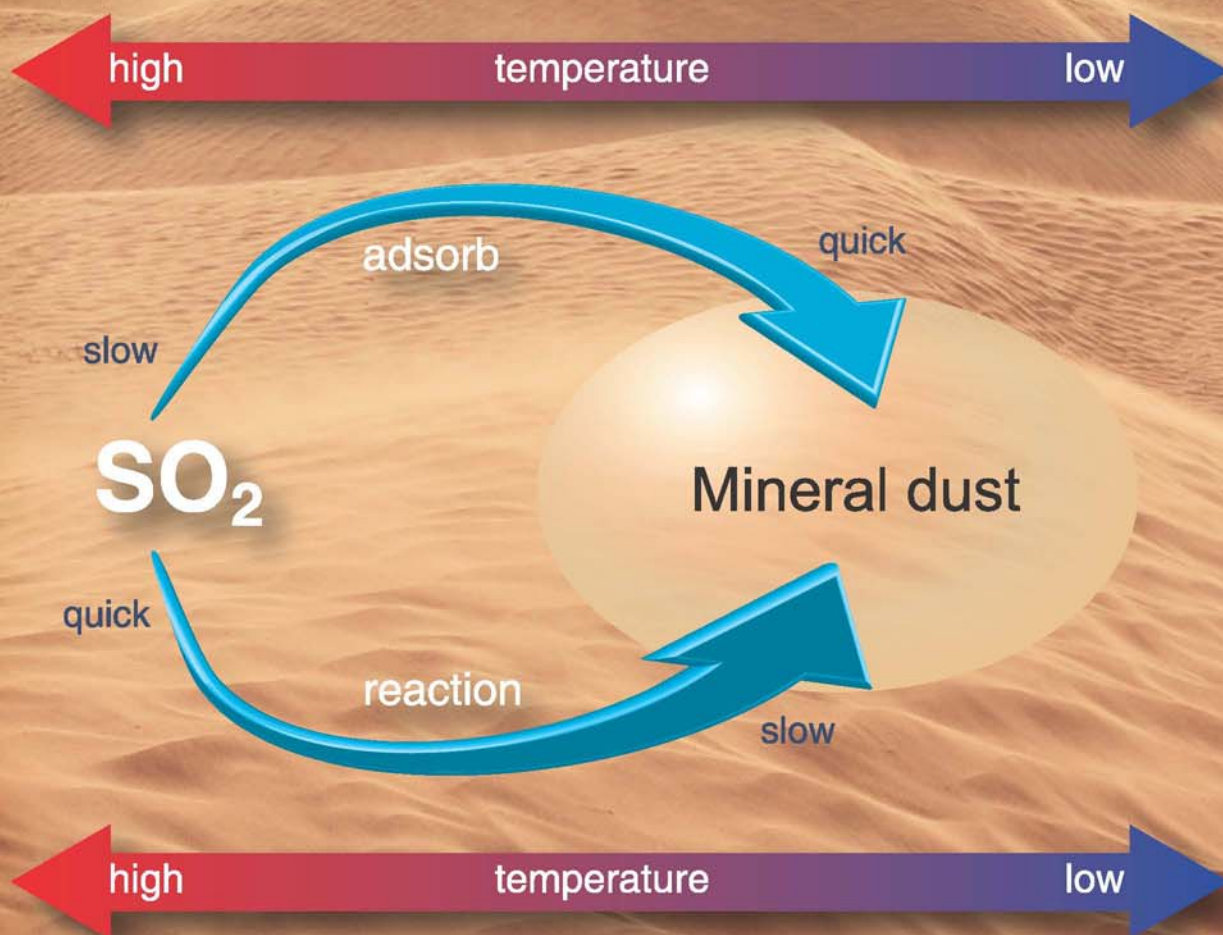


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Chemical characteristics and source apportionment of atmospheric particles during heating period in Harbin, China

Likun Huang^{1,2,*}, Guangzhi Wang^{2,*}

1. School of Food Engineering, Harbin University of Commerce, Harbin 150076, China. E-mail: hk1980@163.com

2. State Key Laboratory of Urban Water Resource and Environment, Harbin Institute of Technology, Harbin 150090, China

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ABSTRACT

Atmospheric particles (total suspended particles (TSPs); particulate matter (PM) with particle size below 10 μm , PM_{10} ; particulate matter with particle size below 2.5 μm , $\text{PM}_{2.5}$) were collected and analyzed during heating and non-heating periods in Harbin. The sources of PM_{10} and $\text{PM}_{2.5}$ were identified by the chemical mass balance (CMB) receptor model. Results indicated that $\text{PM}_{2.5}/\text{TSP}$ was the most prevalent and $\text{PM}_{2.5}$ was the main component of PM_{10} , while the presence of PM_{10-100} was relatively weak. SO_4^{2-} and NO_3^- concentrations were more significant than other ions during the heating period. As compared with the non-heating period, Mn, Ni, Pb, S, Si, Ti, Zn, As, Ba, Cd, Cr, Fe and K were relatively higher during the heating period. In particular, Mn, Ni, S, Si, Ti, Zn and As in $\text{PM}_{2.5}$ were obviously higher during the heating period. Organic carbon (OC) in the heating period was 2–5 times higher than in the non-heating period. Elemental carbon (EC) did not change much. OC/EC ratios were 8–11 during the heating period, which was much higher than in other Chinese cities (OC/EC: 4–6). Results from the CMB indicated that 11 pollution sources were identified, of which traffic, coal combustion, secondary sulfate, secondary nitrate, and secondary organic carbon made the greatest contribution. Before the heating period, dust and petrochemical industry made a larger contribution. In the heating period, coal combustion and secondary sulfate were higher. After the heating period, dust and petrochemical industry were higher. Some hazardous components in $\text{PM}_{2.5}$ were higher than in PM_{10} , because $\text{PM}_{2.5}$ has a higher ability to absorb toxic substances. Thus $\text{PM}_{2.5}$ pollution is more significant regarding human health effects in the heating period.

