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Atmospheric Aerosol Elements over the Inland Tibetan Plateau: Concentration, Seasonality, and Transport

Shichang Kang[1,3*], Pengfei Chen[2], Chaoliu Li[2], Bin Liu[2], Zhiyuan Cong[2,3]

[1] *State Key Laboratory of Cryospheric Science, Cold and Arid Regions Environmental and Engineering Research Institute, CAS, Lanzhou 730000, China*
[2] *Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, CAS, Beijing 100101, China*
[3] *CAS Center for Excellence in Tibetan Plateau Earth Sciences, Beijing 100101, China*

ABSTRACT

Between November 2005 and November 2007, weekly total suspended particle samples were collected at the Nam Co station in the inland Tibetan Plateau (TP). Through inductively coupled plasma mass spectrometry, twenty-nine elements were analyzed and their sources and fluxes were investigated. Mean elemental concentrations were lower than those at the edge of the TP. Some elements, such as Cr, Ni, Cd, and Pb, exhibited high enrichment factors (Cr: 22; Ni: 17; Cd: 23; and Pb: 9), indicating possible anthropogenic influence in this remote region, particularly during the pre-monsoon and monsoon seasons. In addition, an empirical orthogonal function analysis revealed the dominance of crustal-origin elements, rather than anthropogenic elements, in the aerosol. Furthermore, backward air mass trajectories demonstrated that the Nam Co region was mainly influenced by air masses from Central and South Asia. Accordingly, because of dust storms from Central Asia and within the TP, crustal element concentrations, such as of Al, were higher during winter and pre-monsoon seasons than during the monsoon season. By contrast, anthropogenic elements, such as Cr and Cd, were relatively higher during the pre-monsoon and monsoon seasons because of pollutants transported from South Asia, where atmospheric brown clouds are concentrated and biomass combustion is prevalent. Dry deposition of aerosols dominated in the Nam Co region, particularly during the non-monsoon period, which is useful to interpret the elemental records in the TP ice cores and lake sediments.

Keywords: Elements; Aerosol; Atmospheric transport; Deposition flux; Nam Co.

INTRODUCTION

The Tibetan Plateau (TP), the highest and largest low- and mid-latitude plateau in the world, is vital to the climate of the region (Kang *et al.*, 2010; You *et al.*, 2010; Lee *et al.*, 2013). Population is sparse and industrial activity minimal across most of the plateau, and local inhabitants mainly make a living by herding sheep and yaks, making it one of the most remote and pristine regions in the world. However, concerns have been raised in recent years over cryospheric changes caused by global warming (Kang *et al.*, 2010; Bolch *et al.*, 2012; Yao *et al.*, 2012) and chemical contamination from the long-range transport of atmospheric pollutants (Cong *et al.*, 2015; Lüthi *et al.*, 2015), both of which threaten this unique and fragile alpine ecosystem.

Chemical compositions of aerosols from such remote regions as the TP are useful in evaluating the effects of anthropogenic activities on atmospheric environment. Most aerosol chemistry investigations in the TP were short-term and conducted at the edge of the plateau (e.g., Wen *et al.*, 2001; Cong *et al.*, 2007; Meng *et al.*, 2013; Zhao *et al.*, 2013) and Lhasa, the capital city of the Tibet Autonomous Region of China (Zhang *et al.*, 2000; Cong *et al.*, 2011). Therefore, considering the vast area of the TP and the very limited data available for the region, more investigations are required to understand the aerosol chemical composition over the TP, particularly in the remote and inland TP.

The Nam Co region (N30°30′–30°56′, E90°16′–91°03′, 4718 m above sea level (a.s.l.)) is located in the inland TP (Fig. 1) and contains the eponymous lake with an area of 2015 km^2 (Kang *et al.*, 2011). During the monsoon season, the Nam Co region is influenced by the Indian monsoon, which brings moist and warm air from the Indian Ocean. The region is controlled by the Westerly winds during the winter (Byson, 1986; Tang, 1998). The Nam Co Monitoring and Research Station for Multispheric Interactions (Nam

* Corresponding author.
Tel.: +86-0931-4967368; Fax: +86-0931-4967368
E-mail address: shichang.kang@lzb.ac.cn

