

Source apportionment of PM_{2.5} in Beijing in 2004

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Abstract

Based on measurements of fine particulate matter (PM_{2.5}, i.e., particles with an aerodynamic diameter of 2.5 μm or less) in January and August 2004, serious air pollution persists in Beijing. The chemical analysis included organic and elemental carbon, water-soluble ions, and elemental compositions. The positive matrix factorization (PMF) method was used to apportion the PM_{2.5} sources. The sources contributing dominantly to PM_{2.5} mass concentrations are coal combustion in winter and the secondary products in summer. Furthermore, the contributions from motor vehicles, road dusts and biomass burning could not be neglected. The products of biomass burning for winter heating in the area around Beijing could enter the urban area during quasi-quiet weather conditions. In conclusion, some effective control measures were proposed to reduce the PM_{2.5} pollution in Beijing.

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1. Introduction

Recent epidemiologic studies have found an association between ambient fine particulate matter (PM_{2.5}, i.e., particles with an aerodynamic diameter of 2.5 μm or less) and morbidity and mortality [1–3]. The PM_{2.5} problem in Beijing, the capital of China, is very serious, and the mass concentration exceeds 100 μg m⁻³ (e.g., 136 μg m⁻³ in 1999 [4]; 115 and 127 μg m⁻³ at two sites during 1999–2000 [5]; 101 μg m⁻³ in 2000 [6]; 154 μg m⁻³ during 2001–2003 [7]). Wang et al. [7] concluded that almost 79% of the daily PM_{2.5} mass concentrations during 2001–2003 did not meet the US EPA 24-h standard, which is 65 μg m⁻³. The aerosol could also lead the values of the solar ultraviolet irradiance to increase in the troposphere [8]. The air quality in Beijing must be improved to project a positive image of the city to the world as the holder of the 2008 Olympic Games, and the Beijing government is aware of the problem. More recently, Varotsos et al. [9,10] used the detrended fluctuation analysis to particulate measurements in Athens and

East Baltimore, and revealed persistent long-range power-law corrections from 4 h to 9 months for PM₁₀ (particles with an aerodynamic diameter of 10 μm or less) and for lag times from 4 h to 2 weeks for PM_{2.5} fluctuations. Therefore, in the years close to the 2008 Olympic Games, the measures to reduce the PM_{2.5} pollution will be performed, which require a clear, quantitative understanding of its chemical composition and sources. Zheng et al. [6] and Song et al. [11] used the chemical mass balance model (CMB) and the positive matrix factorization (PMF) method to apportion the PM_{2.5} sources in 2000.

The government has enacted many measures to control air pollution in five stages over several years (the Beijing Environmental Protection Bureau [BJEPB], annual reports for 2000–2004) since 1999. However, more pollution control measures will be needed to further reduce air pollution. The consumption of both coal and total energy kept increasing. In 2004, 2.8 × 10⁷ t of coal provided 54.9% of the total energy consumption of 5.1 × 10⁷ t. The number of motor vehicle reached 2.3 million in 2004. The annual SO₂ and NO_x concentrations, 55 and 71 μg m⁻³, respectively, in 2004, were still high by comparison with Western countries. The US national annual SO₂ and NO_x concentrations in 2000 were only about 5 and 20 μg m⁻³, respectively (<http://www.epa.gov/air/airtrends/sulfur2.html>, <http://www.epa.gov/air/airtrends/nitrogen.html>).

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This study showed the chemical composition and source apportionment of PM_{2.5} in Beijing in January and August 2004. The reported results would be very useful for making further policy decisions.

