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Seasonal Variation, Sources and Transport of Aerosols at Lijiang, Southeast Tibetan Plateau

Ningning Zhang[1,2*], Junji Cao[1,3], Rujin Huang[1,4,5], Yuanqing He[2], Qiyuan Wang[1], Chongshu Zhu[1]

[1] *Key Laboratory of Aerosol Chemistry and Physics, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710061, China*
[2] *State key Laboratory of Cryspheric science, Cold and Arid Regions Environmental and Engineering Research Institute, Chinese Academy of Sciences, Lanzhou, 730000, China*
[3] *Institute of Global Environmental Change, Xi'an Jiaotong University, Xi'an, China*
[4] *Laboratory of Atmospheric Chemistry, Paul Scherrer Institute (PSI), 5232 Villigen, Switzerland*
[5] *Centre for Atmospheric and Environmental Studies, Xiamen Huaxia University, Xiamen 361024, China*

ABSTRACT

Aerosol samples were collected during pre-monsoon, monsoon and post-monsoon periods in 2009 in Lijiang, a tourism city located on the southeast Tibetan Plateau, southwest China. To determine the seasonal variation and sources of aerosol species, main elements and water soluble ions were analyzed. The results showed that crustal elements (Si, K, Ca, Ti and Fe) were the main elements with an enrichment whose enrichment factor (EF) value were lower than 10, with the large value (except Ca) occurring during the pre-monsoon period. The EF values of S, Cl, Zn, As, Br, Sb, Pb, and Cu were higher than those of the crustal elements and the large concentration appeared during the monsoon period. Ca^{2+} and SO_4^{2-} were the dominant cation and anion, respectively. The greatest value of total ionic concentration was found during the monsoon period, mainly because of the high concentration of SO_4^{2-} and NH_4^+. Using the positive matrix factorization model, it was found that the main sources of species were from crustal source, transport from south Asia and eastern China, local vehicle emissions and sea salt. Further results indicated that the pollutants mixed with dust, anthropogenic pollutants and biomass burning emissions can be transported to Lijiang from south Asia and Southeast Asia during the pre-monsoon period. In addition, pollutants rich in SO_4^{2-} and heavy metal from the Sichuan Basin and eastern Yunnan Province can also be occasionally transported to Lijiang during the monsoon period. The seasonal differences in chemical composition and transportation pathway may have important implications for regional climate change.

Keywords: Aerosol; Chemical composition; Sources identification; Transport pathway; Lijiang.

INTRODUCTION

Atmospheric aerosol research is an important tool for understanding the atmospheric environment. In particular, quantification of the aerosol properties at remote sites is not only important for assessing the transport of pollution caused by human activities but also for investigating large-scale aerosol effects on biogeochemical cycles and climate change (Qu *et al.*, 2009). The Tibetan Plateau (TP) is the highest plateau in the world and exerts a profound thermal and dynamic influence on the local and global climate (Chan *et al.*, 2006). Furthermore, its location is far from any industrialized area and it has a relatively small population. Therefore the plateau has been frequently chosen as an ideal location to observe the atmospheric environment and to evaluate various impacts of human activities (Kang *et al.*, 2002; Cong *et al.*, 2007) and several aerosol particle studies have been conducted over the TP region (Wake *et al.*, 1994; Cong *et al.*, 2007; Ming *et al.*, 2007; Cao *et al.*, 2009; Wu *et al.*, 2009; Yang *et al.*, 2009; Zhang *et al.*, 2010, 2012a; Zhao *et al.*, 2013; Zhang *et al.*, 2014). For studies aiming to investigate the influences on regional climate change, water resources and glacier retreat by polluted aerosols from south Asia, considerable attention has been focused on the south slope of the TP (Hindman and Upadhyay, 2002; Ramanathan *et al.*, 2007; Bonasoni *et al.*, 2008, 2010; Decesari *et al.*, 2010). However, these studies have not given substantial attention to the southeast TP.

Under the control of prevailing westerlies, the southeast TP and southwest China are located downwind of South Asia. In addition, the southeast TP is also influenced by

* Corresponding author.
E-mail address: zhangnn@ieecas.cn

equatorial-maritime air originating in the Indian Ocean, equatorial-maritime air from the western Pacific and tropical-maritime air originating over the northern Pacific (Araguás-Araguás *et al.*, 1998). Thus, not only the pollutants from south Asia, but also from southeast Asia and east China can be transported to the southeast TP and southwest China (Chan *et al.*, 2006; Qu *et al.*, 2007; Engling *et al.*, 2011). Investigations into the aerosol chemistry of the southeastern TP and its surrounding areas are important for understanding the current impacts of air pollution and for developing scenarios for potential changes in the future.

Lijiang city is located in the southeastern TP, southwest China. Because of its low level of human activities and position far from industrial activities, it was selected as a unique inland precipitation background site during 1987–1989 (Liu *et al.*, 1993). Since 1990, Lijiang has gradually become a world famous tourist site; the number of tourists visiting the city exceeded 7 million in 2009. With the increasing numbers of tourists, the number of vehicles increased from 9,000 in 1995 to 87,000 in 2008. Moreover, in order to encourage tourism, heavy industry development is prevented in Lijiang district, and local farmers and herders are also engaged in tourism-related activities. Now, Lijiang has transformed from a remote village to a tourist city, and the tertiary industry has become the dominant industry. These changes have had an influence on the surrounding environment; for example, the region's water resources (Ning et al., 2007) and precipitation chemistry (Zhang *et al.*, 2012b) have been noticeably influenced. However, there have been few recent reports of the region's aerosol chemistry (Zhang *et al.*, 2007). The purpose of the present study is as follows: (1) to investigate the chemical composition and seasonal variation of aerosol samples, (2) to determine the possible sources of observed species in the atmosphere, and (3) to attempt to evaluate the transport pathways of pollutants.

SAMPLING AND METHODS

Sampling and Analysis

The sampling site was positioned about 5 m above the ground and 1.5 m above the roof of a building, about 300 m away from main road. The building is located in a residential area and there are no industrial emissions around the site. Total suspended particulates (TSP) samples were collected once every two days during the pre-monsoon period (late March to early May), and once a day during monsoon (mid-July to August) and post-monsoon (mid-November to December) periods in 2009. Teflon® Zefluor™ filters (Pall Corporation, Port Washington, NY) at 47 mm diameter and 2 μm pore size were used for sampling. The collector (Zambelli Easy Plus 1, Zambelli srl, Milan, Italy) was driven by 220 V alternating current, and the sampled volume was measured by an in-line flow meter with a mean flow rate of 16.7 L min^{-1}. The ambient temperature and pressure were simultaneously measured for calculating standard (101,325 Pa, 273 K) cubic meters. In total, excluding the samples disturbed by power and precipitation (sampling time of less than 12 h), 23, 36, and 33 TSP samples were selected for pre-monsoon, monsoon, and post-monsoon periods, respectively.

