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The air-borne particulate pollution in Beijing—concentration, composition, distribution and sources

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Abstract

Aerosol samples of $PM_{2.5}$ and PM_{10} were collected in both summer and winter seasons from 2002 to 2003 synchronously at a traffic site, an industrial site, and a residential site in Beijing, which could basically be the representatives over Beijing. Twenty-three elements and 15 ions together with organic carbon and elemental carbon were analyzed systematically for characterization of Beijing aerosol. $PM_{2.5}$ was the major part of the inhalable particles (PM_{10}), as the ratios of $PM_{2.5}/PM_{10}$ were 0.45–0.48 in summer and 0.52–0.73 in winter. SO_4^{2-}, NO_3^-, NH_4^+, organic matter, crustal matter, and element carbon were the six dominant species, which totally accounted for 85.8–97.7% of $PM_{2.5}$. Secondary aerosol (SO_4^{2-}, NO_3^-, and NH_4^+), road dust or/and long-range transported dust from outside Beijing, industry and motor vehicles emission, coal burning were the major contributors to the air-borne particulate pollution in Beijing. Overall, coal burning and the traffic exhausts, plus the dust from the long-range transport, could be the major sources of the aerosol pollution at Beijing. A relatively even spatial distribution of chemical species in $PM_{2.5}$ was found while in PM_{10} a significant variation with the highest concentrations at the industrial site in summer and at the residential site in winter was observed. The concentrations of PM_{10}, $PM_{2.5}$ as well as various chemical species were higher in winter than in summer. The contributions of mineral aerosol from outside Beijing were first estimated with a newly developed element tracer technique, which accounted for 79% and 37% of the total mineral in PM_{10} and $PM_{2.5}$ in winter, and 19% and 20% in summer, respectively. During the dust storm period from 20 to 22 March, it reached up to 97% in TSP, 79% in PM_{10} and 76% in $PM_{2.5}$. This is the technique, firstly, developed for estimating the relative contributions of sources from inside and outside Beijing to the total mineral aerosol and it could provide the basic information in controlling the air-borne particulate pollution at Beijing.

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Keywords: Aerosol; Beijing; PM_{10}; $PM_{2.5}$; Seasonal variation; Spatial variation; Sources; Factor analysis

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1. Introduction

Aerosol particles, especially fine particles with diameter smaller than 2.5 μm, have close relations with

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increased respiratory morbidity and mortality (Dockery and Pope, 1994; Schwartz et al., 1996; Wilson and Suh, 1997; Ostro et al., 1999). Aerosols also affect climate directly by scattering and absorbing radiation or indirectly by changing the depth and albedo of the clouds (Twomey, 1974; Charlson et al., 1992). The direct effect of the regional haze from the anthropogenic aerosols resulted in a 5–30% reduction of the yields of rice and wheat in China (Chameides et al., 1999). The emission of anthropogenic aerosols is increasing sharply with the modern industrialization and motorization over the world, which brings not only regional but also global effects through the long-range transport (Wolf and Hidy, 1997). Aerosol is now receiving worldwide attention, as it would have the potential impacts on the global climate change and the effect on the health to human beings all over the world.

The studies on the characterization of the aerosol in Beijing have been carried out in the last two decades (Winchester and Bi, 1984; Yang et al., 2000; Dong and Yang, 1998; Wang, 1999; Cao et al., 2002). Dust-soil, industry emission, coal burning, vehicle exhaust emission and waste incineration were thought to be the major sources of particulate pollution at Beijing. However, the studies reported were mostly concentrated in TSP/PM_{10}, rarely on $PM_{2.5}$. The composition, sources, spatial and temporal variations of $PM_{2.5}$ are poorly understood. Chen et al. (1994) initiated the investigation of $PM_{2.5}$ and observed that OC, EC, and sulfate, the three major components of $PM_{2.5}$, contributed 66% of the total fine particles, which had the yearly average of $89.7\,\mu g\,m^{-3}$ in Beijing. They inferred with CMB model that diesel exhaust, coal combustion, and re-suspension dust were the major sources of the fine particles in Beijing. He et al. (2001) monitored $PM_{2.5}$ at a residential site and a downtown site in Beijing from July 1999 through September 2000 and observed $PM_{2.5}$ concentrations of $37–357\,\mu g\,m^{-3}$ with significant seasonal variation. They concluded that organic carbon was the most abundant species in $PM_{2.5}$ while sulfate, nitrate, and ammonium also were the major components. Yao et al. (2002) reported that sulfate, nitrate and ammonium were the main ionic species in $PM_{2.5}$, which constituted 44%, 25% and 16%, respectively, of the total mass of water-soluble part. Yao et al. (2003) discussed the various characteristics of particles in different size and the formation mechanism of ions. They speculated that cloud process and non-cloud heterogeneous process were responsible for the formation of sulfate in summer and in spring, respectively. However, the studies mentioned above were based on limited number of samples collected and the concentration, composition, and sources as well as the spatial and temporal variations of Beijing aerosols have been far from the clear characterization.

While several measures in controlling the air pollution in Beijing have been executed since 1990s, such as the use of low-sulfur coal, the partial replacement of coal with natural gas or liquefied petroleum gas, the phase out of leaded gasoline, and the move-out of those highly polluted industry from the city, the air-borne particulate stays at a much higher level than the national air quality standard, as the automobile exhaust has sharply increased since the past decade (Elliott et al., 1997) and coal burning has still been the major energy source. Moreover, Beijing is located in the downwind of Asian dust, which carries those mineral aerosols mixed with pollutants on the pathway and invades Beijing all the time. The long-range transported dust aerosols would further mix with Beijing local aerosols and make Beijing aerosols more complicated. To better understand the characteristics of Beijing aerosols, we continually monitored aerosol samples of PM_{10} and $PM_{2.5}$ synchronously at a traffic site, an industrial site, and a residential site of Beijing for 2 years; analyzed systematically 23 elements and 15 ions together with organic carbon and elemental carbon; and investigated the seasonal and spatial variations. We also developed a new element tracer technique to first distinguish the contributions of mineral aerosol from outside Beijing to the local aerosol in different seasons. This paper reported a comprehensive characterization of Beijing aerosol and provided the scientific background for the further control of air pollution at Beijing.

2. Experimental

2.1. Sampling

Aerosol samples of $PM_{2.5}$ and PM_{10} were collected in both summer and winter seasons from 2002 to 2003 synchronously at three sampling sites, i.e. (1) a traffic site, located in the campus of the Beijing Normal University (BNU) between the 2nd and 3rd ring roads, (2) an industrial site near the Capital Steel Company (CS), and (3) a residential site, Yihai Garden (YH), located near the South 4th ring road. The traffic site is located on the roof (~40 m high) of the 12th floor in the Building of Science and Technology of BNU. The residential site is located on the roof of a residential building roof (~40 m high). The industrial site is located on the roof (~4 m) of a building, which is close to the CS. The three sampling sites could basically be the representatives over Beijing. The location of sampling sites was shown in Fig. 1. PM_{10} and $PM_{2.5}$ aerosol samples were collected with medium-volume samplers manufactured by Beijing Geological Instrument-Dickel Co., Ltd. (model: TSP/PM_{10}/$PM_{2.5}$-2; flow rate: $77.59\,L\,min^{-1}$). Both PM_{10} and $PM_{2.5}$ were daily collected on Whatman® 41 filters (Whatman Inc.,

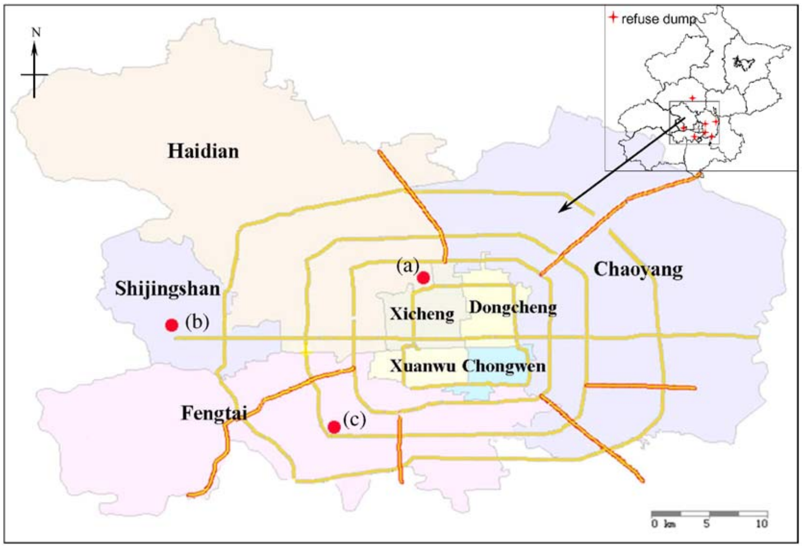

Fig. 1. Map of sampling sites (a: BNU, b: CS, c: YH).

