

# Source Apportionment of Fine Particle PM<sub>2.5</sub> in Beijing, China

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Fine particles with aerodynamic diameter less than 2.5  $\mu\text{m}$  (PM<sub>2.5</sub>) were collected from three sites in Beijing during April, August, and November 2000 and January 2001. After chemical components in samples are analyzed, a chemical mass balance (CMB) receptor model using PAHs as tracers is applied to quantify the source contributions to PM<sub>2.5</sub> in Beijing. The results show that the major sources are coal combustion, fugitive dust, vehicle exhaust, secondary sulfate and nitrate, and organic matter while biomass burning and construction dust contribute only a small fraction. In addition, source inventory in Beijing is used to determine the primary source contributions. The two methods result in comparable results. Source apportionment at three sampling sites presents similar contributions to PM<sub>2.5</sub> although the sites are far away from each other. However, distinct seasonal pattern is presented for the source contributions from coal combustion, fugitive dust, biomass burning, secondary sulfate and nitrate.

Key words: Fine particle; Source apportionment; Beijing

## 1. Background

Airborne particulate matter, especially fine particles, is of concern because of its effects on human health, visibility, and climate in large scale. Fine particles can penetrate into human air tract or even lungs and are harmful to health. Several epidemiological studies <sup>[1-3]</sup> reported a link between elevated particle concentration and increased mortality and morbidity. Additionally, aerosol also influences both atmospheric visibility and climate through the scattering and absorption of solar radiation <sup>[4]</sup>.

China, with economic development and population growth in recent years, is facing with more serious and complicated air pollution that results in very unique chemical transformation and transport process. During the past decades, air pollution in China was mainly caused by the use of coal as fuel for industrial and domestic purposes. SO<sub>2</sub> and particulate matter were among the major pollutants.

Unfortunately, before the existing problems due to coal combustion are resolved completely, other emission sources become more and more important owing to rapid urbanization and industrialization. Fast growth of vehicles in some Chinese mega-cities and economically developed regions, such as Beijing, Shanghai, Guangzhou, Pearl River Delta and Yangtze Delta etc., has led to a sharp increase in concentrations of NO<sub>x</sub>, volatile organic compounds, particulate matter and ozone <sup>[5-7]</sup>. Thus, the combination of coal smog and traffic exhaust results in serious pollution characterized by enhanced photochemical smog, high concentration of fine particles, and poor visibility. The air pollution of this kind is not only a local issue but also regional one in extent.

Beijing is the capital of P. R. China located at the North China Plain with 44 meters above mean sea level in elevation, and a typical Chinese mega-city with a population of over 11 million. This city, while enjoying the rapid development since the 1980s, is bothered with elevated concentration of particulate matter and poor visibility in spite of the adoption of a large number of control measures for particles. Measured PM<sub>10</sub> annual mean mass concentrations <sup>[8, 9]</sup> exceeds the Grade II (100 $\mu\text{g}/\text{m}^3$ ) of China National Ambient Air

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Quality Standard (NAAQS). PM<sub>2.5</sub> concentrations<sup>[8, 9]</sup> are also much higher than 15 $\mu\text{g}/\text{m}^3$ , the recommended standard for ground-level PM<sub>2.5</sub> annual average of USA. The ratio of PM<sub>2.5</sub> concentration to PM<sub>10</sub> concentration (PM<sub>2.5</sub>/PM<sub>10</sub> ratio) ranges between 0.5-0.7 with an average of 0.6, which is about the same as those observed in Europe or USA. Furthermore, PM<sub>2.5</sub> measurements performed along wind direction<sup>[10]</sup> would suggest that anthropogenic pollution in urban area has extended to regional scale in some meteorological condition, and that the whole Beijing-Tianjin area is sometimes covered with large polluted air mass. Studies on visibility were carried out in Beijing, and the anti-correlation between elevated PM<sub>2.5</sub> concentration and visibility was described based on hourly measurements in June of 1999 and January of 2000<sup>[11]</sup>. High level of particulate matter and its adverse effects have inevitably led to increased concern over how fine particles might best be controlled. In order to develop effective control strategies for fine particle pollution abatement and improve air quality in Beijing, knowledge of the relative importance of the various sources that contribute to PM<sub>2.5</sub> is required.