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Introduction

Atmospheric particulate pollution is becoming more and more serious. Many countries and cities have experienced serious ash haze and fog haze weather. This has a significant influence on the urban atmospheric environment and human health, which has received increasing attention in many countries (Godec et al., 2012; Hansen et al., 2010; Kleeman et al., 2009; Tiwari et al., 2012).

Fuel combustion is an important source of atmospheric particulates. Heating processes using coal combustion make a

sizable contribution to particulate matter (PM) concentrations, so particulate matter characteristics during the heating period are very different from those during the non-heating period. In addition, due to the presence of a concave terrain, lower wind speeds, and a relatively stable atmospheric structure in winter, it is difficult for atmospheric particulate matter to disperse, and it accumulates gradually, so that the characteristics of atmospheric particulate matter in the heating period is different from that during the non-heating period (Braniš and ělová, 2010; Kong et al., 2012; Qiao et al., 2010; Wang et al., 2012).

* Corresponding authors. E-mail addresses: hk1980@163.com (Likun Huang)

In several major world cities, the characteristics and source apportionment of atmospheric particulate matter during the heating period and the non-heating period have been studied. However, there are differences in air quality characteristics between other cities and Harbin due to its high latitude, cold climate, and frequent inversion phenomena in the winter season (Dong et al., 2012; Dai et al., 2013; Li et al., 2012; Xu et al., 2012; Shen et al., 2011; Zhang et al., 2012). No detailed investigation has been conducted in Harbin thus far. Accordingly, in order to improve atmospheric quality, protect human health, and reduce ecological damage, it is crucial to investigate the characteristics of particulate matter during the heating period and the non-heating period in order to develop effective air pollution control measures in Harbin. Hence, we collected atmospheric particulate matter (TSP, PM₁₀, and PM_{2.5}) in Harbin in 2010 and 2011 to analyze the chemical characteristics of the particles and further to apportion their main sources by using chemical analysis and the chemical mass balance (CMB) receptor model (Gummeneni et al., 2011).

1. Sampling and analytical methods

1.1. Sample collection

In order to investigate the impacts of heating on ambient air quality, monthly atmospheric particulate matter data, including TSP, PM₁₀, and PM_{2.5}, were continuously collected from August, 2010 to June, 2011. According to the heating period of Harbin, the sample collection was conducted in three stages including “before heating period” from Aug. 2010 to Sep. 2010, “heating period” from Oct. 2010 to Apr. 2011, and “after heating period” from May 2011 to Jun. 2011. The sampling site was located at the South Campus of Harbin University of Commerce as illustrated in Fig. 1. Samples were collected every day, except raining days, snowing days, dust days and other special days. After these abnormal days, sampling was also stopped for one day, because particulate matter characteristics were affected by the weather. At least 15 samples were selected every month for a subsequent chemical and physical analysis. The filters used in this study were 80 mm quartz filters that were initially heated at 900°C before sampling, in order to eliminate the interference of residual organic compounds. Three medium-flow air samplers (Tianhong Intelligent Instrument, Model TH-150, Wuhan, China) were applied to collect TSP, PM₁₀, and PM_{2.5} with the

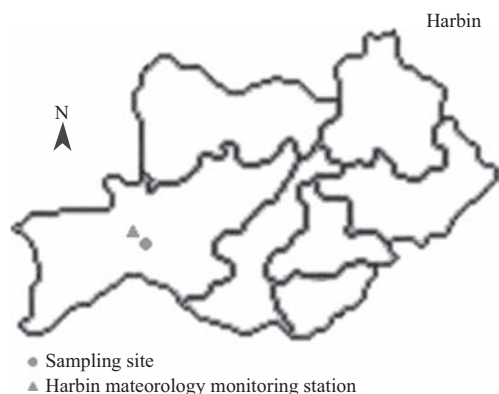


Fig. 1 – Locations of sampling site and meteorology monitoring station in Harbin, China.

same flow rate of 100 L/min and sampling time of 24 hr every time. Overall sampling volume and duration were automatically recorded. The sampling height was 8 m above the ground. Near the sampling site, there was no substantial pollution source or obvious obstacles. The sampling approach was consistent with the requirements of national standards.

1.2. Chemical analysis

1.2.1. Ionic species

Before and after sampling, the quartz filters were temporarily stored at 4°C and then transported back to the Central Laboratory of Harbin Institute of Technology (HIT) for further conditioning, weighing, and chemical analysis. Before conducting the chemical analysis, the filter samples were initially cut into 8 identical parts, four parts of which were used for the analysis of metallic elements, ionic species, total carbon and elemental carbon, respectively, while the remaining parts were used for other chemical analysis (Tsai et al., 2006).

A one eighth segment of the quartz filter to be analyzed for ionic species was put into a 50-mL PE bottle for each sample. Distilled-de-ionized water was added into each bottle for ultrasonic vibration for approximately 120 min. An ion chromatography system (Dionex, Model 100, USA) was used to analyze the concentration of major anions (F⁻, Cl⁻, NO₃⁻, and SO₄²⁻) and cations (Na⁺, NH₄⁺, K⁺, Mg²⁺, and Ca²⁺) (Yuan et al., 2004). Method detection limits were obtained from duplicate analysis of predefined quality control solutions. The method detection limits of F⁻, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH₄⁺, K⁺, Mg²⁺, and Ca²⁺ were 0.013, 0.024, 0.033, 0.021, 0.017, 0.04, 0.039, 0.031, and 0.033 µg/mL, respectively. The averaged recovery efficiencies ranged from the lowest of 90% for Na⁺ to the highest of 110% for Cl⁻ with an overall average of 97% (Huang et al., 2011).

