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Short communication

Source identification and health impact of $PM_{2.5}$ in a heavily polluted urban atmosphere in China

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ABSTRACT

Positive matrix factorization (PMF) was applied to identify the types of $PM_{2.5}$ sources and corresponding mass contributions to $PM_{2.5}$ mass concentrations using $PM_{2.5}$ measurements obtained from Dec. 2007 to Oct. 2008 in Jinan, which is a highly polluted city in China. The reconstructed mass concentrations from six sources matched the observations, and the resolved sources constituted 98.91% of the $PM_{2.5}$ mass concentrations. Secondary sources, the major source contributor, accounted for 55.15% of $PM_{2.5}$ mass concentration, while several other sources, including coal burning (20.98%), soil dust (9.30%), motor vehicles (6.06%), biomass burning (4.55%), and industry (2.87%), contributed a total of 43.76%. The non-carcinogenic risk estimates showed the elemental risk for Mn, Co, S and Cr were high, with values larger than 0.1 being observed for the three groups. The total elemental risk in Jinan for the three groups was higher than 1, and the largest risk was present for children between the age of 6–12 followed by that for children between 2 and 6 years of age, indicating that citizens in Jinan, particularly children, faced more serious potential non-carcinogenic health risks.

1. Introduction

$PM_{2.5}$ is an important pollutant in the atmosphere and proved to make greater contribution to visibility reduction (Ghim et al., 2005), incidence of respiratory diseases (Hong et al., 2002) and global climate change (Sloane et al., 1991) than larger size particle. The concentration of $PM_{2.5}$ in China is much higher than that in America and Europe (Cheng et al., 2011a) and $PM_{2.5}$ has become the principal pollutant in most urban areas in China (Zhang et al., 2009). However, the study associated with chemical characterization and source apportionment of urban $PM_{2.5}$ in China, which is important for understanding the atmospheric processes of $PM_{2.5}$, mainly conducted in super cities such as Beijing (Duan et al., 2006) and Shanghai (Ye et al., 2003), where the big factories has been relegated to the smaller cities and the particulate pollution has been lightened (China Statistical Yearbook 2009). Very limited information has been reported concerning the levels, sources and atmospheric chemical processes of $PM_{2.5}$ in the cities with rapid economic development and responding high emission intensities of air pollutants.

Since 2004, our team has been studying the $PM_{2.5}$ pollution in Shandong Province (mainly in Jinan), where the emission intensities of SO_2, NO_x and PM rank first in China and in the world (Zhang et al., 2009), due to this province's rapid economic development. Significant progress has been made in our understanding of chemical characterization of $PM_{2.5}$ in Jinan. Cheng et al. (2011a) identified the inter-annual variations of $PM_{2.5}$ and major secondary ions (NO_3^-, SO_4^{2-} and NH_4^+). Cheng et al. (2011b) studied the size-fractionated characteristics of aerosol chemical compounds. Gao et al. (2011) reported a highly time-resolved measurement of water-soluble ions in $PM_{2.5}$. Yang et al. (2012) conducted a systematic study on chemical components of $PM_{2.5}$ in Jinan. Wang et al. (2012) studied the secondary formation of inorganic aerosols in the droplet mode through heterogeneous aqueous reactions under haze conditions.

Based on the previous work, which is mainly related with the chemical characteristics of $PM_{2.5}$ and the formation mechanism of secondary aerosols, the question, then, is how much the potential health impact of $PM_{2.5}$ is and how to reduce the loading of $PM_{2.5}$, which is not only meaningful for taking proper measures to decrease $PM_{2.5}$ pollution in Jinan, but also be helpful for the

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developing country with flourishing industry, where the concentration of $PM_{2.5}$ is also very high (Mönkkönen et al., 2005).

PMF is a newly developed source apportionment model, which has been widely applied to assess $PM_{2.5}$ sources in urban regions of the United States of America (Qin et al., 2006; Kim and Hopke, 2008; Rizzo and Scheff, 2007) and Europe (Viana et al., 2008), however, its application in the heavily polluted developing country is very rare. In this paper, we apply PMF model to study $PM_{2.5}$ source apportionment and choose three highly sensitive groups: children (aged 2–6 years and 6–12 years) and older adults (70 years) to study the potential health impact of $PM_{2.5}$. This result can provide reference and enlightenment for the application of PMF and evaluating the health impact of $PM_{2.5}$ in heavily polluted developing countries.

