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Elemental compositions of $PM_{2.5}$ and TSP in Lijiang, southeastern edge of Tibetan Plateau during pre-monsoon period

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ABSTRACT

$PM_{2.5}$ and total suspended particulate (TSP) samples were collected at Lijiang, southeastern Tibetan Plateau, China. Sixteen elements (Al, Si, S, K, Ca, Cr, Mn, Ti, Fe, Ni, Zn, As, Br, Sb, Pb and Cu) were analyzed to investigate their elemental compositions during the pre-monsoon period. The results showed that Ca was the most abundant element in both $PM_{2.5}$ and TSP samples. The enrichment factors (EFs) of Si, Ti, Ca, Fe, K and Mn were all below 10 for both $PM_{2.5}$ and TSP, and these elements also had lower $PM_{2.5}$/TSP ratios (0.32–0.34), suggesting that they were mainly derived from crustal sources. Elements Cu, Zn, S, Br and Sb showed strong enrichment in $PM_{2.5}$ and TSP samples, with their $PM_{2.5}$/TSP ratios ranging from 0.66 to 0.97, indicating that they were enriched in the fine fractions and influenced by anthropogenic sources. Analysis of the wind field at 500 hPa and calculations of back trajectories indicated that Al, Si, Ca, Ti, Cr, Mn and Fe can be influenced by transport from northwestern China during the dust-storm season, and that S, K, Ni, Br and Pb reached high concentrations during westerly transport from south Asia. Combined with the principle component analysis and correlation analysis, elements of $PM_{2.5}$ samples were mainly from crustal sources, biomass burning emissions and regional traffic-related sources.

1. Introduction

The Tibetan Plateau is the Earth's highest plateau, and exerts profound thermal and dynamic influences on local and global climate as well as on atmospheric circulation of the Asian Monsoon System (Chan, Wong, Li, Chan, & Zheng, 2006). Because it is located far from any industrialized area and is lightly populated, the plateau has been chosen as an ideal location to observe the atmospheric environment and to evaluate various impacts of human activities (Cong, Kang, Liu, & Wang, 2007; Kang et al., 2002). To date, several aerosol particle studies have been conducted over the Tibetan Plateau, for example at Wudaoliang and Waliguan located northeast of the plateau (Liu, Zhang, & Shen, 1997; Wen, Xu, Tang, Zhang, & Zhao, 2001; Zhang, Arimoto, Cao, An, & Wang, 2001); on Muztagh Ata Mountain located in the northwest of the plateau (Cao et al., 2009; Wu, Xu, Zhang, Gao, & Yao, 2009); in the Himalaya located in the south of the plateau (Cong, Kang, Dong, Liu, & Qin, 2010; Li, Kang, & Cong, 2007; Shrestha, Wake, & Dibb, 1997); on Mt. Gongga and Mt. Yulong located in the southeast of the plateau (Yang et al., 2009; Zhang, He, Cao, Ho, & Shen, 2012; Zhang, He, Theakstone, & Pang, 2010); and at Nam Co located in the center of the Plateau (Cong et al., 2007). These studies have mainly focused on aerosol chemistry at remote sites over the plateau, and have given less attention to the transitional region at the plateau margin, in particular for the suburban environment near the city.

Lijiang city is located on the southeastern edge of the Tibetan Plateau, southwestern China, and is adjacent to south Asia and southeast Asia. Previous studies have found that pollutants related to human activities and biomass burning emissions from south Asia and southeast Asia not only influence the local atmospheric environment, but can also influence southwestern China by long-range transport (Chan et al., 2006; Engling et al., 2011; Zheng et al., 2007). Lijiang is a famous city for tourism. Increasing numbers of tourists are visiting the city, consequently influencing the environment, e.g., its water resources (Ning & He, 2007) and precipitation chemistry (Zhang et al., 2012). However, there have been no studies on the elemental composition of aerosol in the rural regions of Lijiang, particularly in terms of $PM_{2.5}$. The purpose of the present study is: (1) to investigate the elemental composition of

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TSP and $PM_{2.5}$ samples and (2) to determine the possible sources of elements in the atmosphere during the pre-monsoon period.

2. Methods

2.1. Site description

Lijiang city is located on the southeastern Tibetan Plateau, northwestern Yunnan Province, China (Fig. 1). Because of the attraction of the Old Town, Lijiang was listed as a World Heritage site in 1997. It is also a historic and cultural city of great value and importance (UNESCO World Heritage Centre 1997), and has gradually developed as a tourist center. In 2008, there were approximately 150,000 permanent residents in Lijiang city and over 6 million visiting tourists. In order to encourage tourism, heavy industry development has been limited in Lijiang district; however, the number of vehicles has grown rapidly in association with tourism development. Statistics indicate that the total number of vehicles increased from 9000 in 1995 to 87,000 in 2008 (Lijiang Statistics Yearbook), and vehicle exhausts have been regarded as the major local pollution source.

2.2. Meteorology

The climate of the Lijiang region is controlled by the Asian southwestern monsoonal circulation from May to October, and by the southern branch of the westerly circulation from November to April of the following year. During our sampling period, the prevailing circulation in the study area was dominated by the southern branch of westerlies and the plateau winter monsoon (Fig. 1).