1.2.2. Metallic elements

One eighth of the quartz filter was soaked in 15 mL mixed acid (V_{HNO₃}:V_{HClO₄} = 3:7) and placed on an electrical heating plate at 150–200°C for at least 2-hr digestion until the solution boiled and clarified. During the digestion period, distilled-de-ionized water was added into the residual solution twice or more in order to completely dissolve the metallic elements. The residual solution was then diluted to 50 mL with 0.5 mol/L HNO_{3(aq)} and stored in a polyethylene (PE) bottle. After conducting the above steps, the metallic contents were measured with an inductively coupled plasma-atomic emission spectrometer (ICP-AES, Perkins Elmer, Model 400, USA) (Yuan et al., 2004). The metallic elements analyzed for this study included Al, As, Ba, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, S, Si, Sr, Ti, V, and Zn, with the detection limits of 0.050, 0.014, 0.002, 0.028, 0.002, 0.007, 0.007, 0.009, 0.050, 0.029, 0.016, 0.030, 0.004, 0.030, 9.000, 1.500, 0.010, 0.009, 0.200, and 0.004 µg/mL, respectively.

1.2.3. Carbonaceous contents

Total and elemental carbon (TC and EC) contents of each quartz filter were determined with an elemental analyzer (EA, Fison, Model CHNS/O 1108, Italy). One eighth of each filter sample was heated in advance in a 340°C oven for 100 min to expel the organic carbon (OC) content, and then fed into the elemental analyzer to obtain the elemental carbon (EC) content. Another one eighth of each filter was fed directly into the

elemental analyzer without pre-heating to obtain the total carbon (TC) content (Lin, 2002). Organic carbon could then be determined by subtracting the elemental carbon from the total carbon.

1.3. Scanning electron microscopy

The scanning electron microscope (SEM) uses a focused beam of high-energy electrons to generate a variety of signals at the surface of solid specimens. The signals that derive from electron-sample interactions reveal information about the sample, including external morphology (texture), chemical composition, and crystalline structure and orientation of materials making up the sample. In most applications, data are collected over a selected area of the surface of the sample, and a 2-dimensional image is generated that displays spatial variations in these properties. Areas ranging from approximately 1 cm to 5 μm in width can be imaged in a scanning mode using conventional SEM techniques (magnification ranging from 20 \times to approximately 30,000 \times , spatial resolution of 50 to 100 nm).

1.4. Source apportionment

In this study, the software CMB8.2 based on CMB was used to estimate the contribution of emission sources by determining the best-fit combination of emission source and chemical composition profiles based on reconstructing the chemical composition of ambient samples. The basic calculation principle of model is as follows:

$$C_i = F_{i1}S_1 + F_{i2}S_2 + \dots + F_{ij}S_j + \dots + F_{iJ}S_J \quad (i = 1 \dots I; j = 1 \dots J) \quad (1)$$

where, C_i ($\mu\text{g}/\text{m}^3$) is the concentration of element i measured at the receptor site of atmospheric particulate matter; F_{ij} ($\mu\text{g}/\text{m}^3$) is the concentration of element i measured at the source j of emitted particulate matter; S_j is the contribution ratio of particulate matter emitted from source j ; I is the number of elements; and J is the number of emission sources.

When I is greater than J , a group of results can be obtained by using the equation based on the effective variance and the least squares, which results in the contribution ratio of each source. The model needs the data from the chemical profile of emission sources and the elemental concentrations of receptor samples. Based on these data the model can calculate the apportionment of each source (Huang et al., 2011; Marcazzan et al., 2003).

2. Results and discussion

2.1. Variation of atmospheric particulate matter concentrations during sampling period

Because of the low air temperature during winter in the north of China, heating by coal burning is necessary to increase the indoor temperature; coal combustion results in a significant increase in particulate concentrations. In this study, according to the heating period in Harbin, the year is divided into three periods, including the before heating period (August, September

and early October), the heating period (late October, November, December, January, February, March, and early May) and the after heating period (late April, May, June). Average particulate matter concentrations of these periods are shown in Fig. 2.

As shown in Fig. 2, concentrations of TSP, PM_{10} and $\text{PM}_{2.5}$ in the heating period were 201.62, 155.05 and 107.20 $\mu\text{g}/\text{m}^3$, respectively. These were 1.5–2 times higher than those of the non-heating period, especially mass concentrations of $\text{PM}_{2.5}$. Fine particles were primarily from coal combustion, which is related to the properties and combustion conditions of coal. In addition, the particulate concentration of the Before Heating Period was slightly higher than the After Heating Period due to the different seasonal environment factors. During the Before Heating Period, Heating Period and After Heating Period, the values of $\text{PM}_{2.5}/\text{PM}_{10}$ were 0.64, 0.69 and 0.56, $\text{PM}_{2.5}/\text{TSP}$ were 0.47, 0.53 and 0.42, and $\text{PM}_{10}/\text{TSP}$ were 0.73, 0.77 and 0.74, respectively. The three ratios in the heating period were higher than during the non-heating period. Among them, $\text{PM}_{2.5}/\text{PM}_{10}$ was the most obvious. This showed that the fine particle fraction ($\text{PM}_{2.5}$) was the main component in PM_{10} during the heating period, while the increase of PM_{10-100} was relatively weak. Therefore, limiting fine particles is an effective method to reduce the particulate matter concentrations during the winter heating period.

