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Organic carbon and elemental carbon associated with PM_{10} in Beijing during spring time

Renjian Zhang[a,b], Kin-Fai Ho[b,c,*], Junji Cao[b], Zhiwei Han[a], Meigen Zhang[d], Yan Cheng[e], Shung Cheng Lee[c]

[a] Key Laboratory of Regional Climate-Environment Research for Temperate East Asia, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
[b] SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710075, China
[c] Department of Civil and Structural Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong
[d] State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China
[e] Department of Environmental Science and Technology, School of Human Settlements and Civil Engineering, Xi'an Jiaotong University, No.28 Xianning West Road, Xi'an, Shaanxi 710049, China

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ABSTRACT

A continuous observation of organic carbon (OC) and elemental carbon (EC), and PM_{10} was conducted at an urban site of Beijing to investigate the characterization of carbonaceous aerosols during spring time. The mean value and standard deviations of OC, EC, PM_{10} concentration, and OC/EC ratio were $13.5 \pm 7.0\ \mu g\ m^{-3}$, $7.1 \pm 4.1\ \mu g\ m^{-3}$, $187.8 \pm 136.9\ \mu g\ m^{-3}$, and 2.0 ± 0.4, respectively. OC, EC, and total carbonaceous aerosols (TCA) in PM_{10} account for $9.3 \pm 5.7\%$, $4.7 \pm 2.7\%$, and $19.6 \pm 11.6\%$, respectively. Good correlations ($R^2 = 0.7$) between OC and EC were observed in spring season. Average OC concentrations are $13.5\ \mu g\ m^{-3}$ in both daytime and nighttime. Average EC concentrations in daytime ($7.4\ \mu g\ m^{-3}$) are slightly higher than those in nighttime ($6.8\ \mu g\ m^{-3}$). Both OC and EC concentrations reach maximum value in morning time (07:30–10:30) due to motor vehicles during the traffic rush hour. PM_{10}, OC, and EC concentration increase while $PM_{2.5}$ concentration, OC/EC ratio, $PM_{2.5}/PM_{10}$, and TCA/PM_{10} ratio decrease in dust period in Beijing. During the observation period, the carbonaceous aerosols from motor vehicle and coal combustion accounted for 76% and 24%, respectively. It shows that the motor vehicle represents the dominant emitter of carbonaceous aerosols associated with PM_{10} in Beijing during spring time.

1. Introduction

Carbonaceous aerosols play an important role in the atmosphere as they are involved in health effects [1–5], radiative transfer [6–9], and atmospheric chemistry. Previous study has demonstrated that they could completely prevent precipitation from clouds [10]. Meanwhile carbon can be present in ambient air as organic carbon (OC) and elemental carbon (EC), both of which are the important contributors of particulate matter (PM) in the urban atmosphere [11–15]. While OC is a complex mixture of thousands of different organic compounds, containing polycyclic aromatic hydrocarbons and other components [16–17]; EC is a mixture of graphite-like particles and light-absorbing organic matter. EC may intervene in some important chemical reactions involving atmospheric SO_2, NO*x*, O_3, and other gaseous compounds [18]. Therefore, it is obviously important to study of the physical and chemical characteristics of carbonaceous aerosols in the atmosphere.

Beijing is a city with a large population (with approximately 15 million inhabitants) and has a large number of motor vehicles (over two million), both of which consume a lot of energy. The city's energy consumption is expected to increase by 50% in 2020 as compared to the level of 1995 [19]. Although the local government tried with various means to reduce atmospheric pollution for the 2008 Olympic game in Beijing, particle pollution (particularly for PM_{10}) remains serious and difficult to control, and many current research interests take place in Beijing [17,20–27]. Duan et al. [22] identified that organic aerosols constituted up to 23% of PM_{10} in winter based on a 1.6 ratio for OC to organic matter (OM). Secondary OC was estimated at >40% and as high as 64% of total OC in summer based on a minimum OC/EC ratio of 1.5. Additionally, vertical distribution of OC and TC were determined using thermal optical reflectance (TOR) following the IMPROVE protocol on a DRI Model 2001 Thermal/Optical Carbon Analyzer [23]. OC and EC in $PM_{2.5}$ showed complex vertical distributions and distinct layered structures up the meteorological tower with elevated levels extending to the 100, 200 and 300 m heights. Moreover, dust events occur frequently in

* Corresponding author at: Department of Civil and Structural Engineering, The Hong Kong Polytechnic University, Hung Hom, Hong Kong. Tel.: +852 27664811; fax: +852 23346389.
E-mail address: cehokf@polyu.edu.hk (K.-F. Ho).

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Beijing, which results in high aerosol loading in spring time [28]. In order to study the characteristics of OC and EC in PM_{10} continuous observations were conducted in Beijing during spring time in 2004.

The purpose of this study is to determine the daily/diurnal variations of OC and EC and to identify source contribution of carbonaceous aerosols in Beijing during spring time.

2. Methodology

2.1. Observation site

Measurements of carbonaceous aerosol were conducted at an urban area of Beijing which lies between the North Third Ring Road and the North Fourth Ring Road in Beijing (39°58′N, 116°22′E) (Fig. 1). This observation site was set up to study the particulate and gaseous pollutants [28]. The instrumentations used in this study were installed on the roof of a two-storey building (10 m above ground). A meteorological tower, 50 m to the north of this observation site, was also used to collect data on wind speed and direction.

2.2. Equipment

2.2.1. PM_{10} mass concentration

The Tapered Element Oscillating Microbalance (TEOM) 1400a series, manufactured by Rupprecht and Patashnick was used for the continuous measurement of PM_{10} mass concentration. It is a US EPA equivalent method providing PM_{10} or $PM_{2.5}$ mass concentrations with a flow rate of 16.7 l min^{-1} [29]. TEOM is widely used in different aerosol studies [30–32]: it has a high accuracy with $\pm 1.5\ \mu g\ m^{-3}$ and high resolution with $0.1\ \mu g\ m^{-3}$. However, TEOM records slightly lower concentration values than those determined by reference samplers due to the heated inlet.