Meteorological parameters including air temperature, air pressure, relative humidity, wind speed and wind direction were measured simultaneously during the sampling period (Table 1). There was very little precipitation during our sampling period (see Fig. 2), which could decrease the elemental mass concentration by wet scavenging effects, especially in TSP samples. Using the U, V-wind data downloaded from http://www.esrl.noaa.gov/psd/data/, the wind field at 500 hPa was plotted by Grads (Version 1.8) in Fig. 1.

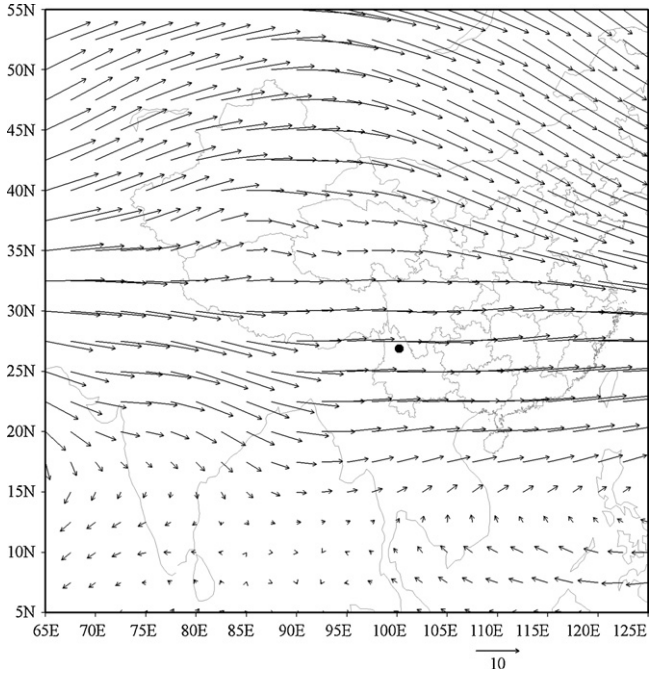

Fig. 1. Location of Lijiang city (black dot) and average wind field of 500 hPa during sampling period.

Table 1
Statistic meteorological status during sampling period.

Parameters	Mean	Max.	Min.
Temperature (°C)	15.7	20.7	8.7
Pressure (hPa)	762.2	767.3	757.9
Humidity (%)	44.7	81	29
Wind speed (m/s)	3.9	7.6	1.6

2.3. Sampling and analysis

The sampling site was set about 5 m above the ground and 1.5 m above the roof of the Mt. Yulong Glacier and Environmental Monitoring Station building. There are some residential but no industrial emissions around the building, represented as a rural environment. $PM_{2.5}$ and TSP sampling was carried out during the pre-monsoon period (21 March to 16 May, 2009), once every two days, respectively. The collector (Zambelli Easy Plus 1, Zambelli srl, Milan, Italy) was driven by 220 V alternating current. Teflon® Zefluor™ filters (47 mm in diameter) with 2 μm pore size (Pall Corporation, Port Washington, NY) were used for sampling. The volume of air sampled was measured by an in-line flow meter with a mean flow rate of 16.7 L/min. Meanwhile, ambient temperature and pressure were converted to the standard (101,325 Pa, 273 K) cubic meters. Totally, 25 $PM_{2.5}$ and 24 TSP samples were collected for analysis. The filter cartridges were packed in clean plastic bags and transported in air-tight containers. After sampling, the filters were removed from the cartridges and placed in pre-cleaned air-tight methacrylate bottles. Samples and seven blank filters were taken with care to minimize contamination both in the field and in the laboratory before analysis in the Institute of Earth Environment, Chinese Academy of Sciences (Xi'an).

The concentrations of elements in $PM_{2.5}$ and TSP samples were determined by energy dispersive X-Ray fluorescence (ED-XRF) spectrometry using the PANalytical Epsilon 5 XRF analyzer (PANalytical B.V., Almelo, The Netherlands), which uses three-dimensional polarizing geometry with 11 secondary targets (CeO_2, CsI, Ag, Mo, Zr, KBr, Ge, Zn, Fe, Ti, and Al) and one Barkla target (Al_2O_3) that supplies a good signal-to-background ratio, permitting the low detection limits. The X-ray source is a side window X-ray tube with a gadolinium (Gd) anode, which is operated at an accelerating voltage of 25–100 kV and a current of 0.5–24 mA (maximum power: 600 W). Characteristic X-radiation is detected by a germanium (Ge) detector (PAN 32). Each sample was irradiated for half an hour and a laboratory blank Teflon filter sample was also analyzed to evaluate analytical bias. The elements that were determined by the ED-XRF method include Al, Si, S, K, Ca, Cr, Mn, Ti, Fe, Ni, Zn, As, Br, Sb, Pb, and Cu with detection limits ($\mu g/cm^2$) of 0.115, 0.093, 0.032, 0.007, 0.007, 0.003, 0.014, 0.005, 0.011, 0.003, 0.008, 0.000, 0.006, 0.033, 0.015 and 0.010, respectively. Quality assurance/quality control (QA/QC) procedures were described in Xu et al. (2012).

3. Results and discussion

3.1. Elemental composition

Fig. 2 shows the temporal variations of all detected elemental mass concentrations in $PM_{2.5}$ and TSP, while statistical results for the element concentrations of $PM_{2.5}$ and TSP during the pre-monsoon period in 2009 are shown in Table 2. The $PM_{2.5}$/TSP ratio