



## Chemical composition and sources of PM<sub>2.5</sub> and TSP collected at Qinghai Lake during summertime



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### ABSTRACT

PM<sub>2.5</sub> and total suspended particulate (TSP) samples were collected from June to September 2010 at Qinghai Lake, northeastern Tibetan Plateau. The concentrations of major water-soluble ions, 10 elements, elemental carbons (ECs) and organic carbons (OCs) were quantified. Mass concentrations of PM<sub>2.5</sub> and TSP were  $21.27 \pm 10.70 \mu\text{g m}^{-3}$  and  $41.47 \pm 20.25 \mu\text{g m}^{-3}$ , respectively, and the mean ratio of PM<sub>2.5</sub>/TSP was 0.51. The greatest anion and cation in both PM<sub>2.5</sub> and TSP samples were  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$ , respectively. Crustal elements, such as Ca, Fe and K, were the main elements in our aerosol samples, and their enrichment factors (EFs) were lower than 10. EFs for Pb, As, and Zn were greater than 10, indicating that they were influenced by anthropogenic sources. EC concentrations were  $0.33 \pm 0.17 \mu\text{g m}^{-3}$  and  $0.47 \pm 0.28 \mu\text{g m}^{-3}$ , whereas OC concentrations were  $1.49 \pm 0.63 \mu\text{g m}^{-3}$  and  $2.30 \pm 0.95 \mu\text{g m}^{-3}$  in PM<sub>2.5</sub> and TSP, respectively. Based on the calculated ratios of EC/TC and  $\text{K}^+/\text{EC}$ , most of the ECs were found to be related to biomass burning emissions. Because of the pollution from local sources, the ratios of OC/EC were  $4.77 \pm 1.32$  and  $5.23 \pm 1.39$  in PM<sub>2.5</sub> and TSP, respectively, which were lower than those of other remote sites. Salt particles produced by the salty lake reacted with acid gases and caused most of the nitrates and a small fraction of sulfate to be distributed in coarse mode; Cl deficit was also observed in our aerosol samples. Results of backward trajectories and correlation analysis show that the concentrations of  $\text{SO}_4^{2-}$ , OCs, As, Pb, and Zn, were influenced by the long-distance transport from eastern China.

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### 1. Introduction

Atmospheric aerosol research is an important tool for atmospheric environment research. Aerosol particles can scatter and absorb incoming solar radiation and thermal radiations emitted from the earth surface, act as condensation nuclei for cloud formation, and participate in heterogeneous chemical reactions. However, uncertainties in the magnitudes of their effects on the earth's interconnected systems remain numerous because of limited knowledge on aerosol chemical composition, optical and physical properties and lack of information on temporal and geographical distributions, particularly for aerosol studies

related to rural and remote regions. Knowledge on aerosol properties at remote sites is important not only for assessing the transport of pollution caused by human activities, but also for investigating large-scale aerosol effects on biogeochemical cycles and climate changes (Qu et al., 2009).

As the highest plateau in the world, the Tibetan Plateau exerts profound thermal and dynamic influences on local and global climate and on the atmospheric circulation of the Asian Monsoon System (Chan et al., 2006). The Tibetan Plateau is far from any industrialized areas and is lightly populated; thus, it is typically chosen as an ideal location for monitoring the background atmospheric environment and for evaluating the various effects of human activities (Kang et al., 2002; Cong et al., 2007). Until now, numerous studies related to aerosol have been conducted on the

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plateau, with most of them concentrated at the southern Tibetan Plateau and the influences on the atmospheric environment of the Tibetan Plateau by South Asia, including Himalaya, which is located south of the plateau (Shrestha et al., 1997; Li et al., 2007; Ming et al., 2007; Cong et al., 2010) and at Mt. Gongga and Mt. Yulong located southeast of the plateau (Yang et al., 2009; Zhang et al., 2010, 2012). However, only a few studies have been conducted at the northern plateau, such as at Muztagh Ata Mountain located northwest of the plateau (Cao et al., 2009a; Wu et al., 2009) and at Wudaoliang and Waliguan located northeast of the plateau (Liu et al., 1997; Wen et al., 2001).