2. Methodology

2.1. Sampling site and chemical analysis

$PM_{2.5}$ filter samples were daily collected between Dec. 2007 and Oct. 2008 by a Multiple Reference Ambient Air Sampler (16.7 l min^{-1}, RAAS2.5-400 Thermo Electron Corporation, USA) in the campus of Shandong University. The number of sampling was 37, 3, 17, 18, 17, 29, 20, 15 in December 2007, January, February, March, April, June, September and October in 2008 respectively. A detailed description of $PM_{2.5}$ sampling site, process and chemical analysis of major water-soluble ions (SO_4^{2-}, NO_3^- and NH_4^+), OC/EC and trace elements can be found in Yang et al. (2012).

2.2. PMF analysis and elemental risk calculations

EPA PMF3.0 was utilized to apportion contributions to the ambient $PM_{2.5}$ levels from the major emission sources in Jinan, Shandong Province. The detailed information on principles and usages of the model can be found in the user manual (USEPA, 2008) and related literature (Paatero, 1997).

It is important to combine source identification and elemental risk evaluations. In this study, representative elemental components of $PM_{2.5}$ were applied to calculate elemental risk. According to the method described by the US EPA (1989), several steps were used to develop the risk evaluation for a special compound, including calculations for 95% upper confidence limit (UCL) of the arithmetic mean concentration, due to the uncertainty associated with the estimated true average concentrations, evaluating the exposure, and calculating the risk.

3. Results and discussion

3.1. Model evaluation

In this study, 156 samples with 26 species and a signal-to-noise ratio greater than 0.2 were considered for the input, and PMF was run multiple times while varying the number of sources, bootstrap and fpeak, which affects the rotations of matrices for source compositions and contributions. Six sources were resolved with a resultant fpeak of 0.1, while Qrobust (3977.0) and Qtrue (4076.3) were almost equal to Qtheory (156 * 26 = 4056), were physically reasonable and could be explained by the potential sources in Jinan (USEPA, 2008). A comparison of the daily calculated $PM_{2.5}$ mass concentrations from six sources with the measured mass concentrations is shown in Fig. 1. The squared correlation coefficient (R^2) of 0.98, a slope of 1 and an intercept of 0 μg m^{-3} indicated that the reconstructed mass concentrations were consistent with the measured mass concentrations, and the resolved six sources

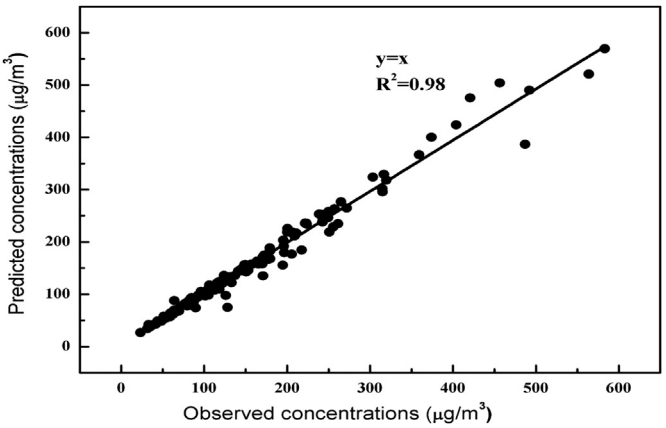

Fig. 1. Predicted versus observed $PM_{2.5}$ mass concentrations in Jinan.

effectively accounted for most of the variability of $PM_{2.5}$ mass concentrations.

3.2. Source apportionment

The six identified source profiles and the time series plots of the estimated daily contribution from each source for $PM_{2.5}$ are described in Figs. 2 and 3, respectively. Table 1 represents the annual and seasonal variations of predicted contribution to $PM_{2.5}$ from each source.

