



Chemical and strontium isotope characterization of rainwater in Beijing during the 2008 Olympic year

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ABSTRACT

To improve the air quality during the 2008 Beijing Olympic Games, the Beijing Municipal Government enforced a series of emission control measures. This provided a valuable case study to evaluate the effectiveness of these aggressive measures on mitigating air pollution and improving the atmospheric environment. In this paper, we report the first results of the chemical and Sr isotopic compositions of the rainwater in Beijing during the 2008 Olympic year. The ionic composition of samples show that Ca^{2+} and NH_4^+ were the dominant cations, accounting for about 76–97% of the total cations, and that SO_4^{2-} and NO_3^- were the major anions, accounting for 61–91% of the total anions measured. Using Na as the indicator of marine origin and Al as that of terrestrial inputs, the proportions of sea salt and terrestrial elements were estimated from elemental ratios. The calculated results indicate that the major chemical components were mainly of non-sea-salt origin. Good correlation between Ca^{2+} and Sr^{2+} ($R^2 = 0.85$) in rainwater samples indicates the potential of Sr as a provenance tracer for Ca. Sr^{2+} concentrations in rainwater samples ranged from 0.01 to $2.87 \mu\text{mol l}^{-1}$, with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from 0.7092 to 0.7109. All of the samples had a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio higher than that of seawater. The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic and elemental ratio systems show that the data set of rainwater was mainly distributed between the seawater end-member (~ 0.70917) and the soil dust end-member (0.7111–0.7115), and the analysis indicate that the effect of anthropogenic inputs could decrease in 2008. Comparisons of the chemical composition of the rainwater in different periods show that all chemical components, except NH_4^+ , had the lowest concentration levels during the Olympic Game period. The VWM concentrations of major ions in the rainwater decreased significantly during the Olympic period by up to 29% for SO_4^{2-} , 39% for NO_3^- , 38% for Ca^{2+} , 51% for Mg^{2+} , 57% for K^+ , and 44% for Na^+ , compared with the pre-control period. The concentrations of almost all species decreased during the 2008 Olympics, most likely due to both strict emission controls and favorable meteorological conditions.

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1. Introduction

The 29th Olympic and Paralympics Games were held between August 8th and September 17th, 2008, in Beijing, People's Republic of China. To achieve good air quality

and maintain clean air throughout the Olympic Games, the Beijing Municipal Government enacted a series of emission control measures and regulations before and during the Games. These included improvement of the energy structure and reduction of local power generation by importing electricity from the surrounding areas, reduction of emissions by relocating heavy polluters (e.g., the Capital Steel Company), and installing desulphurization facilities in factories around

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Beijing. Additionally during the Olympics period, the government introduced strict vehicular emission standards (equivalent to the Euro-IV standards), reduced the number of on-road private cars by half (~1.5 million) through an odd/even plate number rule, and suspended construction activities as well as imposed strict dust controls at construction sites (Mijling et al., 2009; Wang et al., 2009a, 2009c, 2010). Concurrently, the neighboring administrative regions implemented local emission control measures to reduce air pollutants that could influence the environment of Beijing (Stone, 2008; Wang et al., 2010). This provided a valuable opportunity to evaluate the effectiveness of these drastic measures on mitigating air pollution and improving the atmospheric environment in a mega-city.

A number of papers have been published on the results of surface and satellite measurements during the Beijing Olympics, all indicating sharp decreases in the concentrations of the measured pollutants in Beijing during the period of the Olympics (Cermak and Knutti, 2009; Mijling et al., 2009; Witte et al., 2009; Wang et al., 2009a, 2009b, 2009d, 2010). To date most of these published studies have focused on the levels and variations in air quality and air pollutants during the Games. However, few results on the chemical composition of the precipitation and the effects of the drastic emission control measures on the hydrochemistry of the rainwater have been reported. During the rainy season from April to October of 2008, rainwater samples at an urban site in Beijing were collected, and the chemical and strontium isotopic composition of the rainwater was measured. Here, we report the data and results from the analysis of this dataset, thus providing insights into the changes in rainwater chemistry due to natural factors and human intervention programs.

Because the Sr isotopic ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) are relatively unaltered during the biological and geological processes or low-temperature water/rock reactions, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios have been successfully used as source tracers for sediment provenance, chemical weathering, and long-range transport of aerosols (Gallet et al., 1996; Gaillardet et al., 1999; Kanayama et al., 2002; Widory et al., 2010). Sr isotope studies of atmospheric aerosols have been carried out mainly for the soluble components of aerosols in rainwater (Herut et al., 1993; Nakano and Tanaka 1997; Han and Liu, 2006; Xu and Han, 2009; Cheng et al., 2010). Nakano and Tanaka (1997) described a mixing model of three components: Asian dust, seasalt, and local soil, for Sr isotopes in precipitation in Japan. Cheng et al. (2010) studied $^{87}\text{Sr}/^{86}\text{Sr}$ coupled with chemical ratios of rainwater to track the long-range transport of Asian dust and to estimate the relative contribution of the long-range transported Sr and seasalt Sr in rainwater in Taiwan. More extensive $^{87}\text{Sr}/^{86}\text{Sr}$ applications in precipitation, however, are limited because of the low dissolved Sr concentration in precipitation and other complications (Nakano and Tanaka 1997; Xu and Han, 2009; Cheng et al., 2010).

In this paper, we first present and discuss the chemical characterization and Sr isotope signature of the rainwater and identify possible sources that contributed to its chemical composition. We then discuss the changes in the chemical composition of rainwater samples before, during, and after the Olympics, and evaluate the effects of the emission and traffic intervention programs on the chemical composition changes of the rainwater.

