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Atmospheric wet deposition of trace elements to central Tibetan Plateau

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ABSTRACT

To investigate trace elements in wet precipitation over the Tibetan Plateau (TP), a total of 79 event-based precipitation samples were collected from September 2007 to September 2008 at Nam Co Station. Samples were analyzed for concentrations of Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb using inductively coupled plasma-mass spectrometry (ICP-MS). The annual volume-weighted concentrations of elements were generally comparable to other background sites, and much lower than urban areas. The enrichment factors (EF) showed that, in comparison with the Tibetan soils, the wet precipitation had elevated concentrations of Cr, Co, Ni, Cu, Zn, Cd and Pb, probably indicating their anthropogenic origins. Other elements (Al, Fe, Mn and V) with enrichment factor value of <10 may derive mainly from crustal sources. The principal component analysis further confirmed the two different groups of elements in wet deposition samples. The backward trajectories were calculated for each precipitation event using the NOAA HYSPLIT model. The results indicated significant differences of EF for trace elements of anthropogenic origin between the summer monsoon and non-monsoon seasons. The data obtained in the present study indicated that pollutants can affect remote high altitude regions like the Tibetan Plateau through long-range transport, especially in the summer monsoon season.

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1. Introduction

Since the 19th century, atmospheric trace elements have increased significantly due to human activities, especially industrial processes and fossil fuel combustion [\(Nriagu, 1996\)](#page-6-0). Trace elements can undergo long-range transport through the atmosphere and deposit in remote regions far away from populated areas ([Kyllonen et al., 2009\)](#page-6-0). Wet precipitation can efficiently scavenge trace elements present in air to the terrestrial or aquatic surface ([Kim et al., 2000; Shimamura et al., 2006; Nelson et al., 2008\)](#page-6-0). Long-term excessive inputs of such elements may impose burden on ecosystems and human health through various biogeochemical cycles.

Over the last few decades, intensive monitoring programs of trace metals in precipitation have been carried out worldwide, which have mainly focused on the chemical characteristics, deposition fluxes and long-term temporal trends ([Halstead et al., 2000;](#page-6-0) [Wong et al., 2003; Gabrielli et al., 2008; Garcia et al., 2009; Kyllo](#page-6-0)[nen et al., 2009](#page-6-0)). According to these studies, the loadings and sources of trace metals in precipitation have great spatial variability over different locations, which is mainly caused by different

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meteorological conditions and the emission patterns of pollutants. Therefore, more in-depth studies, especially on the regional level, of trace metal pollution in the air, are needed for the assessment of their impact on ecosystems.

The Tibetan Plateau (TP) has been regarded as a sensitive region to anthropogenic impact due to its unique landform, fragile ecosystem, and special monsoon circulation [\(Qiu, 2008](#page-6-0)). However, until now, there have been only a very few studies regarding the chemistry of wet precipitation over the TP, partly due to the difficulty of field sampling there. The remoteness, high altitude and harsh weather conditions make a continuous sampling campaign very challenging. Moreover, existing works on the TP have only dealt with major ions in wet precipitation while trace elements have not been studied [\(Zhang et al., 2003; Li et al., 2007](#page-6-0)).

Recently, several studies have elucidated changes of trace elements related to anthropogenic activities in snow and ice cores from the Tibetan Plateau and its surrounding area, such as eastern Tien Shan, Mt. Muztagh Ata in the eastern Pamirs ([Li et al., 2006\)](#page-6-0), and Mt. Qomolangma (Everest) in the Himalayas [\(Kang et al., 2007;](#page-6-0) [Lee et al., 2008; Hong et al., 2009; Kaspari et al., 2009\)](#page-6-0). Impurities in ice cores come mainly from the atmosphere through wet and dry deposition. Therefore, the patterns of trace elements in yearround wet precipitation can be of value to the interpretation of trace metals preserved in snow and ice core.

The Tibetan Plateau is the source region of the 10 largest rivers in Asia, including the Yangtze River, Yellow River, Yarlung Tsangpo

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(Brahmaputra), Ganges River, Indus River, etc., which are the water sources for about 40% of the world's population. Recently, elevated concentrations of several potentially harmful metals have been found in these large rivers [\(Huang et al., 2008](#page-6-0)). Because all these rivers are both snow- and rain-fed, further study is needed to determine whether there is a link between the chemical composition of wet precipitation in the upper stream catchment region and the pollution of river water.