Qinghai Lake, which is the largest saline lake in China, is located at a transitional zone of the semiarid climate. Qinghai Lake has elicited attention worldwide because of a series of ecological and environmental deteriorations, including decrease in lake water level and increase in salinity and desertification (Xu et al., 2010). Previous studies at Qinghai Lake have mainly focused on the paleoclimate and paleoenvironment information from Qinghai Lake sediment (Liu et al., 2002; Henderson et al., 2003) and water chemistry (Xu et al., 2006). However, the aerosols at Qinghai Lake have not been investigated. Thus, the current study aims to (1) investigate the chemical composition of  $PM_{2.5}$  and the total suspended particulate (TSP) samples, and (2) investigate the possible sources of components in the atmosphere during summertime.

## 2. Experimental methods

### 2.1. Sampling site

Qinghai Lake, the largest closed inland saline lake in China with an area of approximately 4400 km<sup>2</sup>, is located in a structural intermountain depression on the northeastern corner of the Tibetan Plateau (Fig. 1). The lake catchment lies within the northernmost boundary of the Asian summer monsoon. Based

on stable oxygen isotope data, the Qinghai Lake area was found to be climatically controlled by the southwest summer monsoon in summer, associated with a large proportion of annual precipitation and lesser amounts of precipitation in winter dominated by westerly disturbances (Wei and Gasse, 1999). Qinghai Lake has developed into a saline lake (mean salinity of 14.1 mg·L<sup>-1</sup>) with Na–Mg–SO<sub>4</sub><sup>2-</sup>–Cl<sup>-</sup> brine type and alkaline water (pH level of 9 to 9.4) because of the excess evaporation of water (c. 1000 mm·a<sup>-1</sup>) over precipitation (mean annual precipitation over the catchment is c. 400 mm) (Henderson et al., 2003).

Aerosol samples were taken on an atmospheric monitoring tower (N36.977°, E99.899°, 3200 m asl) approximately 10 m above ground level. The tower is located on the peninsula of northwestern Qinghai Lake (Fig. 1), which is a scenic region far from intense human activities and heavy industries. This site is an ideal background station.

### 2.2. Aerosol collection and analysis

$PM_{2.5}$  and TSP samples were collected synchronously from June to September 2010. Each sample was run for 120 h. High-volume samplers (Andersen Instruments/GMW) operated at 1.00 m<sup>3</sup> min<sup>-1</sup> were used to collect  $PM_{2.5}$  samples on 20.3 × 25.4 cm Whatman quartz microfiber filters (QM/A; Whatman, Middlesex, UK). TSP samples were collected using an RP2025 air sampler (Thermo, USA) with 47 mm-diameter Whatman quartz microfiber filters operated at a flow rate of 16.7 L min<sup>-1</sup>. It is noticed that high flow rate will cause the concentration of volatile species to be relatively lower than low flow rate, so the characteristic of volatile species was not discuss in this paper. Overall, 18 TSP and 18  $PM_{2.5}$  samples were collected.

Both  $PM_{2.5}$  and TSP sampling filters were pre-combusted at 800 °C for 3 h prior to use and were maintained in a condition of 50% RH (relative humidity) and 25 °C for over 24 h. Then, the filters were weighed prior to sampling using a

