Available online at www.sciencedirect.com
ScienceDirect
Journal of Environmental Sciences 2012, 24(1) 112–115
JOURNAL OF ENVIRONMENTAL SCIENCES
ISSN 1001-0742
CN 11-2629/X
www.jesc.ac.cn

Vertical characteristics and source identification of PM_{10} in Tianjin

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Abstract

Ambient PM_{10} (particulate matter with a diameter less than 10 μm) concentrations were measured on a 255 meter tower in Tianjin, China. The samples were collected at four vertical levels (10, 40, 120 and 220 m). Vertical characteristics for PM_{10} samples were studied. The results showed that the concentrations of PM_{10} and constituent species had a negative correlation with the sampling height. The highest concentrations of PM_{10} and species were obtained at the 10 m level, and the lowest concentrations were measured at the 220 m level. For the fractions of species to total mass, SO_4^{2-} and NO_3^- had higher values (fraction) at greater height; while Ca had a higher fraction at lower height. Possible source categories for the PM_{10} ambient dataset were identified by the principal component analysis method. The possible source categories included crustal dust, vehicles, cement dust, and incineration as well as secondary sulfate and nitrate sources. Analysis of meteorological factors on PM_{10} concentrations indicated that wind speed and inversion may be the main factors contributing to different concentrations of PM_{10} at different heights.

Key words: vertical characteristic; PM_{10}; sources; principal component analysis

DOI: 10.1016/S1001-0742(11)60734-1

Introduction

Particulate matter pollution has attracted much attention due to its adverse effects on visibility, human health and even the global climate (Watson, 2002; Schwartz et al., 1996; Tie et al., 2009; Nel, 2005). The mass concentration of particles as well as their chemical compositions in different areas have been monitored and studied (Nriagu, 1989; Kleeman et al., 1999; Querol et al., 2004; Turpin et al., 2000; Wang et al., 2008; Zhang et al., 2007). There have been several studies on the distribution and dynamic characteristics of atmospheric pollutants (Kim et al., 2004; Bi et al., 2005; Ding et al., 2005; Meng et al., 2008), while there were only a few studies on the vertical characteristics of particulate matter (Rubino et al., 1998; Chen and Mao, 1998; Chan and Kwok, 2000). However, the vertical variability of particulate matter is very important to the study of ambient air, because particulate matter emitted from the activity of local factories, traffic and residents, as well as regional emission, could transport, transform, dilute and deplete in the boundary layer. Therefore, understanding the vertical structure of particulate matter is important not only for the in-depth understanding of the physical and chemical processes of particulate matter, but also for the government to be able to take effective countermeasures. In this work, the vertical characteristics of PM_{10} were studied in Tianjin, China. The PM_{10} samples were collected from a meteorological tower at four sampling sites of heights: 10, 40, 120 and 220 m.

On the other hand, to understand the sources of the particulate matter, the vertical variability of the constituents of particulate matter was also studied, and possible source categories were identified using the principal component analysis (PCA) method.

1 Materials and methods

1.1 Sampling site description

Tianjin (38°34′N–40°15′N, 116°43′E–118°04′E) is a large city near the capital of China, adjacent to the Bohai Sea. The area of the city is 11200 km^2 and the population is over ten million. Tianjin is a large industrial city and the largest seaport in northern China. Its temperate monsoon climate causes four distinct seasons, with an average annual temperature of 11.4–12.9°C and annual precipitation of 520–660 mm.

PM_{10} samples were collected from a meteorological tower, set up by the Meteorological Bureau of Tianjin. The tower is located in a large residential and commercial area. PM_{10} samplers were set up at the heights of 10, 40, 120, and 220 m on the tower.

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1.2 Ambient sampling method

The samples were measured from Aug 24 to Sep 12 in 2009. In this study, concentrations of chemical species in PM_{10} were analyzed. Samples were collected for 24 hr every day using medium-volume PM_{10} samplers. Two samplers were used in parallel to collect airborne particles on quartz-fiber filters for subsequent ionic/carbon component analysis, and on polypropylene-fiber filters for elemental analysis. The sampling method was referred to our previous works (Shi et al., 2009a, 2009b).

1.3 Chemical analysis

Some elements (Si, Al, Mn, Mg, Ca, K, Cu, Zn, Pb, Cr, Co, Cd and Fe) were analyzed by ICP (IRIS Intrepid II, Thermo Electron, USA) (Baldwin et al., 1994; Watson et al., 1999). These elements were extracted from the polypropylene membrane filters using a laboratory system (ETHOS E, Milestone, Italy). The quartz fiber filters were cut into pieces for respective analysis of ionic and organic carbon (OC). Water soluble Cl^-, NO_3^- and SO_4^{2-} were extracted with an ultrasonic extraction system (AS3120, AutoScience, China) and analyzed by ion chromatography (DX-120, DIONEX, USA) (Carvalho et al., 1995; Chow and Watson, 1999). OC was analyzed with a carbon elemental analyzer (Vario EL, GmbH, Germany) (Zhao et al., 2006).