Maidstone, UK) for element and ion analysis; in addition, $PM_{2.5}$ sample was also collected synchronously on quartz filter (Whatman Inc., Maidstone, UK) every three days for organic carbon (OC) and element carbon (EC) analysis. All those filters were weighed before and after sampling with an analytical balance (Sartorius 2004MP, reading precision 10 μg) after stabilizing in constant temperature($20 \pm 5\,^{\circ}C$) and humidity($40 \pm 2\%$). All the procedures were strictly quality-controlled to avoid any possible contamination of the samples.

For study on the sources of aerosols, soil samples were also collected at 7 typical sites in both geographical distribution and various sources: downtown areas of second ring road, of third ring road, of fourth ring road, a place near a gas station, a residential area, an industry area, and a construction area. For comparison, the soil samples were also collected at Dingling in suburb of Beijing, Duolun in Inner Mongolia, and Fengning in Hebei province.

2.2. *Chemical analysis*

2.2.1. *Element analysis*

The sample filters were digested at 170°C for 4 h in high-pressure Teflon digestion vessel with 3 ml concentrated HNO_3, 1 ml concentrated HCl, and 1 ml concentrated HF. After cooling, the solutions were dried, and then diluted to 10 ml with distilled-deionized water. Total 23 elements (Al, Fe, Mn, Mg, Ti, Sc, Na, Eu, Ce, Sr, Ca, Co, Cr, Ni, Cu, Pb, Zn, Cd, V, S, As, Se, and Sb) were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES, Model: ULTIMA, JOBIN-YVON Company, France). The detailed analytical procedures were given elsewhere (Zhuang et al., 2001, 2003). The soil samples were sieved (500 screen meshes, particles of $<30\,\mu m$ pass through), and then analyzed using the same procedures as mentioned above.

2.2.2. *Ion analysis*

Eleven inorganic ions (SO_4^{2-}, NO_3^-, F^-, Cl^-, NO_2^-, PO_4^{3-}, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}) and 4 organic acids (acetic, formic, oxalic, and methylsulfonic acid (MSA)) were analyzed by Ion Chromatography (IC, Dionex 600) that consists of a separation column (Dionex Ionpac AS 11), a guard column (Dionex Ionpac AG 11), a self-regenerating suppressed conductivity detector (Dionex Ionpac ED50) and a gradient pump (Dionex Ionpac GP50). The details were given elsewhere (Yuan et al., 2003).

2.2.3. *Carbon analysis*

Organic carbon (OC) and element carbon (EC) were analyzed with C/H/N elemental analyzer (Germany Vario EL, C/H/N model) by using a modified method (Chi et al., 1999). The quartz filters were oxidized at 450 and 950°C, for measurement of OC and total carbon (TC), respectively. EC was calculated from the difference between TC and OC. The detailed procedures were given elsewhere (Dan et al., 2004).

3. Results and discussion

3.1. *PM_{10} and $PM_{2.5}$ concentrations*

PM_{10} and $PM_{2.5}$ mass concentrations in both summer and winter seasons at Beijing were shown in Table 1. The average concentrations of PM_{10} in summer at the

Table 1
Average mass concentrations (μg m^{-3}) of PM_{10} and $PM_{2.5}$ ratio of $PM_{2.5}/PM_{10}$ at different site

Site	Species	Summer				Winter			
		Mean(SD)	Min	Max	N	Mean(SD)	Min	Max	N
BNU	PM_{10}	172.2(101.9)	23.9	461.5	22	184.4(130.5)	29.4	446.1	20
	$PM_{2.5}$	77.3(55.7)	15.8	216.2	20	135.7(96.6)	24.0	349.2	20
	$PM_{2.5}/PM_{10}$	0.45(0.22)	0.08	0.96	20	0. 73(0.16)	0.30	0.99	18
CS	PM_{10}	170.0(66.7)	51.1	310.8	22	287.7(155.7)	40.5	573.2	18
	$PM_{2.5}$	82.2(49.5)	12.2	169.5	22	140.8(73.9)	21.3	301.3	20
	$PM_{2.5}/PM_{10}$	0.48(0.21)	0.06	0.87	22	0.52(0.15)	0.26	0.84	18
YH	PM_{10}	150.1(56.9)	63.7	275.7	21	292.7(172.7)	81.6	631.9	18
	$PM_{2.5}$	75.4(45.6)	14.6	179.7	21	182.2(120.8)	43.5	460.9	20
	$PM_{2.5}/PM_{10}$	0.47(0.17)	0.19	0.69	21	0.61(0.09)	0.45	0.84	17

three sites (BNU, CS, and YH) were 172.2, 170.0 and 150.1 μg m^{-3} which were lower than those of 184.4, 287.7 and 292.7 μg m^{-3}, respectively, in winter. The average concentrations of $PM_{2.5}$ in summer at three sites were 77.3, 82.2 and 75.4 μg m^{-3}, respectively, which were much lower than those of 135.7, 140.8 and 182.2 μg m^{-3}, respectively, in winter. The concentrations of PM_{10} exceed the National Ambient Air Quality Standard of China (limit of 150 μg m^{-3} as "light pollution") at any site in each season. The concentrations of $PM_{2.5}$ were ~5 times as high as the limit (annual average of 15 μg m^{-3}, US Environmental Protection Agency (US EPA), 1997, no standard in China yet) in summer and 8–11 times in winter, greatly exceed the USA Ambient Air Quality Standard regulated by USA Environmental Protection Agency. These results indicated clearly that the airborne particulate pollution has been very serious at Beijing and it is much worse in winter than in summer. It can be seen from the discussion below that the coal burning and the traffic exhausts, plus the dust from the long-range transport, could be the major sources of the aerosol pollution at Beijing.

The ratio of $PM_{2.5}/PM_{10}$ was also shown in Table 1. The average ratio of $PM_{2.5}/PM_{10}$ in winter was 0.73, 0.52 and 0.61 at BNU, CS and YH sites, and in summer was 0.45, 0.48, and 0.47, respectively. Evidently, fine particles, $PM_{2.5}$ accounted for about a half of the inhalable particles in summer and much more in winter at Beijing. Such high proportion of fine particles implied that the particulate pollution at Beijing would have a great effect on the health of human being and possibly to the global environmental change, as Beijing is the pathway for the long-range transport of Asia dust that could reach north Pacific and further to west America each year, especially during the spring dust storm period (Zhuang et al., 2001, Sun et al., 2004).

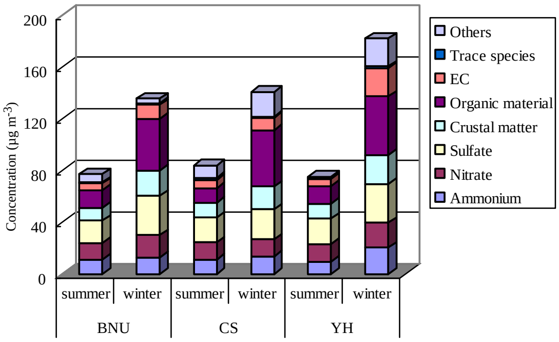

Fig. 2. Average composition in $PM_{2.5}$ at BNU, CS, and YH sites.