2. Methodology

2.1. Background and sampling site description

Beijing is located in the northern part of the North China Plain, not far from the western slopes of the Yanshan Mountains, about 150 km to the Bohai Sea. With a typical continental monsoon climate of a northern temperate zone, there are four distinct seasons in Beijing (Xu and Han, 2009). Beijing, the capital of the People's Republic of China, is the nation's political, economic, and cultural center. The metropolis is one of the world's most crowded cities with a population that had exceeded 17 million by the end of 2007. Furthermore, the number of private cars had exceeded 3.2 million by the end of the same year. Since the 1980s, the city has experienced rapid economic growth and urbanization along with continued expansion of urban transportation, resulting in severe air pollution (Zhang et al., 2000; Yao et al., 2003; Streets et al., 2007; Zhang et al., 2007; Wang et al., 2008;). Simultaneously during this period, the increase of NO_x emissions from automobile exhaust gases and a decrease in alkaline ion concentrations (mainly Ca^{2+}) have resulted in significant acidification of the rainwater in Beijing (Xu and Han, 2009). Beijing is close to many heavily populated, urbanized, and industrialized cities as shown in Fig. 1a, including the Tianjin Municipality, Shijiazhuang in Hebei Province, Taiyuan and Datong in Shanxi Province, and Jinan in Shandong Province, all being typical industrial, coal-burning cities (Streets et al., 2007; Zhao et al., 2009). In these areas, emission controls on stationary sources and vehicles are not as stringent as those in Beijing; moreover, these surrounding areas can contribute significantly to Beijing's air pollution (Streets et al., 2007). In addition, Beijing is also frequently affected, particularly in the springtime, by dust storms emanating from the desert and loess areas several hundred kilometers to the northwest (Sun et al., 2004; Xie et al., 2005; Yang et al., 2007; Xu and Han, 2009).

The sampling site is located in a residential area in the northern part of Beijing between the Third and Fourth Ring Roads of the city (Fig. 1b). The distance from the sampling site to the 2008 Olympic Game stadiums is less than 2 km. The sampler was placed on the roof of an approximately 15 m-high building in the Institute of Geology and Geophysics, Chinese Academy of Sciences (CAS). The rainwater samples were collected manually from the beginning of each rain event with a funnel sampler that was located approximately 120 cm above the roof. The sampler and container were pre-cleaned with acid (2–3 N HCl), rinsed with Milli-Q water ($18.2 \text{ M}\Omega \cdot \text{cm}$), and then dried prior to use. To prevent contamination from dry deposition, special attention was paid to opening the sampler as quickly as possible after the onset of rainfall. Forty-three rainwater samples were collected during the sampling period April to October, 2008.

2.2. Analytical methods

The pH and conductivity values were immediately measured at the end of the rain events with a portable pH and salt conductivity meter. The rainwater samples were then filtered through $0.22 \mu\text{m}$ Millipore membrane filters and the filtrate was separated into two aliquots. One was stored

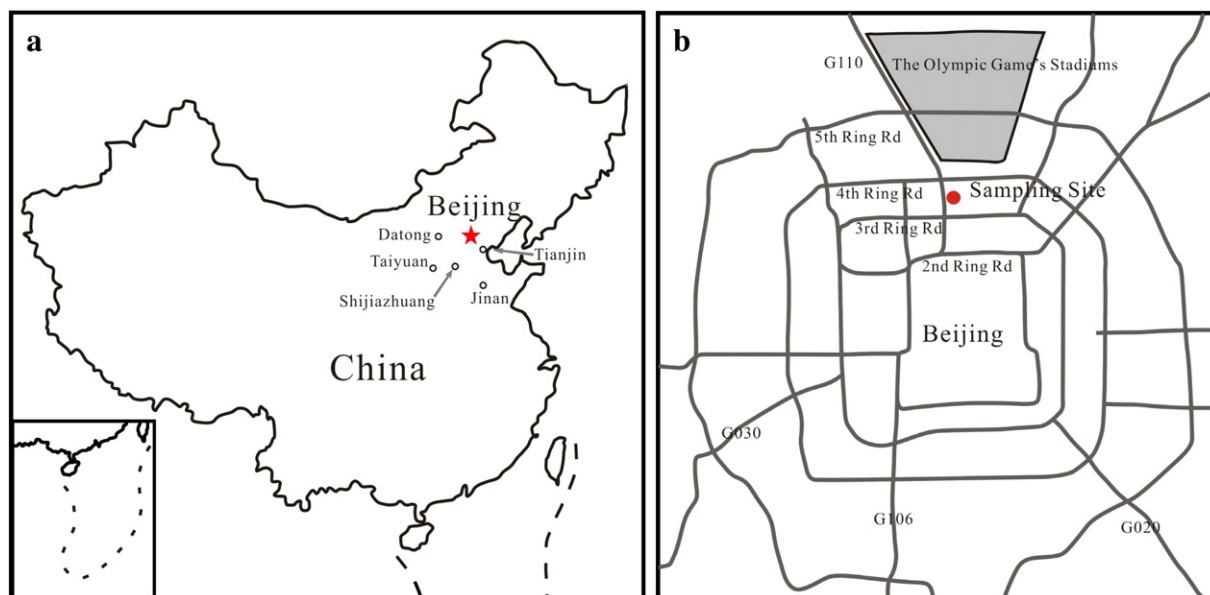


Fig.1. Map showing Beijing city and surrounding areas (a) and the sampling site (b).

directly in a polyethylene bottle to measure anions; the other was acidified with double sub-boiling distilled HNO_3 (6 M) to $\text{pH} < 1.6$ and stored in a polyethylene bottle to measure cations and trace element concentrations (Al and Sr), and Sr isotopic ratios. All containers were previously washed with high-purity HCl and rinsed with Milli-Q water and dried. Major anions (Cl^- , F^- , SO_4^{2-} , and NO_3^-) were measured by using an ionic chromatography (Dionex DX-120). NH_4^+ concentrations were determined by spectrophotometry using the Nessler method. Major cations (K^+ , Na^+ , Ca^{2+} , Mg^{2+}) were determined by ICP-AES (Thermo's IRIS Intrepid II). The concentrations of Al and Sr were measured by ICP-MS (VG POEMS III) in the State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Wuhan. Reagent and procedural blanks were determined in tandem with the sample treatment; the analytical precision was better than $\pm 5\%$.