2.2. Variation of chemical composition in atmospheric particulates during the sampling period

2.2.1. Variation of ion composition

Coal combustion releases a large amount of pollutants, which can change the chemical composition of particulate matter (Lee and Nguyen, 2013). Table 1 shows the variation of ionic composition in TSP, PM_{10} and $\text{PM}_{2.5}$ during the three periods. Analysis of data revealed that concentrations of major ions (K^+ , Mg^{2+} , F^- , Cl^- , SO_4^{2-} and NO_3^-) in the heating period were higher than those in the non-heating period. Among them, K^+ , F^- , SO_4^{2-} and NO_3^- were from fuel combustion, while K^+ came mainly from burning biomass, such as straw, in the suburbs, and F^- , SO_4^{2-} and NO_3^- were from coal combustion within the urban area. In particular, SO_4^{2-} and NO_3^- were significantly higher. This may be due to the high content of S and N in coal, which can generate SO_2 and NO_x in the air to form secondary aerosols during the heating period. Mg^{2+} and Cl^- were formed in manufacturing processes. Those were produced in higher concentrations in the heating period, which was related to industrial activity, and varied with the change of seasons because of the increasing demand for seasonal products. The concentrations of Ca^{2+} and Na^+ in the heating period were less than those in the non-heating period. The main reason was that the two ions came mainly from natural sources. PM coming from dust and soil has a much higher concentration during the non-heating period. This can be related to the seasonal weather. However, dust and soil were usually covered with snow and fallen leaves and not easily raised in wintertime with low wind speed. The concentrations of NH_4^+ after the heating period were higher than those of the other two periods. This could be related to seasonal characteristics. After the heating period, with spring coming and plants growing quickly, a large amount of NH_4^+ is released. Therefore, the concentration of NH_4^+ increased greatly at this time. Generally, the effects of fuel

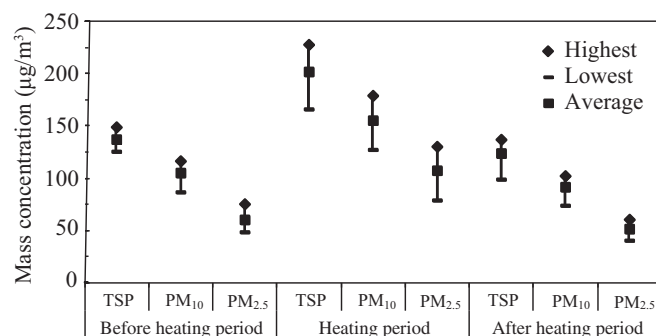


Fig. 2 – PM concentration during heating and non-heating period.

combustion on SO_4^{2-} and NO_3^- concentrations were more significant than for other ions during the heating period.

2.2.2. Variation of elemental composition

Coal combustion releases various elements that can lead to variation in the components of atmospheric particulates during the heating period, especially toxic heavy metals. As shown in Table 2, as compared with the non-heating period, Mn, Ni, Pb, S, Si, Ti, Zn, As, Ba, Cd, Cr, Fe and K had a relatively higher mass concentration in atmospheric particulates during the heating period. In particular, the high concentrations of Mn, Ni, S, Si, Ti, Zn and As in $\text{PM}_{2.5}$ were very prevalent. One reason was that $\text{PM}_{2.5}$ has a strong absorption capacity for metals; the other was that coal combustion releases a large number of fine particles with more metal content than coarse particles (Herrera et al., 2012). Moreover, concentrations of Na, Al, Ca and Mg had no variation in the heating and non-heating period, because they are the main elements in the earth's crust, and their major sources were natural emissions. Sr was found primarily in the PM_{10-100} , but the concentration was low, which suggested that Sr pollution was mainly from natural dust particles or perhaps an exogenous sandstorm. The element V barely registered in the entire sampling period, demonstrating that sources of this element's pollution did not exist in Harbin, or are released in a very low mass concentration. Cu had significant changes only in TSP, and its

concentration in the heating period was higher than in the non-heating period, while it had little change in PM_{10} and $\text{PM}_{2.5}$. The particles having Cu were large particles, and most came from fly ash, which is generated in coal burning.

2.2.3. Variation of carbonaceous compounds

The low atmospheric pressure in Harbin during winter can easily lead to hazy weather and result in the rapid accumulation and formation of organic carbon particles. In addition, organic carbon (OC), which contains a wide range of molecular forms and volatilities, can be formed during incomplete combustion of coal or through secondary chemical reactions from its gaseous precursors in the atmosphere (Rengarajan et al., 2011). So the concentration of organic carbon is higher during the winter heating period. Thus, it is necessary to study the carbon component characteristics of particulate matter in the heating and non-heating period.

Table 3 shows the characteristics of TC, EC, OC and OC/EC in TSP, PM_{10} and $\text{PM}_{2.5}$. Results showed that the mass concentration of OC in the heating period was 2–5 times higher than in the non-heating period. The mass concentration of EC in those stages

Table 1 – Ion concentration in particulate matter (PM) during heating and non-heating period (unit: $\mu\text{g}/\text{m}^3$).

