Long-term monitoring and source apportionment of $PM_{2.5}/PM_{10}$ in Beijing, China

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Abstract

During 2001–2006, $PM_{2.5}$ (particle matter with aerodynamic diameter less than 2.5 microns) and PM_{10} (particle matter with aerodynamic diameter less than 10 microns) were collected at the Beijng Normal University (BNU) site, China, and in 2006, at a background site in Duolun (DL). The long-term monitoring data of elements, ions, and black carbon showed that the major constituents of $PM_{2.5}$ were black carbon (BC) crustal elements, nitrates, ammonium salts, and sulfates. These five major components accounted for 20%–80% of the total $PM_{2.5}$. During this period, levels of Pb and S in PM remained rather high, as compared with the levels in other large cities in the world. Source apportionment results suggest that there were 6 common sources for $PM_{2.5}$ and PM_{10}, i.e., soil dust, vehicular emission, coal combustion, secondary aerosol, industrial emission, and biomass burning. Coal combustion was the largest contributor of $PM_{2.5}$ with a percentage of 16.6%, whereas soil dust played the most important role in PM_{10} with a percentage of 27%. In contrast, only three common types of sources could be resolved at the background DL site, namely, soil dust, biomass combustion, and secondary aerosol from combustion sources.

Key words: $PM_{2.5}$; PM_{10}; monitoring; source apportionment

Introduction

There has been growing concern on the high levels of airborne particulate matter in Beijing in the recent years, and inhalable particulate has long been implicated to have adverse health impacts such as respiratory diseases and increased mortality (Dockery and Pope III, 1994; Donaldson *et al*., 1998). Atmospheric particles through long-range transportation from China mainland to the Pacific can affect the global radiation energy balance (Liu and Peter, 2002) as well as influence the primary productivity of the Ocean and the global carbon budget (Zhuang *et al*., 2001) According to the data of Environmental Situation Bulletin (BJEPB, 2008), all the annual average concentrations of inhalable paritcles PM_{10} (particle matter with aerodynamic diameter less than 10 microns) in Beijing over the period of 1999–2006 exceed the national second-class air quality standard of 100 mg/m^3 (MEP, 1996). A number of articles have been published in the recent years dedicated to the characterization of particulate matter in Beijing (He *et al*., 2001; Sun *et al*., 2004). However, their monitoring activities usually lasted for only one or two years. Limited samples and short monitoring time made deliberate source identification and trend estimation difficult. Systematic and long term records of particulate matter mass and

composition for a developing country like China will help to enhance and create scientific knowledge necessary to address the worsening air quality. In particular, it will help to understand the nature of the particulate pollution in a city in relation to local sources, long-range transport, and atmospheric transformation processes that is essential for the formulation of effective air quality management strategies.

In this article, we report the results of PM characterization over a period of six years, overlapping the implementation period of major air pollution control measures in Beijing. Although several studies on PM source apportionment have been conducted in Beijing (Song *et al*., 2007; Zhang *et al*., 2007), the lack of an ideal background site as well as the lack of long-term monitoring dataset were the common problems. Our source apportionment was based not only on our six-year long-term dataset at an urban sampling site at the Beijng Normal University (BNU), but also on a comparison with the data from a remote background site in Duolun (DL), about 250 km north in the upwind direction of Beijing, where the sand dunes are the nearest to Beijing and threathen its air quality. Such a comparison can throw some light on the differentiation of local soil dust and the dust transported from remote sources like DL.

1 Materials and methods

1.1 Sampling and analysis

The $PM_{2.5}/PM_{10}$ samples were collected during the summertime and wintertime (about one month, respectively) from 2001 to 2006 on the roof of the building of Science and Technology in Beijing Normal University (BNU), which is about 40 m high. The BNU campus is located near a main arterial road of Beijing – the North Third Ring Road. In 2006, $PM_{2.5}/PM_{10}$ samples were also collected at a remote background site in Duolun, which is located about 250 km north from the center of Beijing and is in its upwind direction during the dry season.