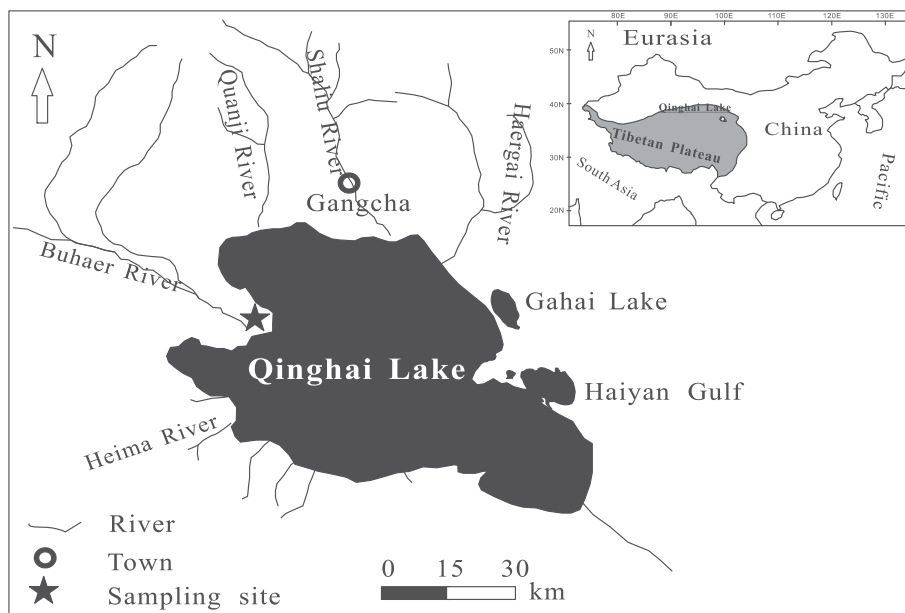


Fig. 1. Location of Qinghai Lake and sampling site.

Sartorius MC5 electronic microbalance (Sartorius, Göttingen, Germany) with sensitivity of 1 µg. After sampling, both PM<sub>2.5</sub> and TSP filters were conditioned and weighed again to determine the mass concentration of the filtered particles. The filters were then cut into portions for individual analysis. Blank samples were also analyzed and the sample results were corrected by averaging the blank sample concentrations.

An ion chromatograph (Dionex Corp., DX-300, USA) was used to measure the main water soluble ions. Concentrations of cation (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, and Ca<sup>2+</sup>) were determined with a CS12A column (Dionex Corp.), using 20 mM methanesulfonate as an eluent, and for anions (SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>), they were separated on an AS11-HC column (Dionex Corp.), using 20 mM KOH as the eluent. The method detection limits (MDLs) for the ions were Na<sup>+</sup> (4.6 µg·L<sup>-1</sup>), NH<sub>4</sub><sup>+</sup> (4.0 µg·L<sup>-1</sup>), K<sup>+</sup> (10.0 µg·L<sup>-1</sup>), Mg<sup>2+</sup> (10.0 µg·L<sup>-1</sup>), Ca<sup>2+</sup> (10.0 µg·L<sup>-1</sup>), SO<sub>4</sub><sup>2-</sup> (20.0 µg·L<sup>-1</sup>), NO<sub>3</sub><sup>-</sup> (15.0 µg·L<sup>-1</sup>) and Cl<sup>-</sup> (0.5 µg·L<sup>-1</sup>). Standard Reference Materials produced by the National Research Center for Certified Reference Materials, China, were analyzed for quality assurance purposes (Shen et al., 2009).

Elements (K, Ca, Ti, Cr, Mn, Fe, Ni, Zn, As, Pb) were determined by Energy Dispersive X-Ray Fluorescence (EDXRF) spectrometry using PANalytical Epsilon 5 XRF analyzer (PANalytical B.V., Almelo, The Netherlands), and the specific laboratory methods are the same as in Zhang et al. (2012). Each sample was irradiated for half an hour and a laboratory blank Quartz filter sample is also analyzed to evaluate analytical bias. The elements that were determined were K, Ca, Ti, Cr, Mn, Fe, Ni, Zn, As, Pb with the detection limits (µg·cm<sup>-2</sup>) of 0.007 (K), 0.007 (Ca), 0.005 (Ti), 0.003 (Cr), 0.014 (Mn), 0.011 (Fe), 0.003 (Ni), 0.008 (Zn), 0.000 (As) and 0.015 (Pb).