In order to determine the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio, approximately 100 ml of each sample was first prepared by completely evaporating it to dryness in a Teflon vessel in an ultra-clean laboratory. The residue was then dissolved in distilled 2.0 N HCl, and Sr and Rb were separated from the samples on a cation-exchange column loaded with Dowex 50 W \times 8 200–400 mesh resins by elution of 2.0 N HCl. In most cases, this technique would result in Sr fractions with only small Rb interferences. Total blanks for Sr were less than 100 pg and were negligible with regard to the total Sr analyzed in the rainwater samples (Xu and Han, 2009). After chemical separation, the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratio was then determined using a Finnigan MAT 262 multiple collector mass spectrometer in the State Key Laboratory of Lithospheric Evolution, the Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing. Reproducibility was verified by periodic determinations of the NBS-987 standard. The average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of this standard for 25 determinations was 0.710227 ± 0.000020 (2σ , $n = 25$) during the course of the study.

3. Results and discussion

3.1. Ionic composition and pH value

The measured parameters (pH and EC), concentrations of the major ions aluminum and strontium, and $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios of rainwater are given in Table 1. The volume-weighted mean (VWM) value of the ionic compositions of rainwater samples and related statistical analyses are also shown in Table 1. The pH values of rainwater samples ranged between 3.99 and 7.88, with a VWM value of 5.32. The frequency distribution of rainwater pH is shown in Fig. 2. The pH value of 5.60 represents the reference level. More than half of rainwater samples had pH values greater than 5.60, while about 42% of the samples had pH values less than 5.60. According to previous studies, naturally-existing CO_2 , NO_x , and SO_2 can dissolve in clouds and water droplets, resulting in pH values of rain in a clean atmosphere between 5.0 and 5.60 (Charlson and Rodhe, 1982; Galloway et al., 1993). Rainwater samples with pH values above 5.60 may indicate certain inputs of alkaline substances into the precipitation in a study area (Xu and Han, 2009). Alkaline rainwater commonly occurs in the beginning and the end of the rainy season, while acidic rainwater mainly occurs in the middle of the rainy season (July to September) in Beijing.

Table 1 shows the order of cation abundance as $\text{Ca}^{2+} > \text{NH}_4^+ > \text{Mg}^{2+} > \text{Na}^+ > \text{K}^+$. Ca^{2+} was the most abundant cation, ranging from 17.2 to 2990 $\mu\text{eq l}^{-1}$, with a VWM concentration of 291 $\mu\text{eq l}^{-1}$. NH_4^+ was the next most abundant cation, ranging from 7.65 to 677 $\mu\text{eq l}^{-1}$, with a VWM concentration of 179 $\mu\text{eq l}^{-1}$. Ca^{2+} and NH_4^+ together accounted for about 76–97% of the total cations measured. Also shown in Table 1 the order of anion abundance was $\text{SO}_4^{2-} > \text{NO}_3^- > \text{Cl}^- > \text{F}^-$. Among the anions, SO_4^{2-} was the dominant anion, with concentrations ranging from 60.7 to 1687 $\mu\text{eq l}^{-1}$ and a VWM concentration of 270 $\mu\text{eq l}^{-1}$. The next most abundant anion was NO_3^- , with concentrations

Table 1Concentrations of major ions (in $\mu\text{eq l}^{-1}$), Al and Sr (in $\mu\text{mol l}^{-1}$), $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, volume-weighted mean (VWM) value and related statistical analyses of rainwater in Beijing.

Sample number	Date mm-dd-yy	pH	EC $\mu\text{s cm}^{-1}$	F ⁻ $\mu\text{eq l}^{-1}$	Cl ⁻ $\mu\text{eq l}^{-1}$	NO ₃ ⁻ $\mu\text{eq l}^{-1}$	SO ₄ ²⁻ $\mu\text{eq l}^{-1}$	NH ₄ ⁺ $\mu\text{eq l}^{-1}$	Ca ²⁺ $\mu\text{eq l}^{-1}$	Mg ²⁺ $\mu\text{eq l}^{-1}$	K ⁺ $\mu\text{eq l}^{-1}$	Na ⁺ $\mu\text{eq l}^{-1}$	Al ³⁺ $\mu\text{mol l}^{-1}$	Sr ²⁺ $\mu\text{mol l}^{-1}$	$^{87}\text{Sr}/^{86}\text{Sr}$	2 σ
BJR-01	4-11-2008	6.05	102	11.4	80.5	179	417	349	276	40.1	8.93	23.6	11.5	0.30	0.710080	0.000015
BJR-02	5-14-2008	6.71	188	36.7	123	403	603	569	508	69.7	19.5	22.6	11.3	0.43	0.709934	0.000014
BJR-03	5-27-2008	7.88	380	90.8	165	658	1367	635	1858	207	80.5	70.9	12.7	1.44	0.710041	0.000018
BJR-04	6-01-2008	7.23	254	74.7	270	1079	1687	592	2990	387	100	204	13.1	2.87	0.710143	0.000017
BJR-05	6-03-2008	6.79	100	18.8	88.4	140	381	289	812	72.3	22.9	21.2	6.16	0.32	n.d.	
BJR-06	6-04-2008	7.10	115	10.5	64.9	174	284	282	545	73.3	12.8	8.61	0.54	0.26	0.709823	0.000016
BJR-07	6-13-2008	3.99	120	10.6	88.9	169	355	289	138	31.8	24.1	2.26	0.40	0.12	n.d.	
BJR-08	6-17-2008	4.24	39	3.89	59.2	87	140	137	72.1	11.7	4.32	6.04	1.75	0.03	n.d.	
BJR-09	6-20-2008	6.45	196	29.8	88.9	293	750	447	939	140	31.2	17.9	0.52	0.51	n.d.	
BJR-10	6-23-2008	6.82	88	8.42	73.1	114	328	167	363	49.1	9.92	16.0	0.16	0.33	0.710847	0.000019
BJR-11	6-25-2008	7.08	192	36.6	108	560	752	327	885	147	19.6	34.8	0.55	1.07	n.d.	
BJR-12	6-26-2008	6.24	94	16.1	74.7	261	411	235	375	83.9	7.03	8.5	1.09	0.25	0.710412	0.000022
BJR-13	6-27-2008	6.64	71	17.0	68.8	107	287	209	310	36.8	5.93	10.5	0.23	0.19	n.d.	
BJR-14	6-28-2008	6.77	116	22.3	81.1	209	419	255	518	60.1	9.80	13.6	0.18	0.44	0.710159	0.000016
BJR-15	6-29-2008	5.83	75	15.3	69.1	142	306	188	289	69.1	4.19	3.96	0.35	0.17	n.d.	
BJR-16	6-30-2008	5.15	233	33.1	136	598	891	677	485	90.8	22.0	24.0	0.39	0.33	0.709805	0.000019
BJR-17	7-01-2008	5.64	150	10.7	80.1	206	661	584	203	43.3	9.77	4.13	0.49	0.10	n.d.	
BJR-18	7-05-2008	6.02	84	10.2	65.6	117	332	318	160	28.1	0.61	1.78	0.48	0.07	0.709931	0.000021
BJR-19	7-08-2008	4.11	72	5.02	61.4	91.2	253	229	93	7.24	0.28	1.87	3.54	0.03	n.d.	
BJR-20	7-09-2008	5.52	140	5.70	64.4	155	481	552	165	26.2	8.34	6.04	0.47	0.08	0.709507	0.000017
BJR-21	7-29-2008	6.78	62	7.91	70.7	68.3	237	271	52.0	13.6	3.79	15.2	0.32	0.04	n.d.	
BJR-22	7-31-2008	6.86	40	5.77	63.7	45.3	111	182	139	8.81	1.99	5.61	0.37	0.05	0.710291	0.000024
BJR-23	8-10-2008	4.11	41	3.94	55.8	49.4	144	115.9	17.2	3.67	2.97	2.98	1.34	0.01	0.709512	0.000016
BJR-24	8-11-2008	4.27	16	4.50	55.4	39.4	109	64.4	52.6	9.71	0.03	0.22	0.10	0.03	n.d.	