	TSP			PM_{10}			$\text{PM}_{2.5}$		
	B	H	A	B	H	A	B	H	A
Ca^{2+}	9.68	3.90	7.32	7.49	2.19	5.69	1.04	0.43	0.61
K^+	1.22	1.46	0.54	0.67	1.27	0.44	0.50	1.00	0.25
Mg^{2+}	0.38	0.45	0.40	0.18	0.31	0.19	0.06	0.08	0.05
Na^+	2.34	1.98	2.83	1.36	1.43	1.90	0.30	0.28	0.36
NH_4^+	13.86	17.23	25.58	12.44	15.40	22.38	9.90	11.37	17.09
F^-	0.42	1.97	0.16	0.28	1.21	0.13	0.09	0.65	0.03
Cl^-	3.11	4.37	2.41	2.25	4.12	2.10	1.61	2.86	1.28
SO_4^{2-}	13.66	27.57	8.83	9.86	19.41	7.37	8.87	14.84	5.39
NO_3^-	11.97	24.95	9.75	9.89	17.33	8.22	7.32	13.16	5.83
Σ/PM	0.48	0.42	0.48	0.44	0.40	0.52	0.49	0.42	0.60

B: before heating period; H: heating period; A: after heating period.
TSP: total suspended particles; PM_{10} : particulate matter with particle size below 10 μm .

$\text{PM}_{2.5}$: particulate matter with particle size below 2.5 μm .

Table 2 – Elemental concentration in particulate matter (PM) during heating and non-heating period (unit: $\mu\text{g}/\text{m}^3$).

	TSP			PM_{10}			$\text{PM}_{2.5}$		
	B	H	A	B	H	A	B	H	A
Mn	0.13	0.29	0.18	0.08	0.24	0.08	0.05	0.18	0.03
Na	2.77	2.88	3.10	2.61	2.50	2.76	2.22	1.99	2.25
Ni	0.01	0.04	0.01	0.01	0.04	0.01	0.01	0.03	0.01
Pb	0.72	1.06	0.25	0.67	0.71	0.14	0.28	0.52	0.12
S	3.67	7.99	2.52	2.04	6.13	2.09	0.65	5.44	1.75
Si	2.56	5.46	0.89	1.89	3.37	0.44	0.91	2.88	0.38
Sr	0.10	0.08	0.00	0.09	0.02	0.00	0.00	0.01	0.00
Ti	0.47	0.62	1.33	0.43	0.61	0.37	0.36	0.60	0.36
V	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Zn	0.50	1.12	0.37	0.43	1.10	0.36	0.27	1.02	0.26
Al	3.69	3.46	4.48	1.88	2.01	2.07	0.44	0.57	0.48
As	0.01	0.04	0.01	0.01	0.04	0.01	0.01	0.04	0.01
Ba	0.12	0.18	0.14	0.10	0.17	0.07	0.05	0.15	0.05
Ca	11.12	8.11	9.78	9.42	7.32	6.91	4.77	4.79	4.42
Cd	0.00	0.03	0.00	0.00	0.02	0.00	0.00	0.02	0.00
Cr	0.08	0.10	0.07	0.08	0.10	0.06	0.07	0.10	0.06
Cu	0.24	0.26	0.12	0.18	0.26	0.11	0.10	0.17	0.11
Fe	3.11	4.23	2.59	2.12	3.14	2.25	0.64	1.53	0.68
K	1.43	2.23	1.44	1.33	2.18	1.09	1.30	1.91	0.59
Mg	1.50	1.84	2.14	0.99	1.23	1.24	0.74	0.85	0.77
Σ/PM	0.23	0.20	0.23	0.24	0.21	0.22	0.23	0.21	0.24

showed basically not much change. OC/EC ratios were 8–11 during the heating period in Harbin, which were much higher than other Chinese cities' data (OC/EC = 4–6), so it can be attributed to the increasing amount of coal combustion. According to the data for TC/PM, carbon was the main component that caused the increase of the particulate matter mass.

Furthermore, by comparing the proportional distribution of the carbon components in TSP, PM₁₀ and PM_{2.5}, organic carbon was richer in PM_{2.5} and OC/EC reached 10.80. This indicates that the particles formed by the secondary reactions of organic gasses were mostly fine particles, which had a higher surface area than coarse particles and stronger absorption capacity compared to coarse particles (Aldabe et al., 2011). This is the reason for the high OC concentrations.

Accordingly, OC was much higher in Harbin than in other cities due to high levels of coal combustion during the heating period, including contributing influences of coal properties, the combustion efficiency of coal, and weather patterns.

2.3. Microscopic form of atmospheric particulates by SEM

Influenced by the emission source properties, the morphology of atmosphere particulates is different. In addition, after two stages of reaction, absorption and mutual attachment, the particles' morphology is changed and differs from source samples. The micro-morphology of source samples can be clearly seen as individual particles, while receptor samples can be seen as the aggregation of particles in SEM. The micro-morphologies of PM₁₀ and PM_{2.5} in different magnifications are shown in Fig. 3.

Observing the surface morphology and distribution characters of particles by scanning electron microscopy, the pollution resources were qualitatively analyzed on the basis of sample data. Fig. 3 shows that PM₁₀ values are derived from soil dust and mineral particles from their dense texture, mostly flakes and irregular shapes, with more than a single source; PM_{2.5} values are derived from combustion particles or secondary organic particles with more rounded, smooth surfaces, and lumps or chain structures.

The surface morphology of atmospheric particles is very different during heating and non-heating periods. Coal combustion is the main way to heat the indoor air in Harbin. The amount of coal used is relatively large in December and

January. Therefore the samples in December and September were analyzed by the use of SEM to show the difference between heating and non-heating periods. The results are shown in Fig. 4.

As can be seen from Fig. 4, the characteristics of particle size, apparent morphology and aggregation state have obvious differences between the two periods. During the non-heating period, the particulates have some apparent characteristics with different shapes, dense texture, large size, light-dark contours, and clear boundary. Individual particles can be seen. Some coarse particles with irregular shape are from natural sources and some fine particles with smooth surface are from dust and organic particles. During the heating period, more floc particles can be seen by SEM, with uniform chain distribution and loosely clumped distribution. They mainly come from chemical polymerization of volatile organic compounds of small size.