2. Methods

2.1. Data description

In January and August 2004, PM_{2.5} was sampled at six sites in Beijing. The locations are shown in Fig. 1, and comprised five urban sites, abbreviated PKU, OLC, SJS, TZ, and LX, and one rural site, MT. Site PKU was located on the roof of a five-story building on the campus of Peking University, which was surrounded by residential apartments and electronics companies. Site OLC was located on the ground in the Olympic Center, and reflected the air quality in the area where the 2008 Olympic Games will be held. Site SJS was on the fourth floor of a building belonging to the Capital Steel Corporation Limited, within the Shijingshan industrial area, and represented industrial pollution. Site TZ was on the 10th floor of a building in Tongzhou, which represented southeastern Beijing and was surrounded by residences. Site LX was on the ground in Fangshan, in southwestern Beijing. As the background site, site MB was near the Ming Tombs, north of Beijing, where fewer people lived.

The samples were collected at 2-day intervals, from 11 to 19 January and from 11 to 19 August 2004, to represent winter and summer. The sampling periods were 12 h at PKU, OLC, SJS, and LX and 24 h at TZ and MT. Three parallel filters (two Teflon filters and one quartz fiber filter, 47 mm in diameter) for each sampling were collected simultaneously. The mass concentrations were measured first. Then, organic carbon (OC) and

elemental carbon (EC) were detected using a thermal/optical carbon analyzer (Sunset Laboratory, OR, USA), and sulfate, nitrate, chloride, and ammonium were analyzed using ion chromatography (IC). The metal elements were determined using inductively coupled plasma mass spectrometry (ICP-MS). In addition, some organic speciation obtained monthly was ascertained using GC/MS, but was not included in the PMF analysis.

2.2. Model description

Paatero and Tapper [12] and Paatero [13] developed the PMF model. The general receptor-modeling problem can be written as

$$X = GF + E, \quad (1)$$

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

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

where $i = 1, \dots, n$ samples; $j = 1, \dots, m$ elements; $k = 1, \dots, p$ sources. The matrices, G , F and E are estimated from the known matrix X in PMF. G and F have the relative units. To find the number of sources, it is necessary to test different of sources and find the optimal one with the most physically reasonable results. To decrease the rotational freedom, PMF applies non-negative 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

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

where s_{ij} is the uncertainty estimate in the j th element measured in the i th sample. The standard deviations for each data point, x_{ij} , are calculated at each iteration step using

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

where c_1 and c_2 are user-specified parameters: c_1 takes the value of the detection limit (Paatero [13]) and c_2 is adjusted to 0.15 in this study, since $Q(E)$ is close to the number of individual elements of X .

In the PMF, the parameters $FPEAK$ and the matrix $FKEY$ are used to control the rotation when finding a more reasonable solution [13], although rotational ambiguity is always a problem in factor analysis methodology. PMF was run many times with different $FPEAK$ values to determine the range within which the objective function $Q(E)$ values in Eq. (3) remains relatively constant. 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. Sometimes

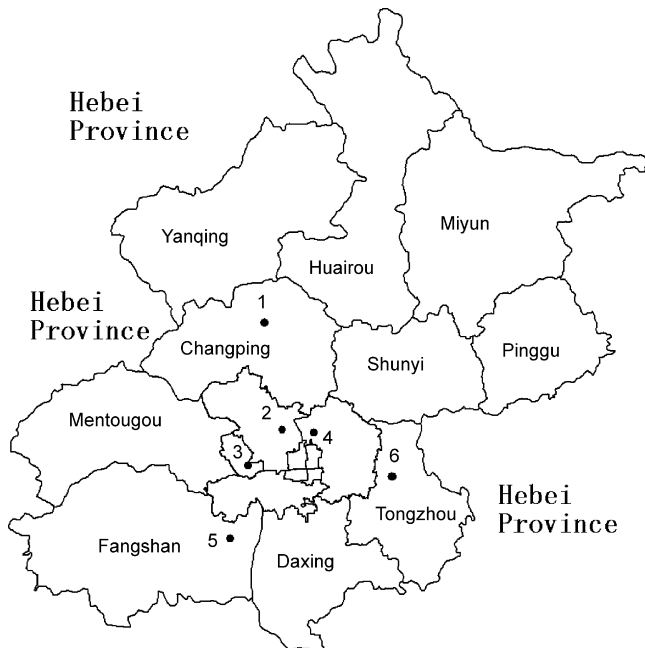


Fig. 1. Locations of the sampling sites in Beijing: 1, MT; 2, PKU; 3, SJS; 4, OLC; 5, LX and 6, TZ.

an entry in matrix F (the source profile) conflicts with *a priori* information indicating that the value should be small or zero. Setting the corresponding control variable in $FKEY$ to a positive value can reduce the entry in matrix F or even force it to zero.