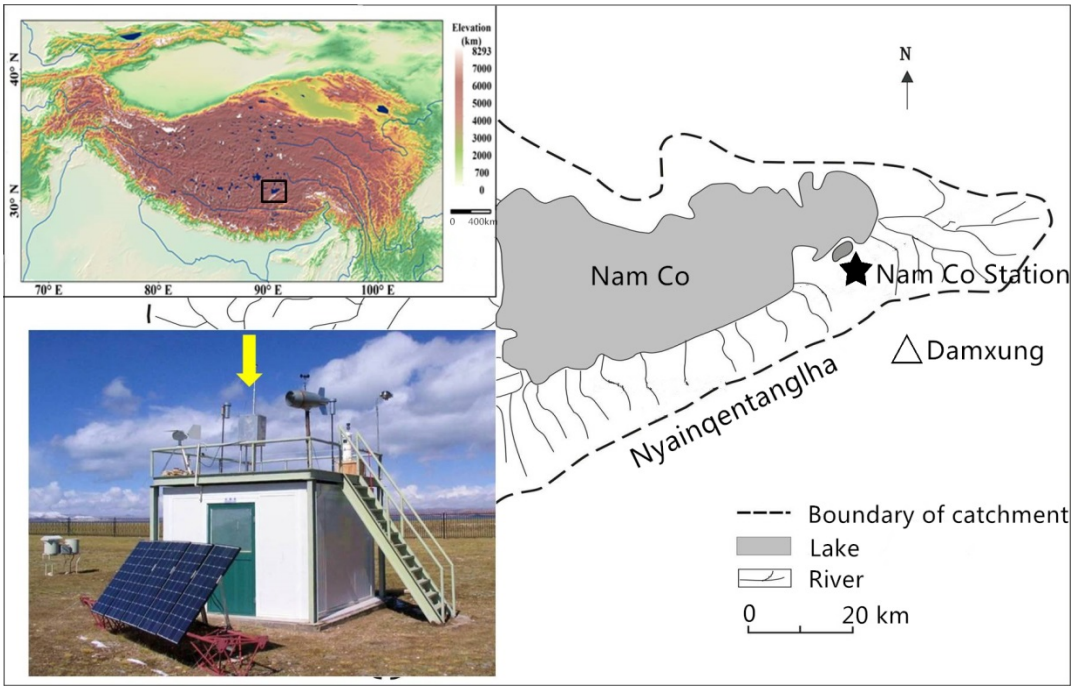

Fig. 1. Location of the sampling site (Nam Co station) in the inland Tibetan Plateau. The TSP sampler is marked by arrow.

Co station; N30°46.44′, E90°59.31′, 4730 m a.s.l.), located on the southeastern shore of Nam Co Lake, was established in June 2005 by the Chinese Academy of Sciences (Kang *et al.*, 2011). The station is far from anthropogenic emission sources and thus is ideal for observing long-term atmospheric environmental changes in the high-elevation regions and investigating how transported anthropogenic pollution influences remote regions.

Some studies have been conducted at this remote station, and low levels of aerosol optical depth (Cong *et al.*, 2009b) and carbonaceous aerosol have been reported (Wan *et al.*, 2015). The elemental compositions of precipitation (Cong *et al.*, 2010), aerosols (Cong *et al.*, 2007; Li *et al.*, 2007b), and snow (Huang *et al.*, 2013; Yu *et al.*, 2013) have indicated the presence of long-range transported pollutants in the Nam Co region (Xia *et al.*, 2011; Lüthi *et al.*, 2015), with partial contributions from local emissions (Li *et al.*, 2007a; Zhang *et al.*, 2012b; Chen *et al.*, 2015). However, the temporal coverage of previous researches were short, with most studies focusing on only one season, and elemental deposition data for this region, which is essential for studying aquatic bioavailability and historical variation (Hsu *et al.*, 2005; Kocak *et al.*, 2005), is limited. Thus, long-term investigations are necessary to comprehensively understand the atmospheric environment, potential pollutant sources, and their deposition flux in this remote region. In this work, we report the elemental composition of 2-year aerosol samples collected between November 2005 and November 2007, the elemental characteristics (such as seasonal variations and potential pollutant sources) and dry deposition fluxes, in order to assess the atmospheric environment in the inland TP. Furthermore, this work could also provide basic information for interpreting paleo-environmental records retrieved from ice cores and lake sediments.

METHODS

Sampling Site Meteorology

The Nam Co station is situated at the southeast shore of Nam Co Lake. The local population in the Nam Co region is about one thousand and the density within a 25 km distance from the station is less than 1 person per km^2 (Cong *et al.*, 2009b). The local residents mainly make a living by herding sheep and yaks, and produce limited atmospheric pollutants in the vicinity of the station. It is relatively isolated from major industrial sources and populated areas. The major sources for pollution are biomass (mainly yak dung) burning and vehicle emission caused by tourism surrounding the station (Chen *et al.*, 2015). Damxung is the largest town near the station (around 50 km away) with a population of 2600 and without industrial activities, which is separated by Nyainqentanglha mountains from the station (Fig. 1). Meteorological parameters (air temperature, wind speed and direction, precipitation, air pressure, and relative humidity) were observed using the automatic weather station at the Nam Co station. During the sampling period, the mean air temperatures were –1.6, 8.7, –4.2, and –7.4°C for the pre-monsoon (March–May), monsoon (June–September), post-monsoon (October–November) and winter (December–February) seasons, respectively, and the highest and lowest monthly temperature occurred in July (8.8°C) and December (–13.6°C). The annual mean wind speed was 4.0 m s^{-1}, and the highest monthly wind speed occurred in January. Southwesterly winds prevailed during the pre-monsoon season and winter and southerly winds were dominant during the monsoon and post-monsoon seasons. Annual precipitation was 414.6 mm, with the majority occurring during the monsoon season; 64% of the precipitation occurred at night, and local convective precipitation was frequent (You *et al.*,

2007). The annual mean air pressure and relative humidity were 571.2 hPa and 52.6%, respectively (Kang *et al.*, 2011).

Field Sampling Methods

Ninety-eight total suspended particle (TSP) samples were collected on polytetrafluoroethylene (PTFE) filters with a pore size of 0.4 μm and diameter of 47 mm. The field sampling was similar to that described in Cong *et al.* (2007). In brief, the sampler was placed on the flat roof (4 m above ground) of the monitoring house at the Nam Co station with a vacuum pump operating at an average flow rate of 16.7 L min^{-1} (Fig. 1). The pump was equipped with an automatic air mass flow meter (Taihe Automation Control and Instrument Corp.) and calibrated to an accuracy of 1.5%. Each sample, containing an air volume of 120–150 m^3, was collected for 7 days. The air volume was converted to the standard condition (25°C, 1000 mb) according to the ambient conditions in the Nam Co region. The samples were individually placed in small Petri dishes (Millipore Corp.) and stored at 4°C until extraction and chemical analysis.