2.4. Source apportionment

2.4.1. Contribution species and amount of pollution sources by CMB

In this study, we applied the CMB8.2 receptor model and source profile from Europe, the United States and Taiwan to analyze sources (Yuan et al., 2004) in order to investigate the contrast of contribution characteristics of pollution sources during the heating and non-heating periods. The results of the analysis are shown in Table 4 and Fig. 5.

As shown in Table 4, the analytical results of PM₁₀ and PM_{2.5} from the CMB suggested that the values of degrees of freedom were all over 10, the correlation coefficients were between 0.64 and 0.77, and the proportions of analysis were 80.48%–92.90%, which agreed with the analytic precision. The analysis of data revealed that there were eleven pollution sources, which were soil, dust, traffic, incinerators, coal combustion, the cement industry, the steel industry, the petrochemical industry, secondary sulfate, secondary nitrate, and secondary organic carbon respectively. As seen in Table 4, the contribution amounts of pollution sources were different in the two periods. For the two kinds of particles in the table, the pollution sources such as soil, dust, incinerators, the cement industry, the steel industry and the petrochemical industry were not influenced significantly by

Table 3 – Carbon component in particulate matter (PM) during heating and non-heating period.

	TSP			PM ₁₀			PM _{2.5}		
	B	H	A	B	H	A	B	H	A
TC concentration (μg/m ³)	13.34	26.95	12.00	12.41	22.95	9.52	6.69	17.70	5.48
EC concentration (μg/m ³)	2.69	2.86	3.82	2.45	2.15	2.61	1.47	1.55	1.62
OC concentration (μg/m ³)	10.65	24.10	8.18	9.96	20.80	6.90	5.22	16.15	3.86
OC/EC ratio	4.05	8.82	2.22	4.07	9.99	2.70	3.83	10.80	2.37
TC/PM ratio	0.10	0.13	0.10	0.12	0.15	0.10	0.11	0.17	0.11
EC/PM ratio	0.02	0.01	0.03	0.02	0.01	0.03	0.02	0.01	0.03
OC/PM ratio	0.08	0.12	0.07	0.10	0.14	0.07	0.09	0.16	0.08

TC: total carbon; EC: elemental carbon; OC: organic carbon.

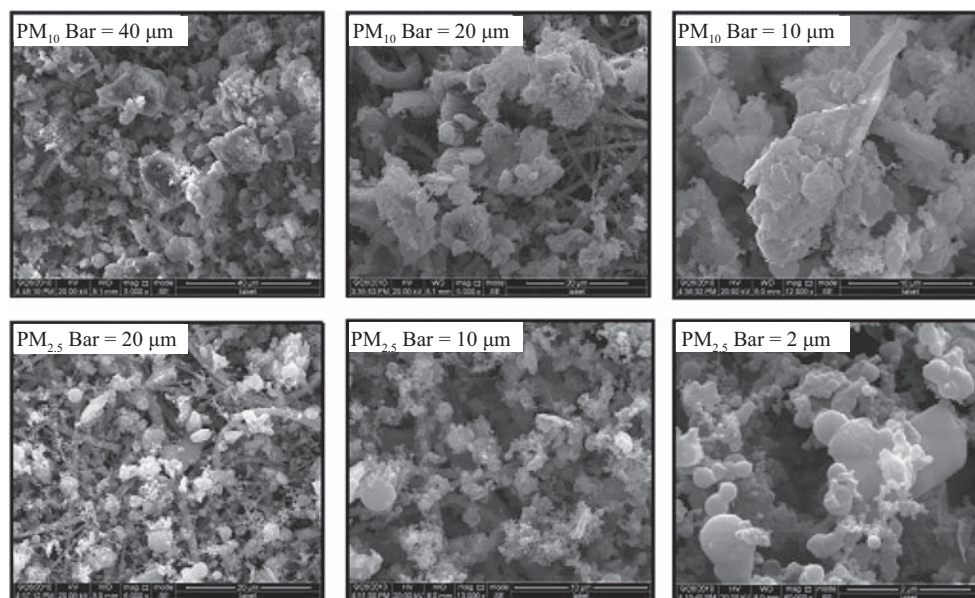


Fig. 3 – Morphological characteristics of PM_{10} and $PM_{2.5}$.

heating. However, soil and dust were greatly influenced by the seasons, and the other sources were significantly influenced by industry. Generally, when the characteristics and scale of industry were invariant, the content of pollution was relatively stable, but the release of gaseous inorganic and organic compounds may form secondary aerosol particles. On the other hand, pollution sources such as traffic, coal combustion, secondary

sulfate, secondary nitrate, and secondary organic carbon were influenced greatly by heating, so the coal combustion had the greatest effect. The contribution of traffic dust during the heating period was mainly due to low temperatures and increasing automobile emissions. At the same time, solid particulate matter and gaseous pollutants also increased; particulate matters were in the form of polymers. Secondary sulfate, secondary