The filter cartridges were packed in clean plastic bags and transported in airtight containers. After sampling, the filters were removed from the cartridges and placed in pre-cleaned airtight methylene bottles. Samples and seven blank filters were taken to minimize contamination both in the field and in the laboratory before analysis at the Institute of Earth Environment, Chinese Academy of Sciences (Xi'an).

In order to analyze the major ionic and elemental concentrations, the samples and blank filters were cut into two pieces. Half of each filter sample was used to determine the major soluble ions (SO_4^{2-}, NO_3^-, Cl^-, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}), using aqueous extracts of the filters by ion chromatography (IC, Dionex 500, Dionex Corp, Sunnyvale, CA). The other half was determined by Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometry using the PANalytical Epsilon 5 XRF analyzer (PANalytical B.V., Almelo, The Netherlands). The elements that were determined by the EDXRF method include Al, Si, S, Ca, K, Mn, Ti, Fe, Ni, Zn, As, Br, Pb, Sb and Cu with the detection limits (μg cm^{-2}) of 0.115, 0.093, 0.032, 0.007, 0.007, 0.014, 0.005, 0.011, 0.003, 0.008, 0.000, 0.006, 0.015, 0.033 and 0.010, respectively. The specific laboratory methods for ion and element detection are the same as described in Zhang *et al.* (2012a), and the aerosol concentrations reported in this study have been adjusted to remove the blank concentration value.

Meteorology

The climate at Lijiang is mainly dominated by the southwesterly summer monsoon (from May to October) and controlled by the southern branch of the westerly jet in winter (from November to the following April) (Pang *et al.*, 2006). During the sampling periods, the climate of Lijiang was controlled by western wind during pre-monsoon and post-monsoon period, and a greater wind speed was observed during post-monsoon period (exceeding 22 m s^{-1}) than that during pre-monsoon period (less than 13 m s^{-1}) at the level of 500 hPa (Fig. 1). During the monsoon period, a low wind speed and a high precipitation and relative humidity were observed (Fig. 1, Table 1). In addition, the summer vacation occurs during the monsoon period, and an increased number of people travel to the Lijiang area (Table 1). This may cause increased emissions of anthropogenic pollutants into the atmosphere.

Satellite Data and Backward Trajectory

In order to investigate the influences on aerosol chemistry in Lijiang by the long-range transport, satellite data and NCEP/NCAR reanalysis data were used. Satellites aerosol products including Moderate Resolution Imaging Spectrometer (MODIS) and the level-2G UV aerosol index (AI) derived from the Ozone Monitoring Instrument (OMI) were used for aerosol optical depth (AOD) and dust aerosol inversion, and were taken from the following online source: (http://disc.sci.gsfc.nasa.gov/giovanni/overview/index.html). The NCEP/NCAR reanalysis data were used for wind field analysis which was also taken from an online source (http://www.esrl.noaa.gov/psd/data/composites/day/). The fire sites distribution data were contributed by: https://firms.modaps.eosdis.nasa.gov/firemap/. It can be seen that the fire

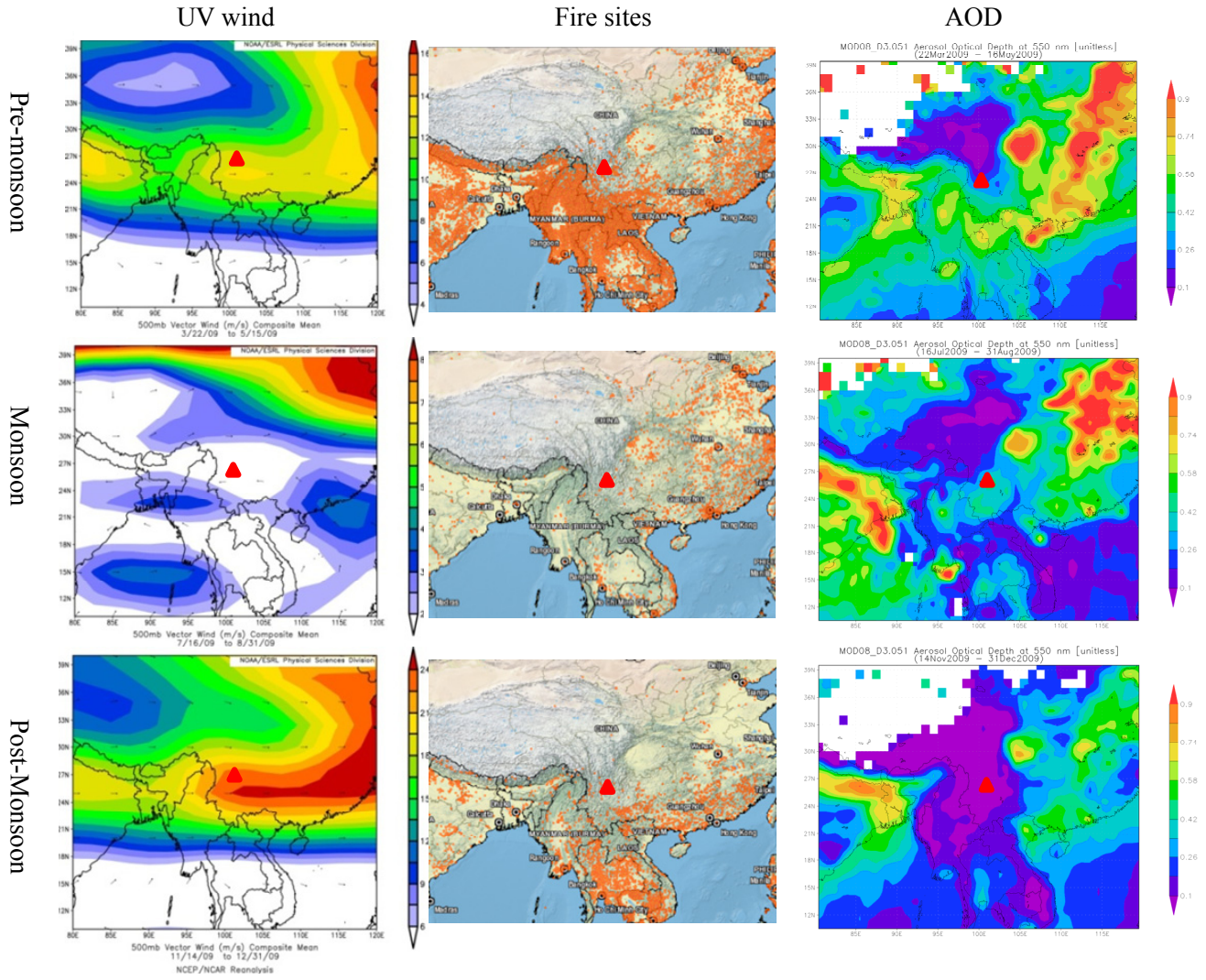

Fig. 1. UV wind at 500 hPa, fire sites and AOD distribution around Lijiang (red triangle).