The objectives of the present study are to: (1) investigate the concentrations of trace elements in wet precipitation in the Tibetan Plateau; (2) estimate wet deposition fluxes of trace elements in the region; (3) identify anthropogenic origins of trace elements in wet deposition; and (4) provide a reliable database of trace elements in precipitation for future temporal trend study.

2. Sampling and analysis

2.1. Site description

Nam Co Station for Multisphere Observation and Research (briefly Nam Co Station, N30°46.44′, E90°59.31′, 4730 m a.s.l.) is situated at the SE shore of Nam Co Lake in the central TP (Fig. 1). The landscape surrounding Nam Co Station mainly consists of high mountains, glaciers, lake and grassland, which are representative of major geomorphologic features of the TP, and prone to be fragile regarding climate change and anthropogenic impacts. The Nam Co

Fig. 1. Location map for the sampling site at Nam Co, central Tibetan Plateau.

Station is relatively isolated from major industrial sources and populated areas. Due to the harsh climate, the local population density within a 25 km distance from the station is less than 1 person per $km²$. The local inhabitants mainly make a living by herding sheep and yak, and produce very little atmospheric pollutant emissions in the vicinity of the station. According to aerosol optical measurement result, the annual mean value of aerosol optical depth (0.05 at 500 nm) at Nam Co is relatively lower than or comparable to other remote sites like Mauna Loa and Dome C, Antarctica ([Cong](#page-6-0) [et al., 2009\)](#page-6-0). Therefore, as one of the most pristine stations in the AERONET network, Nam Co represents a clean continental background site for atmospheric environmental monitoring.

Generally, the Nam Co region is subject to two main atmospheric currents. In summer, the region is under the influence of the Indian Monsoon which is characterized by relatively higher temperature and humid weather with prevailing southerly winds ([You et al., 2007](#page-6-0)). While in other seasons, large scale atmospheric circulation patterns over the region are mainly dominated by westerlies, resulting in limited precipitation. The mean annual air pressure and temperature is 571.2 hPa and 0° C, respectively. The minimum temperature occurs in December, and the maximum in July. The mean annual relative humidity is 52.6%, and annual wind speed is 3.99 m s^{-1} . The annual precipitation is around 450 mm, with the majority of precipitation occurring in the summer monsoon season. The record of precipitation events and amounts at Nam Co Station during the sampling campaign is shown in Fig. 2.

2.2. Sampling

All precipitation events from September 2007 to September 2008 were collected with a total of 79 precipitation samples. Precipitation samples were obtained by an automated precipitation sampler (SYC-2, Laoshan Electronic Instrument Complex Co., Ltd.). The schematic of this type of sampler can be found in the literature [\(Reeve, 2002](#page-6-0)). Briefly, the sampler consists of a rain sensor, rain container and a dust preventing cover. When there was rain, the rainfall sensor would activate the cover to open automatically, thereby exposing the container to wet precipitation. The container was closed when the sensor became dry after the rain. The amount of precipitation in each event was also recorded simultaneously by the sampler. The wet precipitation was stored in a HDPE plastic bag. The conductivity and pH were determined immediately after sampling at the Nam Co Station. Precipitation samples were transferred into pre-cleaned HDPE bottles, and kept frozen at the station and during transport to laboratory.

Fig. 2. Variation of amount of precipitation from September 2007 to September 2008 at Nam Co Station.

The total sampled precipitation was 490.3 mm, accounting for 95.3% of the total precipitation (514.5 mm) in the investigated period. For the purpose of quality assurance and quality control (QA/ QC) of the monitoring results, field blanks were also evaluated monthly. During the field blank sampling procedure, deionized water was flushed through the sampler and was then collected as a field blank solution. Extreme care was taken during the collection, handling, and storage of precipitation samples to minimize contamination. Detailed sampling protocol has been described elsewhere by [Li et al. \(2007\).](#page-6-0)

2.3. Chemical analysis

In the laboratory, precipitation samples were filtered through 0.4 μ m polycarbonate membrane, and then acidified to pH < 2 with ultra-pure $HNO₃$. The concentrations of 11 elements (Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb) were measured directly by inductively coupled plasma-mass spectrometry (ICP-MS, X-7 Thermo Elemental) at the Institute of Tibetan Plateau Research in Beijing. Elemental concentrations were quantified using external calibration standards. An analytical standard was analyzed after the initial calibration, and after every 10 samples.