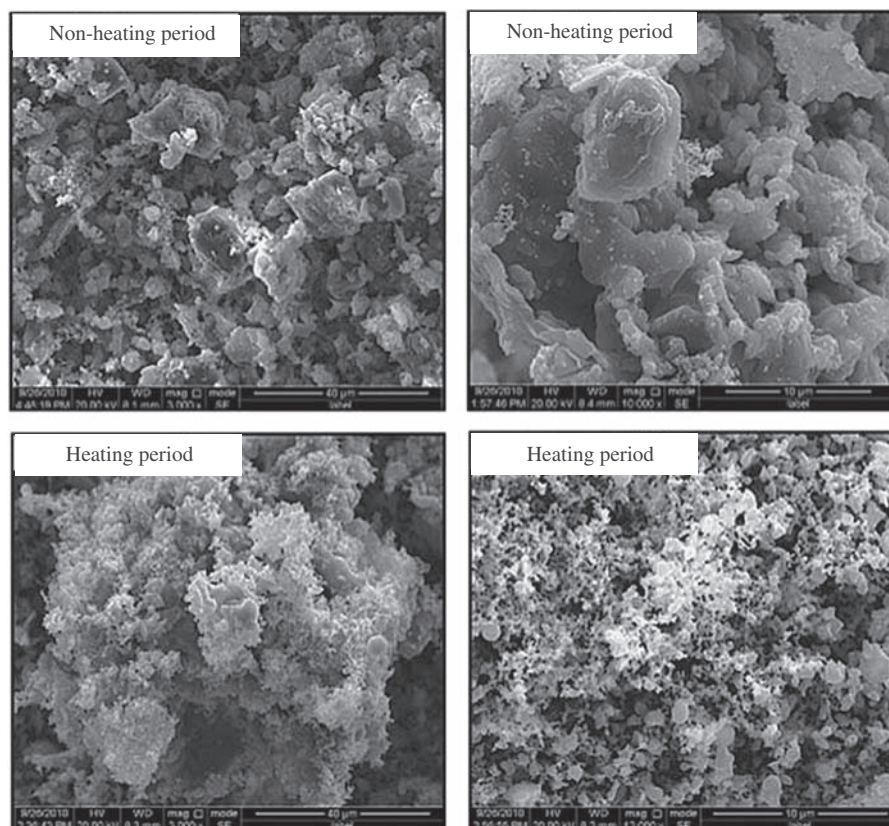


Fig. 4 – Morphological characteristics of atmospheric particles during heating and no-heating.

Table 4 – Source apportionment of particulate matter (PM) during heating and non-heating period (unit: $\mu\text{g}/\text{m}^3$).

	PM ₁₀			PM _{2.5}		
	B	H	A	B	H	A
Soil	5.32	6.14	4.01	2.15	1.98	2.68
Dust	16.25	14.52	13.15	9.13	8.65	7.25
Traffic	12.68	15.15	10.12	1.60	2.10	2.68
Incinerator	6.21	7.46	5.35	2.82	5.20	3.24
Coal combustion	11.21	30.36	10.12	6.25	24.12	7.56
Cement industry	3.12	2.84	3.85	1.64	1.38	1.88
Steel industry	4.31	5.24	3.25	2.35	3.35	1.96
Petrochemical industry	13.58	12.76	14.57	9.56	9.38	9.22
Secondary sulfate	10.14	23.18	8.32	6.97	18.95	4.51
Secondary nitrate	6.21	10.63	7.63	4.94	8.94	5.70
Secondary organic carbon	1.25	5.96	1.59	1.02	4.08	1.10
Sum of identified sources	90.28	134.23	81.96	48.44	88.14	47.77
Others	14.72	20.82	9.52	11.75	19.06	3.65
Degrees of freedom	10	11	10	13	11	12
R ² square	0.77	0.76	0.67	0.64	0.77	0.69
Mass (%)	85.98	86.57	89.59	80.48	82.22	92.90

nitrate, and secondary organic carbon were the secondary particles formed through photochemical reactions. Their gaseous precursors were SO₂, NO_x and organic gaseous compounds that

came from coal combustion, chemical industry, transportation and natural source emissions. Some secondary particles were from pollution sources directly.