2.2.2. Continuous observation of organic and element concentrations

The concentrations of particulate EC were continuously detected by a thermal analysis method using ambient carbon particulate monitors (ACPM), Model 5400 manufactured by Rupprecht & Patashnick Co. Inc. This instrument performs a thermal evolution analysis of carbonaceous material [31,33]. Sampling interval was set to 3 h in the exemplary case presented here. The sample was collected on impactors instead of filters to reduce positive artifacts due to adsorption of organic vapor. Analysis of the samples is performed in a closed cycle, which includes the impactor, a NDIR CO_2 sensor, and an afterburner, which heats the gas stream to 750 °C to burn all remaining material. The amounts of carbonaceous substances evolved at 280 and 340 °C were defined as organic carbon (OC) and 750 °C as total carbon (TC), and the difference between the two is the EC. Detailed description of this instrument has been given in the previous studies [31,34–36]. Meanwhile R&P 5400 has a high resolution of $0.25\ \mu g\ m^{-3}$ and it was calibrated by Beijing Branch of Rupprecht & Patashnick Co. Inc. before it was deployed. The thermal method used by the R&P 5400 is one of the major techniques for carbonaceous aerosols measurements [1]. However, it suffers from negative artifact problems caused both by the missing collection of particles smaller than 0.14 μm and by the evaporation of the organic gases [34]. For detailed description of R&P 5400 instruments see Duan et al. [22] and Zhang et al. [37].

The data of PM_{10}, OC, EC and wind were collected during the observation period which was 15 March to 13 April 2004. The PM_{10} concentrations are 1 h averages, while OC/EC concentrations are 3 h averages. An automatic meteorological observation tower was used to observe the temperature (T-IAP-3, Institute of Atmospheric Physics), humidity (RH-IAP-3, Institute of Atmospheric Physics), and the speed and direction of wind (EC9-1, Changchun Weather Instrument Research Institute) [28].

3. Results and discussion

3.1. Concentration of OC, EC, and PM_{10}

The annual emission rates of PM_{10} over North China Plain estimated for the year 2006 were shown in Fig. 2, which are given at http://www.cgrer.uiowa.edu/EMISSION_DATA_new/data/intex-b_emissions/. These include emissions from power plants, industry, residential biofuel/fossil fuel combustion, and transportation. PM_{10} emission from the south of Beijing area was high.

Table 1 gives the average concentrations of OC and EC determined in this study and the results are compared with other studies. The OC concentration in PM_{10} ranged from 3.4 to $37.8\ \mu g\ m^{-3}$ with average concentration of $13.5 \pm 7.0\ \mu g\ m^{-3}$ while the EC concentration in PM_{10} ranged from 1.4 to $30.3\ \mu g\ m^{-3}$ with average concentration of $7.1 \pm 4.1\ \mu g\ m^{-3}$. As Beijing has switched from residential coal usage to natural gas or central steam, lower levels of OC and EC were observed when compared to other northern cities in China [37]. The concentrations of OC and EC during winter were 1.6 and 1.3 times higher than those during spring, respectively. Low mixing height and low precipitation during winter contributed to the high carbon loading during cold season. As summarized in

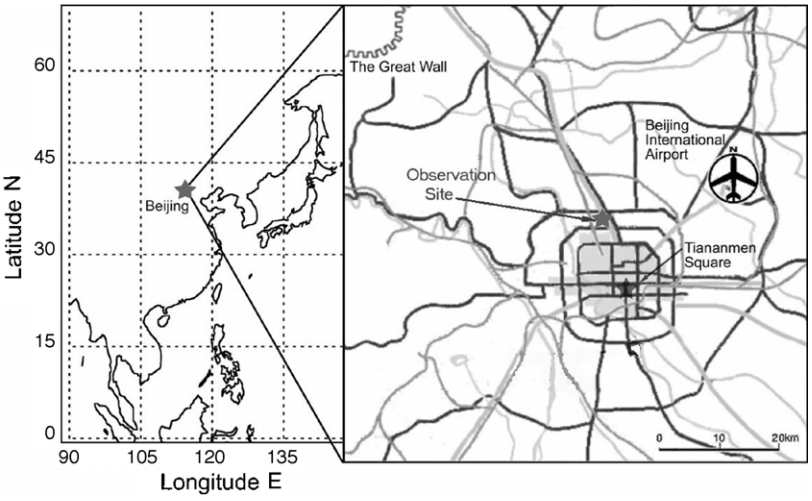

Fig. 1. Map of the sampling location in Beijing.

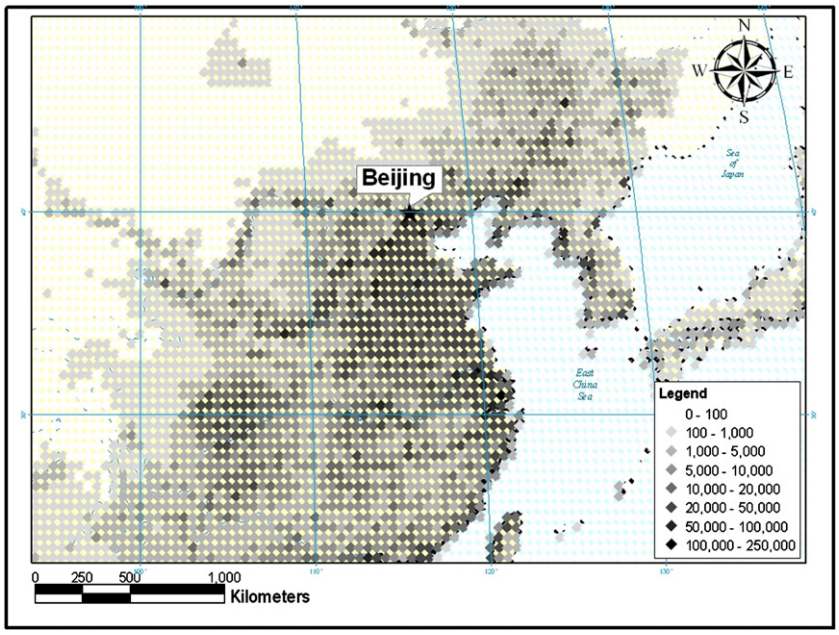

Fig. 2. The annual emission rates of PM_{10} over North China Plain estimated for the year 2006. Location of Beijing is shown as asterisk. (tonnes year^{-1} 0.5 $^{\circ-1}$cell). (available at the website http://www.cgrer.uiowa.edu/EMISSION_DATA_new/data/intex-b_emissions).