OCs and ECs were analyzed for eight carbon fractions (OC1, OC2, OC3, and OC4 at 120 °C, 250 °C, 450 °C, and 550 °C, respectively, in a helium (He) atmosphere; OP, a pyrolyzed carbon fraction determined when reflected or transmitted laser light attained its original intensity after oxygen (O<sub>2</sub>) was added to the analysis atmosphere; and EC1, EC2, and EC3 at 550 °C, 700 °C, and 800 °C, respectively, in a 2% O<sub>2</sub>/98% He atmosphere) with a DRI Model 2001 Thermal and Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA) following the IMPROVE-A (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Cao et al., 2007).

### 3. Results and discussion

As shown in Table 1, the mean mass concentration of the PM<sub>2.5</sub> samples was 21.27 ± 10.70 µg m<sup>-3</sup>, ranging from 9.57 µg m<sup>-3</sup> to 24.52 µg m<sup>-3</sup>. No sampling day exceeded the U.S. National Ambient Air Quality Standard (NAAQS) (35 µg m<sup>-3</sup> for 24 h period). The mass concentrations of the TSP samples ranged from 16.60 µg m<sup>-3</sup> to 75.87 µg m<sup>-3</sup>, with a mean of 41.47 ± 20.25 µg m<sup>-3</sup>, which is close to the PM<sub>10</sub> mass concentration in Waliguan (46.71 µg m<sup>-3</sup>) and Xining (64.25 µg m<sup>-3</sup>) from May to June 2007 (Tang et al., 2010) and in Lhasa (51.8 ± 42.5 µg m<sup>-3</sup>) from September 2007 to August 2008 (Cong et al., 2011). However, the mean mass concentration of the TSP samples was significantly less than the TSP concentration in urban regions, such as in Lanzhou (310.1 µg m<sup>-3</sup>) in summertime (Wang et al., 2009) and in Xian (270.8 ± 110.5 µg m<sup>-3</sup>) in normal days (Shen et al., 2009). Both TSP and PM<sub>2.5</sub> mass concentrations indicated that the Qinghai Lake region had relatively low atmospheric particulate matter loading.

#### 3.1. Water soluble ions

The concentrations of the detected ions, including the ionic concentrations of aerosols from other sites for comparison, are shown in Table 1. Generally, SO<sub>4</sub><sup>2-</sup> showed the greatest mass concentration in both PM<sub>2.5</sub> and TSP samples. The mean concentrations of SO<sub>4</sub><sup>2-</sup> were 4.45 and 5.04 µg m<sup>-3</sup>, which account for 80 ± 11% and 60 ± 11% of total detected ionic mass in the PM<sub>2.5</sub> and TSP samples, respectively. The high percentage of SO<sub>4</sub><sup>2-</sup> may be caused by long-distance transport from eastern China. Moreover, NH<sub>4</sub><sup>+</sup>, which has a substantial part in ionic loading in remote areas, was not detected in our samples. The mass concentration of NH<sub>4</sub><sup>+</sup> was greater than that of SO<sub>4</sub><sup>2-</sup> at Waliguan (Table 1), which is a background monitoring site approximately 130 km away from our sampling site in linear distance. The concentration of NO<sub>3</sub><sup>-</sup> was the second biggest in both PM<sub>2.5</sub> and TSP samples. Our sampling period was the tourism rush season at Qinghai Lake, wherein most tourists were transported by various vehicles. Therefore, NO<sub>x</sub>, which can be derived from the combustion of fossil fuels, can be transformed to particulate NO<sub>3</sub><sup>-</sup> through photo-oxidation reaction. Interestingly, NO<sub>3</sub><sup>-</sup> had a PM<sub>2.5</sub>/TSP ratio of