Inductively Coupled Plasma Mass Spectrometry

Teflon scissors were used to cut the PTFE filters into two pieces for chemical analyses. One half was used for the major and trace element determination of 29 elements (Na, Mg, K, Ca, Li, Be, B, Al, Ti, V, Cr, Fe, Mn, Co, Ni, Zn, Ga, As, Rb, Sr, Y, Cd, Cs, Ba, Tl, Pb, Bi, Th, and U) through inductively coupled plasma mass spectrometry (X-7; Thermo-elemental, USA) at the laboratory of the Institute of Tibetan Plateau Research; the other half was archived. The digestion process was as reported in Wu *et al.* (2009). In brief, the collected filters were initially placed in a Teflon high-pressure digestion vessel with super-pure HNO_3-HF at 150–190°C. These vessels were then sonicated for 20 min and digested in an oven at 190°C for 24 h. The solutions were dried at 150°C. One mL HNO_3 was added and evaporated to dryness and this procedure was repeated twice. Rh and Re solutions were used as internal standards.

Quality Control

The Chinese Loess Reference Material (GBW07408) was used for quality control. The detection limit for each element was calculated as three times the standard deviation of blanks analyzed. It was found that the recoveries ranged from 78.9% for Cr to 121% for Cd (Table SI-1). For precision, the corresponding relative standard deviation (RSD) values of element concentrations measured in the reference material were less than 5% of most elements, except 13% of Cd. In addition, to test the agreement between different portions of the same filter, the exactly same procedures were used to digest and measure the 4 quarters of one filter for each element, the RSD of the 4 parts was less than 15%. The final concentrations were corrected with those of filter blanks. Four samples were not used because of sampling problem (e.g., filter broken, pump broken).

Enrichment Factors Calculation

Enrichment factors (EFs) are frequently used to differentiate the elements of crustal and anthropogenic origins (Al-Momani *et al.*, 2005; Ragosta *et al.*, 2008). EFs were calculated using the following equation:

$$EF_X = \frac{(C_X / C_R)_{aerosol}}{(C_X / C_R)_{crust}} \quad (1)$$

where X represents the element of interest, EF_x and C_x are the enrichment factor and concentration of X, and C_R is the concentration of a reference element. Subscripts *aerosol* and *crust* refer to particles in the aerosol samples and the crustal material, respectively. Al was the reference element for EF calculation, and the elemental composition of < 20 μm particles in the Tibetan topsoil were used as the elemental composition of the crustal material (Li *et al.*, 2009). This database considers the potential influence of regional dust and has been widely adopted in TP studies (Cong *et al.*, 2010; Lee *et al.*, 2011; Huang *et al.*, 2013).

Empirical Orthogonal Function

The interelement relationships in the Nam Co aerosols were investigated through an empirical orthogonal function (EOF) analysis, which was proved more accurate in assessing the behavior of multiple variants than simple linear regressions are (Meeker *et al.*, 1995). The EOFs were rotated using the Varimax criteria, with the number of factors to rotate determined by the common criteria of the number of eigenvalues of the correlation matrix greater than one. EOF analysis was conducted by Matlab 7.0.

Dry Deposition Fluxes Estimation

In this study, the dry deposition fluxes of different elements at the Nam Co station were estimated as follows:

$$F = V_d \times C_x \quad (2)$$

where V_d and C_x are the dry deposition velocity and atmospheric concentration of the element species, respectively (Hsu *et al.*, 2010). Elemental deposition velocities are used for calculating the dry deposition fluxes on the basis of their sources. Generally, anthropogenic (e.g., Cr, Ni, Zn, Cd, Pb, and Bi) and crustal elements usually contain fine and coarse particles, respectively. In this study, mean velocities of 0.36 cm s^{-1} and 1.00 cm s^{-1} were adopted for anthropogenic and crustal elements, respectively (Chen *et al.*, 2008).

RESULTS AND DISCUSSION

Elemental Concentrations

Table 1 presents the mean elemental concentrations of aerosols (mean ± standard deviation and range) collected at the Nam Co station during the 2-year study period together with data from other remote sites. Na, Mg, K, Fe, Ca, and Al were the dominant elements, with concentrations ranging from 98.9 (Na) to 352 (Al) ng m^{-3}, likely because of their abundance in the Earth's crust. Be, Cd, Tl, Bi, Th, and U were present in low concentrations in the order of several picograms per cubic meter. The elemental concentrations were much lower at Nam Co than those at other remote sites, such as Waliguan and Mt. Yulong, located at the

Table 1. A statistical summary of aerosol elemental concentrations (ng m^{-3}) determined at the Nam Co Station and comparison with those from other remote sites (ng m^{-3}).