Secondary sulfate and nitrate was characterized by the highest loading of sulfate, ammonium, OC and high nitrate concentrations. Secondary source is the major contributor, accounting 55.15% of $PM_{2.5}$, indicating that an enhanced secondary conversion may exist in the atmosphere of the heavily polluted area. The secondary source contribution in Jinan is similar with that in urban site of New York (Qin et al., 2006), Hong Kong (Lee et al., 1999) and Chicago (Rizzo and Scheff, 2007), while higher than that in urban site of Seattle (Kim and Hopke, 2008) and Beijing (Song et al., 2006). Table 1 clearly shows that the secondary source in Summer (69.37%) was much higher than that in spring (39.79%), autumn (64.78%) and winter (51.44%). In summer, high temperatures, relative humidity and high solar radiation favour the formation of secondary sulfate (Seinfeld, 1986). Secondary ammonium nitrate is semi-volatile at high temperatures and the concentration of secondary nitrate in summer was lower (Seinfeld, 1986). Therefore, in summer, the secondary source was primarily secondary sulfate and secondary OC.

A factor which could be attributed to coal combustion had high concentrations of Cl, Br, Pb, As, Mn and Cu mixing with medium concentrations of OC, SO_4^{2-}, NO_3^- and Fe. High concentration of Cl was also probably associated with coal burning (Song et al., 2006). Coal is still a primary energy sources in Shandong Province. In 2007, coal consumption was 3.9×10^7 t, which constituted 79.4% of the total energy and is greater than the ratio in Beijing (47.5%). Continued increases in coal consumption and inadequate pollution control devices during burning led to the increase of particulate matter concentrations and coal burning contributions. The annual mean burning contribution was 20.98%, which is slightly higher than that in Beijing (19%) (Song et al., 2006). Seasonal variation of coal burning contribution indicated that peak contribution appeared in the heating period (December 31.41%, February 22.86% and March 33.03%), followed by autumn (12.93%) and the lowest contribution was observed in summer (9.37%), as expected.

The soil dust source was identified by high concentrations of crustal elements Al, Si, Ca, Mg, and Ti, representing wind-blown crust dust and re-suspended road dust. The annual mean soil dust source contribution to $PM_{2.5}$ was 9.30%, similar to the PMF results in

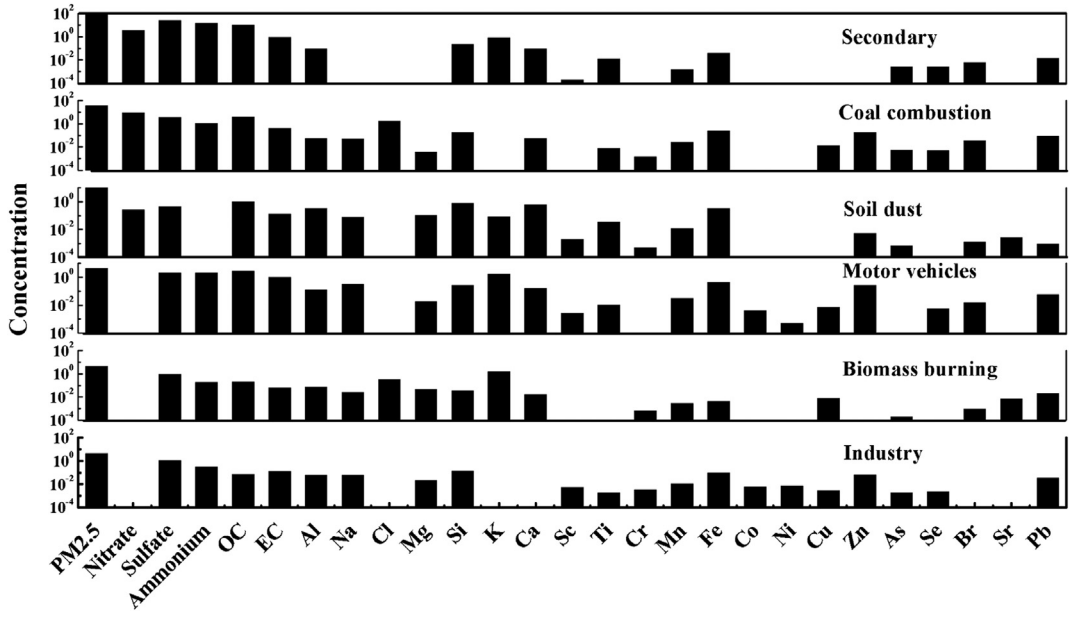

Fig. 2. Source profiles deduced from the PMF analysis of $PM_{2.5}$ for secondary, coal combustion, soil dust, motor vehicles, biomass burning and industry.