The purpose of this study is to quantify the source contributions to PM<sub>2.5</sub> concentration in Beijing by source inventory and CMB model; to make a comparison of the results from these two methods mentioned above; and to investigate the spatial and seasonal variation of source contributions.

## 2. Experimental Methods

### 2.1 Sampling sites

The airborne PM<sub>2.5</sub> samples used in this study were collected at three sites: College of Chemistry of Beijing Union University (BUU), Chinese Academy of Preventive Medicine (CAPM), and Chinese Research Academy of Environmental Sciences (CRAES).

College of Chemistry of Beijing Union University, located at Fatou town in the southeast of Beijing, is 2 kilometer away from Eastern Fourth Ring-road and 500 meter away from chemical industry zone in the east. The sampling location was on the roof of the Main Teaching Building without high-rise buildings

nearby. The two roads in the east and in the north had not much traffic.

Chinese Academy of Preventive Medicine, situated in the south downtown of Beijing, is west of Eastern Secondary Ring-road and 300 meter away from Guangming Bridge. Sampling instruments were placed on the southern side of the roof of the three-story Office Building. The sampling site is adjacent to Panjiayuan East Street in the east and is surrounded by high-rise buildings in the southern and northern directions.

Chinese Research Academy of Environmental Sciences is located at the suburbs in the northeast of Beijing, and around 100 meter away from Lishui Bridge in the south. The sampling location was on the northern side of the roof of the four-story Science & Research Building. In the west, the building is 100 meter from Beiyuan Street that is a busy spine roadway connecting urban area with Changping and the residential area in the northern part of the city. However, the trees between the building and the street decrease the effect of vehicle exhaust on the sampling site.

### 2.2 Ambient sampling and analysis

Atmospheric sampling took place at the three sites over April 25-30, 2000 (spring), August 18-25, 2000 (summer), October 30-November 4, 2000 (autumn), and January 9-14, 2001 (winter). Because there were not enough identical instruments for sampling in the tree sites simultaneously, different instruments were used including MOUDI-100 impactor (MSP Company), A-245 dichotomous sampler (CIREE Company of USA), PM<sub>2.5</sub> sampler (Beijing Geological Instrument Factory) and self-developed sampler. Teflon filter and Quartz fiber filter were used for the determination of different chemical species.

The meteorological conditions were different during the four sampling periods. On April 25 of 2000, air quality was deteriorated by sand storm, and then sunny days were followed. In the sampling period of August, rainy or cloudy days were frequent, which failed to embody the typical meteorological features of high temperature and strong sunshine in summer. Over November 1-4, fine weather was occurred and the wind speed was slow. After snowing, it remained sunny or sunny/cloudy during the sampling period of January, 2001.

Chemical analytical method for ambient samples was described extensively elsewhere <sup>[10]</sup>. Briefly, inorganic elements and ions in PM<sub>2.5</sub> collected by Teflon filters were quantified by induced coupled plasma-atomic emission spectrometry (ICP-AES) and ion chromatography (IC) respectively. Organic and element carbons (OC and EC) in PM<sub>2.5</sub> on quartz fiber filters were determined by means of optical/thermal techniques. Organic compounds including polycyclic aromatic hydrocarbons (PAHs) were determined by gas chromatography/mass spectrometry (GC/MS).