	Nam Co (ng m^{-3})		Waliguan[a]	Yulong[b]	King Sejong[c]	Jungfraujoch[d]	Nord[e]
	Mean ± S.D.	Range					
Li	0.38 ± 0.41	0.0084–2.13					
Be	0.013 ± 0.012	0.0005–0.068			0.00073		
B	1.47 ± 1.58	0.0054–7.35					
Na	98.9 ± 113	4–571	541–1150				
Mg	106 ± 167	2.9–1166	1430–3230				
Al	352 ± 426	9.2–2473	2110–3410	1099.47	1.875		37.9
K	130 ± 147	5.04–883	1190–2200			2.4–17	23.2
Ca	277 ± 396	6.78–2558	2010–4280			4.1–47	29.7
Ti	23 ± 26	0.7–135	106–220	149.94		0.35–3.3	1.8
V	0.61 ± 0.68	0.006–3.55	3.02–5.88	1.76	0.037		0.1
Cr	3.65 ± 3.28	0.0547–17		3.16	0.114		0.08
Fe	207 ± 256	8–1418	27.5–52.9	1258.31		2.8–26	19.4
Mn	5.43 ± 5.41	0.2057–29.9	1720–3910	36.03		0.19–0.37	0.5
Co	0.114 ± 0.105	0.0057–0.619	0.63–1.84		0.0018		
Ni	1.49 ± 1.37	0.0899–7.6		2.95	0.826		0.1
Zn	2.39 ± 1.78	0.0005–9.4	9.5–26.3	71.75	0.133	0.59–16	1.3
Ga	0.102 ± 0.12	0.0026–0.69			0.0006		0.02
As	0.4 ± 0.4	0.0184–1.98	0.70–3.37	3.61			0.06
Rb	0.769 ± 0.84	0.0264–4.89	6.2–14.9		0.0061	0.006–0.038	0.08
Sr	2.01 ± 3.18	0.0349–23.09			0.684	0.024–0.18	0.4
Y	0.127 ± 0.149	0.0032–0.87				0.003–0.056	
Cd	0.01 ± 0.01	0.0000–0.04			0.00142		
Cs	0.15 ± 0.16	0.0028–0.71			0.0002		
Ba	2.55 ± 2.91	0.1091–18.27		12.67	0.0106		
Tl	0.009 ± 0.009	0.0002–0.056		149.94	0.0003		
Pb	0.57 ± 0.5	0.0221–2.34		12.37	0.041	0.23–5.4	0.6
Bi	0.008 ± 0.009	0.0003–0.081			0.0003		
Th	0.08 ± 0.09	0.0018–0.49					
U	0.027 ± 0.031	0.0007–0.173			0.0004		

[a] Aerosols collected at Waliguan, northeast of Tibetan Plateau (TP); 1992–1995 (Wen *et al*., 2001).
[b] Aerosols collected at Mt. Yulong, southeast of TP, January–February 2010 (Zhang *et al*., 2012a).
[c] Aerosols collected at King Sejong Station, Antarctic Peninsula, 2000–2001 (Mishra *et al*., 2004).
[d] Aerosols collected at the High Alpine Research Station, Europe, 1997 (Streit *et al*., 2000).
[e] Aerosols collected at Nord Station, Arctic, 2008–2010 (Nguyen *et al*., 2013).

northeastern and southeastern edge of the TP, respectively (Wen *et al.*, 2001; Zhang *et al.*, 2012a). However, compared with those in the Antarctic (Mishra *et al.*, 2004), Arctic (Nguyen *et al.*, 2013; VanCuren *et al.*, 2012), high Apls (Streit *et al.*, 2000), and Andes (Gaiero *et al.*, 2013; Cereceda-Balic *et al.*, 2012), the major elements with crustal origin had higher concentrations at Nam Co, indicating that the Nam Co atmosphere was mainly influenced by crustal dust. However, some anthropogenic originated trace elements (e.g. Cr, Ni, Pb, As, and Zn) levels at Nam Co were slightly higher than those reported at other remote sites, such as the Nord Station (20 m a.s.l.) in Northeast Greenland (Nguyen *et al.*, 2013) and Jungfraujoch station (the High Alpine Research Station: 3580 m a.s.l) (Streit *et al.*, 2000), suggesting more anthropogenic effects in the Nam Co region.

Enrichment Factors

Fig. 2 shows the EFs of aerosol elements during different seasons. The annual average EFs (Fig. 2(a)) of elements such as Be, Na, Mg, K, Ca, Fe, Ga, Rb, Sr, Y, and Ba were approximately 1, suggesting that these elements were predominantly of crustal origin. However, the EFs of Cr, Ni, Cd, Zn, Pb, and Bi exceeded 10 during the pre-monsoon and monsoon seasons, indicating that these elements partly originated from anthropogenic emissions. This differed from such remote regions as Taklimakan desert and Xinjiang Gobi which have different industrial emission sources from the neighboring regions as indicated by higher EF values for different elements (e.g., Sb, Mo, As, Sn and Co) (Zhang *et al.*, 2014). It was also different from heavily polluted cities like Beijing which reported very high EF values for Cd (407), Zn (86), and Pb (138) (Schleicher *et al.*, 2012), suggesting they had more effects of pollutant sources. The EFs of elements varied seasonally and were high during the pre-monsoon and monsoon seasons (Figs. 2(b) and 2(c)), indicating increased anthropogenic influence during these periods (Cong *et al.*, 2010; Huang *et al.*, 2013). The