Beijing (Song et al., 2006). The highest soil dust contribution appeared in March (21.42%), which could be due to frequent sandstorms from Saharan dust based on the backward trajectory analysis (Cheng et al., 2011a). Fig. 3 shows a sharp increasing value of soil contributions to $PM_{2.5}$ on March 1–2, 2008, which was due to large dust storm sweeping across eastern China during this period. The high contribution of soil dust in February (6.99%) may be related with higher wind speed (2.81 m s^{-1}), which ranked second to that in March (3.00 m s^{-1}) and could lead to stronger soil dust.

The motor vehicle source was represented by high concentrations of EC, Pb and Fe, as well as medium concentrations of OC, Cu, Zn, K and Mn. EC primarily arose from engines; Cu, Fe, Mn and Pb typically exist in brake wear dust, and the Pb is present in motor and fuel oil combustion; Fe, Zn and K are in tailpipe emissions (Begum et al., 2004; Karnae and John, 2011). The annual mean mass contribution of motor vehicles was 6.06%, which closely approximates the 7% derived by Zheng et al. (2005) in Beijing. Seasonal trends of contributions in Fig. 3 depicted higher contributions of motor vehicle source in June (8.33%) and September (8.45%), which may be due to the use of air-conditioner in car and gave rise to the exhaust emission.

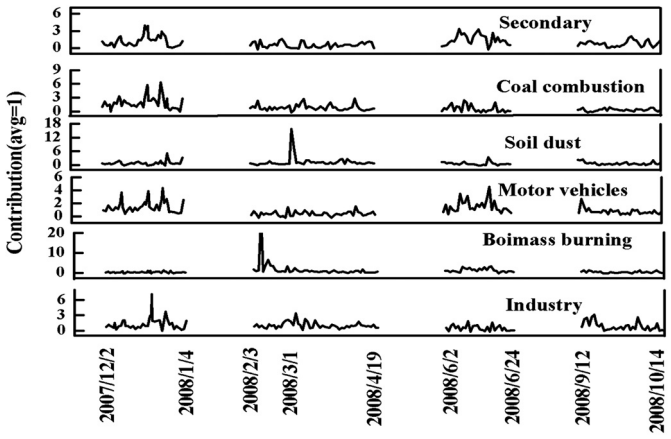

Fig. 3. Daily source contributions deduced from the PMF analysis of $PM_{2.5}$ for secondary, coal combustion, soil dust, motor vehicles, biomass burning and industry.

Biomass burning was composed of high mass fractions of K and Sr, mixing with medium loading of Cu, Mg and Pb. High loading of K primarily came from stalk burning, and the incineration of sludge also emitted K, Pb, Cu, As and Cr (Moon et al., 2008). Agriculture cultivation in suburban areas near Jinan is prevalent, and farmers burn stalk to fertilize the soil in summer. As expected, the contribution of biomass burning was high in June (4.83%). The highest contribution of biomass burning happened in February, 2008 (16.94%), which was intimately associated with the very largely setting off firecrackers for celebrating Chinese New Year (Wang et al., 2007).

The factor characterized by high loading of Ni, Cr, Co, and Sc mixing with other element species, such as Zn, Pb and Fe, appeared to be closely associated with industry sources. Ni and Cr are typically used as indicators of residual fuel oil combustion, which occurs in oil-fired power plants and steam boilers (Karnae and John, 2011). This source indicated that in the northeastern region of this site, residual oil remained a common fuel for industrial sources. Zn, Fe and Pb arise primarily from the steel mill located northeastern Jinan. The annual mean industry source contribution was small (2.87%), and the analyses showed larger source contributions in spring and autumn.

3.3. Health risk estimated from chemical components of $PM_{2.5}$

Based on the source profiles for elemental components of $PM_{2.5}$, eleven elements were used to assess the possible non-carcinogenic risks for public health. The calculated health risk for each element is described in Table 2.

The study showed that elemental risk larger than 0.1 had adverse health effects on children (De Miguel et al., 2007). As shown in Table 2, the non-carcinogenic elemental inhalation risk in $PM_{2.5}$ higher than 0.1 in Jinan was due to exposure to Mn, Co, S and Cr for the three groups. Higher risk values (>0.1) for children and adults were found for soil and road dust derived Mn and As (Kong et al., 2012) in Dongying and for soil derived Pb and As (Hu et al., 2011) in Nanjing. The risk of As in this study was approximately 0.01 for the three groups, which was lower than that in Nanjing (Hu et al., 2011) and higher than that in Luanda, Angola (Ferreira-Baptista and De

Table 1
Annual and seasonal source contributions to $PM_{2.5}$ mass concentrations in Jinan, China from Dec. 2007 to Oct.2008 (%).