### 2.3 Source tests and source profiles

The source emission profiles used in the present study were obtained from source tests in

Beijing <sup>[10]</sup> and some literatures <sup>[12, 13]</sup> that provided the emission rates of OC and EC, particle-phase PAHs, elements and ions for the major sources (See Table 1). Soil dust, road dust and wind-blown dust are combined as fugitive dust since they have similar source profiles. The documented emission rates of K, OC and EC, Al, Ca, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> for wood combustion <sup>[13]</sup> were used as biomass burning source profile because no study on fine particle emission from crop straw burning in Beijing could be referred to. Secondary sulfate and nitrate are formed in atmosphere, and their source profiles were assumed as ammonium sulfate and ammonium nitrate respectively.

Table1 Source profiles of PM<sub>2.5</sub> (aerodynamic diameter less than 2.5 μm) (unit: μg/g)

	Coal combustion [10,11]	Vehicle exhaust [10,11]	Construction dust [10,11]	Fugitive dust [10,11]	Biomass burning [13]	Secondary sulfate	Secondary nitrate
Fluoranthene	1186	2828.4 *	157.33	44.66	0.0	0.0	0.0
Pyrene	490	6512.3 *	93.20	29.58	0.0	0.0	0.0
Benz[a]anthracene	1083	645.31	27.02	9.81	0.0	0.0	0.0
Chrysene	1794	1053.1	66.15	26.78	0.0	0.0	0.0
Benzo	3755	456.66	93.00	34.22	0.0	0.0	0.0
[k+b]fluoranthene							
Benzo[e]pyrene	627	504.17	84.13	30.44	0.0	0.0	0.0
Benzo[a]pyrene	1048	617.05	31.68	8.34	0.0	0.0	0.0
perylene	365	85.03	3.38	2.36	0.0	0.0	0.0
Indeno	1061	223.01	28.07	8.84	0.0	0.0	0.0
[1,2,3-cd]pyrene							
Benzo	836	232.61	30.45	9.40	0.0	0.0	0.0
[ghi]perylene							
Dibenzo[a,h]anthracene	240 *	39.96	1.12	0.27	0.0	0.0	0.0
OC	24800	390000	0.0	186800	484000	0.0	0.0
EC	10700	365000 *	0.0	15700	28600	0.0	0.0
Al	47800 *	2160	42600	73100 *	110	0.0	0.0
Ca	4400	3373	300000 *	46500	100	0.0	0.0
K	11000	2111	16100	12100	16700 *	0.0	0.0
NO <sub>3</sub> <sup>-</sup>	3147	64400	0.0	1100	2500	0.0	775000 *
SO <sub>4</sub> <sup>2-</sup>	263600	28000	0.0	11000	2500	727000 *	0.0
NH <sub>4</sub> <sup>+</sup>	200	15500	0.0	100	1500	273000	226000

注：\*tracer

Source inventory is based upon a compilation of the measured emission factors from given source types and an integration of all activities for those sources across the studied region. The way to construct a source inventory is relatively explicit, and such inventories are widely available

in many countries. However, in the case of particulate matter, some components such as resuspended soil and dust are extremely difficult to quantify with accuracy and are subject to seasonal factors which are inadequately understood. Thus, whilst the source inventories can be regarded with some confidence for the major primary sources such as traffic and power

plant emissions, quantification of fugitive emissions and estimation secondary sources of sulfate and nitrate formed in the atmosphere are highly problematic. In addition, since emissions from different source categories often occur at different heights, their impacts on airborne particle concentrations are not directly proportional to their relative source strengths, which is not given full consideration in source inventories<sup>[14]</sup>.

### 3. Methodology

Three approaches can be employed to evaluate the contributions of source emissions to airborne particles: source inventory, dispersion models, and receptor models. Dispersion models use emissions data and fluid mechanically explicit transport calculations to predict pollutant concentrations at specific air monitoring locations<sup>[15, 16]</sup>. Different from receptor models, these models are definitive enough to identify contributions of individual sources within a class and have been used to develop optimal control strategies for air pollutants for a long time. However, the estimations made by dispersion models sometimes contain much uncertainty since the inventory which these models rely upon is inaccurate for some sources as mention above<sup>[17]</sup>.