Previously calibrated medium volume samplers (Beijng Geological Instrument-Dicked Co., Ltd., China) were empolyed for $PM_{2.5}$ and PM_{10} collection at a constant flow of 77.59 L/min. The sampling time lasted for about 12 h for each sample. Mixed fibre filters (Whatman 41, UK) were used in this campaign for mass, BC, and inorganic chemical species determinations. After sampling, these filters were immediately placed in polyethylene plastic bags and stored in refrigerator, and were then weighed under constant temperature ($20 \pm 5°C$) and humidity (40% $\pm$ 2%), using an analytical balance (Sartorius 2004MP, Germany) with a reading precision of 10 μg.

An inductively coupled plasma spectroscopy and atomic emission spectroscopy (ICP-AES) (Ulttma, Jobin Yvon Co., Ltd., France) were used for the determinations of 23 elements, including Al, As, Ca, Ce, Cd, Cr, Cu, Co, Eu, Fe, Mg, Mn, Na, Ni, Pb, S, Sc, Sr, Se, Sb, Ti, V, and Zn (Zhuang *et al.*, 2001). Ionic Chromatograph (Model 600, Dionex, USA) was empoyed for the measurements of 10 anions (Cl^-, F^-, SO_4^{2-}, NO_3^-, $C_2O_4^{2-}$, PO_4^{3-}, NO_2^-, $HCOO^-$, MSA, CH_3COO^-) and 5 cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+) according to the procedure developed by Yuan *et al.* (2003). A smoke stain reflectometer (Model 43D, Diffusion Systems Ltd., UK) was employed for black carbon (BC) analysis.

Data quality assurance, including inter-laboratory comparison of Standard Reference Material (SRM) measurements and sampler performance, has been taken into account.

1.2 PMF modeling description

The technique of positive matrix factorization (PMF), developed by Paatero and Tapper (1994), was adopted for source apportionment. Owing to the non-negative factor results and appropriate error estimations, it has been widely used in recent years (Anttila *et al.*, 1995; Lee *et al.*, 1999; Hedberg *et al.*, 2005; Rizzo and Scheff, 2007). The general receptor model of PMF can be written as Eq.(1).

$$X = G \times F + E \tag{1}$$

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. The matrices, G, F, and E are estimated from the known matrix X in PMF. To find the right source numbers, it is necessary to test different sources with different settings of parameters and find an optimal one giving the most physically reasonable results.

Since the PM level in Beijing is generally quite high, sampling periods were constrained to less than 24 h and were not consistent. Thus, all raw data ($PM_{2.5}/PM_{10}$) were normalized to a 24-h basis before receptor modeling. In the input data set, the data for species with below detection limit (BDL) values were replaced with values corresponding to one-half of the appropriate analytical detection limit, and the missing species data were replaced with values corresponding to four times of the arithmetic mean of relative species.

After excluding species that were frequently present at concentrations below BDL values, 24 species out of 36 species were chosen for the BNU $PM_{2.5}/PM_{10}$ factor analysis: BC, NO_3^-, SO_4^{2-}, Cl^-, $C_2O_4^{2-}$, NH_4^+, K^+, Na, Mg, Al, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Co, S, and Pb. The total number of samples was 216 and 223 for the BNU $PM_{2.5}$ and PM_{10} analysis, respectively, and the DL $PM_{2.5}/PM_{10}$ samples were pretreated in the same way.

2 Results and discussion

2.1 Trend of seasonal variation of $PM_{2.5}/PM_{10}$ levels in Beijing in 2001–2006

As seen in Fig.1, the $PM_{2.5}$ levels in Beijing in these years remained rather high, and are considerably higher than the average daily value of 65 μg/m³ recommended by United States Environmental Protection Agency (USEPA). However, it is interesting to note that the $PM_{2.5}$ concentration in wintertime has decreased somewhat from 2001 to 2003 and has retained a steady level since 2004. In view of the soaring increase in urban population and vehicular population in these years, the steady $PM_{2.5}$ level in recent years implies that the fuel shift from coal to natural gas for space heating in urban areas may compensate the increment in vehicular emissions. On the contrary, longer and stronger solar irradiation during summer time can favor the photochemical formation of secondary aerosol particles, which are the major constituents of $PM_{2.5}$ (Sun, 2005) that lead to the higher $PM_{2.5}$ levels during summertime.
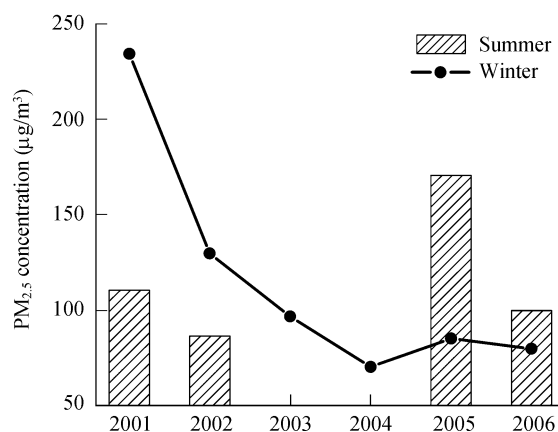