3.2. Chemical composition

As aerosols are mixtures of various compounds, the chemical composition of aerosol refers to those major species of those compounds that mixed in such particles. The chemical composition of the aerosols at Beijing has been speculated and the composition scheme (Fig. 2) was illustrated based on the principle of mass balance with these data generated from element, soluble ion, OC, and EC measurements. Three assumptions were made for this composition analysis as below: (1) Organic material was obtained by multiplying OC concentrations by a factor of 1.2, accounting for hydrogen and oxygen in the organic compounds. As the real chemical composition of organic compounds in aerosols has been poorly identified, the factor used in this calculation is controversial, although the conversion factors of 1.2 or 1.4 are used in the literature (Ho et al., 2003; Ye et al., 2003). In our case if a factor of 1.4 is used, it will lead to the total mass balance to be over 100%. (2) The crustal matter was calculated as the sum of those common oxides of aluminum, silicon, calcium, iron, titanium, magnesium, potassium and sodium. The concentration of Si was estimated on the basis of Si/Al concentration ratio (4.0) obtained from (Zhang et al., 2003; Taylor and McLennan, 1985), as the concentration of Si is not available in this study due to the limitation of the instruments. (3) Trace species were estimated by summing up all elements measured except those mentioned above in crustal matter, i.e. Al, Si, Ca, Fe, Ti, Mg, K, and Na. The results showed that SO_4^{2-}, NO_3^-, NH_4^+, organic material, crustal matter, and element carbon were the six dominant species in fine particle, $PM_{2.5}$, at three sites in both summer and winter seasons, which contributed 85.8–97.7% of the total mass concentration of $PM_{2.5}$. Specifically, SO_4^{2-} accounted for 16.4-26.1%, NO_3^- 9.6–17.5%, NH_4^+ 9.4–14.3%, organic material 13.3–30.9%, crustal matter 12.5–15.5%, and element carbon 6.7–12.0%. If assuming all of those SO_4^{2-} were present as ammonium sulfate, then sulfate would account for 32.6 and 25.3% of the total $PM_{2.5}$ mass concentration in summer and in winter, respectively. The sum of SO_4^{2-}, NO_3^-, and NH_4^+ contributed, in average, 53.9% and 39.3% of the total $PM_{2.5}$ mass concentration in summer and in winter, respectively. It is evident from such a composition in the aerosols in Beijing that the coal burning and the traffic exhausts were two major sources of the aerosols in Beijing.

The percentages of mass concentrations of OC and EC in the total $PM_{2.5}$ mass concentration were higher in winter than in summer, while sulfate and nitrate were higher in summer than in winter at all sites. For example, at YH site, OC and EC accounted for 17.8 and 7.9% of the total $PM_{2.5}$ mass concentration in summer, respectively, which were much lower than 24.7% and 12.0% in winter. The higher percentage of organic matter in winter could be closely related to the higher coal consumption for heating in this season. On the contrary, SO_4^{2-}, NO3$^-$, and NH_4^+ contributed 26.1, 17.5 and 12.9% of the total $PM_{2.5}$ mass concentration in summer, respectively, which were higher than 16.4, 10.6, and 11.1% in winter. The higher light intensity and the high temperature in summer favor the photochemical reaction in the atmosphere and lead to such a result that the secondary pollutants were the major source of the

Table 2
Concentrations (standard deviations in parentheses) of elements and ions in summer at three sites (Co, Cd and V as ng m^{-3}, others as μg m^{-3})

Species	BNU		CS		YH	
	PM10	PM2.5	PM10	PM2.5	PM10	PM2.5
NH_4^+	11.9(9.39)	10.4(7.74)	12.1(8.53)	11.0(6.59)	10.8(7.75)	9.75(6.78)
K^+	1.74(2.18)	1.20(1.32)	1.77(1.56)	1.53(1.24)	1.68(1.77)	1.21(1.64)
F^-	0.38(0.30)	0.11(0.11)	0.37(0.16)	0.06(0.04)	0.36(0.20)	0.10(0.06)
Cl^-	2.69(2.32)	1.93(1.50)	2.91(2.04)	1.98(1.12)	2.29(1.45)	1.69(1.30)
NO_3^-	21.1(17.3)	12.2(12.6)	18.6(13.1)	13.3(9.26)	18.2(12.6)	13.2(10.3)
SO_4^{2-}	24.8(22.3)	16.0(17.3)	24.9(19.8)	19.2(14.0)	25.4(20.4)	19.7(16.2)
As	0.02(0.02)	0.01(0.01)	0.04(0.06)	0.04(0.06)	0.03(0.03)	0.02(0.04)
Cr	0.04(0.02)	0.02(0.01)	0.04(0.02)	0.03(0.04)	0.02(0.01)	0.03(0.07)
Zn	0.33(0.26)	0.32(0.21)	0.69(0.33)	0.63(0.29)	0.42(0.26)	0.27(0.12)
Sr	0.02(0.02)	0.02(0.004)	0.05(0.02)	0.02(0.01)	0.05(0.04)	0.01(0.01)
Pb	0.11(0.09)	0.11(0.05)	0.22(0.11)	0.20(0.10)	0.11(0.08)	0.10(0.05)
Ni	0.04(0.03)	0.06(0.03)	0.05(0.05)	0.06(0.06)	0.05(0.03)	0.07(0.07)
Co	ND	3.85(0.60)	2.91(1.67)	1.56(1.00)	1.64(0.96)	0.54(0.48)
Cd	2.43(2.81)	3.69(1.99)	4.36(3.62)	3.47(2.28)	2.69(2.21)	1.70(1.01)
Fe	3.73(2.36)	0.65(0.32)	6.16(2.29)	1.91(0.99)	3.26(1.56)	1.00(0.50)
Mn	0.11(0.06)	0.03(0.02)	0.18(0.09)	0.08(0.04)	0.09(0.03)	0.05(0.02)
Mg	2.04(1.09)	0.22(0.08)	1.62(0.52)	0.19(0.18)	1.46(0.07)	0.29(0.12)
V	ND	19.0(5.45)	97.5(11.7)	59.0(27.6)	57.2(31.0)	57.1(26.5)
Ca	9.05(5.21)	0.75(0.35)	8.79(3.27)	1.02(0.34)	5.78(3.00)	0.96(0.61)
Cu	0.05(0.04)	0.04(0.03)	0.05(0.04)	0.04(0.05)	0.06(0.04)	0.07(0.08)
Ti	0.33(0.21)	0.03(0.01)	0.27(0.08)	0.04(0.01)	0.22(0.10)	0.05(0.03)
Al	5.33(3.55)	0.53(0.24)	4.77(2.02)	0.49(0.15)	3.36(1.77)	0.68(0.35)
Na	1.60(1.10)	0.39(0.23)	1.07(0.65)	0.25(0.31)	0.92(0.84)	0.27(0.18)
S	7.83(7.18)	5.89(4.84)	8.32(6.73)	6.53(4.80)	8.18(6.82)	6.68(4.91)
OC	ND	11.5(3.7)	ND	9.3(3.2)	ND	11.2(3.8)
EC	ND	5.2(2.4)	ND	6.6(3.2)	ND	5.9(2.6)

ND means not determined.

Table 3
Concentrations (standard deviations in parentheses) of elements and ions in winter at three sites (Co, Cd and V as ng m^{-3}, others as μg m^{-3})