Receptor models assess the contributions from various sources based on observations at sampling sites, the "receptor"<sup>[17, 18]</sup>. The techniques include the chemical mass balance (CMB) approach and some purely statistical approach such as Principle Component Factor Analysis (PCA). CMB requires a detailed knowledge of the composition of emissions from all individual source types which, however, is rarely practicable in an urban area with many potential and diverse sources. One of the limitations of multivariate factor analysis is that large numbers of ambient air samples must be collected and analyzed and that statistically independent sources tracers are required for each major source type. CMB approach used in this study infers source contributions based on elements, ions, OC/EC, and PAHs in PM<sub>2.5</sub> by determining the best-fit linear combination of chemical composition of emission source profiles needed to reconstruct the measured chemical composition of ambient samples. The technique can be expressed by the following set of linear equations

$$C_{ik} = \sum_{j=1}^m \alpha_{ij} S_{jk}$$

where  $C_{ik}$  is the concentration of chemical species  $i$  in fine particles at receptor site  $k$ ;  $\alpha_{ij}$  is the relative concentration of chemical constituent  $i$  in the fine particle emissions from source  $j$ ;  $S_{jk}$  is the mass contributed to total fine particulate at receptor site  $k$  originating from source  $j$ .

## 4. Results and Discussion

### 4.1 Chemical composition of PM<sub>2.5</sub>

The complicated chemical composition of fine particles would give some hints of their sources. The elements, such as Pb, As, V, Ni, Cd, may be considered originating from anthropogenic sources. On the other hand, the crustal elements, such as Al, Si, Fe, often exhibit a predominant occurrence in natural source (soil dust). Some inorganic ions such as sulfate and nitrate indicates the atmospheric transformation of SO<sub>2</sub> and NO<sub>x</sub>. Element carbon (EC) derives from some primary sources, for example combustion. The sources of organic carbon (OC) involve primary sources (such as cooking and biomass burning) and formation of gas-phase organic compounds in atmosphere.

The annual average chemical composition of PM<sub>2.5</sub> in Beijing is illustrated in Figure 1. Among the species detected, OC is the largest fraction in PM<sub>2.5</sub>, accounting for 31.8% of PM<sub>2.5</sub> mass concentration. Crustal components, SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> constitute 9.6%, 7.2% and 4.7% of average PM<sub>2.5</sub> mass concentration respectively. NH<sub>4</sub><sup>+</sup>, EC and other elements account for relative small portion of PM<sub>2.5</sub>. Compared with some cities of other countries<sup>[14,19-21]</sup>, Beijing has much higher concentrations of OC, sulfate, and nitrate. This would imply that secondary aerosol formed due to strong local emission of VOC, SO<sub>2</sub> and NO<sub>x</sub> and high oxidation capacity of atmosphere would be responsible for large fraction of PM<sub>2.5</sub> mass concentration. Besides, high concentration of mineral elements indicates another important source of PM<sub>2.5</sub>---soil dust.

### 4.2 Fine particle mass apportionment

#### 4.2.1 Source contributions from source inventory

The pollution sources of PM2.5 in Beijing were classified in three categories: stationary sources (fuel combustion in various purpose), mobile sources and fugitive sources (construction dust, paved and unpaved road dust, industrial dust, etc.).

After characterization of source emission and measurement of their emission factors, PM2.5 source inventory of 1999 as a reference year was developed within the eight districts of Beijing (1040 km<sup>2</sup>).