Table 1, the OC concentrations in PM_{10} are lower than those measured at Los Angeles (18.5 $\mu g\,m^{-3}$, [38]), Pearl River Delta Region (19.7 $\mu g\,m^{-3}$, [11]), Xi'an (43.3 $\mu g\,m^{-3}$, [18]), and they are similar to those measured at Seoul (11.1 $\mu g\,m^{-3}$, [39]; 10.3 $\mu g\,m^{-3}$, [40]), Kaohsiung (10.4 $\mu g\,m^{-3}$, [41]), however, they are higher than those measured at Helsinki (3.0 $\mu g\,m^{-3}$, [42]). EC concentrations are comparable to those measured at urban areas described above (6.2–10.0 $\mu g\,m^{-3}$), however, they are higher than those in Helsinki (1.2 $\mu g\,m^{-3}$, [42]) and Kaohsiung (4.0 $\mu g\,m^{-3}$, [41]), but lower than that in Xi'an (15.0 $\mu g\,m^{-3}$, [18]).

According to Turpin and Lim [43], the amount of urban organic matter (OM) can be estimated by multiplying the amount of OC by 1.6. Thus, the total carbonaceous aerosol (TCA) was calculated by the sum of OM and EC (TCA = 1.6 × OC + EC). Fig. 3 shows the daily variations of OC, EC, OC/EC ratio, PM_{10} and TCA/PM_{10} ratio (%) in Beijing from 15 March to 12 April 2004. PM_{10} mass concentrations ranged from 12.8 to 986.2 $\mu g\,m^{-3}$, with average concentration of 187.8 ± 136.9 $\mu g\,m^{-3}$, which has exceeded 100 $\mu g\,m^{-3}$, the Second Annual Ambient Air Quality Standard of PM_{10} by China Environmental Protection Administration. As indicated by the results, PM_{10} pollution in Beijing during spring is still serious and requires enhanced attention in order to work out strategies for a more effective control of the pollutants. The OC, EC, and TCA account for 9.3 ± 5.7%, 4.7 ± 2.7%, and 19.6 ±11.6% of PM_{10}, respectively. And the contributions of OC and EC to PM_{10} in spring are lower than that in winter.

3.2. Diurnal variation of OC, EC, PM_{10} concentrations during spring 2004

Table 2 presents the diurnal variations of OC, EC, PM_{10} and OC/EC ratio in Beijing during the spring time of 2004. The PM_{10} concentrations in daytime are obviously higher than that at night since turbulence is usually more active during daytime than at night [44] which can lift more dust particles and cause high ambient PM_{10} concentrations. Average OC concentrations are 13.5 $\mu g\,m^{-3}$ in both daytime and nighttime. Average EC concentrations in daytime (7.4 $\mu g\,m^{-3}$) are higher than that in nighttime (6.8 $\mu g\,m^{-3}$). Both OC and EC concentrations have their maxima between 07:30 and 10:30 due to motor vehicles in the morning traffic rush hour. However, low OC/EC ratios in daytime were observed.

3.3. Impact of meteorological factor

The standard meteorological parameters including wind speed and direction, air temperature and relative humidity were routinely measured. Fig. 4 presents the wind rose diagram measured

Table 1
Comparison of OC, EC concentration ($\mu g\,m^{-3}$) and OC/EC value with results from other areas.

Season/date		OC ($\mu g\,m^{-3}$)	EC ($\mu g\,m^{-3}$)	OC/EC	Reference
Beijing, China	January 2004	21.2	8.9	2.3	[37]
Beijing, China	March and April 2004	13.5	7.1	2.0	This study
Sub-Beijing, China	September–December 2002	21.2	7.3	3.0	[22]
Downtown Beijing, China	July 1999–September 2000	21.5	8.7	2.5	[20]
Los Angeles, USA	November and December 1987	18.5	7.3	2.5	[38]
Seoul, Korea	June 1994	11.1	8.4	1.3	[39]
Seoul, Korea	June 1994	10.3	8.4	1.3	[40]
Kaohsiung, Taiwan	November 1998–April 1999	10.4	4.0	2.6	[41]
Helsinki, Finland July	July 2000–July 2001	3.0	1.2	2.5	[42]
Pearl River Delta Region	January and February, 2002)	19.7	7.8	2.5	[11]
Xi'an,China	September and October, 2004	43.2	15.0	3.2	[18]
Uji, Japan	June 1998–December 1999	3.0	6.2	0.5	[31]

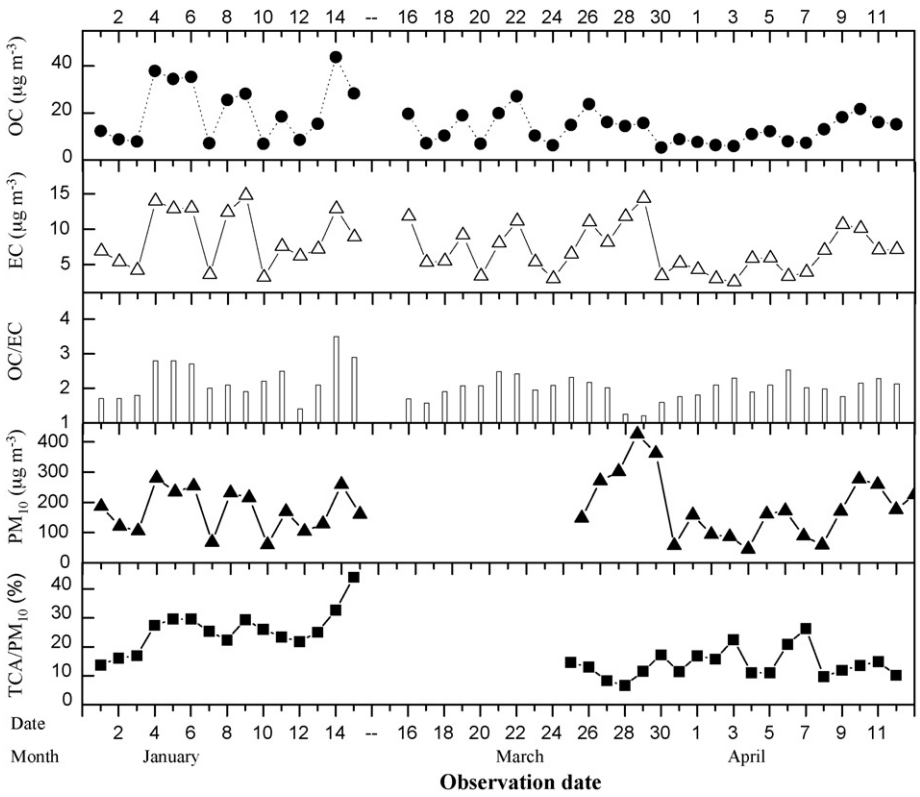

Fig. 3. Time series of day-averaged OC, EC, OC/EC ratio, PM_{10} mass, TCA/PM_{10} in Beijing from 15 March to 12 April 2004.