After excluding species that were frequently present at concentrations below the detection limit (BDL), 22 species were selected for the PMF analysis in 2004: OC, EC, NO_3^- , SO_4^{2-} , NH_4^+ , Cl, Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, and Pb. The sampling number was 98. In this data set, the data for species with BDL values were replaced with values corresponding to one-half of the appropriate analytical detection limit [14]. Si, an important element in Beijing PM_{2.5} [6], was not measured. It was estimated roughly in the apportionment of the PM_{2.5} mass concentrations by multiplying Ca by 1.9, based on the 2000 measurements [11]. The OC was multiplied by 1.4, the average molecular weight per carbon weight, to obtain the mass of organics when entering the PM_{2.5} reconstructed mass.

3. Results and discussion

3.1. Chemical composition

Table 1 presented the chemical mass compositions at the six sites individually and averagely in January and August 2004. Fig. 2 showed the monthly average of chemical contributions. The mass concentrations at MT were obviously low both in January and August, as few people lived there and this site is far away from the downtown (Table 1). The mass concentration of organics at each site was higher in January than in August. They could be emitted from coal combustions, which is still prevalent for winter heating in Beijing. The PM_{2.5} mass with the chemical compositions at LX were the highest. This site was located in Fangshan, where many factories were settled there. Fangshan is also close to Hebei Province, the biomass burning from the farmers could also enter Beijing through Fangshan [15].

In winter, the fluctuations in air pollution in Beijing are often dominated by periods of strong cold air intrusion, which occur every 4 days on average [16]. These cold fronts, with their

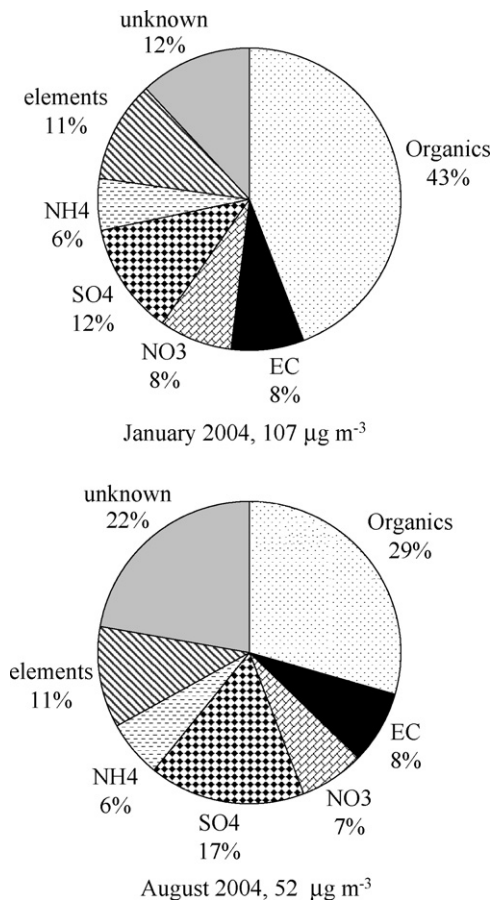


Fig. 2. PM_{2.5} chemical composition in Beijing in January and August 2004.

accompanying high winds, can sweep the air pollutants from the city. After they pass, the pollution begins to re-accumulate under quasi-quiescent conditions until the arrival of the next cold front. The sampling in January 2004 likely captured a typical pollution episode. The wind speed at 1400 h (Beijing time) was 2.0, 2.2, 2.2, 2.5, 1.0, 5.4, and 4.3 m s^{-1} from 13 to 19 January (meteorological records of the Beijing Observatory Station). Cold air