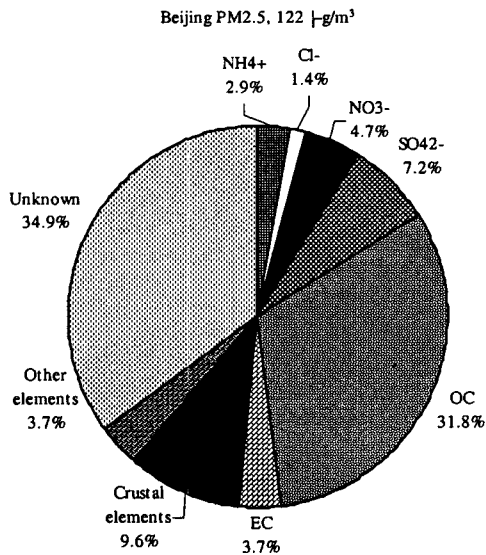


Fig. 1 Chemical composition of PM2.5 in Beijing

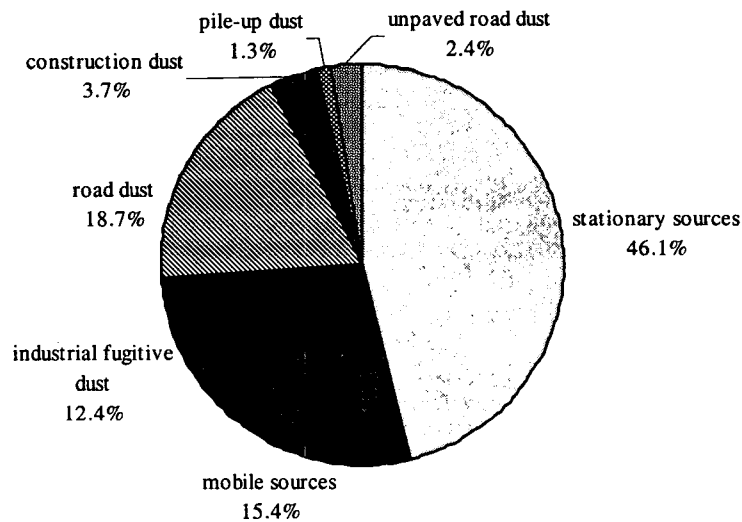


Fig. 2 Source inventory of PM2.5 in Beijing

According to the source inventory presented in Figure 2, all of the sources in Beijing discharge 53266 ton PM<sub>2.5</sub> per year, of which stationary sources accounted for 46.1%, mobile sources 15.4% and fugitive sources 38.5%. Obviously, the stationary and fugitive sources are the major primary sources of PM<sub>2.5</sub> in Beijing. Within the stationary source category, point sources and area sources contribute 18.0% and 28.2% to the total emission respectively. As for fugitive sources, road dust and industrial fugitive dust constitute 18.7% and 12.4% of PM<sub>2.5</sub> emission while construction dust and unpaved road dust only contribute a minor part.

#### 4.2.2. Source contributions from CMB model

Seven major sources of PM<sub>2.5</sub> are identified by CMB model (see Figure 3). They are coal combustion, vehicle exhaust, construction dust, fugitive dust, biomass burning, secondary sulfate and nitrate, and organic matter, which is consistent with the result computed by positive matrix factorization (PMF)<sup>[8]</sup>. These sources totally contributed 72.5% of PM<sub>2.5</sub> mass concentration.

Fugitive dust and coal combustion, with the contributions of 18.1% and 16.4% respectively, are largely responsible for PM<sub>2.5</sub> mass concentration in Beijing. The higher contribution of fugitive dust in Beijing, compared with those in some cities overseas<sup>[22, 23]</sup>, would be attributed to dry climate and low level vegetative coverage. Since coal has by far been used as important fuel in China, it is unsurprising to observe the remarkable effect of coal combustion on urban air pollution. Also, the high contribution of coal combustion in Beijing would indicate there will be a long way to go to control this source effectively.

Using PAHs as tracers makes the estimation of vehicle exhaust contribution possible because the PAHs source profiles for vehicle exhaust and road dust have less linearity than the inorganic element profiles. The result from CMB model shows the contribution from vehicle emission is 5.6%, much lower than the contribution of 15.5% obtained by employing vehicle/road dust source profile of inorganic elements<sup>[10]</sup>. If the difference between 15.5% and 5.6% is served as the rough estimate of the contribution of road dust, it is apparent that

the effect of road dust on PM<sub>2.5</sub> is significant.