Fig. 1 $PM_{2.5}$ seasonal variation at site of Beijng Normal University (BNU), China.

PM$_{2.5}$ is of more concern in this study, since there is no routine monitoring of PM$_{2.5}$ in China and it has larger health impacts. Generally speaking, the levels of Pb in PM$_{2.5}$ decreased abruptly since 2001, indicating that the efforts of phasing out leaded gasoline did work (Fig.2). Nevertheless, the daily levels of Pb fluctuated significantly from 7.1 to 2,370 ng/m^3 with a mean of 180 ng/m^3 (Sdandard Division 233). Although they are lower than the figures in the previous reports (Sun *et al*., 2006), they are considerably higher than the values in other cities (Chow *et al*., 1996; Gao *et al*., 2002; Huang *et al*., 1996). In view that leaded gasoline has been banned since 1997 in Beijing, there may be some unknown minor Pb sources, such as illegal leaded gasoline used in vehicles from remote cities, industrial activities, coal combustion, and re-entrainment of lead-contaminated soil particles. Further investigation is needed for clear source identification.

Regarding the pollution element As, a descending trend was observed (Fig.2), which may be explained by the reduction of coal consumption in the urban areas, as coal combustion is the main source of As in Beijing. However, coal is still used in the suburban areas and in nearby cities, therefore, As can still be detected.

There seems to be a good correlationship between the total sulfur content and the sulfate content in PM$_{2.5}$ (R^2 = 0.68). It implies that the major source of sulfur is coal combustion. High concentrations of S frequently appear in wintertime (Fig.2).

The ratio of NO$_3^-$/SO$_4^{2-}$ has been used in China as an indicator of the relative importance of mobile source vs. stationary coal combustion source (Wang *et al*., 2005). Arimoto *et al*. (1996) ascribed high NO$_3^-$/SO$_4^{2-}$ ratios to the predominance of mobile source over stationary source of pollutants. Here, the average ratios of NO$_3^-$/SO$_4^{2-}$ in PM$_{2.5}$ varied from 0.34 to 1.11 with an average value of 0.63 (data not shown). There is a slow increasing trend of the NO$_3^-$/SO$_4^{2-}$ ratio, suggesting that the contribution of mobile emissions in Beijing is increasing. However, the ratio in general retained a value less than 1. This implies that coal combustion still remains to be a primary emission source.
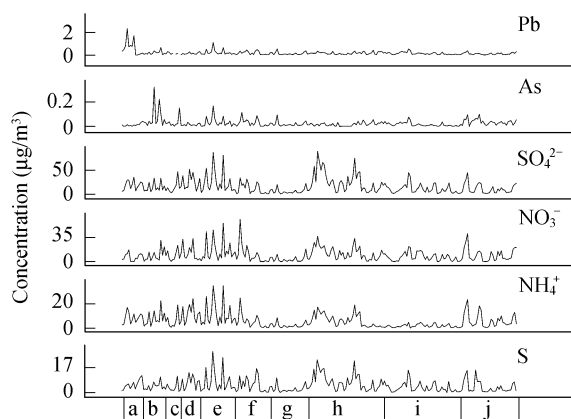

Fig. 2 Temporal variation of important pollutants in PM$_{2.5}$ at the BNU site during 2001–2006. (a) 2001 summer; (b) 2001 winter; (c): 2002 summer; (d) 2002 winter; (e) 2003 winter; (f) 2004 winter; (g) 2005 summer; (h) 2005 winter; (i) 2006 summer; (j) 2006 winter.