Table 1
Average concentration ($\mu\text{g m}^{-3}$) of chemical species in PM_{2.5} in January and August 2004

Site	Month	PM _{2.5}	Organics	EC	NO_3^-	SO_4^{2-}	NH_4^+	Elements
PKU	January	100.4	44.4	7.1	9.2	12.6	5.4	10.3
OLC	January	97.2	40.4	7.1	6.0	11.0	5.4	9.6
MT	January	76.9	34.2	5.5	7.1	9.8	5.3	7.5
TZ	January	111.2	52.0	8.8	9.1	14.5	5.6	12.9
SJS	January	116.2	52.6	8.7	8.9	13.5	7.3	12.0
LX	January	126.5	55.8	11.4	9.5	14.3	6.5	14.9
Average	January	106.7	47.1	8.3	8.3	12.7	6.0	11.4
PKU	August	51.4	15.1	3.8	4.0	9.3	3.2	5.9
OLC	August	58.8	17.5	4.0	2.3	7.0	3.1	5.0
MT	August	25.9	6.9	1.4	1.8	5.5	2.1	2.9
TZ	August	46.5	15.3	3.7	4.7	8.0	2.8	4.6
SJS	August	52.8	16.8	3.9	3.8	9.4	3.7	5.8
LX	August	62.0	16.3	6.0	4.9	11.2	3.9	8.0
Average	August	52.3	15.3	4.0	3.7	8.7	3.3	5.7

arrived in Beijing on 18 January. The PM_{2.5} concentrations were high on 13, 15, and 17 January and decreased rapidly on 19 January. In January 2004 the organics dominated aerosol mass, with a share of 43% (Fig. 2). These could be emitted from coal burning for winter heating.

In August 2004, the PM_{2.5} concentration in August 2004 was $52 \mu\text{g m}^{-3}$. The frequent rain that occurred during the samplings in August 2004 likely washed out the ambient particles (the meteorological records showed that it rained on 11, 12, 15, and 16 August). In August 2004 the organics dominated aerosol mass, with a share of 29% (Fig. 2). However, as the seasonal heating stopped, they could be the parts of secondary organic carbons (SOC) from photochemical reactions. The percentage of SO_4^{2-} in 2004 was high of 17% (Fig. 2) due to the stronger solar radiation in summer could accelerate the transformation rate of SO_2 .

It should be noticed that the atmospheric nitrate and sulfate could be harmful to the cultural heritage in Beijing, such issue had been investigated by Ferm et al. [17,18] in Greece.

3.2. Source apportionment

After numerous runs, no rotation was selected ($FPEAK=0$) as the $Q(E)$ values remained relatively constants, and six possible particulate sources were identified and could be explained the reasonable results. The six probable sources were identified

as biomass burning, secondary sulfate, secondary nitrate, coal combustion, motor vehicles, and road dust. However, SO_4^{2-} was unreasonably high in the chemical profiles of biomass burning, and motor vehicle emissions. The $FKEY$ values were set to 9 to reduce these sulfate values markedly. Fig. 3 presented the identified source profiles. Table 2 listed the source contributions at the six sites individually and averagely during the two sampling periods.

The first source was coal combustion, reflected in the high OC, EC, SO_4^{2-} , and Cl [6,11,19] (Fig. 3). The mass contribution and percentage in January 2004 were $40.6 \mu\text{g m}^{-3}$ and 38%, respectively, which were much larger than the values of $5.9 \mu\text{g m}^{-3}$ and 11% in August 2004, because coal burning for heating contributed much to the PM_{2.5} pollution in winter, and stopped in summer. The minimum contribution was found at MT where few people live there.