BJR-25	8-14-2008	5.47	9.0	2.31	52.8	26.2	60.7	46.1	58.3	4.86	1.07	1.83	0.23	0.01	n.d.	
BJR-26	8-15-2008	5.25	32	7.12	54.0	60.7	166	101	96.0	7.24	1.69	11.9	0.92	0.03	0.709184	0.000023
BJR-27	8-18-2008	6.49	85	26.8	89.9	333	518	76.3	1344	153	16.3	21.7	0.34	0.49	0.709556	0.000019
BJR-28	8-21-2008	4.85	40	6.67	65.1	77.5	184	169	52.0	12.0	3.58	2.39	2.51	0.04	0.709461	0.000021
BJR-29	8-26-2008	4.67	164	27.0	88.9	405	743	329	818	58.4	14.0	13.0	0.40	0.67	0.710011	0.000018
BJR-30	8-27-2008	4.49	46	6.73	63.9	93.3	192	145	51.6	18.2	2.43	4.17	3.78	0.09	n.d.	
BJR-31	8-29-2008	5.81	205	49.3	122	505	727	467	939	151	20.8	36.3	4.45	0.56	0.710039	0.000016
BJR-32	8-30-2008	6.22	106	21.7	75.3	163	464	181	630	53.4	7.85	13.1	1.02	0.25	n.d.	
BJR-33	9-07-2008	4.72	75	9.67	64.5	116	355	195	227	15.1	3.25	7.61	7.19	0.20	0.710893	0.000022
BJR-34	9-09-2008	5.04	55	10.3	68.3	106	262	150	242	19.7	3.71	7.00	2.58	0.08	0.709781	0.000019
BJR-35	9-10-2008	5.45	25	3.3	55.3	49.0	137	82.4	148	5.51	1.10	1.65	0.13	0.03	n.d.	
BJR-36	9-15-2008	6.25	548	16.3	74.9	187	327	209	375	67.2	7.01	5.70	0.79	0.17	n.d.	
BJR-37	9-16-2008	5.10	70	12.8	69.8	138	301	227	282	33.3	9.08	7.17	1.21	0.14	0.710344	0.000015
BJR-38	9-19-2008	4.18	83	8.49	63.4	101	302	208	107	17.9	7.62	5.57	6.48	0.07	0.709951	0.000015
BJR-39	9-21-2008	4.19	50	4.41	58.9	69.9	188	163	35.8	4.20	3.22	1.57	1.79	0.03	0.709407	0.000017
BJR-40	9-24-2008	6.01	23	4.22	55.0	33.9	97.2	70.8	174	12.7	1.05	0.65	0.10	0.05	0.709475	0.000019
BJR-41	10-05-2008	6.31	115	18.5	90.7	359	412	7.65	1247	195	16.3	27.9	0.26	0.32	0.709808	0.000020
BJR-42	10-09-2008	6.10	145	29.2	106	591	516	348	625	118	29.2	41.0	17.9	0.40	0.710216	0.000016
BJR-43	10-23-2008	6.25	129	21.9	81.1	541	532	57.0	1053	116	23.5	19.9	19.7	0.43	0.709826	0.000017
Min		3.99	9.0	2.31	52.8	26.2	60.7	7.65	17.2	3.67	0.03	0.22				
Max		7.88	548	90.8	270	1079	1687	677	2990	387	100	204				
Mean		5.75	116	18.1	82.7	230	423	260	483	64.5	13.6	17.6				
Median		6.01	88.0	10.7	70.7	142	332	209	282	40.1	7.85	8.52				
S.D.		1.03	99.8	18.2	37.6	221	322	173	570	73.9	19.3	32.1				
VWM		5.32	72.8	10.5	67.8	139	270	179	291	38.5	6.69	8.51				
VWMBO		5.76	89.4	11.6	71.8	155	316	262	307	43.6	8.82	10.2				
VWMDO		4.96	57.6	9.2	63.3	94.8	224	142	189	21.2	3.79	5.73				
VWMAO		5.38	81.4	12.0	72.7	240	308	97.7	579	81	10.8	13.2				

Min = minimum; Max = maximum; S.D. = standard deviation; n.d. = not determined; 2σ = isotope errors correspond to 2 standard deviation; VWM = volume-weighted mean; VWM_{BO} = volume-weighted mean concentration before the Olympics; VWM_{DO} = volume-weighted mean concentration during the Olympics; VWM_{AO} = volume-weighted mean concentration after the Olympics.