2.2 Source apportionment of Beijing PM$_{2.5}$/PM$_{10}$ by PMF

We performed testing runs with different number of factors to find an optimal one with the most physically meaningful results. The results given by 8 and 6 factors for PM$_{2.5}$ and PM$_{10}$, respectively, are the most reasonable and easy for interpretation. An important output is the explained variation (EV), which is dimensionless and indicates how important each factor is in explaining the observed matrix. The value of EV ranges from 0.0 to 1.0, from no explanation to complete explanation. In the results of the BNU data, about 86.3% of the observed PM$_{2.5}$ data can be explained by the eight factors, and 88.6% of the observed PM$_{10}$ data can be explained by the six factors. The following are the common factors for both PM$_{2.5}$ and PM$_{10}$:

Source 1 represents the local soil-related sources with typical crust elements, i.e., Al, Fe, Mn, Mg, and Ca, including air-borne road dust and fugitive dust. The high EV value of Ca may be associated with the intensive construction activities in Beijing in recent years. This is an important source of Beijing aerosol.

Source 2 is dominated by SO$_4^{2-}$, NH$_4^+$, and NO$_3^-$, representing the sources of secondary ionic particles. The secondary sulfates are formed by photochemical reactions, especially in summer with strong solar radiation and high temperature (Seinfield and Pandis, 1998). It was also reported that high humidity will promote the formation of secondary sulfates (Yao *et al*., 2003). Nitrates are mainly formed by the oxidation of NO$_x$. Low temperature will favor the formation of ammonium nitrate aerosol (Li *et al*., 2004). This can explain the high contribution of nitrates in wintertime. For PM$_{2.5}$, secondary sulfates and secondary nitrates were resolved as two independent sources, whereas for PM$_{10}$, they were emerged as one secondary aerosol source.

Source 3 is rich in Zn, Pb, Cu, and BC. Zn possibly comes from the fuel burning or tire abrasion (Pacyna, 1986), exist in the mobile exhaust (Huang *et al*., 1994). Zn has been used as the fingerprint of mobile vehicle emission sources in recent PMF analysis (Li *et al*., 2004). Cu may come from the braking system and the pump system (Lee *et al*., 1999). Pb may have been a minor additive for low-grade fuel produced by out-of-dated oil refineries in China, as leaded-gasoline has been phased out since 1999 in China. In source 3, we also found a medium contribution of BC. Hence, we ascribe source 3 to the mobile source.

Source 4 is rich in K, representing the source of biomass-burning emissions. It includes most likely household combustion of agricultural residues and firewood, plus little incineration. As we explained earlier, the EC/OC (elemental carbon/organic carbon) data were rare, and were not taken simultaneously. For this reason, we did not include the EC/OC data.

Source 5 is a minor industrial emission source, characterized by As, V, Cd, Mn, and Fe, probably from industrial smelting emissions. Lee *et al*. (1999) also reported a similar factor for the Hong Kong's PM source apportionment.

Source 6 is dominated by BC, S, and Cl. BC and S usually come from coal combustion. It was also found that Cl was associated with coal burning during wintertime (Yao *et al.*, 2002). Therefore, source 6 can be ascribed to coal combustion.

Besides the above-mentioned six common sources, there is an unknown source for $PM_{2.5}$. In this source, Na has the highest EV value. A similar source having high EV values of Na and Si has been ascribed to non-local dust (Xie and Liu, 2007; Zhao and Hopke, 2004, 2006). Since we used hydrofluoric acid for sample pre-treatment, we did not have Si data. However, in this factor, we found some other elements like Cl and S with high EV values correlated with sodium. The non-local dust in Beijing PM is mainly transported from Inner Mongolia (Xie and Liu, 2007), where there are several dried salinas and the main sodium-containing constituents are $Na_2SO_4 \cdot 10H_2O$ and $NaCl$ (Xu, 1993; Li *et al.*, 1990). Therefore, particles transported from these remote sources are characterized by their contents of sodium sulfate and sodium chloride. Consequently, it seems reasonable to ascribe this factor as the non-local dust. In PM_{10}, this source possesses only a small weight, and cannot be resolved by PMF.