Species	BNU		CS		YH	
	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$
NH_4^+	13.7(12.0)	12.9(10.7)	19.1(13.4)	13.3(7.18)	18.9(12.1)	20.3(10.4)
K^+	1.88(1.75)	1.94(1.80)	3.25(2.76)	1.88(1.33)	3.21(2.62)	4.23(2.10)
F^-	0.98(0.66)	0.58(0.30)	1.47(0.78)	0.58(0.33)	1.30(0.94)	0.59(0.43)
Cl^-	7.17(4.02)	6.38(3.45)	9.80(3.78)	6.57(2.07)	9.66(6.12)	7.36(4.69)
NO_3^-	18.7(18.4)	17.0(15.4)	23.2(19.2)	13.5(9.17)	27.3(19.9)	19.3(13.7)
SO_4^{2-}	34.5(32.8)	30.4(25.4)	40.5(35.4)	23.1(17.1)	46.0(39.1)	29.9(23.4)
As	0.06(0.07)	0.05(0.05)	0.09(0.06)	0.05(0.03)	0.08(0.06)	0.06(0.04)
Cr	0.04(0.06)	0.02(0.01)	0.008(0.007)	0.03(0.01)	0.03(0.02)	0.03(0.02)
Zn	0.68(0.65)	0.58(0.56)	0.87(0.73)	0.49(0.29)	1.14(0.85)	0.73(0.50)
Sr	0.06(0.03)	0.02(0.01)	0.09(0.04)	0.01(0.01)	0.11(0.06)	0.03(0.02)
Pb	0.37(0.37)	0.31(0.33)	0.46(0.40)	0.27(0.17)	0.49(0.42)	0.32(0.23)
Ni	0.11(0.11)	0.08(0.06)	0.09(0.07)	0.06(0.05)	0.13(0.08)	0.08(0.05)
Co	5.56(1.99)	3.58(1.46)	4.41(1.81)	1.14(0.83)	5.13(2.95)	2.85(1.47)
Cd	15.2(20.6)	11.2(15.9)	15.0(1.84)	7.63(7.38)	21.9(25.6)	10.6(11.8)
Fe	2.62(1.35)	1.04(0.59)	4.97(2.41)	1.06(0.38)	4.17(2.57)	1.18(0.89)
Mn	0.11(0.06)	0.08(0.05)	0.21(0.12)	0.10(0.04)	0.19(0.11)	0.10(0.06)
Mg	0.95(0.58)	0.32(0.16)	1.38(0.79)	0.28(0.09)	1.31(0.90)	0.33(0.18)
V	7.36(5.00)	2.49(2.08)	29.2(9.04)	23.2(19.3)	18.0(9.86)	12.6(10.3)
Ca	4.57(1.92)	1.67(0.68)	14.9(8.48)	2.04(0.58)	13.6(9.57)	2.44(1.81)
Cu	0.11(0.11)	0.08(0.06)	0.09(0.07)	0.05(0.03)	0.12(0.07)	0.07(0.05)
Ti	0.24(0.11)	0.07(0.03)	0.36(0.17)	0.06(0.02)	0.34(0.18)	0.080.07)
Al	4.05(2.13)	1.11(0.55)	6.08(2.87)	0.85(0.44)	5.54(3.18)	0.99(0.63)
Na	1.86(1.16)	1.15(0.79)	2.56(1.46)	1.14(0.93)	2.52(1.82)	1.28(1.10)
S	10.0(10.7)	8.71(8.57)	14.0(13.0)	7.77(6.32)	14.6(12.6)	9.44(7.55)
OC	ND	33.2(26.4)	ND	36.3(22.8)	ND	37.5(11.2)
EC	ND	11.0(9.4)	ND	9.8(6.8)	ND	21.9(12.1)

ND means not determined.

aerosols, as SO_4^{2-}, NO_3^-, and NH_4^+ accounted for much higher percentage of the total mass concentration in the summer. It must be noted that the crust matter in average accounted for 13.0–15.5% of the total $PM_{2.5}$ mass concentration in summer and 12.5–14.5% in winter with a relatively even spatial distribution at three sites over Beijing. This result indicated that the dust, including those from local re-suspended soil and those transported from out of the city, was also one of the major sources of the fine particles in Beijing, where the long-ranged transported dust from Asia desert and from the dry and semi-dry areas often invaded and passed through.

3.3. Spatial variations

PM_{10} showed clear spatial variations in winter. The concentrations of PM_{10} at YH site were the highest of the three sites of different types. Much more coal in winter at the dense population residential area, such as YH site, was used for heating, which would produce more pollution aerosols. Also, the northwest wind in winter would bring the industrial pollutants from CS downwind to the YH site, making it the most polluted site of the three sites in the winter. At the industrial site CS, the increased coal consumption in winter plus the industry emission both led to its pollution level being higher than the traffic site, BNU. In summer YH site showed the least concentration of PM_{10} compared with the other two sites, indicating that this residential site has less particulate pollution sources in this season. At the traffic site, BNU, the higher $PM_{2.5}/PM_{10}$ ratio than that at the other two sites might be likely due to the greater traffic flow at this site that emitted more exhausts and, in turn, to transform to more fine particles.

However, the concentrations of $PM_{2.5}$ at different sites had no big difference both in summer and in winter. $PM_{2.5}$, normally to be defined as fine particles, could be transported to a much longer range for its longer residence time in the atmosphere. Thus, $PM_{2.5}$ fine particles could likely inter-mix or intra-mix well with each other in the atmosphere and lead the spatial distribution to be even.

The concentrations of various chemical species at a specific location are mainly determined by the distance from their sources, which would reflect the emissions from those point-sources. However, for those elements with multi-sources, their spatial variations were significantly different in different seasons. Tables 2 and 3 showed the concentrations of various chemical species in PM_{10} and $PM_{2.5}$. The average concentrations of Al in PM_{10} were 5.33, 4.77 and 3.36 $\mu g\,m^{-3}$ in summer, and 4.05, 6.08, and 5.54 $\mu g\,m^{-3}$ in winter at BNU, CS and YH sites, respectively. The highest concentration of Al at BNU site in summer was likely due to the re-suspension of road dust by wind and motor vehicles as well as the construction dust from construction sites nearby. While in winter the highest concentration of Al at CS site could be related to the coal fly ash from this industry company plus the re-suspended road dust. CS site showed the highest concentration and YH site appeared to be the lowest for elements Fe, Zn and Pb both in PM_{10} and $PM_{2.5}$ in summer. The average concentration of Zn in PM_{10} and $PM_{2.5}$ at CS in summer was 0.69 and 0.63 $\mu g\,m^{-3}$, respectively, both higher than those at the other two sites. For Pb, the concentration at CS site was nearly twice higher than that at the other two sites. As the CS site is located at a place close to the CS, the largest industrial emission of pollution elements, such as Zn and Pb with elements such as Fe and Mn, it is no wonder that CS site showed the highest concentration for these elements compared to other sites. YH site is a residential area that is relatively clean in summer, thus it showed the lowest concentration of these pollution elements among the three sites. In winter, CS site still showed the highest concentration of Fe, this no doubt was related to the industrial emission. However, YH site showed the highest concentrations of Zn and Pb in winter. This might be due to the coal consumption plus the industrial pollutants brought by northwest wind from upwind area, e.g. CS. Although the leaded gasoline has been phased out since 1997 at Beijing, the concentration of Pb in aerosol at Beijing is still considerably high compared with that in the cities of USA. The highest concentration of Pb at the industrial site, CS, indicated that the industrial emission might be an important source of Pb in Beijing, although the exact sources of this high Pb in the aerosol was unclear at this stage. $PM_{2.5}$, as mentioned above, had relatively even spatial distribution, so did for the various chemical species. For example, in winter the average concentrations of Fe were 1.04, 1.06, and 1.18 $\mu g\,m^{-3}$ at BNU, CS, and YH sites, respectively, which showed no big difference over Beijing. Sulfate and nitrate also showed relatively even distribution at the three sites over Beijing in both summer and winter seasons, as both sulfate and nitrate were mainly transformed by the heterogeneous or homogeneous reactions of their precursors such as SO_2 and NO_x, which would not strongly affected by those point-source emissions. This could reveal that the long-range transported aerosol might be one of the important sources of sulfate and nitrate in Beijing aerosol.

3.4. Temporal and seasonal variations

Fig. 3 shows the concentrations of six elements (Fe, Ca, Al, Zn, Pb and Cu) and three ions (NH_4^+, NO_3^- and SO_4^{2-}) in PM_{10} and $PM_{2.5}$ collected at YH site as a function of the time when samples were collected. The concentrations of these elements and ions showed significant variations from day to day. The concentrations in PM_{10} were in the range 0.47–10.60 $\mu g\,m^{-3}$ for Fe, 0.71–35.76 $\mu g\,m^{-3}$ for Ca, 0.35–12.82 $\mu g\,m^{-3}$ for Al, 0.038–3.45 $\mu g\,m^{-3}$ for Zn, 0.0041–1.74 $\mu g\,m^{-3}$ for Pb, 0.016–0.27 $\mu g\,m^{-3}$ for Cu, 1.26–40.60 $\mu g\,m^{-3}$ for NH_4^+, 3.57–65.60 $\mu g\,m^{-3}$ for NO_3^-, and 3.30–121.11 $\mu g\,m^{-3}$ for SO_4^{2-}. Similarly, these elements and ions in $PM_{2.5}$ varied greatly, for example, the concentrations varied in the range 0.29–2.36 $\mu g\,m^{-3}$ for Al, 0.067–1.96 $\mu g\,m^{-3}$ for Zn, and 3.49–77.09 $\mu g\,m^{-3}$ for SO_4^{2-}. The temporal variations are mainly due to the meteorological factors, such as temperature, relative humidity, and wind speed, which favor or adversely affect the dispersion of pollutants. In addition, changes in source emission strength with time also affects these variations.