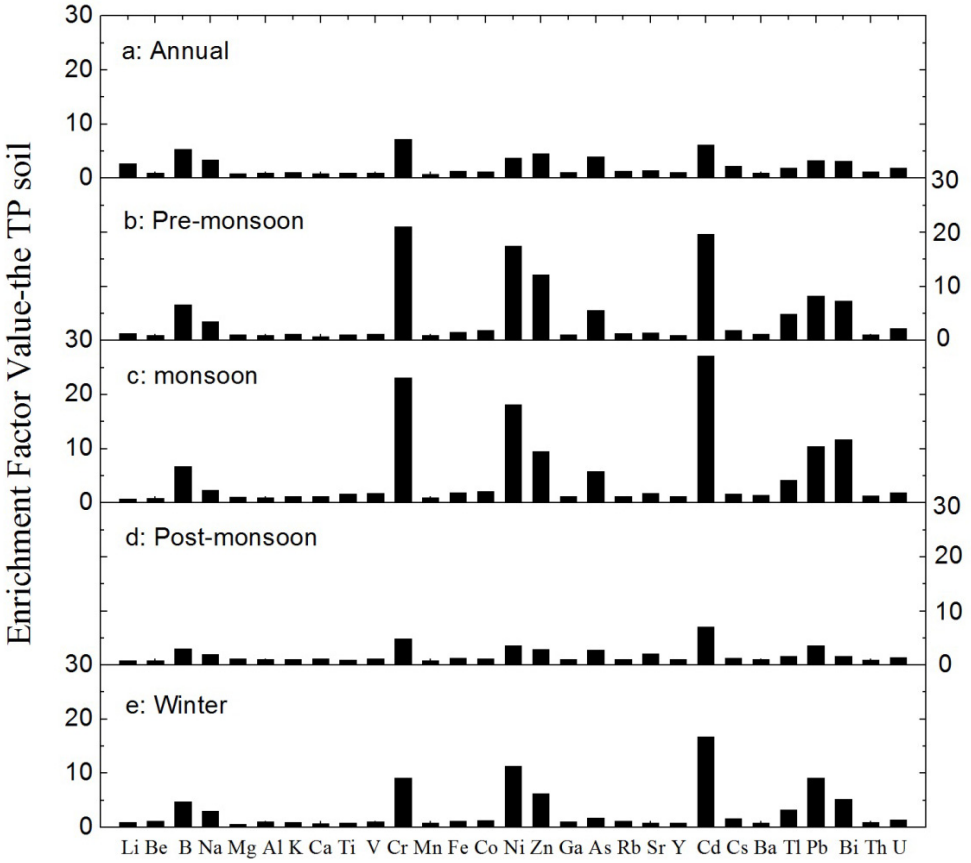

Fig. 2. Enrichment factors of elements in aerosols for annual average (a) and different seasons (b–e).

high EFs of these trace elements during the pre-monsoon season were consistent with the concentration of atmospheric brown clouds (ABCs) in South Asia (Ramanathan *et al.*, 2007). Thus, these pollutant-related elements with high EFs in the Nam Co region were possibly long-range transported from South Asia into the TP (Lüthi *et al.*, 2015) and were partly influenced by local emissions (Chen *et al.*, 2015). For example, Chen *et al.* (2015) reported that yak dung combustion could also emit aerosols with high elemental EF values (e.g., Cd, Zn, and Pb). However, it is hard to distinguish the source between long-range transport and local emissions based only on EF values.

Associations among Elements

Four EOF factors of contribution on whole samples are shown in Table 2. In this study, EOF1 accounted for 71.9% of the total variance in the elements' time series. Most major crustal elements (Al, K, Fe, Na, Mg, and Ca) and trace crustal elements (e.g., Li, Be, Ga, Rb, Sr, Ba, and Y) were strongly loaded on EOF1 (Table 1), which is suggestive of a shared origin and consistent with their EFs being nearly 1 (Fig. 2). Cr, Ni, and Zn dominated EOF2 with little loading on the other crustal elements (Table 2). Considering the high EFs in these elements (Fig. 2), the group of EOF2 elements clearly originated from anthropogenic sources which might be long-range transported from South Asia as mentioned above. For example, Cr is dominated by the emission from the fossil fuel combustion, steel industry or solid waste dumping, while Ni is regarded as an indicator of emission from fuel burning and vehicular emissions (Pacyna, 1984). And Zn may also be derived from similar sources, or other traffic-related sources (Chueinta *et al.*, 2000). EOF3 accounted for 6% of the total variance and was loaded primarily on Cd and Bi, which also exhibited high EFs, indicative of an anthropogenic source different from that of EOF2 elements, such as the combustion activities of local Tibetan residents (Li *et al.*, 2012a; Chen *et al.*, 2015). This phenomenon was also observed in an investigation of major ions in local precipitation (Li *et al.*, 2007a). EOF4 was mainly loaded on As, thus suggesting a unique source or transport pathway different from those of the other elements. Concentration of As is naturally high in the TP soil which may cause high levels in the atmosphere of the study region (Li *et al.*, 2009), and Guo *et al.* (2008) also reported that there existed large numbers of As-enriched geothermal springs in the southern TP. As-enriched sediments around these springs might be another contributor for high As concentration in the atmosphere of the region.

Potential Source Regions

According to the EOF associations, Al, Cr, Cd, and As were selected to represent variations in the EOF groups during the sampling period (Figs. 3 and SI-1). A trajectory model (HYSPLIT) was used to investigate the 5-day air mass backward trajectories over the Nam Co region (http://ready.arl.noaa.gov/HYSPLIT.php) (Fig. 4). The cluster

Table 2. Empirical orthogonal function (EOF) associations for elements in the Nam Co aerosols.

Elements	EOF1	EOF2	EOF3	EOF 4
Li	82.4	–0.3	–2.6	3.5
Be	91.1	–1.2	–0.7	0.5
B	46.9	0.1	0.0	2.3
Na	78.6	–1.9	–3.0	2.3
Mg	90.4	–0.1	–1.6	–1.9
Al	**98.0**	–0.3	–0.5	–0.5
K	**98.5**	–0.3	–0.7	0.0
Ca	92.9	–0.4	–0.7	–1.8
Ti	82.1	–0.2	0.0	–1.4
V	88.9	0.4	–0.6	0.0
Cr	2.9	**88.9**	0.0	0.0
Mn	95.9	–0.1	–0.2	–2.0
Fe	96.6	0.1	–0.4	–0.1
Co	93.6	2.7	–0.7	–0.6
Ni	14.5	**55.6**	0.2	–6.9
Zn	11.1	**41.9**	3.7	7.1
Ga	98.0	–0.3	–0.4	–0.4
As	35.6	5.1	–0.1	**26.2**
Rb	97.1	–0.5	–0.5	0.1
Sr	89.7	0.0	–1.5	–2.4
Y	97.0	–0.4	–0.1	–0.1
Cd	36.8	0.0	**46.5**	–5.7
Cs	70.3	–2.0	0.1	9.2
Ba	96.7	0.0	–0.3	–1.4
Tl	45.4	–1.1	24.2	–0.6
Pb	65.1	0.0	22.0	–1.6
Bi	5.6	–2.7	**61.8**	7.8
Th	95.3	–0.4	0.0	0.8
U	87.8	0.0	–0.3	2.2
Total	71.9	7.2	6	3.1