Table 2
Diurnal variation of concentration of OC, EC, PM_{10}, in spring in 2004.

Time (h)	0 (22:30–01:30)	3 (01:30–04:30)	6 (04:30–07:30)	9 (07:30–10:30)	12 (10:30–13:30)	15 (13:30–16:30)	18 (16:30–19:30)	21 (19:30–22:30)
OC($\mu g\,m^{-3}$)	13.5 ± 5.5	13.3 ± 6.5	14.3 ± 8.1	14.7 ± 9.1	14.4 ± 7.4	12.8 ± 6.6	12.2 ± 6.1	13 ± 5.3
EC($\mu g\,m^{-3}$)	6.7 ± 3.1	6.5 ± 2.8	7.3 ± 4.1	7.9 ± 5.0	7.8 ± 4.2	7.1 ± 5.3	6.8 ± 4.6	6.6 ± 3.2
OC/EC	2.1 ± 0.4	2.1 ± 0.4	2.0 ± 0.3	1.9 ± 0.4	1.9 ± 0.4	1.9 ± 0.5	1.9 ± 0.4	2.1 ± 0.4
PM_{10}($\mu g\,m^{-3}$)	188.1 ± 103.7	180.0 ± 100.0	172.0 ± 109.3	180.1 ± 146.2	177.7 ± 118.9	200.6 ± 218.9	206.3 ± 119.3	197.3 ± 128.3

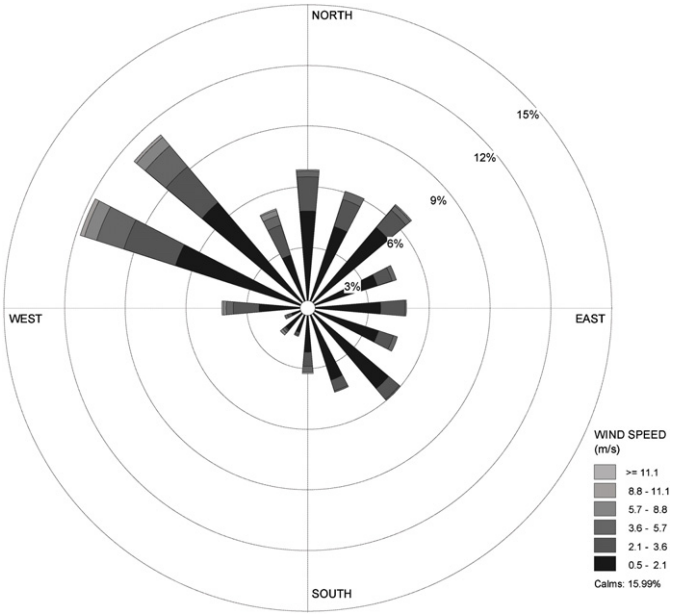

Fig. 4. Wind rose diagram during the sampling period in Beijing (15 March to 13 April 2004).

at the 8 m level of the meteorological tower. The wind was mainly from the northwest sector during the sampling period. North and northwest part of Beijing is mainly surrounded by the mountains while south and southeast of Beijing are heavily populated, urbanized, and industrialized areas such as Tianjin, Hebei province, and Shandong province. High emission of carbonaceous aerosols from southern part of Beijing (Fig. 2) was the major course for the episode day. According to the 24 h back trajectory analysis (Fig. 5a), when the air mass passed over southwest part of Beijing (26 March 2004 and 10 April 2004), high concentrations of air pollutants was observed (e.g. PM_{10} = 272 and 259 $\mu g\,m^{-3}$, respectively). On the contrary, Fig. 5b showed that the clean air masses came from the north during 30 March, 2 April and 6 April when low levels of pollutants were observed (e.g. PM_{10} = 57, 86 and 88 $\mu g\,m^{-3}$, respectively). Moreover, high negative correlations were observed between wind speed and air pollutants (except dust storm period) while fair positive correlations were between RH and air pollutants (except dust storm period). The linear regression results relating air pollutants and wind speed or RH are shown in Table 3. Low daily concentrations of air pollutants were observed when wind speed was high. Wind blows away air pollutants, gases and particulate alike and favors pollutant dilution. Therefore, wind speed, wind direction and RH affect the concentration of particulate pollutants at urban area of Beijing, but no significant relationships were observed between particulate pollutants and temperature.

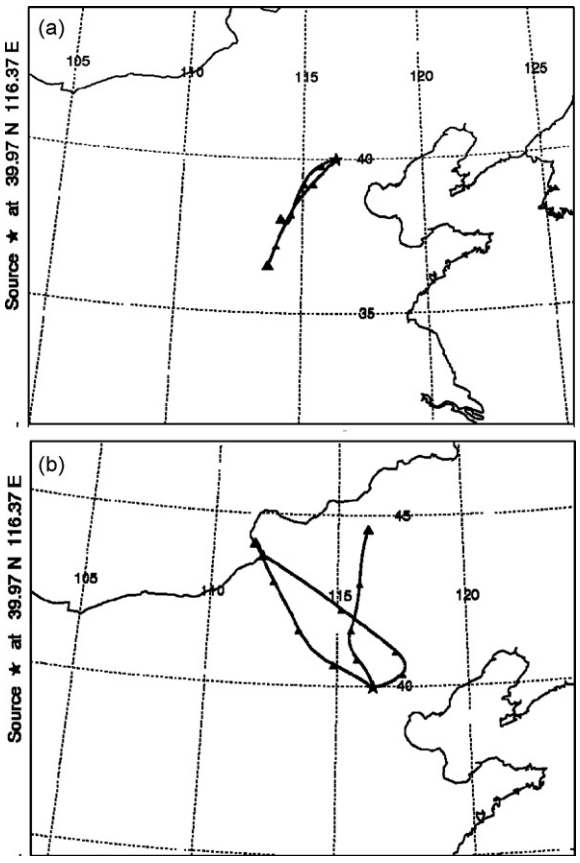

Fig. 5. 24 h air mass back trajectories on (a) 26 March and 10 April (b) 30 March, 2 April and 6 April.

3.4. Relationship of organic carbon and elemental carbon and ratios of OC to EC

The relationship of OC and EC indicates the origins of carbonaceous aerosols [45]. To investigate, the regression between the OC and EC concentrations are shown in Fig. 6 and good OC–EC correlations ($R^2 = 0.7$) were observed for spring season. This suggests that the emissions of OC and EC in Beijing could have similar sources, such as local motor vehicular emission and residential and commercial coal combustion. The slope of regression line in spring was different to that of winter, which indicated different emission sources contributing to the ambient carbonaceous particles.