In addition, construction dust and biomass burning contribute small amount to PM<sub>2.5</sub>, with the contributions of 3.3% and 4.5%, respectively.

Besides primary sources, about 9.6% of PM<sub>2.5</sub> mass concentration is attributed to secondary formation of sulfate and nitrate in atmosphere. Organic matter in fine particles derives from both primary source and organic compounds formed in atmosphere, i.e. secondary organic aerosol (SOA). The contribution of 15.0% from organic matter computed by CMB includes those from the two origins. According to studies carried out by Schauer and Zheng et al.<sup>[22-24]</sup>, the difference between total fine particle OC and those from primary sources, accounting for 19.2%-34.1% of OC in PM<sub>2.5</sub>, is related to SOA. Based on this percentage and measured OC concentration in Beijing (22.4% of PM<sub>2.5</sub>, annual average), it would be inferred that the contribution of SOA to PM<sub>2.5</sub> is about 6.0-10.6%. Summing the contributions of secondary sulfate, secondary nitrate and SOA would lead to a remarkable contribution of 15.6-20.2%, implying that secondary sources might be decisive in improvement of air quality in Beijing.

#### 4.2.3. Comparison of the results from source inventory and CMB model

Table 2 compares the average source contribution estimations calculated by source inventory and CMB model. For easy comparison with source inventory which only provides the information on primary sources, the contributions of primary sources computed by CMB are normalized in the last row of the table.

As seen in Table 2, both source inventory and CMB model suggest that the major primary sources of PM<sub>2.5</sub> in Beijing are coal combustion and fugitive dust while mobile sources contribute a minor fraction of PM<sub>2.5</sub>. The two methods also provide a comparable contribution of about 13-15% for vehicle exhaust. However, as for coal combustion, source inventory gives higher contribution estimation than CMB. This would probably be due to the ignorance of the reduced effect of high chimney emission on ambient PM<sub>2.5</sub> in source inventory. In the case of fugitive dust, the disagreement of the contributions from the two approaches could partly be attributed to the uncertainty in fugitive

dust inventory which needs further improvement. Also, the higher contribution from CMB might

mean that it includes the effect of dust from local area and outside.

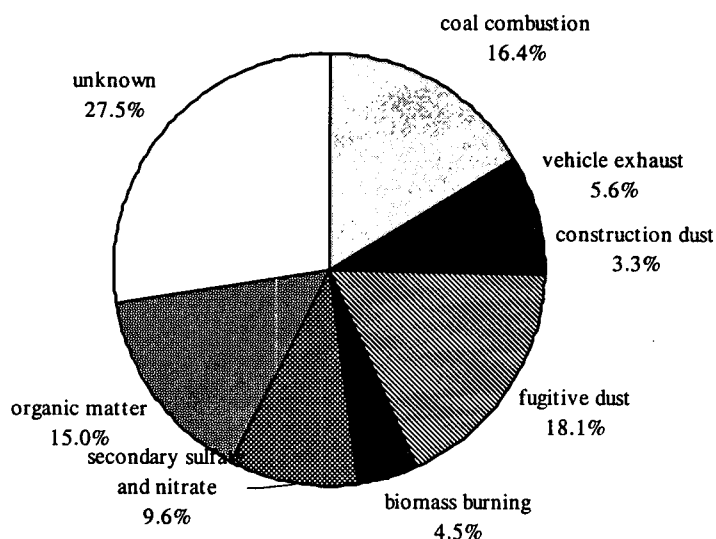


Fig. 3 Source apportionment of PM2.5 in Beijing

Table 2 Comparison of the results from source inventory and CMB model

Source	Source contribution (%)		
	Source inventory	CMB	CMB (normalized)*
Coal combustion	46.1	16.4	37.8
Mobile sources	15.4	5.6	12.9
Fugitive sources	38.5	21.4	49.3
<i>Industrial fugitive dust</i>	12.4	NA	
<i>Road dust</i>	18.7	NA	
<i>Construction dust</i>	3.7	NA	
<i>Pile-up dust</i>	1.3	NA	
<i>Unpaved road dust</i>	2.4	NA	
Biomass burning	NA**	4.5	
Secondary sulfate & nitrate	NA	9.6	
Organic matter	NA	15.0	
Unknown	NA	27.5	
<b>Total</b>	<b>100</b>	<b>100</b>	<b>100</b>