The method detection limits (MDLs), defined as three times the standard deviation of replicate blank measurements, were Al, 0.063 μg L $^{-1}$; V, 0.016 μg L $^{-1}$; Cr, 0.032 μg L $^{-1}$; Mn, 0.005 μg L $^{-1}$; Fe, 0.618 μ g L⁻¹; Co, 0.001 μ g L⁻¹; Ni, 0.036 μ g L⁻¹; Cu, 0.042 μ g L⁻¹; Zn, 0.037 μ g L⁻¹; Cd, 0.001 μ g L⁻¹; and Pb, 0.006 μ g L⁻¹. The accuracy of the analytical protocol was ascertained based on repeated measurement of an externally certified reference solution (AccuTrace Reference Standard). The recoveries ranged from 85% for Cr to 105% for Ni. Regarding the analytical precision, the corresponding RSD values of all element concentrations measured in the reference material were less than 5%. The trace metal amounts found in the field blanks were generally lower than the detection limits or <10% of the amount found in the precipitation sample from Nam Co Station. Thus field blank values were not quantitatively subtracted in the calculation of metal concentrations in samples.

2.4. Back trajectory analysis

To identify the influence of air mass from different transport pathways on the elemental composition of wet precipitation at Nam Co, backward trajectory analysis was conducted for the field sampling period using the HYSPLIT model, developed by NOAA/ ARL ([Draxler and Rolph, 2003\)](#page-6-0). In this study, 5-day back trajectories were calculated at a height of 1000 m above ground for each sampling day. The trajectories were generated using Global Data Assimilation System (GDAS) meteorological archive from National Center for Environmental Prediction (NCEP). The errors accompanying HYSPLIT-generated trajectories were estimated to be in the range of 15–30% of the travel distance, and uncertainty increases with the distance of transport [\(Stohl, 1998](#page-6-0)).

3. Results and discussion

3.1. Data summary

The pH of precipitation samples in this study ranged from 6.42 to 8.95 with a median value of 7.85. All precipitation samples had a pH value higher than 5.6, which is the pH of unpolluted water equilibrated with atmospheric $CO₂$ [\(Charlson and Rodhe, 1982\)](#page-6-0). The pH value of Nam Co precipitation is comparable to those from other arid and semi-arid regions where alkaline airborne dust aerosols could efficiently neutralize the acidity in precipitation. For example, 7.27 was recorded at Urumqi River Valley, NW China ([Zhao et al., 2008\)](#page-6-0), 7.5 at Lhasa ([Zhang et al., 2003\)](#page-6-0), 6.35 at Waliguan (GAW) station in northeastern TP [\(Tang et al., 2000\)](#page-6-0), 7.01 at Dayalbagh, North Central India ([Kumar et al., 2002](#page-6-0)), and 6.4 in Northern Jordan ([Al-Momani, 2003\)](#page-6-0).

The volume-weighted mean (VWM) concentrations, standard deviations of the VWM concentrations, minimum and maximum concentrations of trace elements are shown in [Table 1.](#page-3-0) The standard deviations of the VWM were calculated using the formula from [Galloway et al. \(1984\).](#page-6-0) According to the concentrations, the elements measured can be divided into three groups: Al, Fe and Zn with an average concentration of higher than 1 μ g L⁻¹, Cr, Mn, Cu, Ni, and Pb with concentrations between 0.1 and $1 \mu g L^{-1}$, and V, Co, and Cd with concentrations lower than $0.1~\mu{\rm g}$ ${\rm L}^{-1}.$