Seasonal variations of chemical species were also illustrated in Fig. 3. Overall, the concentrations of those elements and ions measured in winter were ~1–3 times higher than those in summer. Table 4 showed the concentrations of SO_2, NO_2, CO, and O_3 in the period studied. The average concentrations of SO_2, NO_2, and CO in winter were 156, 98.2, and 8.4 $mg\,m^{-3}$, which were 10, 1.5, and 2.9 times higher than those in summer, respectively. It is believed that the industrial emissions would remain reasonably constant throughout the year. Thus, the much higher concentration of SO_2 in winter, which would, in turn, lead to the higher sulfate aerosol in winter, must be from the much higher consumption of the domestic coal for heating in this season. Yao et al. (2002) also found strong correlations between sulfate and SO_2 in winter. The higher concentrations of NO_2 and CO in winter were likely to be from the higher consumption of coal, which would emit more NO_2 and CO, plus the higher emission from the automobiles, which would produce higher exhausts due to cold start in the winter. The higher concentration of nitrate in winter could associate with the lower temperature, which favors the shift from the gas phase of nitric acid to the particle phase of ammonium nitrate (Stelson and Seinfeld, 1982). However, the meteorological variations could also be the factors affecting the degree of the air pollution. Fig. 4 presented those meteorological factors during the period studied. The low wind speed and temperature in winter favor the accumulation of pollutants, while the high temperature in summer favors the air convection and the dispersion of pollutants. In

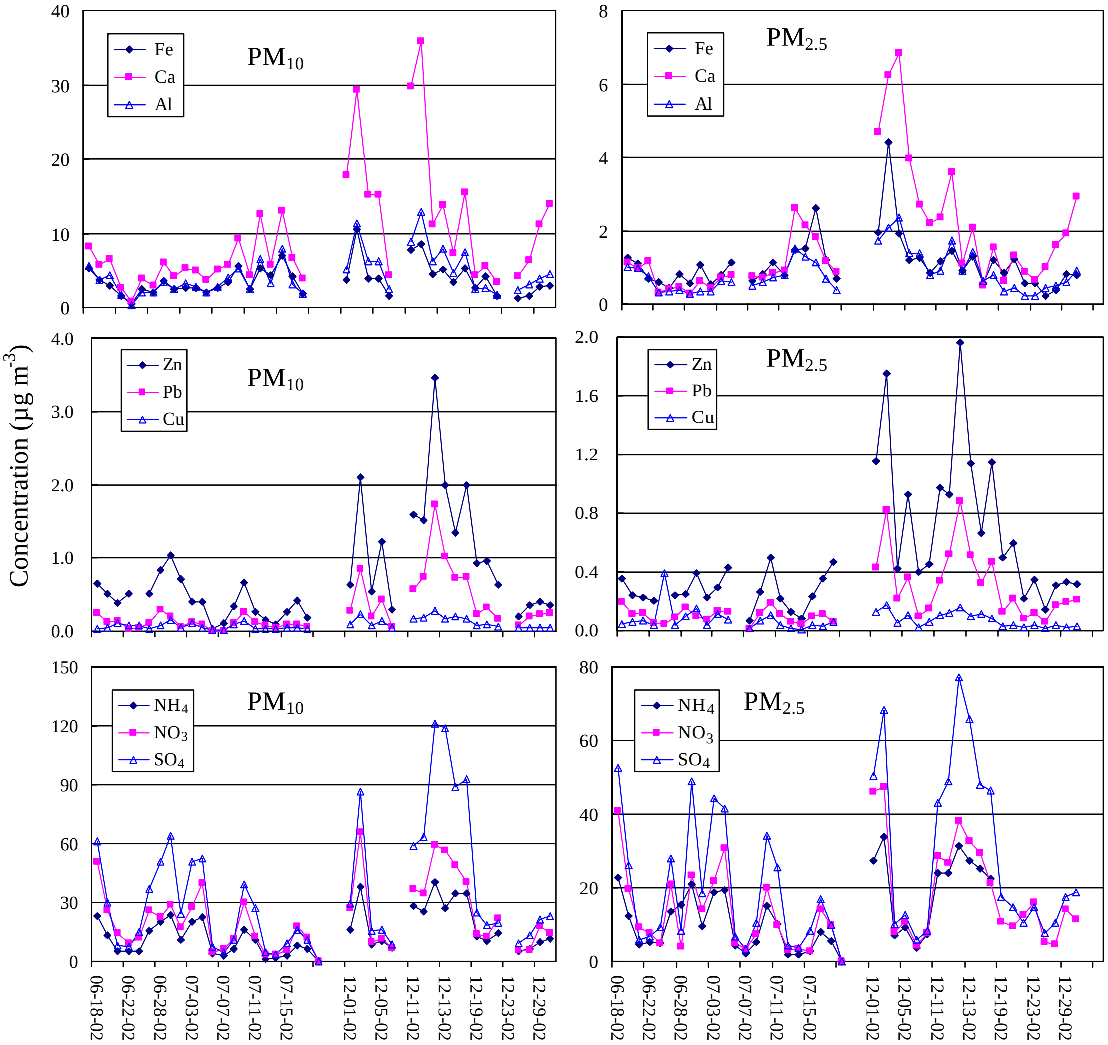

Fig. 3. Variations of selected elements and ions in PM_{10} and $PM_{2.5}$ with time at YH site.

Table 4
Concentrations of SO_2, NO_2, CO and O_3 during sampling period (CO as $mg\,m^{-3}$, others as $\mu g\,m^{-3}$)

Species	Summer					Winter				
	Mean	SD	Min	Max	N	Mean	SD	Min	Max	N
SO_2	15	6	7	33	33	156	55	56	261	31
NO_2	64.4	11.9	38.4	88.8	33	98.2	34.9	38.4	180.8	31
CO	2.9	0.9	1.5	5	33	8.4	6.7	1.9	31.5	29
O_3	93.0	46.3	14.4	177.6	33	20.6	14.3	7.2	50.4	29

addition, the barer surface in winter would re-suspend more dust while more precipitation in summer would wash out more particles. It must be noted that in summer the higher O_3 concentration of $93.0\,\mu g\,m^{-3}$, 4.5 times higher than that of $20.6\,\mu g\,m^{-3}$ in winter, would favor the photochemical reactions and form more secondary particulates, as mentioned above that SO_4^{2-}, NO_3^-, and NH_4^+ accounted for much higher percentage

(26.1, 17.5 and 12.9%) of the total mass concentration in the summer than that (16.4, 10.6, and 11.1%) in winter. The meteorological factors plus the higher concentrations of O_3 in summer determine that the aerosol particles in summer at Beijing could be characterized specifically by the secondary pollutants.

3.5. *Sources of Beijing aerosol—factor analysis*

Factor analysis (FA) was performed with the elemental and ionic data set of PM_{10} and $PM_{2.5}$ at three sites to identify the sources of Beijing aerosols. To maximize the source-identification power, 19 species (As, Zn, Pb, Ni, Cd, Fe, Mn, Mg, Ca, Cu, Ti, Al, Na, NH_4^+, K^+, Cl^-, F^-, NO_3^-, SO_4^{2-}) were selected after excluding those species without enough reliable data for the statistical work due to part of which could be below the detection limit. The results of FA for PM_{10} and $PM_{2.5}$ were shown in Tables 5 and 6, respectively. Four factors were identified for PM_{10}, and they explained 88.3% of total variance of the data set. The communalities for all species were higher than 0.82, indicating that the four factors identified were satisfactory. The first factor that explained the most of the variance (60.5%) was heavily loaded with As, Zn, Pb, Ni, Cd, and Cu, representing the combined sources of industry and motor vehicle emission. The metallurgical processes could produce the largest emissions of Cu, Ni, and Zn (Pacyna, 1998), while exhaust emissions from road vehicles could also contain various amounts of Cu, Zn, and Ni (Pacyna, 1986, Lee et al., 1999). As leaded gasoline has been banned since 1997 in Beijing, the emission of those vehicles should not be the dominant source of Pb in Beijing aerosols. The industrial emission, transport from those areas outside of Beijing, and those re-suspended dust could be the possible sources of Pb, and this needs to be further studied. The second factor showed high loadings for Fe, Mn, Mg, Ca, Ti, Al, and Na, which explained 17.8% of the total variance. This factor was clearly associated with the mineral aerosols that would be likely from the re-suspended road dust and from the long-range transported dust from outside Beijing. The third factor was highly loaded with NO_3^-,

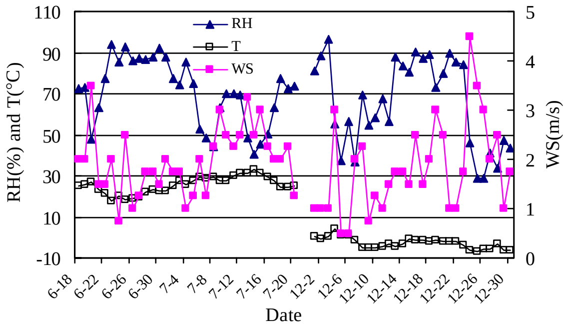

Fig. 4. Meteorological factors during sampling period.