Table 1. Meteorological status and tourist statistics during sampling period.

	Pre-monsoon	Monsoon	Post-monsoon
Temperature (°C)	16.4 ± 2.6	18.3 ± 1.5	8.2 ± 2.7
Total precipitation (mm)	0	345.4	0.2
Pressure (hPa)	762.0 ± 1.8	762.7 ± 2.4	763.4 ± 2.4
Relative humidity (%)	41.8 ± 9.0	80.0 ± 6.3	48.6 ± 12
monthly tourists (× 10^4)	62.3	78.2	59.7

sites were very concentrated around southeast TP during the pre-monsoon period, and the greatest AOD value was also observed during the monsoon period (Fig. 1).

The Hybrid Single-particle Lagrangian Integrated Trajectory (HYSPLIT) model is a powerful tool for modelling air mass trajectory and has been widely used in the field of atmospheric science. In this study, 6 days of backward trajectories were calculated using the internet-based HYSPLIT tool (http://ready.arl.noaa.gov/HYSPLIT.php). The ending location was at the sampling site (26.90°N, 100.21°E) with the altitude of 1000 m above ground level (AGL), and the ending time at 12:00 Beijing time (04:00 UTC).

Sources Apportionment

Positive matrix factorization (PMF) was used to identify possible sources for the main components. PMF is a multivariate factor analysis tool that utilizes error estimates and produces non-negative results (Paatero and Tapper, 1994). The tool also provides factor contributions and factor profiles of the sample data. Here, the PMF 3.0 model of the US EPA (Norris *et al.*, 2008) was used to apportion the source contributions. Species with more than 95% of the concentration below the method detection limit were excluded from PMF analysis. Missing data were replaced by median value of the respective species, and the equation-based

uncertainty was calculated. At the same time, the species with a signal-to-noise ratio smaller than 2 were also excluded.

RESULTS AND DISCUSSION

The temporal variation of the main detected species is presented in Fig. 2 and Table 2. It is clear that the concentration of species related to crustal materials, such as Al, Si, Ti and Fe, shown the greatest value during the pre-monsoon period (Fig. 2, episode I) than in the monsoon and post-monsoon periods, with the exception of Ca^{2+}, which has the largest concentration during the post-monsoon period. In addition, the greatest concentration of some components contributed by anthropogenic sources, such as SO_4^{2-}, NH_4^+, S, Zn, As, Pb and Cu, appeared during monsoon period (Fig. 2, episode II). This indicates that aerosol chemistry during different periods is influenced by various sources at Lijiang.

Elemental Composition

Elemental concentration and comparison between various sampling periods are shown in Table 2 and Fig. 2. It is clear that Al, Si, Ca, Fe and S are the dominant elements at Lijiang during the pre-monsoon, monsoon and post-monsoon

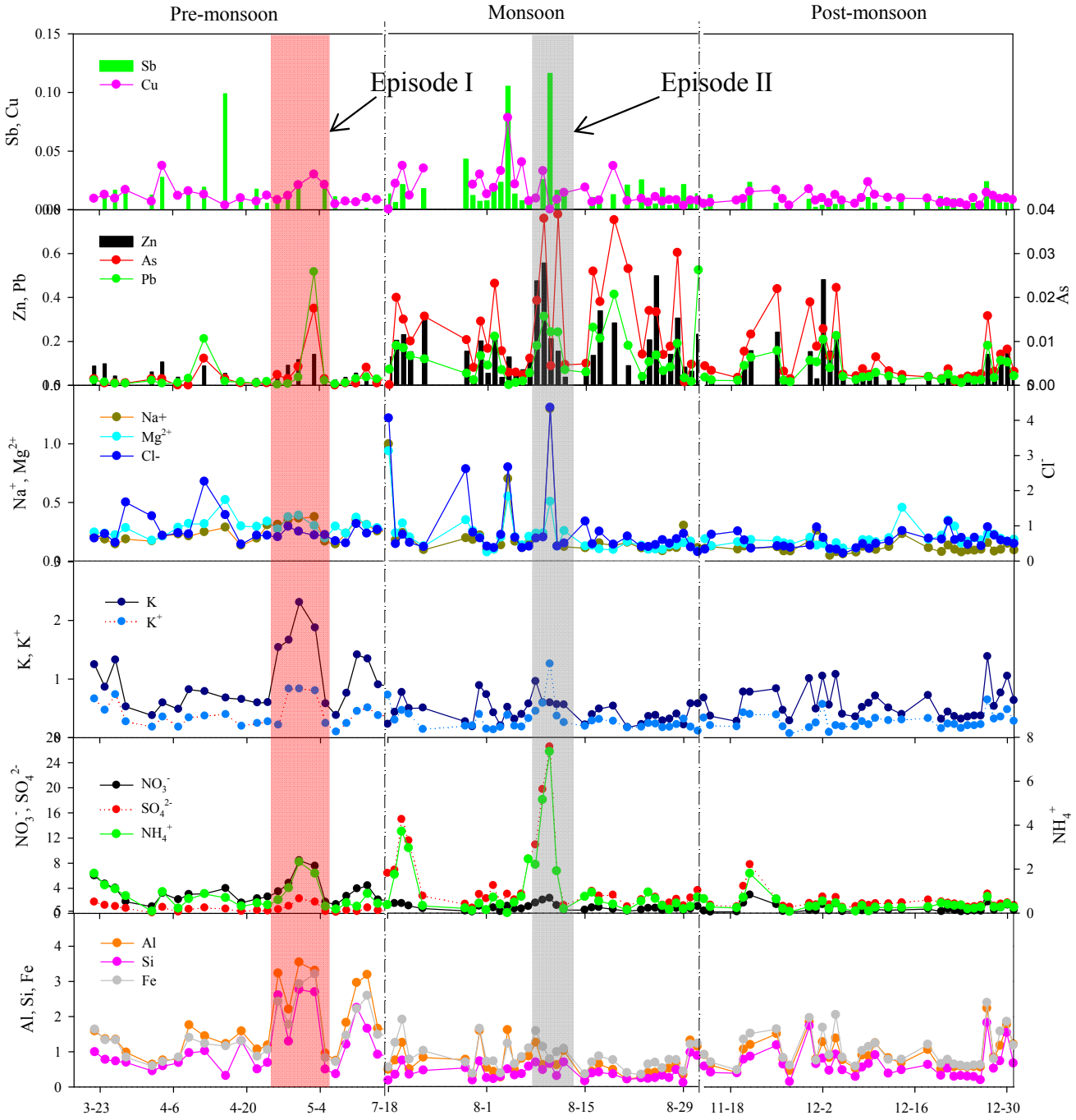

Fig. 2. Concentration of elements and ions during pre-monsoon, monsoon and post-monsoon period at Lijiang.