The results are compared to the data reported for various locations around the world in [Table 2.](#page-3-0) For the crustal elements, such as Al and Fe, the concentrations at Nam Co are substantially lower than at other sites, reflecting a low soil particle loading in the atmosphere. Among those sites, Northern Jordan and Mersin in Turkey exhibit the highest value of crustal elements, which were ascribed to the high aerosol dust in the arid and semi-arid climate ([Al-Momani, 2003; Ozsoy and Ornektekin, 2009\)](#page-6-0). With respect to the anthropogenic elements, such as Cr, Ni, Cu and Pb, the concentrations at Nam Co are generally comparable to the background sites, such as the Aegean Sea, Greece, Nakanoto, Japan and Reston, Virginia, while they are 2–30-fold lower than the urban sites, such as Singapore ([Hu and Balasubramanian, 2003](#page-6-0)) and Mexico City ([Baez et al., 2007](#page-6-0)). To understand the possible spatial variation of trace elements in wet precipitation over TP, the concentrations of trace elements in this work are also compared with the fresh snow from Mt. Qomolangma (Everest), Himalayas ([Zhang et al., 2008\)](#page-6-0). As seen in [Table 2,](#page-3-0) the concentrations of Fe, Mn, Cu and Zn in the precipitation samples were similar to those in fresh snow from the Himalayas. This suggests that there is a relatively homogeneous distribution in the wet precipitation chemistry of the southern part of the TP.

3.2. Estimates of natural versus anthropogenic contributions

The enrichment factor (EF) of elements in precipitation relative to crustal material (e.g., Al, Fe) is often used to evaluate the degree of anthropogenic influence [\(Hu and Balasubramanian, 2003; Kyllo](#page-6-0)[nen et al., 2009](#page-6-0)), and is defined as follows:

$$
EF_X = \frac{(C_X/C_R)_{precipitation}}{(C_X/C_R)_{crust}}
$$

where X represents the element of interest; EF_X , is the enrichment factor of X; C_X , the concentration of X; and C_R , the concentration of a reference element. The precipitation and crust subscripts refer to wet precipitation samples and the average crustal material, respectively.

The main source of uncertainty in EF calculations is the definition of the reference crustal material. Although in most previous studies this was based on the average upper continental crust (UCC) compositions, the types of crustal materials could be different in different locations. Therefore, in this study, the average top soil composition from the Tibetan Plateau [\(Li et al., 2009\)](#page-6-0) was used as the elemental reference instead of UCC to reduce this uncertainty. Aluminium was selected as the reference element for the EF calculation. Elements with an EF value close to unity indicate strong influence of natural components, whereas high values of EF may indicate potential anthropogenic origins. In the present work, elements with an EF > 10 are considered to be of mainly anthropogenic origin.

Table 1

Statistical data for elemental concentrations (µg L^{-1}) in wet precipitation at Nam Co (2007–2008).

^a Volume-weighted mean.

b Volume-weighted standard deviation.

^c Median value.

d Below detection limit.

Table 2

Comparison of the trace element VWM concentration (μ g L⁻¹) with the data reported from various locations around the world.

Means the data is not VWM concentration.

^a [Sakata and Asakura \(2009\)](#page-6-0).

b [Koulousaris et al. \(2009\)](#page-6-0).

 c [Al-Momani \(2003\).](#page-6-0)

d Conko et al. (2004) .

e Baez et al. (2007)

[Baez et al. \(2007\).](#page-6-0)

^f [Hu and Balasubramanian \(2003\)](#page-6-0).

^g [Ozsoy and Ornektekin \(2009\)](#page-6-0).

h [Zhang et al. \(2008\)](#page-6-0).

 \overline{a} [Aizen et al. \(2009\).](#page-6-0)

^j [Kaspari et al. \(2009\)](#page-6-0).

Fig. 3. Average enrichment factors of trace metals in wet precipitations at Nam Co.

Enrichment factor values for trace elements in Nam Co precipitation samples are shown in [Fig. 3.](#page-3-0) Clearly, the EF value varies substantially among different elements, with the lowest for Fe (1.41) and the highest for Zn (357). Overall, the determined elements could be divided into two groups: Non-enriched elements, such as Al (the crustal reference element), Fe, Mn and V with volumeweighted mean EF values of 1–10, indicating soil dust as the dominant source; moderately to highly enriched element with EF values in the range of 10–1000 include Cr, Co, Ni, Cu, Zn, Cd and Pb, reflecting an important contribution from anthropogenic sources.