Table 3
Regression and correlation analysis of air pollutants concentration and wind speed (except dust storm period).

	Regression relationship[a]	R^2
PM_{10}	$PM_{10} = -116.4WS + 348.4$	0.58
	$PM_{10} = 4.0RH - 13.4$	0.44
$PM_{2.5}$	$PM_{2.5} = -61.2WS + 166.6$	0.57
	$PM_{2.5} = 2.5RH - 41.0$	0.61
OC	$OC = -8.5WS + 26.07$	0.67
	$OC = 0.25RH + 1.5$	0.37
EC	$EC = -3.8WS + 12.3$	0.60
	$EC = 0.1RH + 2.0$	0.24

[a] WS: wind speed ($m\,s^{-1}$); RH: relative humidity (%).

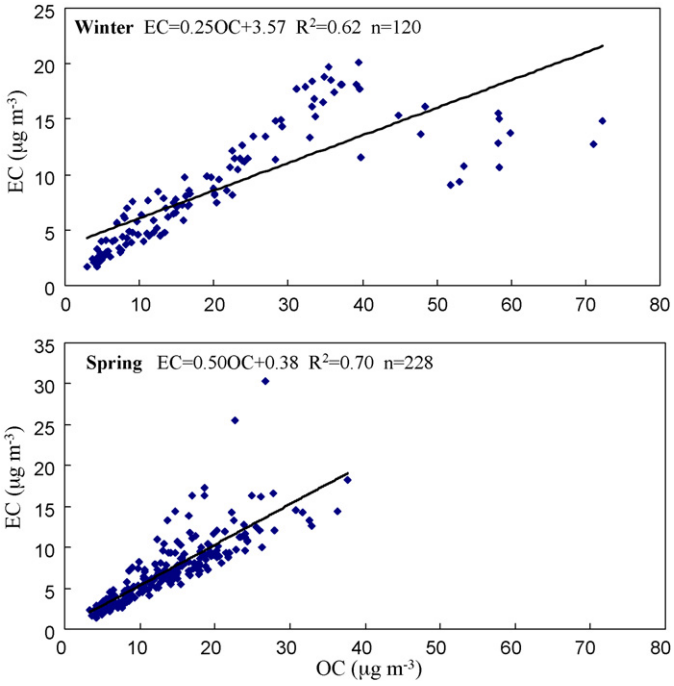

Fig. 6. Relationships between OC and EC concentrations in PM_{10} during winter and Beijing in 2006.

3.5. Source identification of carbonaceous aerosol based on OC/EC ratio

OC to EC ratio is commonly regarded as secondary organic aerosol (SOA) indicator [45]. High OC/EC ratios determined in the atmosphere indicate the formation of SOA or the contributions from residential biomass burning and coal combustion. Moreover, coal combustion with high OC/EC ratio was the major source in the northern cities in China [18] and the ratios exceeding 2.8 have been used to identify SOA formation [46].

China produces and consumes 25% of the world's total coal [47], which constituting about 70% of the national energy budget. Coal will be China's dominant energy source for next 20 years [48] and coal combustion is one important source of atmospheric particles in typical cities in northern part of China during heating period [44,49]. While only limited coal combustion sources are observed in the urban areas of Beijing, it is still widely used in the neighboring areas [50]. Motor vehicle exhaust is another important source for ambient particles [20,50] and this resulted in low OC/EC ratio [37]. Watson et al. [51] reported that OC/EC ratio is 1.1 from motor vehicle and 2.7 from coal combustion. The study by Cao et al. [18] shows that OC/EC ratio from coal combustion is 12. The elevated OC/EC ratios were found during heating seasons with increased primary emissions from residential coal combustion.

The average OC/EC ratios in Beijing were in the range between 0.88 and 2.95 with an overall average of 2.0 for PM_{10}. Therefore, the formation of SOA in Beijing during spring is not significant and the measured OC is mainly from primary emissions. Moreover, spring is not the biomass burning season, and the main primary sources for carbonaceous aerosol are coal combustion and motor vehicles.

The contribution of the two major sources is calculated using OC/EC ratio with the speculation that carbonaceous aerosols are primary emission during spring period. According to Zhang et al. [37], OC/EC ratio was 5.6 for coal combustion in Beijing in winter. In this study, OC/EC ratios in Beijing ranged from 0.88 to 2.95. So 0.88 is used as the OC/EC ratio from motor vehicles sources and 5.6 is the OC/EC ratio from coal combustion sources. Then, a simple

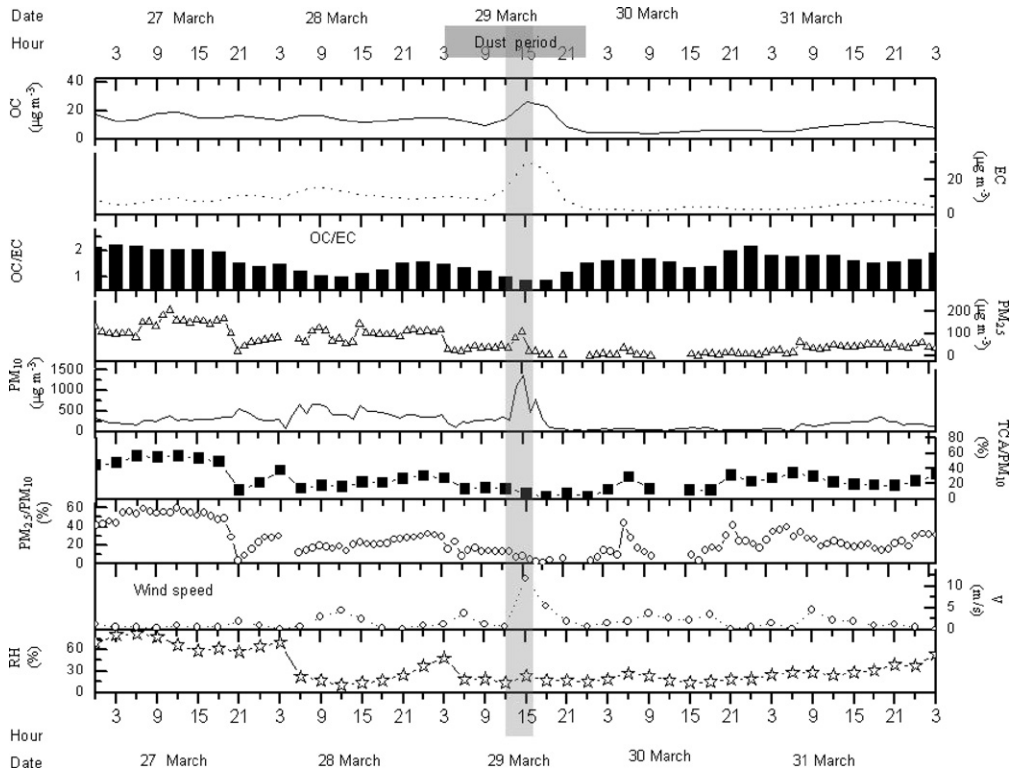

Fig. 7. Time series of 3 h-averaged OC, EC, OC/EC ratio, $PM_{2.5}$, PM_{10}, $PM_{2.5}/PM_{10}$, TCA/PM_{10}, and wind speed, and relative humidity in dust period.