The second source was secondary sulfate, identified by the high sulfate and ammonium concentrations (Fig. 3). Its contribution was $12.6 \mu\text{g m}^{-3}$ and 24% in August 2004, which was much higher than the $9.2 \mu\text{g m}^{-3}$ and 9% in January 2004. The conversion rates of SO_2 are high in summer because the high relative humidity (RH) accelerates the dry conversion processes [20,21]. The spatial distribution for the secondary sulfate was relatively uniform.

The third source was secondary nitrate, which results in high nitrate and ammonium concentrations (Fig. 3). Ammo-

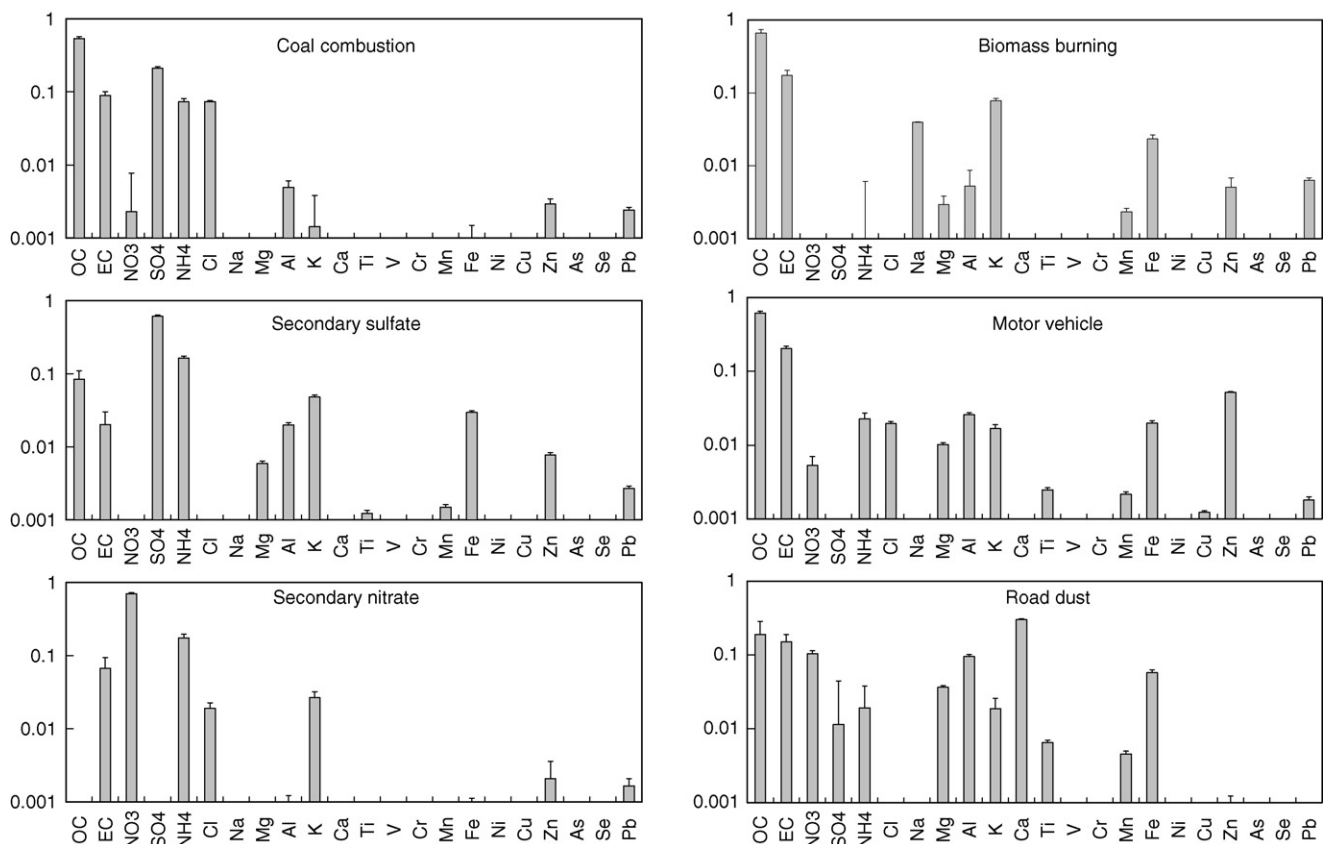


Fig. 3. Source profiles resolved from PM_{2.5} samples for biomass burning, secondary sulfates, secondary nitrates, coal combustion, motor vehicles, and road dust (prediction \pm standard deviation).