When the EFs of an element in each sample are plotted against the concentration of Al (EF-Al diagram), the points should form a horizontal line, independent from Al concentrations for purely crustal elements. On the other hand, the EFs of non-crustal elements would decrease with increasing Al concentration [\(Al-](#page-6-0)[Momani, 2003\)](#page-6-0). In Fig. 4, the plots for Fe, Mn and V with EF less than 10 are relatively constant, indicating their crustal material origins. However, other elements all show high EF values at low Al concentrations, and low EF values at high Al concentrations, indicating the major anthropogenic contributions to these elements in precipitation.

Among the enriched elements, Cr is dominated by emissions from fossil fuel combustion, the steel industry or solid waste dumping ([Wise et al., 2009\)](#page-6-0), while Ni is generally regarded as an indicator of emissions from fuel burning and traffic sources. The major sources of Cu in atmospheric particles are from the combustion of fossil fuels, industrial metallurgical process and waste incineration ([Nriagu, 1996\)](#page-6-0). Zinc may be derived from similar sources, or traffic-related activities ([Adachi and Tainosho, 2004\)](#page-6-0). Since the 1980s, leaded gasoline has been phased out gradually to lower atmospheric Pb pollution, while relatively high levels of Pb still exist in some areas due to coal burning and re-suspension of contaminated soil particles [\(McConnell and Edwards, 2008; Oz](#page-6-0)[soy and Ornektekin, 2009](#page-6-0)). Because of low emissions of industrial pollutants from local sources in the TP, trace elements in the precipitation could be long-range and transported into the Nam Co region by atmospheric circulation, and deposited in the high altitude area (4730 m a.s.l.).

3.3. Wet deposition fluxes of trace metals

The wet deposition flux (μ g m $^{-2}$ a $^{-1}$) of each element was calculated on the basis of the sum of the amounts (elemental concentration multiplied by each precipitation volume) in the precipitation collected for the whole year. The results and data from world literature are listed in [Table 3](#page-5-0).

It is clear that Al, Fe and Mn represented the highest loadings because of their high concentrations in dust derived from crustal materials. The deposition fluxes of Mn, Fe, Cd and Pb at Nam Co were similar to the remote marine site, Fiordland at New Zealand, while Cu and Zn are significantly higher. The deposition fluxes of all of the elements at Nam Co are one to two orders of magnitude lower than other sites close to urban or industrial locations. (see [Table 3](#page-5-0)).

The wet deposition fluxes of elements depend on both the concentrations in the precipitation and the precipitation amount. For example, the concentrations of Cu, Zn, Ni and Pb at Nam Co are comparable to those at Nakanoto ([Table 2\)](#page-3-0), a background site under the influence of long-range transported pollutants from the Asian continent, especially in winter. However, the total wet deposition fluxes at Nam Co were one order of magnitude lower due to the much higher annual amount of precipitation at Nakanoto $(2240 \pm 170 \text{ mm})$ in comparison with Nam Co (514.5 mm) .

3.4. Principal component analysis

Principal component analysis is a multivariate statistical method, which is frequently used to simplify large and complex data sets in order identify potential pollution sources. In this study, VARIMAX rotated principal component analysis was performed (SPSS 13.0) with the elemental data obtained by ICP-MS analysis. But V and Cd were not considered in this analysis, because their concentrations in more than 30% of the samples were below the detection limits. The PCA results are presented in [Table 4](#page-5-0). Two major components were identified with eigen values greater than 1, which explained a sum of 83.54% of the overall variances in the whole data set. Factor loadings considered as significant are marked as bold type in [Table 4.](#page-5-0) The first component is largely associated with Cr, Co, Ni, Cu, Zn and Pb accounting for 46.39% of the total variance, which clearly represents the anthropogenic contribution, such as fossil fuel combustion, traffic emission and metal smelting [\(Al-Momani, 2003; Ozsoy and Ornektekin, 2009; Song](#page-6-0) [and Gao, 2009](#page-6-0)). These elements were all enriched in comparison with crustal materials as shown in [Fig. 3](#page-3-0), indicating a good match between PCA and EF calculations. The second component has a high loading for Al, Mn and Fe, which are typical elements of dust

Fig. 4. The crustal enrichment factor (EF) versus Al concentration plot for trace elements in precipitation.