Table 2. Statistical summary of elemental and ionic concentration (μg m^{-3}) at Lijiang during sampling periods.

	Pre-monsoon		Monsoon		Post-monsoon		Annual	
	average	SD	average	SD	average	SD	average	SD
Al	**1719.63**	897.83	780.12	739.88	944.27	434.25	1070.94	788.40
Si	**1130.46**	742.24	483.22	530.74	648.52	409.81	703.36	609.99
S	1184.73	671.29	**1483.48**	1688.56	679.94	480.84	1122.27	1189.67
K	**970.95**	508.96	479.76	278.21	597.96	276.82	644.63	400.53
Ca	5210.25	1632.74	4296.75	1711.07	**6294.01**	2007.65	5228.14	1989.03
Ti	**165.43**	78.88	118.08	92.05	123.52	61.24	131.64	80.71
Mn	39.57	25.54	**68.25**	57.12	38.87	41.59	50.17	47.10
Fe	**1483.99**	721.88	977.43	790.73	1067.48	505.87	1133.58	708.34
Ni	**2.79**	1.78	1.96	3.36	1.36	0.81	1.93	2.39
Zn	59.74	32.25	**181.98**	132.66	82.49	95.31	115.85	114.64
As	2.01	3.61	**18.99**	22.55	6.05	5.98	10.01	16.17
Br	6.10	4.91	-	-	4.02	2.68	-	-
Sb	14.34	19.95	**19.92**	24.87	7.94	5.48	14.29	19.52
Pb	46.27	108.06	**152.24**	122.32	65.62	58.12	94.30	109.56
Cu	10.11	6.38	**17.87**	21.28	7.50	3.59	12.18	14.40
Na^+	**240.66**	75.23	222.21	252.76	112.00	45.14	187.29	172.96
NH_4^+	789.02	580.27	**1106.69**	1546.58	364.97	306.66	761.22	1067.14
K^+	**402.84**	224.98	299.46	211.02	275.42	125.80	316.68	193.75
Mg^{2+}	**292.48**	75.13	211.05	164.45	183.99	68.82	221.70	123.53
Ca^{2+}	4297.91	1292.09	3643.91	1484.98	**4602.19**	1238.31	4151.14	1404.27
Cl^-	884.66	409.27	**887.40**	965.72	562.49	212.48	770.17	662.92
NO_3^-	**1014.74**	602.35	879.02	504.43	652.33	605.10	835.61	578.26
SO_4^{2-}	3490.80	1870.69	**4913.82**	5557.42	1797.01	1276.81	3440.08	3890.63

Bold means the highest value of species for the three sampling periods; "-" means no data.

periods. The total concentration of crustal elements (Al, Si, Ti, Ca and Fe) is 8.27 μg m^{-3}, which is more than five times greater than the concentration detected at Lijiang (1.52 μg m^{-3}) in 2003 (Zhang *et al.*, 2007). However, the total concentration of heavy metals (Ni, As and Cu) related to anthropogenic sources is 24.21 μg m^{-3}, substantially higher than the 2003 value (14.2 μg m^{-3}) (Zhang *et al.*, 2007). This suggests that the growth magnitude of crustal elemental concentration is larger than that of heavy metal elements, which further implies that the emissions of anthropogenic pollutants are lower than those of crustal sources. Moreover, except for Ca, the crustal elements showed the greatest concentrations during the pre-monsoon period than during the monsoon and post-monsoon periods. The lowest concentrations were observed during the monsoon period. In contrast, the largest concentrations of most anthropogenic elements were found during the monsoon period. These comparisons show that the detected elements were contributed by different sources.

The EF is often used to identify the influences from crustal material and anthropogenic sources, and it is generally defined as follows:

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

where X represents the element of interest, C_X is the concentration of X, and C_R is the concentration of a reference element. The aerosol and crust subscripts refer to particles in the aerosol and crustal material, respectively. The reference elements commonly used are Al, Si, Ti, and Fe. In this study, Al was selected as the reference material, and the average upper continental crust composition of Wedepohl (1995) was used for calculation.

Commonly, elements whose EF value approach unity, suggesting the dominant source for those elements originate from the crust. If the EF value is greater than 10, it indicates that the concentration of the elements is influenced by anthropogenic sources. The EFs of elements in our samples are shown in Fig. 3. It can be seen that EF values of S, Cl, Zn, As, Br, Sb, Pb and Cu are greater than 10 during the three periods, which indicates that the concentration of those elements were mainly contributed by human activities. Especially, the EF value of Sb is greater than 1000, even up to 8700 during the monsoon period.

The highest EF values of most elements were observed during monsoon period, rather than during the pre-monsoon and post-monsoon periods. This was especially the case for those elements with an EF value greater than 10. For instance, the EF value of As during the monsoon period was 5 and 26 times greater than the post-monsoon and pre-monsoon periods, respectively. This indicates that more pollutants related to human activities were emitted or transported to Lijiang during the monsoon period. Southwest China is influenced by various circulations, and it was mainly controlled by the southwest monsoon during the monsoon period. Cong *et al.* (2007) showed that elements related to anthropogenic emissions from south Asia can be transported to the south TP or even central TP during monsoon period. In our study, it has a strongest wet scavenging effect for particles since it is the rain season of monsoon period (Table 1).