Table 2
PM2.5 Source contributions in mass concentrations ($\mu\text{g m}^{-3}$) in Beijing in January and August 2004

Site	Month	Coal consumption	Secondary sulfate	Secondary nitrate	Biomass burning	Motor vehicle	Road dust	Unknown
PKU	January	38.1	6.6	11.3	18.0	7.9	5.9	12.6
OLC	January	34.7	8.0	7.1	11.5	10.5	6.5	18.9
SJS	January	45.7	11.6	11.8	17.6	11.5	6.7	11.4
MT	January	25.3	9.2	9.2	12.2	3.6	6.8	10.5
LX	January	50.0	11.3	12.3	17.9	7.4	12.7	14.9
TZ	January	44.7	8.1	11.4	22.2	7.2	7.6	10.2
Average	January	40.6	9.2	10.5	16.4	8.5	7.8	13.7
PKU	August	6.4	10.2	4.9	8.9	2.3	7.2	11.4
OLC	August	7.0	11.1	2.6	4.0	12.7	2.7	18.6
SJS	August	3.4	14.1	5.6	8.2	9.9	3.5	8.2
MT	August	1.6	9.4	2.0	2.6	3.1	1.6	5.6
LX	August	6.6	17.6	4.6	7.1	10.5	7.2	8.5
TZ	August	10.3	9.8	5.0	8.8	3.0	2.0	7.6
Average	August	5.9	12.6	4.2	6.7	7.8	4.4	10.6

nitrate is semi-volatile at high temperatures [20] and is not expected to be present in aerosols in summer or during the day [21]. Its mass contribution and percentage in January 2004 were $10.5 \mu\text{g m}^{-3}$ and 10%, respectively, which were higher than the values of $4.2 \mu\text{g m}^{-3}$ and 8% in August 2004. Similar with the secondary sulfate, the secondary nitrate was also uniform spatially.

The total percentages of secondary sulfate and nitrate were 19 and 32% in January and August 2004.

The fourth source was biomass burning, which is characterized by a high OC, EC, and K [22] (Fig. 3). Although biomass burning in suburban Beijing is prevalent in spring and fall [23], the amount in January 2004 was surprisingly high at $16.4 \mu\text{g m}^{-3}$ (15%) (Table 2). Levoglucosan, an important organic trace in biomass burning [24], was also very high in January 2004, with an average concentration for the six sampling sites of $3.1 \mu\text{g m}^{-3}$. The level was only $0.31 \mu\text{g m}^{-3}$ in August 2004. In suburban Beijing and the counties in Hebei Province near Beijing, the residents still burn wood and stalks for heating. During the sampling period in January 2004, the weather was quasi-quiet for several days, so that emissions from biomass burning might have entered Beijing via turbulent diffusion and weak advection.

The fifth source was motor vehicle emissions, which are characterized by high OC, EC, and Zn (Fig. 3). The government recently implemented the European Euro-3 standard in China and unleaded gasoline (ULP) started to replace lead-containing fuel in 2000 (<http://www.bjepb.gov.cn/>). Zn is emitted from lubricant oil, brake linings, and tires [25]. The mass contributions of motor vehicles were 8.5 and $7.8 \mu\text{g m}^{-3}$ in January and August 2004, with percentages of 8 and 15%, respectively (Table 2). In 2004, there were 2.3 million motor vehicles. The motor vehicle contribution in Beijing was very high compared to that in New York city [26], which was only $0.52 \mu\text{g m}^{-3}$ in July 2001. Moreover, the number of household vehicles in New York alone, which was 2.98 million in 2000 (<http://www.rightofway.org/research/5borocars2000.htm>), exceeded the total number of vehicles in Beijing in

2004. In other words, emissions from motor vehicles in Beijing are a much larger factor than in New York city. Recent testing showed that 40% of the cars and 70% of the taxis in Beijing failed to meet the most basic western emission standards [27]. The motor vehicle contributions were high in OLC and SJS due to the heavy traffic. However, although the PKU site is surrounded with traffic roads, the contribution was high in January, but not in August.