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Comparison of wet deposition fluxes of trace metals (μ g m⁻² a⁻¹) in different sites around the world.

^a [Halstead et al. \(2000\)](#page-6-0).

 b [Kim et al. \(2000\).](#page-6-0)</sup>

[Sakata and Asakura \(2009\)](#page-6-0).

[Al-Momani et al. \(1998\)](#page-6-0).

[Conko et al. \(2004\).](#page-6-0)

^f [Hu and Balasubramanian \(2003\)](#page-6-0).

Table 4

Factor loadings normalized with VARIMAX rotation.

particles (crustal source). This factor accounts for 37.15% of the total variance in the data set.

3.5. Back trajectories analysis

As described in Section [2.1,](#page-1-0) the seasonality of Nam Co can be roughly divided into the summer monsoon and non-monsoon seasons. The trajectories achieved by HYSPLIT revealed two different sources of air masses at the sampling site (Fig. 5). In the summer monsoon period (late June–early September), slow-moving air masses come from Bangladesh and NE India. In other seasons, most of the air masses are rapidly moving and come from the west. Therefore, the two different air mass pathways over the TP generally correspond to the summer monsoon and a westerly system.

The EF ratios for each element during the summer monsoon and non-monsoon seasons were also calculated. The ratio of EF_{monsoon} to EFnon-monsoon for V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd and Pb were 1.24, 3.37, 1.41, 1.56, 2.97, 3.63, 3.22, 3.42, 2.96 and 2.85, respectively. The results showed a clear seasonal difference for the enrichment of the non-crustal elements. In the summer monsoon season, the Nam Co region received more significant anthropogenic inputs. This seasonal pattern agrees well with the trace element changes recorded in snow pits of the central Himalayas ([Kang](#page-6-0) [et al., 2007; Lee et al., 2008\)](#page-6-0), indicating that there is a similar pattern at a regional scale over the southern TP. Namely, that the summer monsoon circulation can bring considerable pollutants from South Asia to the TP, while air masses belonging to the westerly flow do not carry large quantities of pollutants because they originate and travel far away from the industrialized and heavily populated regions [\(Xiao et al., in press\)](#page-6-0). Atmospheric pollution is serious in South Asia [\(Salam et al., 2008](#page-6-0)). A previous study on elemental composition of aerosols from Nam Co also suggested that several potentially harmful metals may be transported over long distances from South Asia ([Cong et al., 2007](#page-6-0)). Recently, [Xu et al.](#page-6-0) [\(2009\)](#page-6-0) pointed out that extensive black soot aerosols in South Asia could be lofted to the high TP and cause rapid glacier retreat. Therefore, the long-range transport of atmospheric pollutants from South Asia with the summer monsoon circulation as well as its environmental impact need more research.

4. Conclusions

The concentrations and wet deposition fluxes of trace elements were determined at Nam Co, a remote high altitude site in the central Tibetan Plateau from 2007 to 2008. The concentrations of trace

Fig. 5. Frequency plot of 5-day back trajectories for the summer monsoon (August 2008) and non-monsoon periods (January 2008) arriving at Nam Co Station. Back trajectories were run every 6 h using HYSPLIT and the NCEP reanalysis data.

elements in wet precipitation from Nam Co were among the lowest measured values reported worldwide. Their concentrations were also comparable to those in the fresh snow at the summit of the world (Mt. Everest, Himalayas). However, some elements (Cr, Co, Ni, Cu, Zn, Cd and Pb) were significantly enriched in wet precipitation relative to crustal materials, suggesting their potential anthropogenic sources. The current wet deposition fluxes of trace elements at Nam Co were generally lower than at other sites throughout the world, due to the low concentrations of metals and limited precipitation.

The backward trajectories revealed two different sources of air masses arriving at Nam Co over the year corresponding to the summer monsoon and non-monsoon seasons, respectively. Furthermore, the non-crustal elements have much higher EF values in the monsoon season. Therefore, the summer monsoon circulation may bring considerable pollutants from South Asia, while the dominant westerly flow in winter was relatively clean. In general, the results suggested that pollutants have affected this pristine region through long-range transport, especially in the summer monsoon season.

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