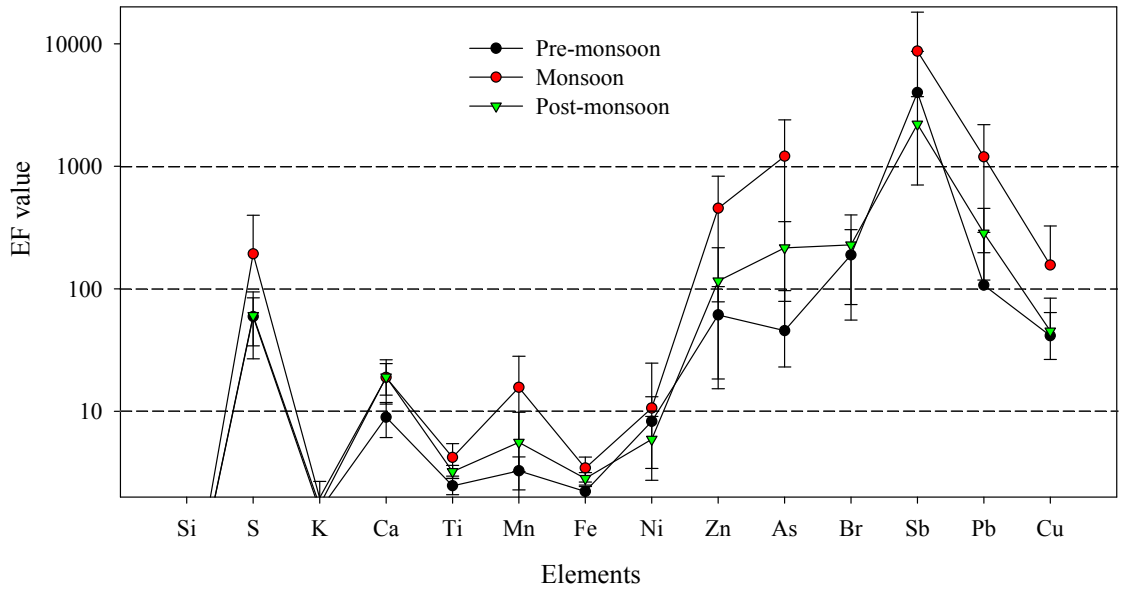

Fig. 3. EF value for selected elements.

However, the greatest concentrations and EF values of some pollution elements occurred during the monsoon period. These elements may have been transported by the southwest monsoon from south Asia.

Ionic Composition

According to decreasing concentrations, ionic sequence is: $Ca^{2+} > SO_4^{2-} > NO_3^- > Cl^- > NH_4^+ > K^+ > Mg^{2+} > Na^+$ (Table 2). Ca^{2+} and SO_4^{2-} were the dominant cation and anion during the whole sampling period, respectively. The correlation coefficients between cation and anion were 0.87 ($P < 0.001$), 0.94 ($P < 0.001$) and 0.85 ($P < 0.001$) during the pre-monsoon, monsoon and post-monsoon periods, respectively. This indicates that the main ions present in the air samples were detected in our filters. Because the local rock is mainly composed of limestone, Ca^{2+} was the main cation during our sampling period, especially during the post-monsoon period, which caused an ion imbalance in the aerosol samples (Fig. 4). This supports the findings of other studies in Lijiang and the south TP (Zhang *et al.*, 2011, Zhao *et al.*, 2013).

The ions SO_4^{2-}, NO_3^- and NH_4^+ (SNA) have a significant correlation with each other (R: 0.73–0.98, P < 0.001) during the whole sampling period, which suggests that they were all influenced by the same source or transportation process. However, the greatest concentration of NO_3^- was observed during the pre-monsoon period and that of SO_4^{2-} and NH_4^+ during the monsoon period, which indicates that there are additional sources of NO_3^- during the pre-monsoon period and SO_4^{2-} and NH_4^+ during the monsoon period, respectively. Previous studies have shown that the largest total ionic concentration occurred during the pre-monsoon period at other sites along the south edge of the TP (Shrestha *et al.*, 2000; Desesari *et al.*, 2010; Zhao *et al.*, 2013). However, our data shows that the largest total detected ionic concentration was observed during monsoon period, which was caused by the large concentrations of SO_4^{2-} and NH_4^+ during this period (Table 2).

For K^+, it is treated as tracer for biomass burning (Andreae, 1983). In our study, K^+ shows an obvious greatest value during pre-monsoon period (Table 2, Fig. 2). This is also a frequent frost fires period in south Asia and north Burma, which is located in the upwind direction of Lijiang (Fig. 1). Thus, the pollutants from biomass burning can be easily transported to Lijiang. Furthermore, the concentrations of Cl^- and Br were also higher during the pre-monsoon period, which may also have originated from biomass burning emissions (Yamasoe *et al.*, 2000).

Source Apportionment

The detected ions and elements (except Br) were analyzed in the PMF model. Accordingly, five factors were selected, and the profile is shown in Fig. 5.

Factor 1 is dominated by Al, Si, Ca, Ti, Fe and part of K^+ and NO_3^-. It can be interpreted as crustal source. It is notable that the largest concentrations of Al, Si, K, Ti, Fe and K^+ occurred during the pre-monsoon period (Table 2), and episode I with high value of crustal elements also occurred during the pre-monsoon period (Fig. 2). This indicates that the high contribution from crustal source mainly occurred during the pre-monsoon period, and may be associated with dust events from India and northwest China.

Zn, As and Pb were highly loaded in Factor 2, the greatest concentrations of Zn, As and Pb in our study were observed during the monsoon period (Table 2, Fig. 2). The correlation coefficients of Zn and As, Zn and Pb, As and Pb were 0.65 ($P < 0.001$, $N = 91$), 0.64 ($P < 0.001$, $N = 91$) and 0.82 ($P < 0.001$, $N = 91$), respectively. Generally, As and Pb are regarded as trace elements for coal burning (Xu *et al.*, 2012). However, Lijiang's main industry is tourism and is mainly powered by hydropower thus coal combustion in the area is limited. The study region is located downwind of north India and near to east China, which are both characterized by heavy air pollution. Cong *et al.* (2007)