	Coal combustion	Biomass burning	Industry	Secondary	Soil dust	Motor vehicles	Unknown
Spring	26.76 (±18.24)	4.03(±2.69)	3.69(±1.60)	39.79(±27.11)	17.72(±16.65)	3.81±(2.39)	4.21(±9.72)
Summer	9.37(±10.63)	4.83(±4.88)	1.37(±2.04)	69.37(±25.47)	6.12(±10.67)	8.33(±10.20)	0.61(±10.86)
Autumn	12.93(±12.57)	2.66(±2.80)	3.19(±2.49)	64.78(±19.95)	8.31(±6.52)	7.04(±3.92)	1.09(±5.44)
Winter	28.27(±13.05)	5.88(±13.83)	2.95(±1.78)	51.44(±20.49)	6.37(±6.68)	5.70(±3.37)	–
Annual	20.98(±16.02)	4.55(±8.85)	2.87(±2.10)	55.15(±25.19)	9.30(±11.32)	6.06(±5.47)	1.08(±7.86)

Table 2
Health risk estimated from chemical elemental components of $PM_{2.5}$.

Elements	Risk		
	Children (2–6)	Children (6–12)	Adult
S	2.65E−01	2.69E−01	2.01E−01
V	1.37E−04	1.39E−04	1.04E−04
Cr	1.68E−01	1.71E−01	1.28E−01
Mn	4.81E−01	4.89E−01	3.65E−01
Ni	9.27E−05	9.41E−05	7.04E−05
Cu	1.66E−04	1.69E−04	1.26E−04
Zn	3.78E−04	3.84E−04	2.87E−04
Pb	1.40E−02	1.42E−02	1.06E−02
As	8.57E−03	8.70E−03	6.51E−03
Cd	1.57E−03	1.59E−03	1.19E−03
Co	4.63E−01	4.70E−01	3.51E−01
Total	1.40E+00	1.42E+00	1.06E+00

Miguel, 2005). The respiratory intake of As might increase the chance of lung cancer mortality, as well as skin and several internal organ cancers, through ingestion (USEPA, 2005), therefore, the risk of As cannot be ignored. The lowest risk element was Ni with the value of 9.41E−5, which fell into the acceptable risk range (1.0E−6 ~ 1.0E−4) (Hu et al., 2011). The estimated risk of Pb in $PM_{2.5}$ was 0.0142, lower than that in Nanjing, and Mexico (Hu et al., 2011; Díaz and Rosa Dominguez, 2009), implying that the concentrations of Pb in the atmosphere in Jinan were relatively lower. The elemental risk according to the impact of heavy metals on health could be cumulative, and the total elemental risk for the three groups was larger than 1 with a value of 1.40 for children of 2–6 years, 1.42 for children of 6–12 years, and a lower value (1.06) for adults, indicating that the combination of elements in $PM_{2.5}$ may pose more serious public health risk. In addition, children were the most sensitive group to develop non-carcinogenic effects, and avoiding possible exposure to these contaminant elements is important. Therefore, it is critical for the government to adopt effective measures to control $PM_{2.5}$ pollution and to improve urban air quality.

4. Summary

The PMF model was successfully conducted to apportion $PM_{2.5}$ from different sources in Jinan, which is a highly polluted city. The PMF model annually resolved 98.91% of $PM_{2.5}$ in Jinan between December 2007 and October 2008, and the six potential source contributions, in descending order, were: secondary (55.15%) > coal combustion (20.98%) > soil dust (9.30%) > motor vehicles (6.06%) > biomass burning (4.55%) > industry (2.87%).

The results of the non-carcinogenic risk assessment indicated that the single elements of Mn, Co, S and Cr gave rise to potential health risks with values higher than 0.1 for the three groups. The combination of all the elemental risks in Jinan for these three groups was higher than 1; furthermore, the risk for children between the ages of 6 and 12 years were largest suggesting that the children in Jinan, were faced with more serious potential non-carcinogenic health risks.

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