source identification equation is given as the following:

$$0.88\gamma + 5.6(1-\gamma) = C_{avg} \quad (1)$$

where γ is the fraction of carbonaceous aerosol of motor vehicle origin and $(1-\gamma)$ is fraction of carbonaceous aerosol of coal combustion origin, C_{avg} is averaged OC/EC ratio. The average OC/EC in the observation period was 2.0 and the calculated contribution of local motor vehicle and coal combustion sources is 76% and 24%, respectively. This shows that motor vehicles were the dominant emitter of carbonaceous aerosols associated with PM_{10} during spring.

3.6. Organic carbon and elemental carbon in dust storm period

Dust storms are serious natural disasters in China and occur frequently in Beijing during spring (March–May) [28,52,53,54]. From 2000 to 2002 these storms had disrupted air traffic, commercial activities, affected air quality and the daily life over broad areas of the country [55,56]. Previous study suggests that dust weather frequency in North China is associated with both the strength of the winter monsoon and the state of El Niño-Southern Oscillation (ENSO) [57]. The weather observations made by the China Meteorological Administration (CMA) classified dust events into four categories: dust in suspension, blowing dust, a dust storm, or a severe dust storm. These categories are based on factors such as horizontal visibility, that is, the horizontal visibilities for dust in suspension, blowing dust, dust storm and severe dust storm are less than 10 km, 1–10 km, 500–1000 m and less than 500 m, respectively. According to this definition, the dust event on 29 March 2004 in Beijing is a blowing dust.

Fig. 7 presents the variations of OC, EC, OC/EC ratio, $PM_{2.5}$, PM_{10}, $PM_{2.5}/PM_{10}$ (%), TCA/PM_{10} (%), wind speed, and relative humidity from 24 to 31 March 2004. Meteorological data showed that the ground wind speed reached a maximum of 11.8 m s^{-}, while the relative humidity decreased to ~17% on 29 March 2004. 1 h-averaged PM_{10} and $PM_{2.5}$ concentration reached maximum of 1363.9 μg m^{-3} and 109.0 μg m^{-3}, respectively during dust storm period. The percentage of $PM_{2.5}/PM_{10}$ reached low value of 8%. From 27 to 29 March, PM_{10} concentration has an increasing trend, while $PM_{2.5}$ concentration has a decreasing trend and reach minimum on 29 March. The percentages of $PM_{2.5}/PM_{10}$ also decrease from 27 to 29 May. When dust event occurred, OC and EC had high values of 26.7 and 30.3 μg m^{-3}, respectively. However, the contributions of total carbon aerosols to PM_{10} were low before and during dust period and increase after dust event. OC/EC ratio reaches their minimum during dust period. This shows that PM_{10}, OC, and EC concentration increase while $PM_{2.5}$ concentration, OC/EC ratio, $PM_{2.5}/PM_{10}$, and TCA/PM_{10} ratio decrease during the dust period.

4. Conclusion

Continuous observations of OC, EC, and PM_{10} were conducted in Beijing, China in order to investigate the characterizations and sources of OC and EC. PM_{10} pollution in Beijing during spring is still serious; the average concentration of PM_{10} has exceeded 100 μg m^{-3}, the Second Annual Ambient Air Quality Standard of PM_{10} by China Environmental Protection Administration. Therefore, enhanced attention was required in order to work out strategies for a more effective control of the pollutants. OC and EC were the dominant species in PM_{10} which account for $9.3 \pm 5.7\%$ and $4.7 \pm 2.7\%$ of PM_{10}, respectively. High daily concentrations of air pollutants were observed, when the air masses came from south or the wind speed was low. However, when the clean air masses came from the north or the wind speeds were strong, low levels of pollutants were observed in the urban area of Beijing. Overall, vehicular exhaust and coal combustion, plus the dust from the long-range transport could be the major sources of the PM_{10} pollution in Beijing during spring time. Based on OC/EC ratio, the contribution of local motor vehicle and coal combustion sources for carbonaceous aerosols are 76% and 24%, respectively. It shows that the

motor vehicle is the dominant emission for carbonaceous aerosols in Beijing PM_{10} in spring and winter. Therefore, vehicular control emission will be the possible strategies to decrease PM_{10} pollution in the atmosphere of Beijing.