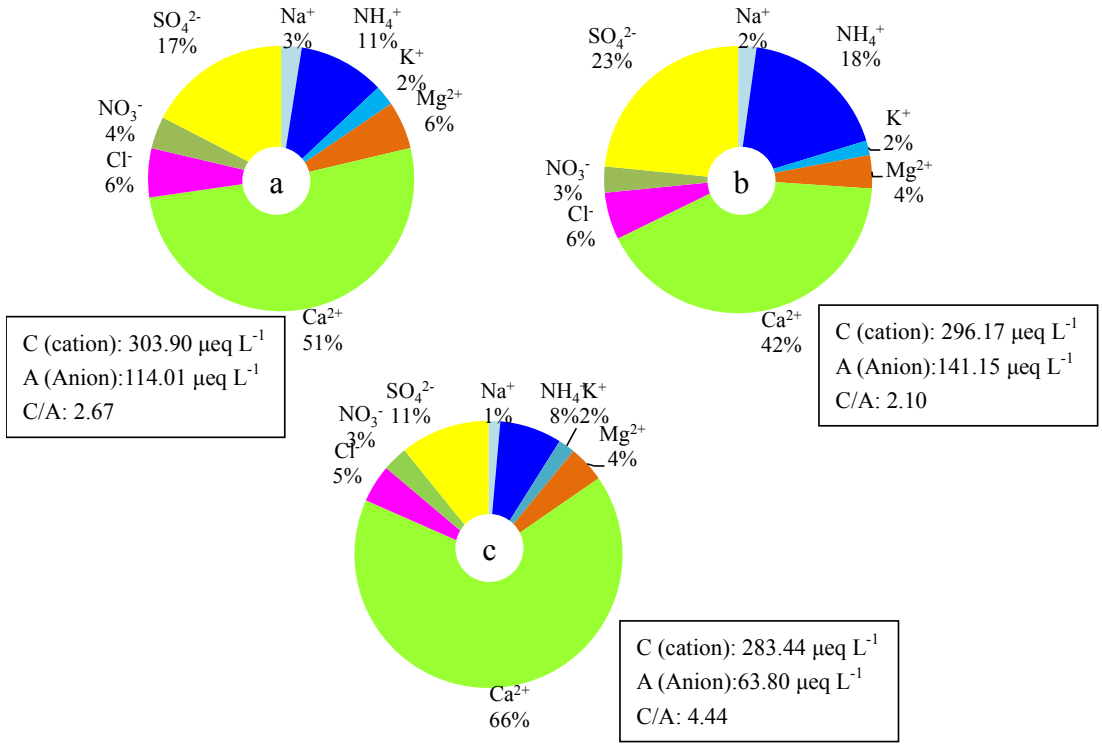

Fig. 4. Percent of ions in pre-monsoon (a), monsoon (b) and post-monsoon (c).

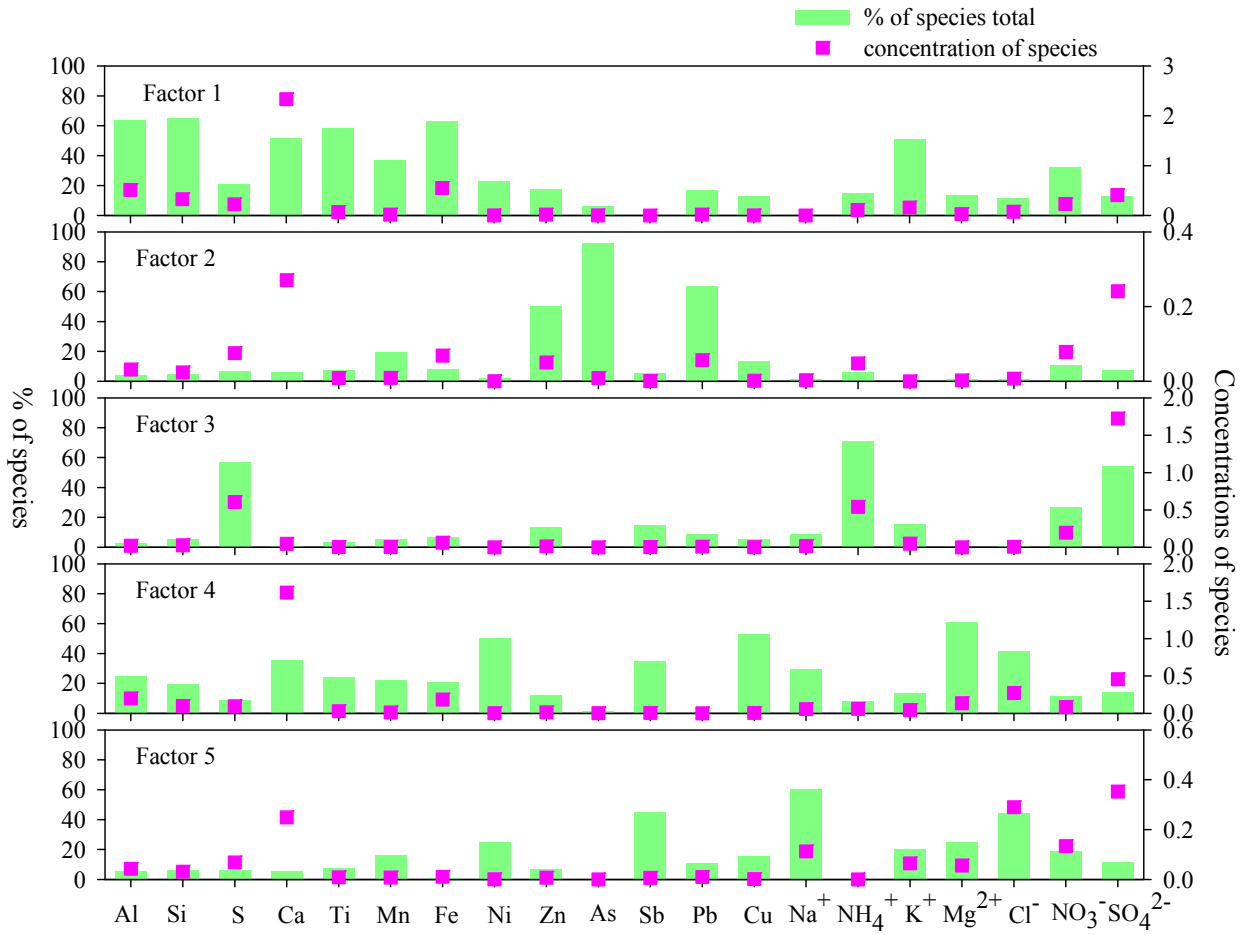

Fig. 5. Source profiles of species calculated by PMF.

have pointed out that anthropogenic pollutants (such as Zn, As, etc.) can be transported to central TP from south Asia. Thus, Factor 2 can be considered as long range mainly transport from south Asia.

Concentrations of S, SO_4^{2-} and NH_4^+ were very high in Factor 3, and their greatest values were also found during the monsoon period as the species in Factor 2. However, they did not belong to the same factor, which suggests that they were contributed by different sources. Qu *et al.* (2009) studied the aerosol at Zhuzhuang, a remote site at southeast TP, and found that pollutants with high concentration of SO_4^{2-} can be transported to southeast TP from eastern China during the monsoon period. These results indicate that Factor 3 can be attributed to pollutants from eastern China.

Factor 4 is enriched in Cu, Ni, Sb, Mg^{2+} and Ca. This is likely to represent the local vehicle emissions. It has been shown that Cu, Ni and Sb can be produced in the vehicle brake wear process (Weckwerth, 2001), and Ca can be produced by the re-suspension process.

The Cl^- and Na^+ concentrations were highly loaded in Factor 5. These ions are commonly regarded as sea salt species. The higher concentrations of Cl^- occurred during the monsoon period, and the concentrations of Na^+ during the monsoon period were similar to the largest values during the pre-monsoon period. The correlation coefficient of Cl^- and Na^+ was 0.92 ($P < 0.001$). This high correlation suggests that Factor 5 can be regarded as the sea salt source.