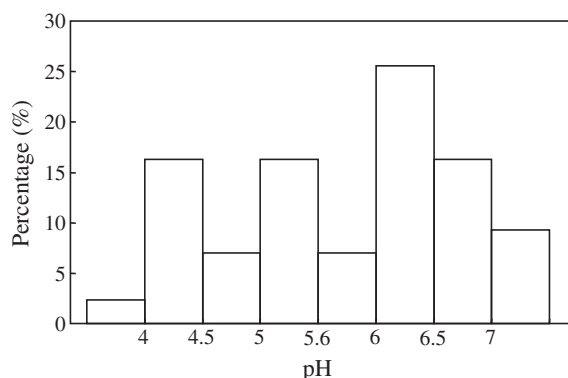


Fig. 2. The frequency distribution of rainwater pH in Beijing in 2008.

ranging from 26.2 to 1079 $\mu\text{eq l}^{-1}$ and a VWM concentration of 139 $\mu\text{eq l}^{-1}$. SO_4^{2-} and NO_3^- were conventional acidic ions in precipitation; their sum accounted for 61–91% of the total anions measured.

3.2. Origins of major ions in the rainwater

In order to find possible association between ions in precipitation and, consequently, the likely sources of pollutions, correlation coefficients (R^2) between ions in precipitation were calculated and are presented in Table 2. Good correlation between SO_4^{2-} and NO_3^- ($R^2=0.84$) was expected because of the co-emission of their precursors SO_2 and NO_x , and would reflect the input of pollutants from fossil fuel combustion. Power plants and vehicles are the most predominant sources of SO_2 and NO_x in Beijing (Wang et al., 2008). Certain correlations exist among cations (Ca^{2+} , Mg^{2+} , K^+ , Na^+), suggesting that these elements have a common source. These elements are common constituents of soil dust in the Beijing area and in the arid and semi-arid regions of northern China (Feng et al., 2001; Yokoo et al., 2004; Xu and Han, 2009). A positive correlation between Ca^{2+} and Sr^{2+} ($R^2=0.85$) in the rainwater samples (Fig. 3.) indicates the potential of Sr as a provenance tracer for Ca. Concentrations of Ca^{2+} in the precipitation samples show good correlations with SO_4^{2-} ($R^2=0.75$) and NO_3^- ($R^2=0.75$), indicating that Ca^{2+} is the main cation that neutralizes the acidity of the precipitation.

Atmospheric aerosols, including sea salts, crustal dust, volcanic dust, biogenic material, and anthropogenic emissions

are the main sources of chemical compositions in rainwater (Roy and Négrel, 2001; Négrel et al., 2007). To determine the sea salt and terrestrial contributions to the rainwater in Beijing, the approach presented by Négrel and Roy (1998) was used. Na was considered as the marine reference (ref) species. Assuming that Al emanates almost from terrestrial input, the measured Na value in the rainwater (rw) was corrected for a continental contribution as follows, where the equation is expressed on a molar basis:

$$\text{Na}_{\text{ref}} = \text{Na}_{\text{rw}} - 0.11 \times \text{Al}_{\text{rw}}. \quad (1)$$

The contribution of the non-sea-salt component (nss) for a given element X is thus:

$$X_{\text{nss}} = X_{\text{rw}} - \text{Na}_{\text{ref}} \times (X/\text{Na})_{\text{sw}} \quad (2)$$

where X represents Cl^- , SO_4^{2-} , K^+ , Ca^{2+} , Mg^{2+} and Sr^{2+} . The elemental ratios $(X/\text{Na})_{\text{sw}}$ were determined according to the composition of seawater given by Berner and Berner (1987). The results of minimum, maximum, and mean values for the non-sea-salt component in the rainwater expressed in percentage are given in Table 3.

The proportion of Cl^- from non-sea-salt origin typically ranged from 13% to 99.6%, with a mean value of 80.8%. The Cl^-/Na^+ molar ratios of the rainwater from Beijing were several times larger than that of seawater ($\text{Cl}^-/\text{Na}^+ = 1.17$; Berner and Berner, 1987), also indicating a significant anthropogenic Cl^- source. The non-sea-salt input of Cl^- could come from various sources of pollution, including automobiles, coal combustion, incinerators, and combustion/decomposition of organo-chlorine compounds. (e.g., Fuzzi et al., 1984; Sanusi et al., 1996; Négrel and Roy, 1998). The non-sea-salt input of SO_4^{2-} ranged from 98.6% to 99.98% in our samples. In China, coal accounts for about 70% of the commercial energy production, leading to large emissions of SO_2 , which, to date, has been the most important precursor of acid rain in China (Aas et al., 2007).

Almost all Ca^{2+} in the rainwater from Beijing was of non-sea-salt origin, with a mean value of 99.8%. Identifying the provenance of Ca^{2+} in rainwater holds great importance to the study of the acid rain problem because this particular element is the dominant cation that neutralizes acid and is indispensable for plant growth (Nakano and Tanaka, 1997; Larssen and Carmichael, 2000). There are two main origins for the Ca^{2+} in rainwater: the first is natural Ca^{2+} , derived from the dissolution of calcium carbonate from soil dust

Table 2

Matrix of correlation coefficients (R^2) of ionic concentrations (in $\mu\text{eq l}^{-1}$) of rainwater in Beijing.