Acknowledgements

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References

[1] H. Cachier, M.P. Brmond, B.M. Patrick, Carbonaceous aerosols from different tropical biomass burning sources, Nature 340 (1989) (1989) 371–373.
[2] A. Molnár, E. Mészáros, H.C. Hansson, H. Karlsson, A. Gelencser, G.Y. Kiss, Z. Krivácsy, The importance of organic and elemental carbon in the fine atmospheric aerosol particles, Atmos. Environ. 33 (1999) 2745–2750.
[3] J.J. Cao, S.C. Lee, K.F. Ho, S.C. Zou, K. Fung, Y. Li, J.G. Watson, J.C. Chow, Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, Atmos. Environ. 38 (2004) 4447–4456.
[4] J.C. Chow, J.G. Watson, L.-W.A. Chen, W.P. Arnott, H. Moosmuller, K.K. Fung, Equivalence of elemental carbon by Thermal/Optical Reflectance and Transmittance with different temperature protocols, Environ. Sci. Technol. 38 (2004) 4414–4422.
[5] J. Wu, Y.Y. Xu, C.B. Fu, R.J. Zhang, M. Dai, Y. Zhu, Comparison of simulating mineral dust aerosols in East Asia by two emission schemes, China Particuol. 4 (2006) 293–299.
[6] M.Z. Jacobson, Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, Nature 409 (2001) 695–697.
[7] S. Menon, J. Hansen, L. Nazarenko, Y. Luo, Climate effects of black carbon aerosols in China and India, Science 297 (2002) 2250–2253.
[8] X.X. Li, Z.X. Shen, J.J. Cao, S.X. Liu, C.S. Zhu, T. Zhang, Distribution of carbonaceous aerosol during spring 2005 over the Horqin Sandland in northeastern China, China Particuol. 4 (2006) 316–322.
[9] F.F. Fu, L.J. Xu, W. Ye, Y.Q. Chen, M.Y. Jiang, X. Xu, Morphology of black carbon aerosols and ubiquity of 50-nanometer black carbon aerosols in the atmosphere, China Particuol. 4 (2006) 323–326.
[10] D. Rosenfeld, Suppression of rain and snow by urban and industrial air pollution, Science 287 (2002) 1793–1796.
[11] J.J. Cao, S.C. Lee, K.F. Ho, X.Y. Zhang, S.C. Zou, K. Fung, J.C. Chow, J.G. Watson, Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period, Atmos. Environ. 37 (2003) 1451–1460.
[12] J.Z. Yu, J.W.T. Tung, A.W.M. Wu, A.K.H. Lau, P.K.K. Louie, J.C.H. Fung, Abundance and seasonal characteristics of elemental and organic carbon in Hong Kong PM_{10}, Atmos. Environ. 38 (2004) 1511–1521.
[13] M.G. Zhang, Modeling of organic carbon aerosol distributions over East Asia in the springtime, China Particuol. 2 (2004) 192–195.
[14] M.G. Zhang, Z.W. Han, L.Y. Zhu, Simulation of atmospheric aerosols in East Asia using modeling system RAMS-CMAQ: model evaluation, China Particuol. 5 (2007) 321–327.
[15] Z.W. Han, R.J. Zhang, Q.G. Wang, W. Wang, J.J. Cao, J. Xu, Regional modeling of organic aerosols over China in summertime, J. Geophys. Res. 113 (2008), doi:10.1029/2007JD009436, D11202.
[16] J.H. Yu, T. Chen, B. Guinot, H. Cachier, T. Yu, W.Q. Liu, X. Wang, Characteristics of carbonaceous particles in Beijing during winter and summer 2003, Adv. Atmos. Sci. 23 (2006) 468–473.
[17] F. Yang, K. He, B. Ye, X. Chen, L. Cha, S.H. Cadle, T. Chan, P.A. Mulawa, One-year record of organic and elemental carbon in fine particles in downtown Beijing and Shanghai, Atmos. Chem. Phys. 5 (2005) 1449–1457.
[18] J.J. Cao, F. Wu, J.C. Chow, S.C. Lee, Y. Li, S.W. Chen, Z.S. An, K. Fung, J.G. Watson, C.S. Zhu, S.X. Liu, Characterization source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China, Atmos. Chem. Phys. 5 (2005) 3127–3137.
[19] D.G. Streets, S. Gupta, S.T. Waldhoff, M.Q. Wang, T.C. Bond, B. Yiyun, Black carbon emissions in China, Atmos. Environ. 35 (2001) 4281–4296.
[20] K.B. He, F.M. Yang, Y.L. Ma, Q. Zhang, X.H. Yao, C.K. Chan, S. Cadle, T. Chan, P. Mulawa, The characteristics of $PM_{2.5}$ in Beijing, China, Atmos. Environ. 35 (2001) 4959–4970.
[21] G.C. Wang, J.H. Bai, Q.X. Kong, A. Emilenko, Black carbon particles in the urban atmosphere in Beijing, Adv. Atmos. Sci. 22 (2005) 640–646.
[22] F.K. Duan, K.B. He, Y.L. Ma, Y.T. Jia, F.M. Yang, Y. Lei, S. Tanaka, T. Okuta, Characteristics of carbonaceous aerosols in Beijing, China, Chemosphere 60 (2005) 355–364.
[23] C.Y. Chan, X.D. Xu, Y.S. Li, K.H. Wong, G.A. Ding, L.Y. Chan, X.H. Cheng, Characteristics of vertical profiles and sources of $PM_{2.5}$, PM_{10} and carbonaceous species in Beijing, Atmos. Environ. 39 (2005) 5113–5124.
[24] Y.L. Sun, G.S. Zhuang, Y. Wang, L.H. Han, J.H. Guo, M. Dan, W.J. Zhang, Z.F. Wang, Z.P. Hao, The air-borne particulate pollution in Beijing–concentration, composition, distribution and sources, Atmos. Environ. 38 (2004) 5991–6004.
[25] F.K. Duan, X.D. Liu, K.D. He, Y.W. Li, S.P. Dong, Characteristics and source identification of particulate matter in wintertime in Beijing, Water Air Soil Poll. 180 (2007) 171–183.
[26] F.K. Duan, K.B. He, Y.L. Ma, F.M. Yang, X.C. Yu, S.H. Cadle, T. Chan, P.A. Mulawa, Concentration and chemical characteristics of $PM_{2.5}$ in Beijing, China: 2001-2002, Sci. Total Environ. 355 (2006) 264–275.
[27] Z. He, Y.J. Kim, K.O. Ogunjobi, J.E. Kim, S.Y. Ryu, Carbonaceous aerosol characteristics of PM2.5 particles in Northeastern Asia in summer 2002, Atmos. Environ. 38 (2004) 1795–1800.
[28] R.J. Zhang, R. Arimoto, J.L. An, S. Yabuki, J.H. Sun, Ground observation of a strong dust storm in Beijing in March 2002, J. Geophys. Res. (2005), doi:10.1029/2004JD004589, 110(D18S06).
[29] L.F. Salter, B. Parsons, Field trials of the TEOM and Partisol for PM_{10} monitoring in the St Austell china clay area, Cornwall, UK, Atmos. Environ. 33 (1999) 2111–2114.