Seasonal Pollution Episodes and Its Implication

Mixture Pollution during Pre-monsoon

During the pre-monsoon period, the frequency of desert dusts originating from western India increased during the later stages, until the onset of the monsoon (Srivastava *et al.*, 2011). During transportation, dust mixes with various anthropogenic aerosols emitted from the Indo-Gangetic Basin region. Those mixture of pollutants labeled by high concentration of nitrate and crustal elements (Rastogi and Sarin, 2005) can be transported to India Ocean, eastern India and the southern Himalayas (Savoie *et al.*, 1987; Lelieveld *et al.*, 2001; Zhao *et al.*, 2013). As Fig. 2 shown, episode I is characterized by a clear peak of crustal elements (Al, Si and Fe) with NO_3^- also present on 26 April, 28 April, 30 April and 3 May. The average concentration of Al, Si, Fe and NO_3^- was 3.07 μg m^{-3}, 2.34 μg m^{-3}, 2.58 μg m^{-3} and 1.89 μg m^{-3}during 26 April to 3 May, which was 2.87, 3.34, 2.28 and 2.25 times higher than the annual average concentration, respectively. Based on the satellite data (Fig. 6), it was found that the largest AOD values occurred at south edge of TP and eastern China, and the largest value of UVAI appeared at south edge of TP and western China during episode I. This suggests that the pollutants present in the south edge of the TP during episode I not only originated from human activities, but also from natural dust. All of the backward trajectories during episode I originated from the south edge of the TP, and the wind direction at 500 hPa also flowed to southwest China from north India. This implies that the pollutants, including anthropogenic emissions and dust, can be transported to southwest China. Qu *et al.* (2008) also found that higher concentrations of NO_3^-, K and Fe were present when the transportation was from the west of the Southwest China region.

The relationships between NO_3^- and Al, Si and Fe during the three periods were shown in Table 3. It is clear that NO_3^- has a highly significant relationship with Al, Si, and Fe during the pre-monsoon period, which is more significant than their relationship during the monsoon and post-monsoon period. This corresponds with the finding that the greatest concentrations of NO_3^-, Al, Si, Ti and Fe concentration occurred during pre-monsoon period (Table 2). In Section Source Apportionment, these species were represented in Factor 1, which indicates that these species were mixed together during transportation processes.

Multiple Pollutants during the Monsoon Period

During the monsoon period, the most impressive phenomenon is the high concentrations of some heavy metal elements, such as Zn, As, Sb, Pb and Cu. Although various sources contributed to these high concentrations (Fig. 5), all heavy metal elements had high concentrations during monsoon period specifically (Table 2). Heavy metals,

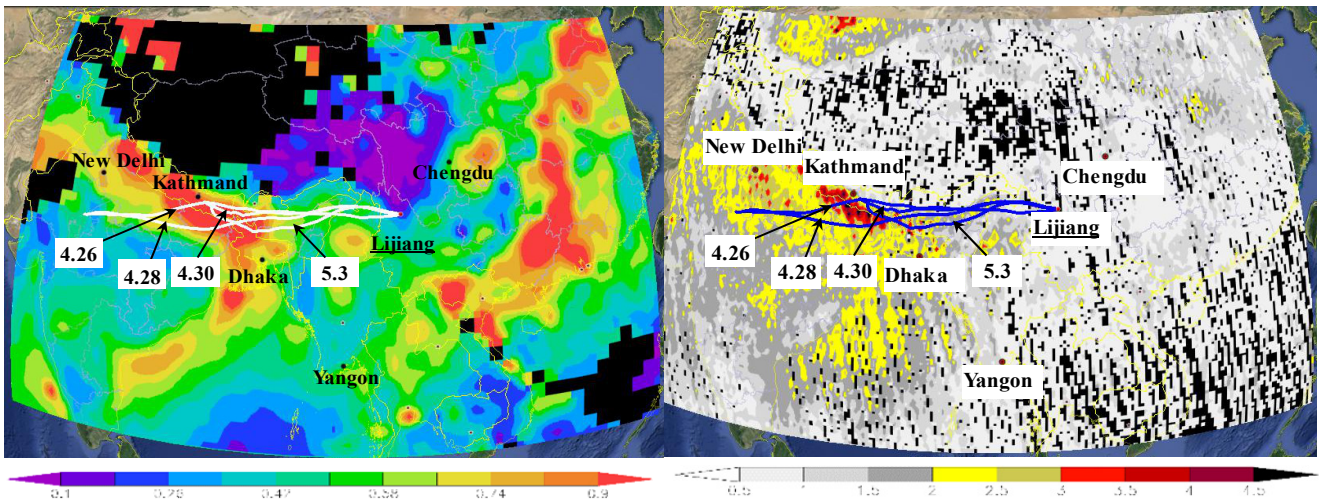

Fig. 6. Distribution of AOD, UVAI, backward trajectories and wind field (500 hp) during 26 April to 3 May.

Table 3. Correlation coefficients between NO_3^-, SO_4^{2-} and crustal elements.

	Pre-monsoon		Monsoon		Post-monsoon	
	NO_3^-	SO_4^{2-}	NO_3^-	SO_4^{2-}	NO_3^-	SO_4^{2-}
Al	**0.88**	**0.71**	0.38	0.08	**0.63**	0.37
Si	**0.86**	**0.67**	0.37	0.13	**0.57**	0.34
Fe	**0.93**	**0.80**	**0.47**	0.17	**0.72**	**0.46**

Bold means P < 0.001.

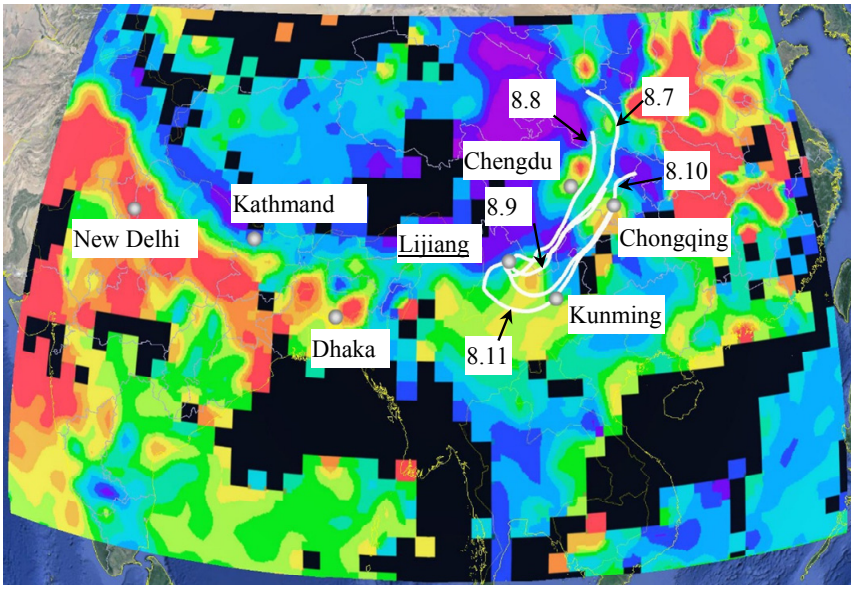

Fig. 7. Distribution of AOD and backward trajectories during 7 to 11 August.