Table 5
Varimax rotated factor loading matrix for chemical species of PM_{10} combined from three sites

Species	Factor 1	Factor 2	Factor 3	Factor 4	Communality
	Industry and motor vehicles	Road dust	Secondary	Incineration/coal burning	
NH_4^+	0.55	0.16	0.76	0.23	0.96
NO_3^-	0.39	0.25	0.85	0.16	0.90
SO_4^{2-}	0.61	0.13	0.73	0.15	0.90
K^+	0.34	0.36	0.70	0.36	0.95
F^-	0.33	0.32	0.24	0.81	0.94
Cl^-	0.43	0.25	0.35	0.77	0.95
As	0.79	0.11	0.25	0.24	0.82
Zn	0.84	0.24	0.35	0.17	0.92
Pb	0.88	0.19	0.30	0.22	0.96
Ni	0.80	0.14	0.28	0.30	0.99
Cd	0.90	0.02	0.14	0.05	0.85
Fe	0.15	0.87	0.16	0.00	0.88
Mn	0.53	0.68	0.19	0.18	0.90
Mg	−0.09	0.92	0.18	−0.03	0.91
Ca	0.11	0.82	0.03	0.43	0.87
Cu	0.76	0.16	0.31	0.27	0.99
Ti	0.17	0.88	0.14	0.28	0.96
Al	0.18	0.90	0.14	0.26	0.96
Na	0.35	0.62	0.22	0.59	0.91
% variance	60.5	17.8	5.1	5.0	88.3

Table 6
Varimax rotated factor loading matrix for chemical species of $PM_{2.5}$ combined from three sites

Species	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Communality
	Secondary	Road dust	Metallurgical emission	Motor vehicles	Nonferrous metal smelter	Incineration/ coal burning	
NH_4^+	0.82	0.12	0.07	0.32	0.30	0.20	0.93
NO_3^-	0.90	0.18	0.08	0.24	0.16	0.17	0.64
SO_4^{2-}	0.83	0.16	0.07	0.29	0.33	0.17	0.84
K^+	0.58	0.12	0.16	−0.04	0.25	0.48	0.89
F^-	0.18	0.35	0.04	0.07	0.25	0.81	0.92
Cl^-	0.31	0.38	0.09	0.21	0.22	0.75	0.94
As	0.20	0.25	0.12	0.11	0.79	0.15	0.72
Zn	0.47	0.09	0.32	0.38	0.56	0.23	0.86
Pb	0.43	0.21	0.23	0.29	0.68	0.33	0.95
Ni	0.27	0.12	0.03	0.89	0.24	0.13	0.92
Cd	0.27	0.03	0.00	0.25	0.80	0.20	0.80
Fe	0.09	0.22	0.95	0.00	0.10	0.05	0.71
Mn	0.30	0.29	0.58	0.21	0.40	0.45	0.89
Mg	0.07	0.86	0.08	0.14	0.28	0.07	0.76
Ca	−0.02	0.74	0.25	0.04	0.06	0.51	0.82
Cu	0.27	0.11	0.04	0.90	0.22	0.11	0.92
Ti	0.28	0.77	0.18	0.03	−0.02	0.29	0.73
Al	0.17	0.84	0.09	0.10	0.14	0.30	0.84
Na	0.29	0.37	0.07	0.21	0.21	0.69	0.76
% variance	53.7	13.3	6.1	5.7	4.9	3.6	87.4

SO_4^{2-}, NH_4^+, and it clearly indicated that this factor was mainly from the coal burning and, in turn, the chemical transformation that formed the secondary aerosols, as it is shown mainly to be ammonium sulfate and ammonium nitrate. The fourth factor had significant loadings for Cl^- and F^-, and this could likely represent the waste incineration and possibly part of the coal burning. Many refuse dumps in Beijing (see Fig. 1) could emit large amounts of pollutants such as HCl and HF and other elements such as Cr, Cu, Pb, and Mn when being smashed or incinerated. These pollutants from waste incineration could be transported to the whole urban area and became one of the important sources of Beijing aerosol. Chueinta et al. (2000) found that Cl^- was possibly from the polyvinylchloride plastic in the trash-burning, while the high concentration of Cl^- in winter was also likely to be associated with coal burning (Yao et al., 2002).

Six factors were retained when applying FA to $PM_{2.5}$, which could explain 87.4% of the total variance. Communalities ranged from 0.64 (for NO_3^-) to over 0.90 for the remaining species. The first factor represented the secondary aerosols with high loadings for NO_3^-, SO_4^{2-}, and NH_4^+, and it explained 53.7% of the total variance. The second factor was associated with Mg, Ca, Ti, and Al, and it could represent the mineral aerosols that would likely be to be from the re-suspended road dust and from the long-range transported dust from outside Beijing as mentioned above. The third factor had high loading for Fe and moderate loading for Mn, and this could indicate the metallurgical emission, which was most likely from the CS at Beijing. As Mn was normally associated with ferrous metal processing (Kumar et al., 2001) and Fe was also enriched in fly ash from coal combustion or natural origin (Gao and Anderson, 2001), these sources could also possibly contribute to this factor. The fourth factor had high loadings for Ni and Cu, and it could represent the source from motor vehicles emission. Residual oil combustion was well known to be the source of Ni and V in aerosols (Swietlicki and Krejci, 1996). As the data for V in this study was not enough to be considered in this statistical work, it was not very clear to interpreter the fourth factor. The fifth factor was loaded with As, Zn, Pb, and Cd, and this could represent clearly the industrial emissions, mostly from those non-ferrous smelters. Lee et al. (1999) resolved a similar factor to the fifth factor discussed here when studying the source of particulate pollutants in Hong Kong. The sixth factor was associated with Cl^- and F^-, and it could represent the sources from the waste incineration and part of the coal burning, as mentioned above.

3.6. *Sources of mineral aerosol from outside Beijing*

We developed a new element tracer technique to estimate, mainly, the relative contributions of mineral aerosol from outside Beijing to the total mineral aerosol.

To be an ideal tracer used in distinguishing the local sources of mineral aerosols from outside the city, it should be derived and emitted stably from both inside city and outside city. To be a source signature it also should be stable and distinct during the transport of the aerosol (Zhang et al., 1996). Although there is neither an ideal tracer nor a specific element that is unique to a specific source region, it is possible to find a pair of elements, whose ratio could distinguish the sources both inside and outside the city. After collecting and analyzing a great number of surface soil samples collected from many typical places over entire Beijing, from many dust source areas, we found that the ratio of Mg/Al in the aerosols collected at Beijing could be the best tracer that met the two basic requirements (the distinct signature and the stability during transport). The detailed principle and the procedures of this technique were given elsewhere (Han et al., 2004).