More details about sampling, treatment and analysis of sources and ambient samples were reported in our previous works (Shi et al., 2009a, 2009b).

2 Results and discussion

2.1 Concentrations and vertical characteristics

Table 1 lists the concentrations of PM_{10} for each sampling site. It can be seen that at 10 m, the highest PM_{10} concentration was obtained with a value of 140 μg/m^3; while at 220 m, the lowest concentration (80 μg/m^3) was obtained. The PM_{10} concentrations had a negative correlation with the sampling height. This is reasonable because PM_{10} is emitted from sources mainly located near the ground surface.

The average values of some abundant species concentrations in the ambient receptor for each sampling site are listed in Table 2. Similar to the PM_{10} concentrations, these species had higher concentrations at the lower height sampling sites and lower concentrations at the higher sampling sites. The species concentrations had a positive correlation with the PM_{10} concentrations.

In addition to the species concentrations, the fractional abundance of species are shown in Table 3. Unlike the case

Table 1 PM_{10} concentrations at each height

Height (m)	PM_{10} concentration (μg/m^3)
10	140
40	120
120	108
220	80

Table 2 Species concentrations at each height

Height (m)	Si (μg/m^3)	Al (μg/m^3)	Ca (μg/m^3)	OC (μg/m^3)	NO_3^- (μg/m^3)	SO_4^{2-} (μg/m^3)
10	8.51	6.32	7.07	13.45	18.03	27.40
40	6.84	5.87	5.09	10.83	16.90	26.14
120	6.39	4.93	4.56	9.59	18.88	25.33
220	4.91	4.01	2.51	7.30	13.27	19.65

Table 3 Species fractions at each height.

Height (m)	Si (%)	Al (%)	Ca (%)	OC (%)	NO_3^- (%)	SO_4^{2-} (%)
10	5.78	4.49	5.39	9.61	12.24	17.84
40	5.61	4.96	4.55	9.03	13.06	19.39
120	5.84	4.67	4.79	8.88	16.08	20.93
220	5.65	4.93	3.02	9.13	13.61	23.01

for species concentrations, SO_4^{2-} and NO_3^- had higher fraction at greater height; while for Ca, a higher fraction was obtained at lower height.

2.2 Source identification

The possible source categories for the PM_{10} samples were identified by PCA method. PCA is a useful statistical technique which can identify the possible source categories, and has been used widely in several cities (Thurston and Spengler, 1985; Hopke, 2003; Guo et al., 2004). PCA can extract some factors after analyzing the ambient dataset. These factors can be associated with different source categories, according to the loading values of the factors.

In this work, the samples from four heights were combined together to construct one ambient dataset. This dataset was analyzed by PCA. Yuan et al. (2006) used a similar combined dataset to determine the source categories in Hong Kong, and acceptable results were obtained. Therefore in this study, a combined dataset was also used.

Generally, varimax rotation should be used when the PCA method is performed, in order to obtain stable results. After varimax rotation, four factors were extracted. The eigenvalues of these extracted factors were all greater than 1.0. The varimax rotated factor loadings from PCA are shown in Table 4.

Table 4 Factor loadings

Species	Factor 1	Factor 2	Factor 3	Factor 4
Si	0.82	0.39	0.15	–0.02
Al	0.77	0.41	0.24	0.08
Mn	0.53	0.10	0.69	0.00
Mg	0.26	0.32	0.76	–0.14
Ca	0.24	0.35	0.80	0.01
K	0.72	0.39	0.17	0.08
Cu	0.08	0.83	0.01	–0.03
Zn	0.50	0.55	0.37	0.05
Pb	0.31	0.64	0.04	0.44
Cr	–0.06	–0.27	0.62	0.33
Co	0.12	–0.06	0.12	0.71
Cd	0.03	0.07	–0.07	0.80
Fe	0.83	0.44	0.09	–0.03
Cl^-	0.04	0.87	0.12	–0.07
NO_3^-	0.88	–0.02	0.14	0.11
SO_4^{2-}	0.85	–0.21	0.15	0.18
OC	0.33	0.79	0.24	–0.02

OC: organic carbon

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The first factor had a high loading for Si, Al, Fe, NO_3^- and SO_4^{2-}. Si, Al, Fe are the source markers for the crustal dust source, while NO_3^- and SO_4^{2-} are the source markers for secondary nitrate and sulfate sources, therefore this factor is likely to be related to mixed crustal dust and secondary sources. The second factor had a high loading for OC, which is the source marker for vehicles. Therefore factor 2 can represent the vehicle source. Factor 3 had a high loading for Ca, the source marker for the cement dust source, therefore factor 3 might be the cement dust source. Factor 4 had a high loading for Cd, therefore this source can be identified as the incineration source.

According to the discussion above, the possible source categories for the ambient PM_{10} in Tianjin might be crustal dust, vehicle, cement dust, and incineration as well as secondary sulfate and nitrate sources.