Note: The numbers represent the percentage of variance associated with the corresponding major elements. Negative values indicate an inverse relationship. The totals represent the percentage of total variance explained by each EOF.

trajectories were calculated using Global Data Assimilation System (GDAS) data at 500 m above ground starting from 00:00 Coordinated Universal Time. The Nam Co region was found to be clearly influenced by the Westerlies during the winter and pre-monsoon seasons and by the Indian monsoon during the monsoon season.

Al concentrations were the highest during the winter and pre-monsoon seasons and gradually decreased during the monsoon season with the lowest level in August (the highest monthly precipitation amount), reflecting a higher loading of crustal aerosol in the atmosphere during the non-monsoon period (Figs. 3 and SI-1). The TP is a source region for dust aerosol (Fang *et al.*, 2004). Generally, the dust storm activity occurs over the TP during the winter and pre-monsoon seasons (Song *et al.*, 2004). For example, on January 20, 2006, satellite data recorded a large dust event over the TP (National Satellite and Meteorological Center, 2006), which corresponded with high Al concentration at the Nam Co station (Fig. SI-1). Therefore, seasonal Al variation in Nam Co aerosols is consistent with dust storm activity over Central Asia, particularly over the TP.

The 5-day backward trajectories backtracked the air masses to South Asia (Nepal, India, and Pakistan) and Central Asia (Iran, Afghanistan, northwestern China). The air masses from central Asia and South Asia traveled thousands of kilometers in 5 days before arriving in the Nam Co region during the winter and pre-monsoon seasons (Fig. 4). Thus crustal aerosols (represented by Al concentrations) may be transported from the arid and semiarid regions of Central and South Asia during these two periods, as suggested by other researches (Han *et al.*, 2004; Kang *et al.*, 2007; Mishra *et al.*, 2014).

Cr concentrations in Nam Co aerosols, representative of such anthropogenic inputs as fossil fuel combustion, steel industry activities, and solid waste dumping (Pacyna, 1984), were high from April to June (or pre-monsoon season) (Fig. 3). The Cr might be from the northern Indo-Gangetic Plain where a 3 km thick brownish layer of pollutants (or ABCs) has been detected because of operation of brick kilns, vehicle emissions, steel industry or solid waste dumping, and biomass burning (Ramanathan *et al.*, 2007; Bonasoni *et al.*, 2010). The air mass backward trajectories suggested that more than half of the air masses at Nam Co were transported from the northern India and Nepal during the pre-monsoon season (Fig. 4), carrying with pollutants to the Nam Co region; this result is consistent with the findings of Lüthi *et al.* (2015).