The sixth source was road dust, which included high levels of the crustal elements Al, Ca, Mg, Fe, and Ti [22] (Fig. 3). The sulfate and nitrate in this source could be caused by the deposition [18]. This contributed $7.8 \mu\text{g m}^{-3}$ (7%) in January and $4.4 \mu\text{g m}^{-3}$ (8%) in August 2004 (Table 2). The contribution was high at LX where the buildings were under construction.

In total, the PMF method resolved 87 and 80% of the PM2.5 in January and August 2004, respectively.

3.3. PM2.5 sources and human health

Mar et al. [28] found that the sulfate and traffic sources of PM2.5 were most consistently associated with cardiovascular mortality in Phoenix, AZ, USA. The sulfate and traffic contributions to PM2.5 in Beijing were 11 (including the primary particles from coal combustion) and $8 \mu\text{g m}^{-3}$ higher than in Phoenix (2 and $6 \mu\text{g m}^{-3}$), respectively [29]. Grahame and Hidy [30] found that coal combustion was an important contributor to mortality risk, and it is unfortunate that the contribution of coal burning in Beijing is so high, especially in winter, accounting for around 40% of the PM2.5 concentrations. In contrast, soil and biomass burning sources were not associated with an increased risk in Phoenix [28]. The chemical composition of road dust in Beijing differed from that in Phoenix because the former included sulfate, OC, and EC, probably as a result of the deposition of coal-burned particles. The related toxicity needs to be studied. Moreover, the effects of individual chemical species, such as black carbon and heavy metal elements, on human health in Beijing should also be examined.

4. Conclusions

PM_{2.5} pollution was still serious in Beijing. The organics were dominant in the two sampling periods in the PM_{2.5} mass concentrations. However, they could be mainly emitted from coal combustion for winter heating and transferred from ambient volatile organic compounds (VOCs) in summer, respectively. The results of source apportionment showed that the coal combustion contributed mostly to PM_{2.5} mass concentration in winter, and the secondary products could be the dominant one in summer. The contributions from biomass burning especially in winter, and motor vehicles and road dusts could not be neglected.

To alleviate the PM_{2.5} pollution in Beijing, some control measures should be performed. Firstly, reducing the coal-fired light-duty boilers used for winter heating and replacing coal-fired power stations with some advanced coal gasification technologies could be the important pathways to control the PM_{2.5} related coal. Further, the reducing SO₂ mainly emitted from coal burning also would lead a lower secondary sulfate in PM_{2.5}. It is a good example that Beijing government had performed lots of control measures on SO₂, such as prohibiting the use of high-sulfur coal, forcing the industry to desulfurize and dedust the emitted smoke in the recent years, the annual SO₂ concentration decreased from 80 μg m⁻³ in 1999 to 55 μg m⁻³ in 2004. Secondly, public traffic, such as subways and buses should be improved to prohibit the private cars increasing in Beijing. The gaseous precursors of NO_x and VOCs from motor vehicles could be transformed to the secondary products (SOC, and secondary nitrate). Grassing and paving on bare surfaces could avoid the dusts entering the air. We noted that the contribution of biomass burning was surprisingly high in January 2004, at 15%. As the sampling period covered a period of quasi-quiet weather, emissions from biomass burning from suburban Beijing and Hebei Province could have entered the Beijing urban area. The Beijing government also must interact with the government of Hebei Province to control biomass burning in the areas surrounding Beijing.

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