2.3 Meteorological factors

According to Fig. 1, it can be seen that three PM_{10} peak concentrations occurred on 25 August, 2 September and 9 September. The first peak and the second peak were different compared to the third one. For the first and second peak, the concentrations at different heights were close. At the third peak, however, the concentrations at different heights showed large differences. The variance of PM_{10} concentrations was related to meteorological factors such as wind speed and inversion. From the wind speed profile in Fig. 2, we can see that on the days when the first and the second peak concentrations occur, the difference in wind speed between the four different heights was not remarkable, so that mixing was good in the lower boundary layers. For the third peak, the wind speed showed large differences with height. As there was an inversion layer above 50 m, the mixing of the PM_{10} concentration was restrained. In addition, the temperatures at different heights on 25 August, 2 September and 9 September are described in Fig. 3, which might be another factor in the PM_{10} peak concentrations.

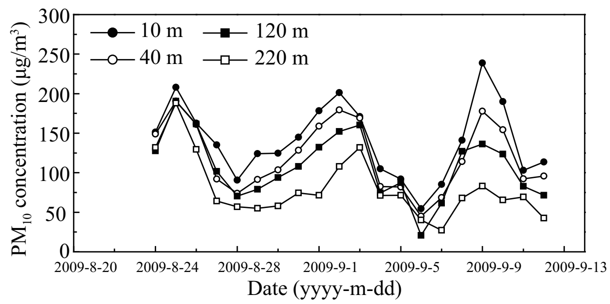

Fig. 1 Total mass concentrations of PM_{10} in different heights during sampling period.

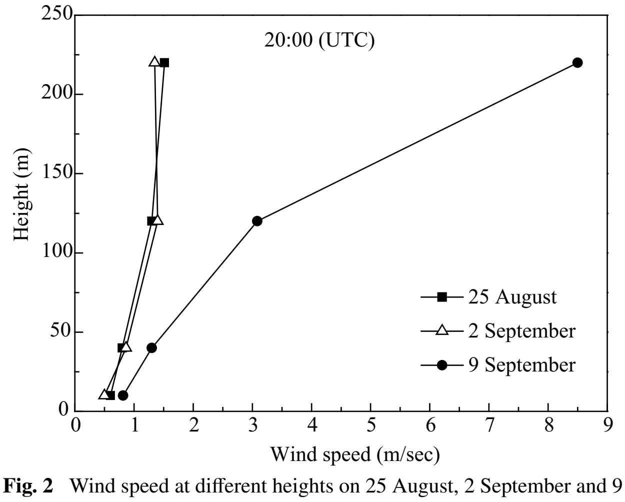

Fig. 2 Wind speed at different heights on 25 August, 2 September and 9 September.

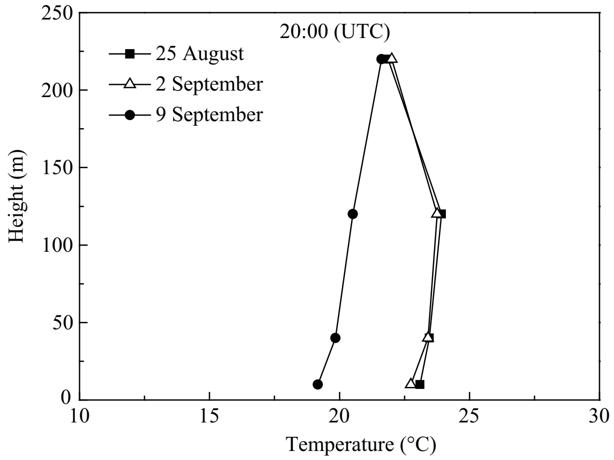

Fig. 3 Temperature at different heights on 25 August, 2 September and 9 September.

3 Conclusions

Vertical characteristics of PM_{10} concentrations were analyzed at four different heights, 10, 40, 120 and 220 m. As detailed in the discussion, the highest concentrations of PM_{10} and constituent species were obtained at the 10 m level; while the lowest concentrations were measured at the 220 m level. The PM_{10} concentrations had a negative correlation with the sampling height. However, the fractional abundance of the species showed different trends: SO_4^{2-} and NO_3^- had higher fraction at greater height; while for Ca, a higher fraction was obtained at lower height. The vertical distribution of the concentration and chemical species of PM_{10} showed remarkable characteristics, which were attributable to different sources, as well as different atmospheric physical and chemical processes for different spatial scales. Potential source categories for PM_{10} from the sampling sites were identified using PCA. Crustal dust, vehicle, cement dust, and incineration as well as secondary sulfate and nitrate sources might be the important sources. The variance of PM_{10} concentrations was also related to meteorological factors, and the results of the analysis showed that wind speed and inversion may be the main factors resulting in the difference of PM_{10} concentrations as a function of height.

Acknowledgments

This work was supported by the Key Projects in the Science & Technology Pillar Program of Tianjin (No. 09ZCGYSF02400), the Innovation Foundation of Nankai University, the Combined Laboratory of the Tianjin Meteorological Bureau and Nankai University, and the

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Fundamental Research Funds for the Central Universities.

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