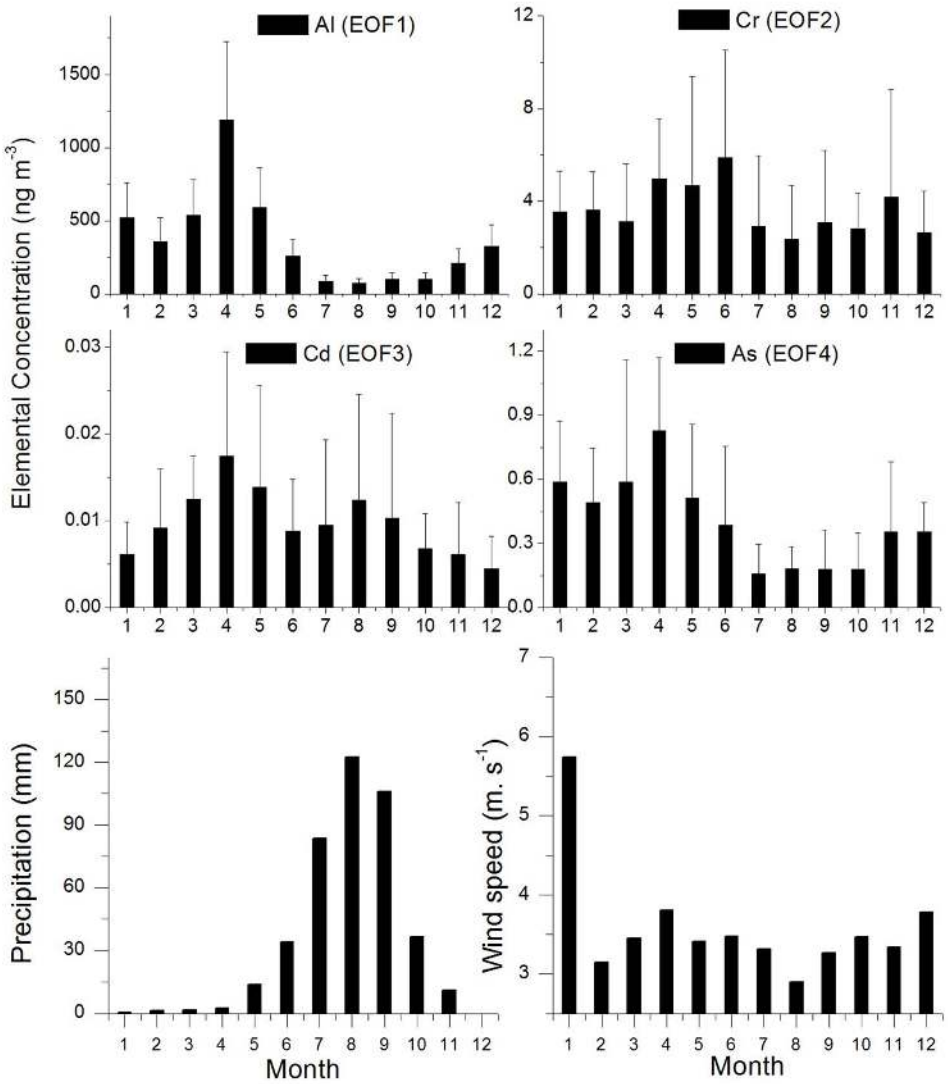

Fig. 3. Variations of monthly average concentrations for selected elements in Nam Co aerosols.

As discussed, Cd might indicate other anthropogenic inputs. High Cd concentrations were recorded during the pre-monsoon and monsoon seasons (Figs. 3 and SI-1). These high Cd concentrations might have the same origin as Cr. During the monsoon season, the Indian summer monsoon dominates the Nam Co region and transports air masses from the India Ocean to South Asia (Byson, 1986; Zheng *et al.*, 1997); this was affirmed by the 5-day backward trajectories that showed the majority of air masses arriving from Bangladesh and the northeastern India in the monsoon season (Fig. 4). Moreover, individual particle analyses suggested that the summer monsoon circulation transported pollutants from South Asia to the inland TP (Cong *et al.*, 2009a). In addition, local residents moving to the southeastern shore of Nam Co Lake from the northern regions utilize large amounts of biomass, particularly yak dung, for cooking and heating. Yak dung combustion releases Cd in high concentrations and EFs, possibly influencing the atmosphere (Chen *et al.*, 2015). Thus, the high Cd concentrations during the pre-monsoon and monsoon seasons can be attributed to both anthropogenic emissions transported from South Asia and local emissions.

The source of As compound was complicated. It had mainly two sources. The first was crustal source, which was similar with typical crustal elements such as Al or K. This phenomenon could be found in EOF1, where As element also showed some contribution (35.6%). The second was contribution of sediments from geothermal springs, which was abnormally enriched of As compounds and widely distributed in the southern TP (Li *et al.*, 2009). The contribution of this kind of sediments was especially high during the dry season (winter and pre-monsoon seasons). This was different from large cities such as Beijing which showed that the bioavailability of As in TSP from anthropogenic sources (e.g., coal combustion) was higher than that of geogenic As (e.g., long-range transported during dust storms) (Schleicher *et al.*, 2011).

Atmospheric pollution is severe in South Asia and is considered a major contributor to the Indo-Asian ABC haze because of high industrial emissions, coal burning, vehicular emissions, and waste incineration (Basha *et al.*, 2010; Hyer and Chew, 2010). Pollutants may reach higher elevations

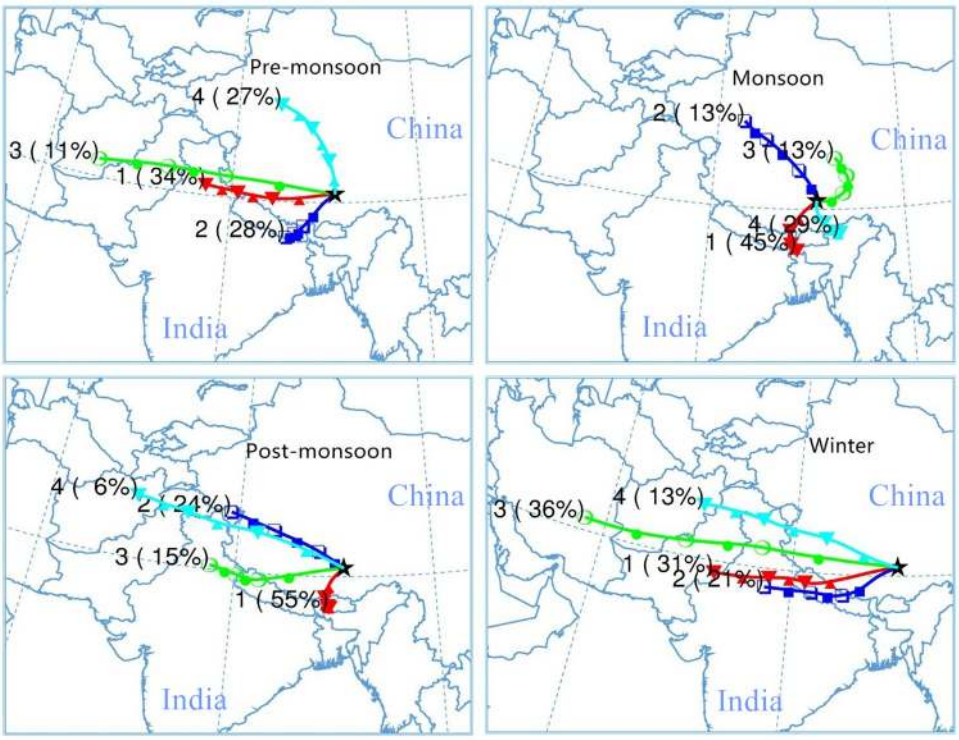

Fig. 4. Cluster-mean 5-day backward air trajectories for the Nam Co region simulated by the HYSPLIT V4.8 model.

or move farther to the inland TP through long-range transport (Ming *et al.*, 2010; Xia *et al.*, 2011; Zhao *et al.*, 2013; Lüthi *et al.*, 2015). In addition, air masses are transported from South Asia year-round (Fig. 4). Thus, considering the limited local pollution sources in the Nam Co region, we suggest that anthropogenic elements in aerosols are mainly transported by air masses from South Asia, particularly when intensified pollution events occur in South Asia during the pre-monsoon season.