It has been believed that the dust from the north and northwest of Beijing are the two main sources of the Beijing mineral aerosols (Ren et al., 2003). The strong north or northwest wind, especially in spring and winter, would carry a lot of dusts and invade Beijing, and those dusts transported from outside Beijing become part of Beijing mineral aerosol. It has been reported that 91.4% particles in the loss of Inner Mongolia can form mineral aerosols, while only 15.6% particles in dust soil from desert areas of Gansu and 7.2% in coal ash of Huhehot, a city located in north of Beijing, can form mineral aerosols (Liu et al., 2002). It is clear that the loess in Inner Mongolia would be the major no-local source of the mineral aerosols in Beijing. Duolun is located in the desert area in Inner Mongolia, an outside source area of the mineral aerosol at Beijing. Fengning in Hebei province, which is located between Beijing and Duolun, is the necessary pathway through which mineral aerosols in Duolun were transported to Beijing. The ratios of Mg/Al in the dust soil of Duolun and Fengning were 0.12 and 0.15, respectively. The average ratio of Mg/Al (0.14) of soil at Duolun and Fengning could be used as the representative of the non-local source from north of Beijing. Dingling, located in northwest of Beijing, is on the pathway of the non-local sources from northwest of Beijing. Huhehot of Inner Mongolia is located in the west-northwest of Beijing. The ratio of Mg/Al in the surface soil collected from Dingling (0.23) is similar to that in the loess collected from Huhehot (0.21), and it is also similar to that in the Loess Plateau at Luochuan of Shanxi province (0.19). Thus, the average ratio of Mg/Al of these loess samples (0.21) could be used as the representative of non-local sources from west/northwest of Beijing. As a tracer to be used to estimate the contribution of the non-local sources, it is acceptable to use the average ratio of Mg/Al (0.175) of soil samples (from the two source areas in west/northwest of Beijing (0.21) and in north of Beijing (0.14)) as the representative of non-local sources of mineral aerosols in Beijing. The ratio of Mg/Al in surface soil collected from those typical places all over Beijing ranged from 0.28 to 0.61, varying by a factor of only 1~2. In addition, the ratios are all higher than that (Mg/Al = 0.175) in non-local sources of mineral aerosols in Beijing, which satisfied the basic requirement to distinguish the non-local source from the local source. Thus, it is reasonable to use the average ratio (0.45) of Mg/Al of those surface soil samples at Beijing to represent the local sources.

We are now able to estimate the relative contributions of sources of mineral aerosols to total mineral aerosols from both inside and outside Beijing. Suppose the contributions of mineral aerosol from inside and outside Beijing to the total mineral aerosol are m and n, if the compositions of mineral aerosol from the source regions are constant during the transport, the following equations are given:

$$(\mathrm{Mg/Al})_{\mathrm{aerosol}} = m \times (\mathrm{Mg/Al})_{\mathrm{local}} + n \times (\mathrm{Mg/Al})_{\text{non-local}}, \tag{1}$$

$$m + n = 1, \tag{2}$$

where $(\mathrm{Mg/Al})_{\mathrm{aerosol}}$ is the average ratio of Mg/Al in aerosols at Beijing, $(\mathrm{Mg/Al})_{\mathrm{local}}$ is the average ratio of Mg/Al of the soil samples collected from all over Beijing, $(\mathrm{Mg/Al})_{\mathrm{non-local}}$ is the average ratio of Mg/Al of the soil samples from sources outside Beijing.

The results of the average relative contributions of mineral aerosol from inside and outside Beijing to the total mineral aerosol were listed in Table 7, which were calculated according to the equations above. In summer, the sources from outside Beijing contributed 19% and 20% of the total mineral aerosol in PM_{10} and in $PM_{2.5}$, respectively. In winter the sources from outside Beijing contributed 79% and 37% of the total mineral aerosol in PM_{10} and in $PM_{2.5}$, respectively. It could also be seen that in the spring of 2002, the sources from outside Beijing accounted for 69% and 76% of the total mineral aerosol in PM_{10} and in $PM_{2.5}$, respectively. Moreover, during the dust storm period from 20 to 22 March, it reached up to 97% in TSP, 79% in PM_{10} and 76% in $PM_{2.5}$, respectively. These results clearly demonstrated that the sources from outside Beijing contributed significantly to the mineral aerosol in Beijing, and much more in spring/winter than in summer. As in spring/winter much more frequent and stronger north or northwest wind bring much more dust from outside Beijing and result in the much greater contributions of non-local sources to the total mineral aerosols at Beijing. Especially, dust storm in spring could carry large amounts of dusts and became the main sources of mineral aerosols in Beijing. It is evident that the results are quite coincident with the actual facts, as shown in the case of dust storm. The results calculated using this technique quantitatively indicated that the air-borne

Table 7
Ratios of Mg/Al in aerosol and soil samples, and contribution of mineral aerosol from outside Beijing to the total mineral aerosol of Beijing

Types	Mean	Min	Max	Contribution (%)
TSP-spring	0.27	0.20	0.34	62
TSP-dust storm	0.24	0.17	0.30	72
PM_{10}-spring	0.25	0.19	0.30	69
PM_{10}-dust storm	0.24	0.22	0.25	72
PM_{10}-summer	0.40	0.25	0.70	19
PM_{10}-winter	0.22	0.12	0.32	79
$PM_{2.5}$-spring	0.23	0.18	0.28	76
$PM_{2.5}$-dust storm	0.24	0.23	0.26	72
$PM_{2.5}$-summer	0.40	0.21	0.72	20
$PM_{2.5}$-winter	0.35	0.12	0.73	37
Beijing soils	0.46			
Dingling soils	0.23			
Duolun soils	0.12			
Fengning soils	0.15			
Luochuan soils[a]	0.19			
Loess in Inner Mongolia[b]	0.21			

[a] Nishikawa et al. (1991).
[b] Liu et al. (2002).

particulate pollution at Beijing were not only from the local sources but also from outside Beijing. This is the technique firstly developed for estimating the relative contributions of sources from inside and outside Beijing to the total mineral aerosol at Beijing and it could provide the basic information in controlling the air-borne particulate pollution at Beijing.

4. Conclusions

The air-borne particulate pollution has been very serious at Beijing and it is even worse in winter than in summer. Fine particles ($PM_{2.5}$) were the major part of inhalable particles (PM_{10}), as the ratios of $PM_{2.5}/PM_{10}$ were 0.45–0.48 in summer and 0.52–0.73 in winter. SO_4^{2-}, NO_3^-, NH_4^+, organic matter, crustal matter, and element carbon were the six dominant species. Sulfate could account for 32.6 and 25.3% of the total $PM_{2.5}$ mass concentration in summer and in winter, respectively. Secondary aerosol (SO_4^{2-}, NO_3^-, and NH_4^+), road dust or/and long-range transported dust from outside Beijing, industry and motor vehicle emission and coal burning were the major contributors to the air-borne particulate pollution in Beijing. Overall, coal burning and the traffic exhausts, plus the dust from the long-range transport, could be the major sources of the aerosol pollution at Beijing. A relatively even spatial distribution of chemical species in $PM_{2.5}$ was found while in PM_{10} a significant variation with the highest concentrations at the industrial site in summer and at the residential site in winter was observed. The concentrations of PM_{10}, $PM_{2.5}$ as well as various chemical species were higher in winter than in summer when aerosol could be characterized by the secondary pollutants. The contributions of mineral aerosol from outside Beijing were first estimated with a newly developed element tracer. The aerosol from outside Beijing accounted for 79% and 37% to the total mineral aerosol in PM_{10} and $PM_{2.5}$ in winter, and 19% and 20% in summer, respectively. This is the technique firstly developed for estimating the relative contributions of sources from inside and outside Beijing to the total mineral aerosol and it could provide the basic information in controlling the air-borne particulate pollution at Beijing.

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References

Cao, L., Tian, W., Ni, B.F., Zhang, Y., Wang, P., 2002. Preliminary study of airborne particulate matter in a Beijing sampling station by instrumental neutron activation analysis. Atmospheric Environment 36, 1951–1956.

Chameides, W.L., Yu, H., Liu, S.C., Bergin, M., Zhou, X., Mearns, L., Wang, G., Kiang, C.S., Saylor, R.D., Luo, C., Huang, Y., Steiner, A., Giorgi, F., 1999. Case study of the effects of atmospheric aerosols and regional haze on agriculture: an opportunity to enhance crop yields in China through emission controls? Process Natural Academy Science (USA) 96 (24), 13626–13633.

Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley, J.A., Hanse, J.E., Hofmann, D.J., 1992. Climate forcing by anthropogenic aerosols. Sciences 255, 423–430.

Chen, Z., Ge, S., Zhang, J., 1994. Measurement and analysis for atmospheric aerosol particulates in Beijing. Research of Environmental Sciences 7 (3), 1–9 (in Chinese).