Ions	F^-	Cl^-	NO_3^-	SO_4^{2-}	NH_4^+	Ca^{2+}	Mg^{2+}	K^+	Na^+	Sr^{2+}
F^-	1									
Cl^-	0.79	1								
NO_3^-	0.77	0.83	1							
SO_4^{2-}	0.85	0.86	0.84	1						
NH_4^+	0.41	0.43	0.36	0.57	1					
Ca^{2+}	0.75	0.77	0.75	0.75	0.16	1				
Mg^{2+}	0.52	0.57	0.69	0.54	0.07	0.93	1			
K^+	0.81	0.87	0.74	0.82	0.35	0.81	0.58	1		
Na^+	0.60	0.88	0.68	0.68	0.22	0.77	0.56	0.81	1	
Sr^{2+}	0.74	0.87	0.75	0.80	0.25	0.85	0.59	0.85	0.91	1

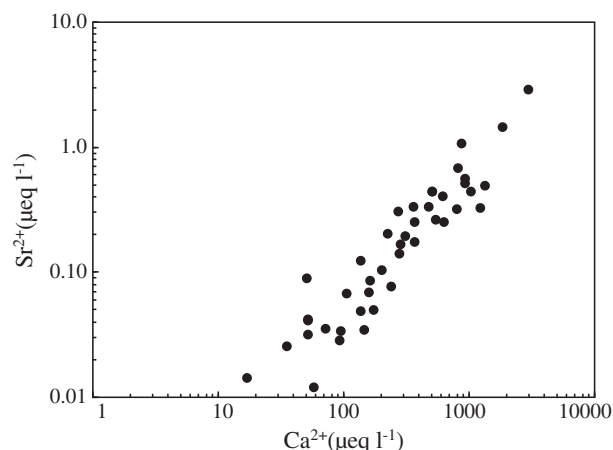


Fig. 3. The correlation between Ca^{2+} and Sr^{2+} ($R^2=0.85$) in the rainwater samples.

and/or carbonate weathering; the second emanates from anthropogenic inputs *i.e.*, industrial and agricultural activities. A previous study indicated that Ca^{2+} in Beijing rainwater emanated mainly from the dissolution of CaCO_3 from soil dust transported either from local areas or from desert and loess areas in northern China (Xu and Han, 2009). The elevated levels of pH and Ca^{2+} and Sr^{2+} concentrations in the spring rains in the East Asian area (*e.g.*, Japan, Beijing) also suggest the dissolution of calcium carbonate from soil dust, which is common in spring (Nakano and Tanaka, 1997; Nakano et al., 2006; Xu and Han, 2009). Calcium can also be produced by human activities, such as building, burning coal, traffic, construction with cement, and other activities. The large turbulences generated by construction can also enhance the spread of aerosols in the atmosphere (Sanusi et al., 1996).

Mg^{2+} and K^+ were also mainly of terrestrial origin, in a range of 64.3% to 99.5% and 82.2% to 99.8%, with mean values of 92.7% and 96.0%, respectively. These low values for marine Mg^{2+} and K^+ show that, like Ca^{2+} , Mg^{2+} and K^+ can also come from soil dust (Bernier and Bernier, 1987). Other non-marine origins of these elements are probably anthropogenic pollution (industrial and agricultural activities) and biogenic aerosols, even in temperate climates (Bernier and Bernier, 1987; Négrel and Roy, 1998). However, major elemental chemistry cannot precisely characterize the various end members. Therefore, Sr isotope compositions are used to obtain additional information concerning the different possible sources in next section.

3.3. Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic composition

Because the strontium isotopic system is a radiogenic one and its ratios vary between different sources (Faure, 1986),

any potential mass dependent isotope fractionation is small, and Ca and Sr exhibit similar geochemical cycles (*e.g.*, Åberg et al., 1989; Åberg, 1995; Bailey et al., 1996). Consequently, Sr isotopes are expected to provide insights into the sources (and their mixtures) of base cations in rainwater, particularly Ca, which is not well-constrained from concentration data alone (Herut et al., 1993; Nakano and Tanaka, 1997; Négrel and Roy, 1998; Schmitt and Stille, 2005; Han and Liu, 2006; Xu and Han, 2009). The Sr concentrations of rainwater samples ranged from 0.01 to $2.87 \mu\text{mol l}^{-1}$, with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from 0.7092 to 0.7109. All of the samples had $^{87}\text{Sr}/^{86}\text{Sr}$ ratios higher than that of seawater, which reflect a contribution from at least one more radiogenic Sr source. The radiogenic source can be either natural or anthropogenic input. The “natural” sources could be attributed to the soil dust, some pioneering researches have demonstrated that the high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the spring rainwater in China and Japan were owing to the dissolution of carbonate in the loess dust ($^{87}\text{Sr}/^{86}\text{Sr}=0.7111\text{--}0.7115$; Nakano and Tanaka, 1997; Nakano et al., 2006; Xu and Han, 2009; Xu et al., 2009; Cheng et al., 2010). The other Sr source could be mixture of various anthropogenic inputs. Widory et al. (2010) determined the origin of total suspended particulates (TSP) and fine particles ($\text{PM}_{2.5}$) in the atmosphere of Beijing by using of coupling both Sr and Pb isotope systematics. The result showed that atmospheric particles from coal combustion had a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70924, the cement factories with a value of 0.71029, and the smelters with the most radiogenic value of 0.71206 (Widory et al., 2010). The emissions from coal combustion were compatible with our estimation of anthropogenic end-member around 0.7090–0.7095 (Xu and Han, 2009). The $^{87}\text{Sr}/^{86}\text{Sr}$ of aerosols from cement factories and smelters were similar to the range for soil dust (0.7111–0.7115), however, we had the reason to

Table 3

Minimum, maximum and mean values for the non-sea-salt (nss) component of rainwater in Beijing.

	nss- Cl^- (%)	nss- SO_4^{2-} (%)	nss- Ca^{2+} (%)	nss- Mg^{2+} (%)	nss- K^+ (%)	nss- Na^+ (%)	nss- Sr^{2+} (%)
Min	13.0	98.6	98.7	64.3	82.2	0.1	93.0
Max	99.6	99.98	99.98	99.5	99.8	20.8	99.9
Mean	80.8	99.6	99.8	92.7	96.0	3.8	98.5

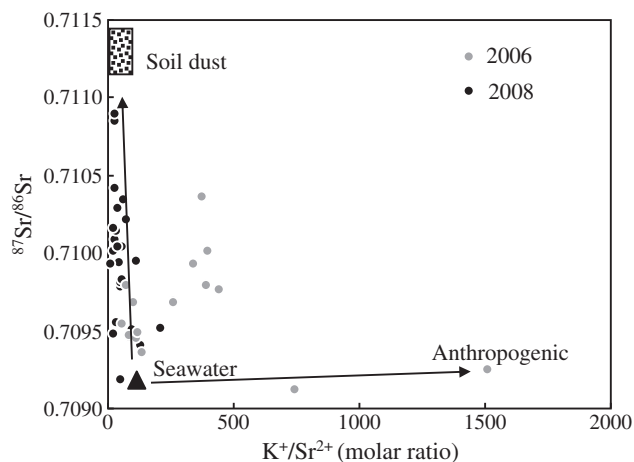


Fig. 4. Variation of Sr isotope ratios against K^+/Sr^{2+} molar ratios in the rainwater samples. The data set of the rainwater from Beijing in 2006 (gray dots) are also plotted in the figure for comparison.

believe that the contributions of Sr from cement factories and smelters reduced in 2008, because some heavy industrial polluters were relocated in 2008 and construction activities were suspended during the Olympic period.