including individual metal elements and metal compounds, can impact human health. In the new Chinese ambient air quality standards (GB 3095-2012), the annual concentration limit for Pb and the reference concentration of As are 500 μg m^{-3} and 6 μg m^{-3} respectively. Obviously, the annual concentration of As exceed the value. In particular, the concentration of As was 18.99 μg m^{-3} during the monsoon period, which is three times higher than the reference value. In addition, Lijiang, as a tourist city, experiences its peak tourist quantity during the monsoon period (Cao *et al.*, 2012). However, the best season for tourists to visit Lijiang should be the post-monsoon period with respect to the concentration of heavy metal elements.

Episode II (7 to 10 August) was also identified by the high concentrations of SO_4^{2-}, NH_4^+, As, Zn and Pb (Fig. 2), which were on average 4.77, 5.62, 2.12, 2.94 and 2.17 times higher than their annual average value, respectively. Fig. 7 shows that the calculated backward trajectories all passed through Sichuan Basin and the eastern Yunnan Province, both of which also had a high AOD value. Sichuan Basin has been considered as the most pollution area in China because of its dense population and unique topography. Using the Community Multiscale Air Quality (CMAQ) model, Zhang *et al.* (2012) showed that Sichuan Basin was one of the maximum sulfate concentration centers in August 2009, because of the increased secondary formation in summer. Eastern Yunnan Province has also been shown to be a regional pollutant source in summer (Qu *et al.*, 2008). Moreover, the high concentrations of As, Zn and Pb occurred along with SO_4^{2-} and NH_4^+, suggesting that As, Zn and Pb can be also transported from east China during some pollution events.

The source apportionment results shows that Factor 2 (mainly As, Zn and Pb) and Factor 3 (mainly SO_4^{2-} and NH_4^+) were from different sources. Actually, the correlation between those species were not significant; for example, the correlation coefficient between SO_4^{2-} and As, Zn, and Pb were 0.02 (P > 0.05), 0.42 (P > 0.01) and 0.27 (*P* > 0.05) during the monsoon period, respectively. This suggests that the pollutants did not originate from the same source and processes, and thus represented multiple pollutant sources during the monsoon period.

Implications

Studies have shown that the TP has been warming over recent years (Jones and Briffa, 1992; Liu *et al.*, 2000; Guo and Wang, 2011). However, the seasonal and spatial warming trends are different. In particular, the northern part of the TP shows the most significant warming, especially during summer time (Guo and Wang, 2011). In contrast, the temperature increase rate in the southeastern TP and southwest China was lower than that of the whole of China during 1961–2010, and the most distinguished increasing trend occurred in winter, followed by autumn; however, spring and summer temperatures did not show any significant increasing trend (Ma *et al.*, 2013).

South Asia is a major source of black carbon, especially during the pre-monsoon period, which has a significant warming effect on climate, and has been shown to cause snow/ice melt over the TP (Xu *et al.*, 2009; Lau *et al.*, 2010; Yasunari *et al.*, 2010). Lau *et al.* (2010) also found that the aerosol mixed with dust and black carbon can heat the mid-troposphere and accelerate the melting of snow on the western TP. In our study, it is also found that dust aerosol mixed with anthropogenic aerosol and biomass burning aerosol can be transported to the southeast TP. Thus, the question is why the greatest warming season in the southeast TP did not appear during pre-monsoon (spring) and why the warming trend in the southeast TP is the lowest of the TP overall. This is may be related to the presence and the mixture status of dust aerosols. Dust can mix with various anthropogenic aerosols during transport and change the aerosol optical properties, which causes radiative impacts and enhances variability (Dey *et al.*, 2008; Mishra *et al.*, 2008).

The implication for regional climate change is also found during monsoon periods (summer). Studies have shown that sulfate has cooling effects on climate change (Lelieveld and Heintzenberg, 1992), and also that anthropogenic aerosols (such as sulfate) have cooling climatic effects in the Sichuan Basin in southwest China (Qian and Giorgi, 2000). Thus, the high concentration of sulfate transported from the Sichuan Basin may slow the warming trend of the southeast TP. In fact, Ma *et al.* (2013) showed that there is no significant warming trend during summer. Thus, the various transport pathways may have important implications for regional climate change by influencing the aerosol chemistry.

CONCLUSION

In this study, seasonal aerosol samples were collected in 2009 at Lijiang on the southeast TP, southwest China. Fifteen elements and eight ions were detected to study the chemical composition, seasonal variation, sources, and transport of atmospheric aerosols. The major results are as follows:

1. Al, Si, Ca, S and Fe were the main detected elements, and $Ca^{2+}$and SO_4^{2-} were the dominant cation and anion, respectively. The EF values of Si, K, Ca, Ti, Fe were lower than 10, indicating that they were mainly from crustal material. The EF values of S, Cl, Zn, As, Br, Sb, Pb and Cu were greater than 10, indicating that they were contributed by human activities. The greatest concentrations of crustal elements appeared during the pre-monsoon period, and the total ions and most of the elements influenced by anthropogenic sources showed the greatest concentrations during the monsoon period.

2. Five factors were calculated by the PMF model: Al, Si, Ca, Ti, Fe and part of K^+ and NO_3^- can be interpreted as crustal source; Zn, As and Pb can be considered as long range transport from south Asia; S, SO_4^{2-} and NH_4^+ can be attributed to pollutants from East China; Cu, Ni, Sb, Mg^{2+} and Ca represent the local vehicle emissions; and Cl^- and Na^+ were regarded as sea salt sources.

3. Based on the combination of MODIS, NCEP/NCAR reanalysis data and back trajectories, it is found that aerosols in this region are polluted by a mixture of dust, anthropogenic pollutants and biomass burning pollutants during the pre-monsoon period, and those pollutants were transported from south and southeast Asia. During the monsoon period, pollutants from Sichuan Basin and eastern Yunnan Province can be transported to Lijiang with high loading of SO_4^{2-}, NH_4^+ and some heavy metal elements.

According to the current results, further studies should be performed to improve understanding of the health effects on tourists to the region by the high concentration of heavy metal elements observed during monsoon period. In addition, research is required to detail the influences of transported chemical components on regional climate change.

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