The $PM_{2.5}/PM_{10}$ samples at the DL site were also used for source apportionment by PMF, and the results show fewer sources at this background site, as expected. There are four factors for DL $PM_{2.5}$ with 82% explanation: i.e., local dust enriched in Mg, Al, Ca, Ti, and Fe, non-local dust characterized by high Na EV value, biomass burning characterized by K^+, and secondary nitrate represented by NH_4^+ and NO_3^-. For DL PM_{10}, similar 4 factors provide the most physically reasonable explanation: namely, local dust characterized by Ca, Ti, Mn, Fe, Mg, and Al, non-local dust with high Na EV value, biomass burning represented by K^+, and secondary ions represented by NH_4^+, NO_3^-, SO_4^{2-}. In fact, in the fourth source of DL $PM_{2.5}$, the secondary nitrate source also contained SO_4^{2-}. Since its EV values are considerably lower than those of nitrates, we ascribed this source to secondary nitrate instead of secondary ions.

For the quantitative estimation of mass contributions of the resolved factors, the observed PM mass concentrations were regressed against the factor scores using MLR (multi-linear regression), and the average source contribution of each factor to the total mass can be derived from this regression process. The annual arithmetic means of the total mass contribution apportionment for different types of sources at these two sites are shown in Fig.3. Dust, combustion activities, and secondary aerosol are the three major sources of $PM_{2.5}/PM_{10}$ in Beijing. Among these sources, coal combustion turned out to be the largest contributor of $PM_{2.5}$ with a percentage of 16.7%, whereas local dust is the primary source of PM_{10} with a percentage of 23%. Regarding the PM samples at the DL site, local dust is the primary source for $PM_{2.5}$ and PM_{10} with mass contributions of 36.2% and 61.7%, respectively.
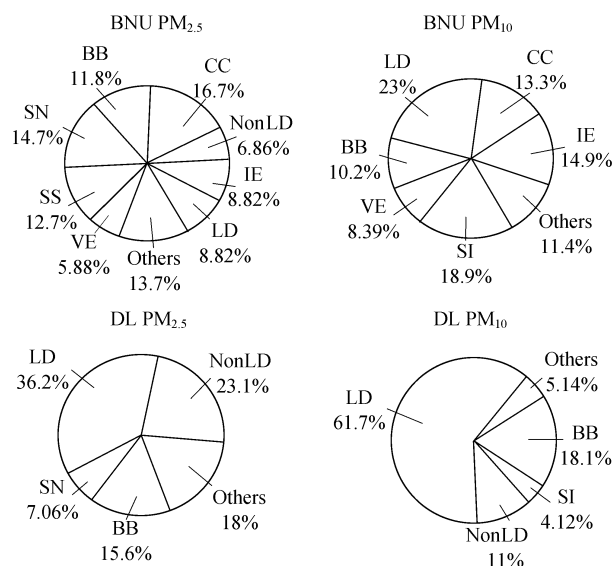

Fig. 3 Source apportionment for $PM_{2.5}/PM_{10}$ at the BNU and Duolun (DL) sites. BB: biomass burning; CC: coal combustion; IE: industrial emissions; LD: local dust; Non-LD: non-local dust; SI: secondary ions; SN: secondary nitrate; SS: secondary sulfate; VE: vehicle emissions. Others: sources that could not be explained by positive matrix factorization (PMF).

3 Conclusions

The $PM_{2.5}/PM_{10}$ samples were collected in summer and winter during 2001–2006 at the BNU sampling site. Totally 23 elements, 15 ions, and BC were determined. The trends of the temporal variations of some important pollutants in $PM_{2.5}$ are discussed. Source apportionment by PMF provides 6 common sources for Beijing $PM_{2.5}$ and PM_{10}, namely, local dust, secondary aerosol, mobile source, biomass burning, coal combustion, and minor industrial emission. Among these sources, soil is the major source of PM_{10}, while particles of secondary origins are the main source of $PM_{2.5}$. Regarding the background site in Duolun, crustal particles are the dominant constitutent of both fine and coarse fractions. Biomass combustion is the most important anthropogenic activity.

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