Fig. 4 shows the variation of $^{87}Sr/^{86}Sr$ ratios against element-to-Sr molar ratios (e.g. K^+/Sr^{2+}) in the rainwater; the data set of the rainwater from Beijing in 2006 are also plotted in the figure for comparison. As seen on the figure, at least three-component mixing is needed to interpret the data, which include sea salts ($^{87}Sr/^{86}Sr = 0.70917$), soil dust (0.7111–0.7115 and lowest K^+/Sr^{2+} ratio), and anthropogenic inputs (0.7090–0.7095 and highest K^+/Sr^{2+} ratio) (Xu and Han, 2009). It is worth noting in Fig. 4 that the data set of rainwater in 2008 was mainly distributed between the sea salt end-member and the soil dust end-member. It could be suggested that the impact of emissions from the anthropogenic activities in 2008 is smaller than that in 2006. This was consistent with the fact that systematic long- and short-term emission control measures and regulations were implemented in 2008 to improve air quality, including relocating heavy industrial polluters (e.g. the Capital Steel Company and other factories in south area of Beijing) and freezing construction activities. From the data and Fig. 4, it was also noted that the $^{87}Sr/^{86}Sr$ ratios of rainwater in 2008 were more radiogenic than that of rainwater in 2006. It was shown that aerosols from coal combustion and automobile exhaust had low $^{87}Sr/^{86}Sr$ ratios, with the value of 0.7092 and 0.7077–0.7083, respectively (Widory et al., 2010; Négrel et al., 2007). Therefore, we suggested that this phenomenon was resulted by some emission control measures, including reducing local thermal power generation, introducing strict vehicular emission standards (Euro IV) and reducing on-road cars by half through an odd/even plate number rule.

3.4. Effects of the emission control measures on chemical compositions of the rainwater

In 2008, the Beijing Municipal Government enacted a series of emission control measures and regulations before

and during the Olympic Games to ensure good air quality during the period of the world athletic competition. Generally, these measures were classified into before, during, and after the full-scale control periods, according to the magnitude and scale of control measures (Wang et al., 2009a). To examine in more detail the effects of the emission control measures on the chemical compositions of the rainwater in Beijing, the annual rain events were also divided into three different periods: before the Olympics (BO, before 19 July); during the Olympics (DO, 20 July–19 September); and after the Olympics (AO, after 20 September). The VWM concentrations and the variation of the ionic composition of the rainwater in the three periods are presented and compared in Table 1 and Fig. 5. The VWM concentrations of Cl^- , SO_4^{2-} , NO_3^- , NH_4^+ , Ca^{2+} , Mg^{2+} , K^+ and Na^+ in the DO period were 4.87, 62.8, 220, 92, 145, 172, 19.3, 3.73 and 5.26 $\mu eq l^{-1}$, respectively. All the chemical components except NH_4^+ had the lowest concentration levels in the DO among the three periods, as shown in Fig. 5. The VWM concentrations of SO_4^{2-} , NO_3^- , Cl^- and F^- in the DO period were reduced by 29.1%, 38.7%, 21.1% and 11.8%, respectively, from values in the BO period. The decrease of SO_4^{2-} and NO_3^- in the DO period may mainly be attributed to the emission control measures, including moving and closing high-emission factories, reducing the number of private cars by half through an odd/even number rule, curtailing the use of 70% of government cars, among other measures. Recent studies have indicated that these measures significantly reduced both SO_2 and NO_x emissions during the 2008 Olympic Game period (Mijling et al., 2009; Wang et al., 2009a, 2010; Wu et al., 2010). The VWM concentrations increased again after the control period ended, with an increase of 153% for NO_3^- and 37.4% for SO_4^{2-} , relative to the DO period. The great increase of NO_3^- concentration after the control period demonstrates that vehicular emissions are the most dominant source of NO_x in Beijing. It has been estimated that transportation was responsible for about 74% of all NO_x emissions in Beijing in 2007 (Wang et al., 2008).

The VWM concentrations of base cations in the DO period decreased by up to 38.3% for Ca^{2+} , 51.3% for Mg^{2+} , 57.0% for K^+ and 43.9% for Na^+ , respectively, compared with the BO