Notes: \*normalization of the contributions from coal combustion, mobile sources and fugitive sources

\*\*not available

#### 4.3 Spatial variation of source contributions

The sampling sites of BUU, CAPM and CRAES in this study are far away from each other in different directions of Beijing. The comparison of source contributions at the sites could give an outline of the spatial variance of PM2.5 origins. As shown in Figure 4, comparable PM2.5 mass concentrations and

similar source contributions are observed at the three sampling locations, which are consistent with what occurred in São Paulo, a mega-city of Brazil [20]. The similarity of PM2.5 source contributions in a large area would indicate that the air pollution caused by fine particle tends to be a regional problem which is quite different from that caused by coarse particles.

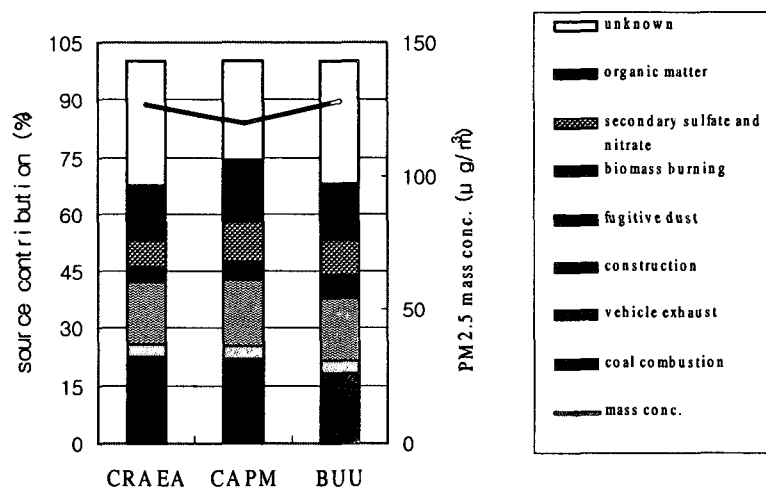


Fig. 4 Source contributions to PM<sub>2.5</sub> at the different sampling sites in Beijing

#### 4.4 Seasonal variation of source contributions

Figure 5 presents the seasonal pattern of contributions from the major sources of PM<sub>2.5</sub>. The importance of coal combustion and fugitive dust changes remarkably in different seasons. The highest source contribution is found in spring for fugitive dust, and in winter for coal combustion. The dry weather in Beijing would take main responsibility for the severe pollution caused by fugitive dust in spring. The meteorological record of 2000 showed that in spring the wind speed was highest while precipitation was lowest, and that days with low wind speed (less than 2m/s) or high humidity (higher than 60%) were scarce [25]. In addition, dry weather in North China since the 1990s and large area of unpaved ground worsen the dust pollution [25]. The influence of sand storm in the first two sampling days was also the cause for highest contribution of dust in spring. In winter, high consumption of coal and unfavorable meteorological condition for dispersion made the contribution of coal combustion rise up, which was similar to what occurred for wood combustion in southeastern cities of USA. The results reported by Zheng [23] showed that the contribution of wood combustion to the total organic carbon concentration in the atmosphere increased during the colder months of October and January due to the high residential wood

burning activities at the time of year.