Dry Deposition Fluxes of Trace Elements

Atmospheric pollutants are removed by both dry and wet deposition, and dry particulate deposition contributes to approximately 60% of all fluxes (Galarneau *et al.*, 2000). Elemental deposition fluxes are essential for evaluating atmospheric loading and input to both the terrestrial and aquatic ecosystems, and for assessing historical atmospheric changes using lake sediments and ice core records. So far, numerous approaches have been used to estimate the deposition velocities of aerosols (Jacobson, 2005; Hsu *et al.*, 2010).

Table 3 compares the dry deposition fluxes of elements at Nam Co with data from other media and sites. The dry deposition fluxes exhibited a pattern similar to that of their concentrations, with high levels observed for Al, K, Fe, Na, Mg, Ca, Li, Be, Ga, Rb, Sr, Ba, and Y, which were loaded on EOF1; this result indicates a heavy atmospheric dust load in the region (Fang *et al.*, 2004). Compared with fluxes in East China Sea (Hsu *et al.*, 2010), most of the fluxes in the Nam Co region were lower or comparable. However, some elements such as Al, Ti, Cr, Fe, and Ni had higher fluxes. Compared with fluxes reported earlier in the Nam Co region (Cong *et al.*, 2010), dry deposition fluxes in this study were higher than those of wet deposition (Table 3) because precipitation occurs mostly during the monsoon period. The total deposition flux in the Nam Co region was higher than that in Zhadang glacier. Such differences might be caused by two reasons: First, the Nam Co region has a relatively low elevation (Li *et al.*, 2007b). Second, the snow pit of Zhadang glacier was not representative of deposition throughout the year because large quantities of snow falling during the monsoon season melted because of the increased temperature in the TP (Kehrwald *et al.*, 2008; Kang *et al.*, 2015). However, the total elemental fluxes at Zhadang glacier were much lower than those in the lake core drilled at Nam Co Lake (Li *et al.*, 2011), implying that atmospheric deposition accounted for only a small proportion of the lake sediment records; a large proportion was derived from the river input to the lake basin (Li *et al.*, 2011). The fluxes reported in this study were rough estimations; nevertheless, they are useful in assessing the atmospheric environment and its impacts on the inland TP ecosystem.

CONCLUSIONS

Aerosol elements at the Nam Co station for a 2-year period showed lower elemental concentrations compared with data from other remote sites in the edge of the TP. The major elements, Na, Mg, Al, K, Ca, and Fe, exhibited similar seasonal variations, suggesting a common crustal origin. High concentrations of these elements occurred during the winter and pre-monsoon season, which corresponds with the dust storm activities in Central Asia and the TP, whereas

Table 3. The estimated dry deposition fluxes (μg m^{-2} yr^{-1}) for elements and compared with other related data.

	Nam Co station			Zhadang Glacier[b]	Lake sediments[c]	East China Sea[d]
	Dry	Wet[a]	Total	Total	Total	Dry
Li	103				20600	
Be	3.28				684	
B	273				14135	
Na	31500					
Mg	34400				6347825	
Al	113100	5510	118610	8989		14600
K	41550					
Ca	87500				33790424	
Ti	7400				547126	2190
V	189	33	222	10	13835	547.5
Cr	415	139	554	17	14139	83.95
Fe	66600	5020	71620		5769425	14235
Mn	1740	297	2037	360	127584	2445.5
Co	36.5	7.1	43.6	6	1951	25.19
Ni	310	97	407	23	8382	87.6
Zn	542	266	808	160	19076	6935
Ga	32.9				3720	24.46
As	101				4475	142.4
Rb	246				40634	511
Sr	650				177516	
Y	40.8				5199	12.41
Cd	1.11	1.8	2.91	0.2	48.5	69.35
Cs	46.3				8643	36.5
Ba	818				96854	511
Tl	1.98				226	12.41
Pb	137	60	197	18	7496	912.5
Bi	1.73				237	
Th	25.7				4445	
U	8.64				2789	

[a] Precipitation samples collected at Nam Co; 2007–2008 (Cong *et al*., 2010); [b] Snow samples collected at Zhadang glacier; 2009 (Huang *et al*., 2013); [c] Total deposition fluxes recovered from the Nam Co Lake core; part of the elemental data was published in Li *et al*. (2011); [d] Dry deposition fluxes in East China Sea; 2005–2007 (Hsu *et al*., 2010).

low concentrations occurred during the monsoon season. Cr, Ni, Zn, Cd, Pb, and Bi had relatively higher EFs during the pre-monsoon and monsoon seasons, indicating the possible influence of anthropogenic emissions. Seasonal variations and air mass backward trajectories revealed that higher concentrations of Cr, Ni, and Zn during the pre-monsoon season might be attributable primarily to long-range transport from the southern Himalayas during ABCs events. However, high Cd concentrations during the pre-monsoon and monsoon seasons could reflect the effect of long-range transported anthropogenic emissions from South Asia and local biomass combustions. Dry deposition fluxes, a large part of which occurred during the non-monsoon period, were much higher than wet deposition fluxes. This work provides a baseline for assessing atmospheric environment and interpreting paleo-environment changes in the TP.

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SUPPLEMENTARY MATERIALS

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

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