[30] A.M. King, New Directions: TEOMs and the volatility of UK non-urban PM_{10}: a regulatory dilemma? Atmos. Environ. 34 (2000) 3211–3212.
[31] R. Höller, S. Tohnoa, M. Kasaharaa, R. Hitzenberger, Long-term characterization of carbonaceous aerosol in Uji, Japan, Atmos. Environ. 36 (2002) 1267–1275.
[32] A. Even, A. Khlystov, G.P.A. Kos, H.M.T. Brink, G. Hoek, M. Oldenwening, J. Hartog, Improvement of BC Measurement with the ambient carbon particulate monitor RP5400, J. Aerosol Sci. 31 (S1) (2000) 897–898.
[33] R.W. Long, N.L. Eatough, N.F. Mangelson, W. Thompson, K. Fiet, S. Smith, R. Smith, D.J. Eatough, C.A. Pope, W.E. Wilson, The measurement of $PM_{2.5}$ including semi-volatile components, in the EMPACT program: results from the Salt Lake City Study, Atmos. Environ. 37 (2003) 4407–4417.
[34] K. Matsumoto, T. Hayanoa, M. Uematsua, Positive artifact in the measurement of particulate carbonaceous substances using an ambient carbon particulate monitor, Atmos. Environ. 37 (2003) 4713–4717.
[35] K. Matsumoto, M. Uematsu, T. Hayano, K. Yoshioka, H. Tanimoto, T. Iida, Simultaneous measurements of particulate elemental carbon on the ground observation network over the western North Pacific during the ACE-Asia campaign, J. Geophys. Res. 108 (D) (2003), doi:10.1029/2002JD002744.
[36] A.D.A. Castanho, P. Artaxo, Wintertime and summertime Sao Paulo aerosol source apportionment study, Atmos. Environ. 35 (2001) 4889–4902.
[37] R.J. Zhang, J.J. Cao, S.C. Lee, Z.X. Shen, K.F. Ho, Carbonaceous aerosols in PM_{10} and pollution gases in winter in Beijing, J. Environ. Sci. China 19 (2007) 564–571.
[38] J.C. Chow, J.G. Watson, E.M. Fujuta, Z. Lu, D.R. Lawson, Temporal and spatial variations of $PM_{2.5}$ and PM_{10} aerosol in the southern California air quality study, Atmos. Environ. 28 (1994) 2061–2080.
[39] Y.P. Kim, K.C. Moon, J.H. Lee, N.J. Baik, Concentrations of carbonaceous species in particles at Seoul and Cheju in Korea, Atmos. Environ. 33 (1999) 2751–2758.
[40] S.S. Park, Y.J. Kim, K. Fung, $PM_{2.5}$ carbon measurements in two urban areas: Seoul and Kwangju, Korea, Atmos. Environ. 36 (2002) 1287–1297.
[41] J.J. Lin, H.S. Tai, Concentrations and distributions of carbonaceous species in ambient particles in Kaohsiung City, Taiwan, Atmos. Environ. 35 (2001) 2627–2636.
[42] J. Viidanoja, M. Sillanpaa, J. Laakia, V. Kerminen, R. Hillamo, P. Aarnio, T. Koskentalo, Organic and black carbon in $PM_{2.5}$ and PM_{10}: 1 year of data from an urban site in Helsinki, Finland, Atmos. Environ. 36 (2002) 3183–3193.
[43] B.J. Turpin, H.J. Lim, Species contributions to $PM_{2.5}$ mass concentrations: Revisiting common assumptions for estimating organic mass, Aerosol Sci. Technol. 35 (2001) 602–610.
[44] X.Y. Zhang, J.J. Cao, L.M. Li, R. Arimoto, Y. Cheng, B. Huebert, D. Wang, Characterization of atmospheric aerosol over Xian in the south margin of the Loess Plateau, China, Atmos. Environ. 36 (2002) 4189–4199.
[45] J.C. Chow, J.G. Watson, L.C. Pritchett, W.R. Pierson, C.A. Frazier, R.G. Purcell, The DRI Thermal/Optical Reflectance carbon analysis system: Description, evaluation and applications in U.S. air quality studies, Atmos. Environ. 27A (1993) 1185–1201.
[46] J.J. Cao, S.C. Lee, J.C. Chow, J.G. Watson, K.F. Ho, R.J. Zhang, Z.D. Jin, Z.X. Shen, G.C. Chen, Y.M. Kang, S.C. Zou, L.Z. Zhang, S.H. Qi, M.H. Dai, Y. Cheng, K. Hu, Spatial and seasonal distributions of carbonaceous aerosols over China, J. Geophys. Res. 112 (2007), doi:10.1029/2006/JD008205, D22S11.
[47] J.G. Liu, J. Diamond, China's environment in a globalizing world, Nature 435 (2005) 1179–1186.
[48] P. Aldhous, China's burning ambition, Nature 435 (2005) 1152–1154.
[49] Z.B. Shi, L.Y. Shao, T.P. Jones, A.G. Whittaker, S.L. Lu, K.A. Bérubé, T.E. He, R.J. Richard, Characterization of airborne individual particles collected in an urban area, a satellite city and a clean air area in Beijing, 2001, Atmos. Environ. 37 (2003) 4097–4108.
[50] M.O. Dan, G.S. Zhuang, X.X. Li, H.R. Tao, Y.H. Zhuang, The characteristics of carbonaceous species and their sources in $PM_{2.5}$ in Beijing, Atmos. Environ. 38 (2004) 3443–3452.
[51] J.G. Watson, J.C. Chow, J.E. Houck, PM2.5 chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in northwestern Colorado during 1995, Chemosphere 43 (2001) 1141–1151.
[52] Z.X. Shen, X.X. Li, J.J. Cao, S. Caquineau, Y.Q. Wang, X.Y. Zhang, Characteristics of clay minerals in Asian dust and their environmental significance, China Particuol. 3 (2005) 260–264.

[53] Z.X. Shen, J.J. Cao, R. Arimoto, R.J. Zhang, D.M. Jie, S.X. Liu, C.S. Zhu, Chemical composition and source characterization of spring aerosol over Horqin sand land in northeastern China, J. Geophys. Res. 112 (2007), doi:10.1029/2006JD007991, D14315.
[54] Z.W. Han, H. Ueda, K. Matsuda, R.J. Zhang, K. Arao, Y. Kanai, H. Hasome, Model study on particle size segregation and deposition during Asian dust events in March 2002, J. Geophys. Res. 109 (2004), doi:10.1029/2004JD004920, D19205.
[55] X.J. Zhou, X.D. Xu, P. Yan, Y.H. Weng, J.L. Wang, Dynamics characteristics of spring sandstorms in 2000, Sci. China D 45 (2002) 921–930.
[56] Z.J. Zhou, G.C. Zhang, Typical intensive dust event in northern China (1954-2002), Chinese Sci. Bull. 48 (2003) 2366–2370.
[57] R.J. Zhang, Z.W. Han, M.X. Wang, X.Y. Zhang, Dust storm weather in China: new characteristics and origins (in Chinese), Quat. Sci. 22 (2002) 374–380.