Figure 2 indicate that for PM_{10} particles the major contributing sources at BNU sites were soil dust and fossil fuel combustion. However, for $PM_{2.5}$ the contribution of fossil fuel combustion became less prominent, while secondary particles and automobile exhaust contributed more in the $PM_{2.5}$ than in PM_{10} .

After running the CMB model many times with an input file in which results in the wet and dry seasons have been averaged, we got relatively satisfactory fitting parameters (Table 8). The dry season represents December, January, February, March, April and May, whereas the wet season includes June, July, August, September, October and November. The source apportionment at the five sites for the whole year and different seasons is shown in Figure 3.

Table 8	Details of the fitting	parameters in	CMB8	model
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Sizo	Season	D ²	Percent 2		Number of
Size		Λ	mass (%)	л	fitting species
PM_{10}	annual	0.94	89.7	2.15	17
	wet	0.92	92.1	4.08	17
	dry	0.93	85.6	3.05	18
$PM_{2.5}$	annual	0.89	83.2	3.21	20
	wet	0.95	70.9	2.17	18
	dry	0.86	101.2	3.06	20



Figure 3 Source apportionment by CMB8 at the BNU site.

From Figure 3, one can see that the percentage contributions of each source were consistent with those of the PMF model. Unlike the PMF model, the average OC and EC concentrations were taken into account in this case, thus the motor vehicle source can be estimated clearly. Fossil fuel combustion sources contribute more in the dry season than in the wet season, while soil dust was a major contributor for PM_{10} particles and did not show much seasonal variation.

4 Conclusions

Aerosol source apportionment for PM2.5 and PM10 particles in Beijing has been conducted by using PMF and CMB8 models. The PMF approach was applied at the urban intensive site, and explained the PM₁₀ and PM_{2.5} data with 8 factors and about 78% 83% of the variation of the most fitted species. Observing the F-explained variation, we found 6 common source-types at the BNU site, which can be assigned as: (1) soil-related dust (crustal elements Fe, Al, Ti, Ca, Mn); (2) fossil fuel combustion including industrial emission (As, Se, Pb, Sb, V); (3) secondary pollutants $(SO_4^{2-}, NO_3^{-}, NH_4^{+})$; (4) biomass burning (K^+, Cl^-, F^-) ; (5) automobile exhaust (CH₃CHOO⁻, CHOO⁻); and (6) industrial sources (Ni, Cu, Cr, etc). Generally speaking, the results of CMB8 model showed similar sources, and the relative contributions of these sources are consistent with those of the PMF model. Because the average OC and EC concentrations were taken into account only in CMB

modeling, the motor vehicle sources were estimated more clearly in this case.

We have identified both regional common sources, namely vehicular emissions, particulate of secondary origin and biomass burning, and country-specific problems, such as sand storms and soil dust that should be addressed to for effective air quality control. Some management recommendations have been formulated as follows.

Extremely fine particulate in dried riverbed in this semiarid area tends to become air-borne during the dry season, and liana-type vegetation covering is recommended. Our data indicate that lead contents in particulate samples were higher than the background levels, although leaded gasoline has been banned since 1999 in Beijing and since 2001 in the whole country. Illegal leaded gasoline can still be found in gas stations in the vicinal provinces and cities off Beijing. Stringent inspection and monitoring of illegal leaded gasoline are recommended for ambient monitoring of lead in air particulate, inspection of gasoline lead content at gas-filling stations and inspection of gasoline lead content of vehicles entering Beijing.

Our findings also indicate that there are terrestrial sources of organic sulfur emissions probably from cropland and land-filling sites in the suburban areas. Hence agricultural area sources of secondary particulate should not be overlooked.

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