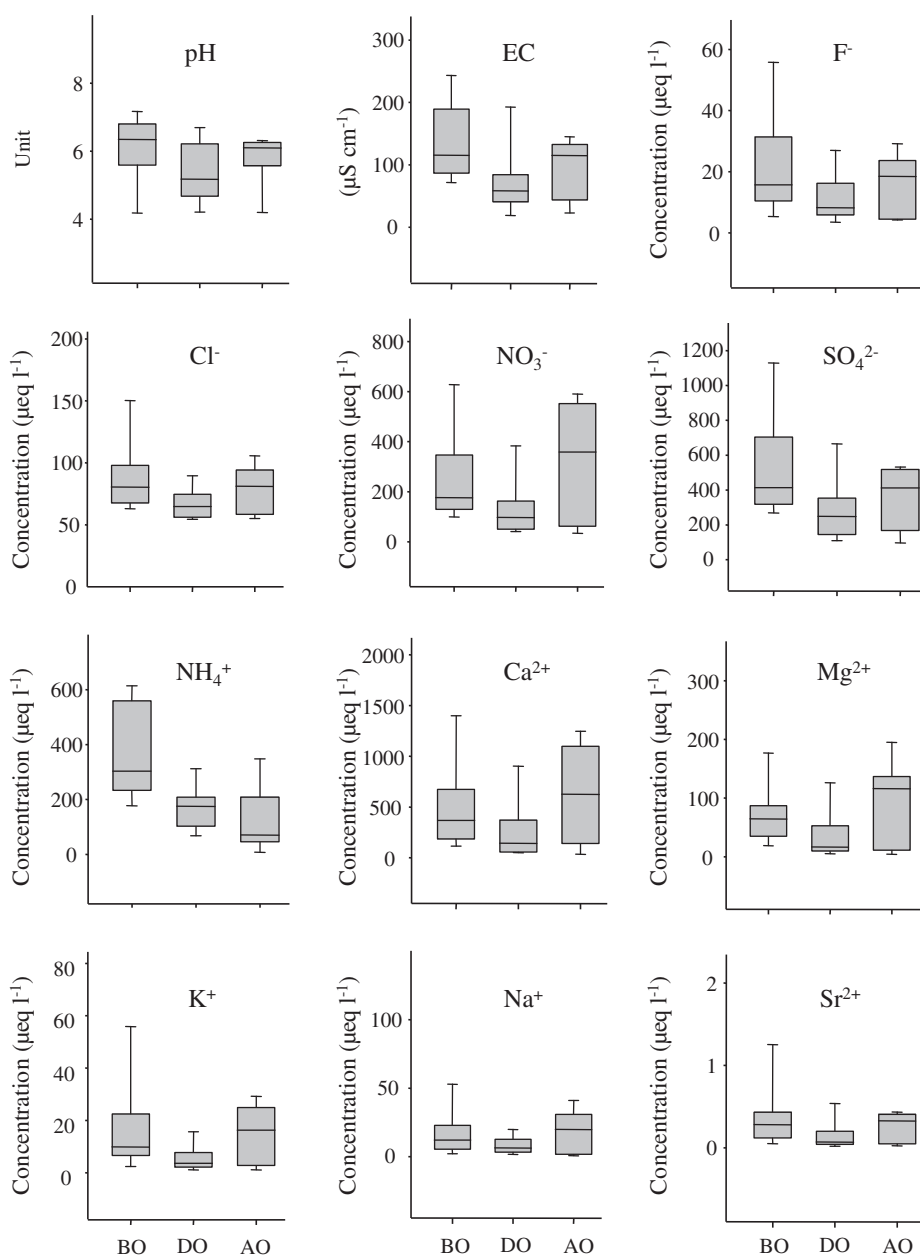


Fig. 5. The comparison of chemical composition of the rainwater before the Olympics (BO, April 11–July 20), during the Olympics (DO, July 20–September 20) and after the Olympics (AO). The upper and lower boundaries of boxes indicate 75th and 25th percentiles; the line within the box marks the median; the whiskers above and below boxes indicate the 90th and 10th percentiles.

period. The large decrease of base cations in the DO period may have resulted from both meteorological and emission control influences because these elements may be derived from both natural (e.g., soil dust) and anthropogenic inputs (building, burning coal, traffic, etc.). Vegetation coverage of bare ground was increased and construction activities were stopped during the 2008 Olympic Game period. Consequently, these control measures could have decreased the spread of soil dust aerosols into the atmosphere, resulting in the reduction of base cations concentration in precipitation. On the other hand, precipitation data show that the amount

of precipitation (225 mm) and the number of rainy days (15 days) during the 2008 Olympic Game period in Beijing were substantially more than those in other years for the same period (Mijling et al., 2009). The humid weather could also have prevented soil dust aerosols from entering into the atmosphere. To quantify the relative importance of the factors responsible for the lower base cations concentrations in the DO period, detailed modeling work is still needed, which is beyond the scope of this study. Among the ionic components, only NH_4^+ had the lowest concentration levels in the AO period. In general, NH_4^+ mainly originates from agricultural

activities and the burning of biomass. It can be attributed the steady decrease of the concentrations of NH_4^+ in the rainwater to the following reasons: (1) no special control measures were enacted for agricultural activities during the 2008 Olympic Game period; (2) the agricultural activities, such as fertilization, mainly concentrate in the springtime in Beijing area.

4. Conclusions

The chemical and Sr isotope compositions of the rainwater in Beijing during the 2008 Olympic Games were determined at an urban site near the Olympic stadiums. The ionic composition shows Ca^{2+} and NH_4^+ were the dominant cations, which accounted for about 76–97% of the total cations, and SO_4^{2-} and NO_3^- were major anions, which accounted for 61–91% of the total anions measured. The calculated results indicate that the major chemical components were mainly of terrestrial origin.

The Sr concentrations of rainwater samples ranged from 0.01 to $2.87 \mu\text{mol l}^{-1}$, with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from 0.7092 to 0.7109. All of the samples had $^{87}\text{Sr}/^{86}\text{Sr}$ ratios higher than that of seawater. The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic and elemental ratio systems could suggest presence of two main sources of Sr in the rainwater, which are the sea salt ($^{87}\text{Sr}/^{86}\text{Sr} = 0.70917$) and the soil dust ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7111\text{--}0.7115$). Compared to the data of the rainwater in 2006, the analysis could imply that the effect of the anthropogenic input on the rainwater decrease in 2008.

Comparison of chemical composition of the rainwater in different periods: before the Olympics (BO), during the Olympics (DO), and after the Olympics (AO), all the chemical components in the rainwater, except NH_4^+ , had the lowest concentration levels in the during Olympic Game period. The VWM concentrations of major ions in this period decreased by up to 29.1% for SO_4^{2-} , 38.7% for NO_3^- , 38.3% for Ca^{2+} , and 51.3% for Mg^{2+} , compared with the pre-Olympic Game period. The concentrations of all species decreased during the 2008 Olympic Games, which can likely be attributed to both strict emission controls and/or favorable meteorological conditions. Since NH_4^+ in the rainwater are mainly from agricultural activities which concentrated in the springtime in Beijing area, and no special control measures were enacted for agricultural activities during the Olympic Game period. Therefore, the concentration of NH_4^+ displayed a steady decreasing tendency from April to October.

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