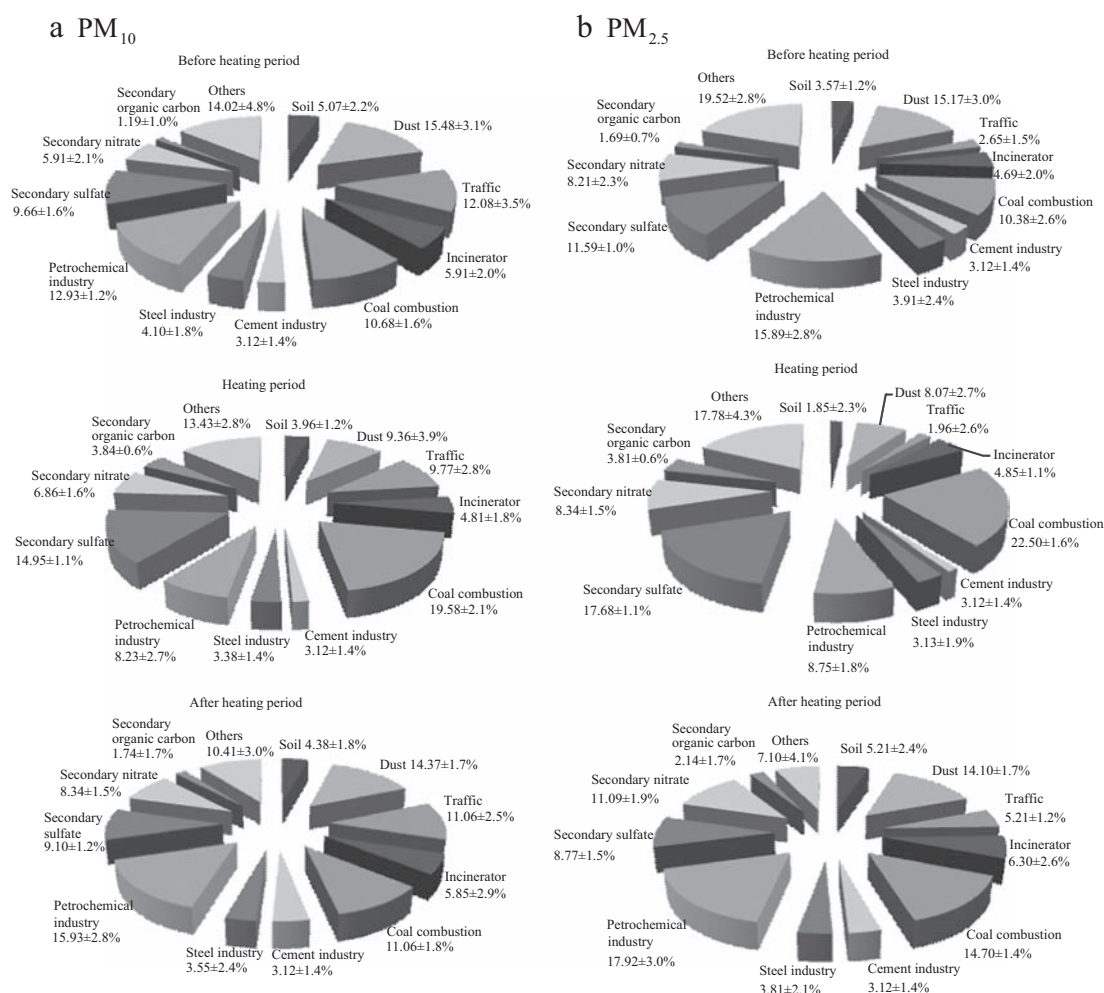


Fig. 5 – Concentration rate of sources to PM₁₀ (a) and PM_{2.5} (b) during the periods of before heating period, heating period, and after heating period.

2.4.2. Contribution ratio of pollution sources by CMB

Fig. 5 shows the contribution ratio of various pollution sources in PM_{10} and $PM_{2.5}$. As seen from the analysis of PM_{10} , before the heating period, dust and the petrochemical industry accounted for the largest percentage of contributions, followed by traffic, coal combustion, and secondary sulfate. In the heating period, coal combustion and secondary sulfate had a higher contribution ratio, followed by traffic, dust, petrochemical industry, and secondary nitrate. After the heating period, dust and the petrochemical industry had a higher contribution ratio, followed by coal combustion, traffic, and secondary nitrate.

According to the analysis of $PM_{2.5}$, dust and the petrochemical industry also accounted for a large percentage of contributions before the heating period, followed by coal combustion, secondary sulfate, and secondary nitrate. In the heating period, coal combustion and secondary sulfate had a higher contribution, followed by the petrochemical industry, dust and secondary nitrate. After the heating period, the petrochemical industry, dust and coal combustion had a larger contribution, followed by secondary nitrate, secondary sulfate and soil. Therefore, both kinds of particles had common characteristics. Coal combustion and secondary sulfate had a larger contribution in the heating period, while they had a lesser contribution in the other periods. This indicated that the amount of coal fuel has a larger effect in the heating period, and it could also have been used as a fuel in the non-heating period, but its amount was much smaller. Secondary sulfate generation had great relevance to coal combustion, so the proportion of contribution was basically identical. Moreover it was noteworthy that the proportion of contribution was relatively small for organic carbon in the entire sampling period, but compared to the non-heating period, the proportional contribution was 5 times greater in the heating period. This revealed that coal combustion or other sources associated with heating released more primary or secondary particulate matter. In the heating period, the proportional contribution of the petrochemical industry and dust was reduced. The main reason was that the primary particulate matters from the two pollution sources were not very volatile, because the decrease of their contribution ratio was caused by an increase of the released particles from other pollution sources. When the gaseous pollutants from the petrochemical industry combined with coal combustion particles, they promoted the formation of secondary particulate matter.

In addition, the contribution ratio of some hazardous pollution sources in $PM_{2.5}$ is higher than in PM_{10} , because $PM_{2.5}$ has a higher ability to absorb toxic substances. Thus $PM_{2.5}$ pollution is more significant to human health effects in the heating period.

3. Conclusions

The results of atmospheric particulate matter concentration showed that $PM_{2.5}/TSP$ was most significant. PM_{10} was primarily composed of $PM_{2.5}$ during the heating period, while the increase of PM_{10-100} was relatively weak.

Chemical analysis indicated that the effects of fuel combustion on SO_4^{2-} and NO_3^- concentrations were more serious than for other ions during the heating period. By comparison with the non-heating period, Mn, Ni, Pb, S, Si, Ti, Zn, As, Ba, Cd, Cr, Fe and K had relatively higher mass concentrations in atmospheric

particulates during the heating period, and in particular, Mn, Ni, S, Si, Ti, Zn and As in $PM_{2.5}$ were very significant. $PM_{2.5}$ had a strong absorption capacity for metals. Coal combustion released a large number of fine particles containing more metal elements than coarse particles. The mass concentration of OC in the heating period was 2–5 times higher than in the non-heating period. EC in the two periods basically did not change much. OC/EC ratios were 8–11 during the heating period in Harbin, which was much higher than the other Chinese cities' data (OC/EC = 4–6).

The results calculated by the CMB receptor model showed that eleven pollution sources were apportioned, of which traffic, coal combustion, secondary sulfate, secondary nitrate, and secondary organic carbon made the greatest contributions. Before the heating period, dust and the petrochemical industry made a larger contribution. In the heating period, coal combustion and secondary sulfate made a higher contribution. After the heating period, dust and the petrochemical industry made a higher contribution. The contribution ratio of hazardous pollution sources in $PM_{2.5}$ is higher than in PM_{10} , because $PM_{2.5}$ has a higher ability to absorb toxic substances. Thus $PM_{2.5}$ pollution is significantly more noteworthy to human health effects in the heating period.

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