Different from coal combustion and fugitive dust, vehicle exhaust and construction dust had stable contributions to PM<sub>2.5</sub> in four seasons. Biomass burning is another source whose emission changes a lot in different seasons. In this study, the contribution of biomass burning is high in spring and autumn. Particularly, the contribution rises up to 8.1% in autumn, which is 2.5 times higher than that in spring and 3.9 times higher than that in winter. Potassium is usually served as a tracer of biomass burning. Duan et al. [26] investigated the seasonal variation of K concentration in PM<sub>2.5</sub> and found that the concentration increased in Beijing in spring and autumn, which is consistent with the results in this study. The reason for that is that crop straw burning activities are frequent in Beijing after harvest in autumn and before ploughing in spring.

The contribution of secondary sulfate and nitrate, owing to the transformation of high-level SO<sub>2</sub> into secondary pollutants by homogenous and heterogeneous reactions and the accumulation of pollutants in stagnant weather, rises up to the highest level of 17% in winter, which is 1.7 times higher than that in spring and 2.5 times higher than that in autumn. However, because rainy or cloudy days dominated the sampling period in summer, the high contribution of secondary sulfate and nitrate is not observed as expected.

The variability of the contribution of organic



matter, with the range of 13.7-15.8%, is insignificant with the seasons. This is different from what observed in southeastern cities in US

where organic matter had highest contribution in July and had strong relation with the concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  [23].

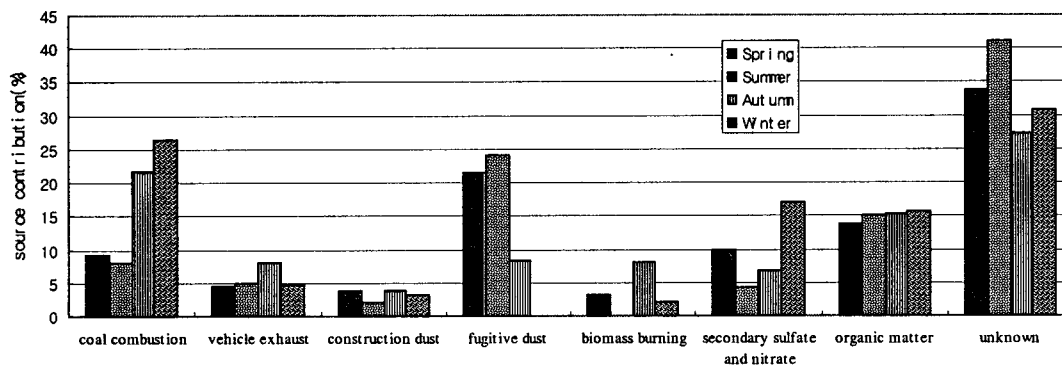


Fig. 5 Seasonal variation of source contributions in Beijing

## 5. Conclusion

The sources of PM<sub>2.5</sub> are explored in this paper by developing source inventory and applying CMB model to ambient samples collected in Beijing. The major primary sources are fugitive dust, coal combustion and vehicle exhaust, which are responsible for the majority of PM<sub>2.5</sub> mass concentration. Construction dust and biomass burning are also detected by CMB model with relative low contributions. Besides, about 9.6% of PM<sub>2.5</sub> mass concentration is assigned to secondary sulfate and nitrate, and 15% to organic matter. As discussed in this paper, 6.0-10.6% of PM<sub>2.5</sub> could further be explained by SOA. Thus, the contribution from secondary sources rises up to 15.6-20.2%, indicating the high oxidation capacity of atmosphere and rapid formation of secondary pollutants in the air. Application of CMB model at the sampling sites gives an insight on the spatial variability of PM<sub>2.5</sub> sources. The results show that the sources present similar contributions to PM<sub>2.5</sub> mass concentrations at the different locations, which might imply air pollution of PM<sub>2.5</sub> has extended to a large scale and is developing a regional problem in Beijing. Additionally, distinct seasonal variability is observed for contributions of fugitive dust, coal combustion, biomass burning and secondary sulfate and nitrate.

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