Chi, X., Di, Y., Dong, S., Liu, X.D., 1999. Determination for organic carbon and element carbon in atmospheric particles. China Environmental Monitor 15 (4), 11–13.

Chueinta, W., Hopke, P.K., Paatero, P., 2000. Investigation of sources of atmospheric aerosol at urban and suburban residential areas in Thailand by positive matrix factorization. Atmospheric Environment 34, 3319–3329.

Dan, M., Zhuang, G., Li, X., Tao, H., Zhuang, Y., 2004. The characteristics of carbonaceous species and their sources in $PM_{2.5}$ in Beijing. Atmospheric Environment 38, 3443–3452.

Dockery, D.W., Pope, C.A., 1994. Acute respiratory effects of particulate air pollution. Annual Review of public Health 15, 107–132.

Dong, J., Yang, S., 1998. Characteristics of the aerosol and study of their sources in Huabei clean area. Environment Chemsitry 17 (1), 38–44.

Elliott, S., Shen, M., Blake, D.R., Lu, R., Russell, A.G., Kao, C.Y.J., Streit, G.E., Zhao, X.P., McCreary, E.I., Rowland, F.S., Brown, M.J., Turco, R.P., 1997. Atmospheric effects of the emerging mainland Chinese transportation system at and beyond the regional scale. Journal of Atmospheric Chemistry 27 (1), 31–70.

Gao, Y., Anderson, J.R., 2001. Characterization of Chinese aerosols determined by individual-particle analysis. Journal of Geophysical Research 106 (D16), 18037–18045.

Han, L., Zhuang, G., Sun, Y., Wang, Z., 2004. Local and non-local sources of airborne particle pollution at Beijing—A new elemental tracer technique for estimating the contributions of mineral aerosols from out of the city. Science in China (Series D), in press.

He, K., Yang, F., Ma, Y., Zhang, Q., Yao, X., Chan, C.K., Cadle, S., Chan, T., Mulawa, P., 2001. The characteristics of $PM_{2.5}$ in Beijing, China. Atmospheric Environment 35, 4959–4970.

Ho, K.F., Lee, S.C., Chan, C.K., Yu, J.C., Chow, J.C., Yao, X., 2003. Characterization of chemical species in $PM_{2.5}$ and PM_{10} aerosols in Hong Kong. Atmospheric Environment 37, 31–39.

Kumar, A.V., Patil, R.S., Nambi, K.S.V., 2001. Source appointment of suspended particulate matter at two traffic junctions in Mumbai, Indian. Atmospheric Environment 35, 4245–4251.

Lee, E., Chan, C.K., Paatero, P., 1999. Application of positive matrix factorization in source apportionment of particulate pollutants in Hong Kong. Atmospheric Environment 33, 3201–3212.

Liu, C., Zhang, J., Liu, S., 2002. Physical and chemical characters of materials from several mineral aerosol sources in China. Environmental Science 23 (4), 28–32 (in Chinese).

Nishikawa, M., Kanamori, S., Kanamori, N., Mizoguchi, T., 1991. Kosa aerosol as eolian carrier of anthropogenic material. The Science of Total Environment 107, 13–27.

Ostro, B.D., Hurley, S., Lipsett, M.J., 1999. Air pollution and daily mortality in the Coachella Valley, California: a study of PM_{10} dominated by coarse particles. Environment Research 81, 231–238.

Pacyna, J.M., 1986. In: Nriagu, J.O., Davidson, C.I. (Eds.), Toxic Metals in the Atmosphere. Wiley, New York.

Pacyna, J.M., 1998. Source inventories for atmospheric trace metals. In: Harrison, R.M., van Grieken, R.E. (Eds.), Atmospheric Particles, IUPAC Series on Analytical and Physical Chemistry of Environmental Systems, Vol. 5. Wiley, Chichester, UK pp., 385–423.

Ren, Z., Gao, Q., Su, F., Wang, Y., Zhang, Z., Yang, X., 2003. The regional characteristics of the atmosphere environment and the impact of dust-storm in Beijing. Engineer Science 5 (2), 49–56 (in Chinese).

Schwartz, J., Dockery, D.W., Neas, L.M., 1996. Is daily mortality associated specifically with fine particles? Journal of Air and Waste Management Association 46, 927–939.

Stelson, W.T., Seinfeld, J.H., 1982. Relative humidity and temperature dependence of the ammonium nitrate dissociation constant. Atmospheric Environment 16, 983–992.

Swietlicki, E., Krejci, R., 1996. Source characterization of the Central European atmospheric aerosol using multivariate statistical methods. Nuclear Instrument and Method in Physics Research Section B 109/110, 519–525.

Sun, Y., Zhuang, G., Yun, H., Zhang, X., Guo, J., 2004. Characteristics and sources of 2002 super dust storm in Beijing. China Science Bulletin 49 (8), 698–705.

Taylor, S.R., McLennan, S.M., 1985. The Continental Crust: Its Composition and Evolution. Blackwells, Oxford, England.

Twomey, S., 1974. Pollution and the planetary albedo. Atomspheric Environment 8, 1251–1256.

US Environmental Protection Agency (US EPA), 1997. National Ambient Air Quality Standards for Particulate Matter, Final Rule. Federal Register. 1997, 62 (138), Part II, EPA, 40 CFR Part 50, 18 July.

Wang, X., 1999. Determination of concentrations of elements in atmospheric aerosol of urban and rural areas of Beijing in winter. Biological Trace Element Research 71–72, 203–208.

Wilson, W.H., Suh, H.H., 1997. Fine particles and coarse particles: concentration relationships relevant to epidemiological studies. Journal of the Air and Waste Management Association 47, 1238–1249.

Winchester, J.W., Bi, M., 1984. Fine and coarse aerosol composition in an urban setting: a case study in Beijing. Atmospheric Environment 18 (7), 1399–1409.

Wolf, M.E., Hidy, G.M., 1997. Aerosols and climate: anthropogenic emissions and trends for 50 years. Journal of Geophysical Research [Atoms.] 102 (D10), 11113–11121.

Yang, S., Dong, J., Cheng, B., 2000. Characteristics of air particulate matter and their sources in urban and rural area of Beijing, China. Journal of Environment Science 12 (4), 402–409.

Yao, X., Chan, C.K., Fang, M., Cadle, S., Chan, T., Mulawa, P., He, K., Ye, B., 2002. The water-soluble ionic composition of $PM_{2.5}$ in Shanghai and Beijing, China. Atmospheric Environment 36 (26), 4223–4234.

Yao, X., Lau, A.P.S., Fang, M., Chan, C.K., Hu, M., 2003. Size distribution and formation of ionic species in atmospheric particulate pollutants in Beijing, China: 1-inorganic ions. Atmospheric Environment 37, 2991–3000.

Ye, B., Ji, X., Yang, H., Yao, X., Chan, C.K., Cadle, S.H., Chan, T., Mulawa, P.A., 2003. Concentration and chemical composition of $PM_{2.5}$ in Shanghai for a 1-year period. Atmospheric Environment 37, 499–510.

Yuan, H., Wang, Y., Zhuang, G., 2003. The simultaneous determination of organic acid, MSA with inorganic anions in aerosol and rain-water by ion chromatography. Journal of Instrumental Analysis 6, 12–16 (in Chinese).

Zhang, X., Zhang, G., Zhu, G., Zhang, D., An, Z., Chen, T., Huang, X., 1996. Elemental tracers for Chinese source dust. Science in China 39 (5), 512–521.

Zhang, X., Gong, S., Shen, Z., Mei, F., Xi, X., Liu, L., Zhou, Z., Wang, D., Wang, Y., Cheng, Y., 2003. Characterization of soil dust aerosol in China and its transport and distribution during 2001 ACE-Asia: 1. Network observations. Journal of Geophysical Research [Atmos.] 108(D9), ACH 3/1–ACH 3/13.

Zhuang, G., Guo, J., Yuan, H., Zhao, C., 2001. The compositions, sources, and size distribution of the dust storm from China in spring of 2000 and its impact on the global environment. China Science Bulletin 46 (11), 895–901.

Zhuang, G., Guo, J., Yuan, H., Zhang, X., 2003. Coupling and feedback between Iron and sulphur in air-sea exchange. China Science Bulletin 48 (11), 1080–1086.