**Table 1**  
Concentration of major ions (µg/m<sup>3</sup>) at Qinghai Lake, and a comparison with other sites.

Sites	Sampling period	Size fraction	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	Reference
Qinghai Lake	Jun–Sep 2010	PM <sub>2.5</sub>	0.13	–	0.12	0.06	0.23	0.07	0.38	4.45	This study
		TSP	0.48	–	0.13	0.26	0.72	0.39	1.3	5.04	
<i>Tibetan Plateau sites</i>											
Mt. Gongga	Dec. 2005–Nov. 2006	PM <sub>10</sub>	0.1	1.7	0.31	0.08	0.68	0.26	0.88	4.74	Zhao et al. (2009)
Waliguan	Oct. 1994	TSP	–	0.33	–	–	–	0.2	0.24	0.22	Yang et al. (1996)
East Rongbuk Glacier	Sep. 2003	TSP	0.04	–	0.02	0.01	0.04	0.1	0.15	0.4	Ming et al. (2007)
Mt. Yulong	May–Jun. 2006	TSP	0.12	–	0.05	0.01	0.09	0.01	0.61	0.05	Zhang et al. (2010)
<i>Urban sites</i>											
Xian	October 2006–September 2007	PM <sub>2.5</sub>	0.9	7.6	1.9	0.2	1.3	4.2	12.0	27.9	Shen et al. (2009)
		TSP	1.4	8.3	2.3	0.9	11.4	4.6	16.1	34.0	
Chongqing	July 2001	TSP	0.1	3.16	1.6	–	4.87	1.76	2.88	13.92	Tao et al. (2006)
Shanghai	Summer 2004	PM <sub>2.5</sub>	0.51	2.44	0.23	0.28	1.55	0.50	2.59	5.43	Wang et al. (2006a)
Beijing	Spring 2001–2004	TSP	2.05	12.4	1.84	0.8	12.16	8.22	22.76	27.25	Wang et al. (2006b)

0.18, indicating that  $\text{NO}_3^-$  was mainly dominant in coarse modal in our samples, in agreement with the report by [Mkoma et al. \(2009\)](#) in East Africa. This result can be attributed to the reactions between airborne gaseous  $\text{HNO}_3$  and crustal particles and salt particles, as well as the correlation between  $\text{NO}_3^-$  and  $\text{Ca}^{2+}$  ( $R = 0.81, P < 0.01$ ) and  $\text{Mg}^{2+}$  ( $R = 0.79, P < 0.01$ ) was significant. Moreover,  $\text{Ca}^{2+}$  was the dominant cation in both  $\text{PM}_{2.5}$  and TSP samples.

Ionic mass concentrations from previous studies at different locations, including the sites in the Tibetan Plateau and the urban area, were listed for comparison. All detected ions in our TSP samples were lower than the corresponding ionic mass concentration collected in the urban area, except for the  $\text{Na}^+$  concentration in our TSP sample that was greater than that in Chongqing ([Tao et al., 2006](#)). However, all ionic concentrations in the TSP were greater than those studied in the Tibetan Plateau, except for the  $\text{K}^+$  concentration that was lower than that in Mt. Gongga. Furthermore, compared with the ionic concentration observed at Waliguan, the concentrations of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ , as secondary aerosols, were greater than the values in the TSP samples, particularly the  $\text{SO}_4^{2-}$  concentration, reported at Waliguan. These results indicate that the ionic concentration at Qinghai Lake was influenced by anthropogenic sources.

### 3.2. Elemental composition

The overall average values of the concentration of detected elements in the  $\text{PM}_{2.5}$  and TSP samples, including the elemental concentrations from other sites, are listed in [Table 2](#). Ca, Fe, and K related to crustal matter were the main elements found in both  $\text{PM}_{2.5}$  and TSP samples. Significant correlations between Ca and Fe ( $R = 0.98, P < 0.01$ ) and Ca and K ( $R = 0.99, P < 0.01$ ) were found in the TSP samples. To identify the influences of anthropogenic sources, the EFs of the elements were calculated as follows:

$$\text{EF}_X = (C_X/C_R)_{\text{aerosol}} / (C_X/C_R)_{\text{crust}}$$

where X represents the selected element,  $C_X$  is the concentration of X, and  $C_R$  indicates the concentration of a reference element. The aerosol and crust subscripts refer to the ratios in the aerosol samples and crustal material, respectively. Al, Si, Ti, and Fe are commonly used as reference elements. In this study, Fe was selected as the reference material, and the elemental composition of the average upper continental crust was obtained from [Wedepohl's \(1995\)](#) work.

[Fig. 2](#) shows the EFs of the elements in our samples. In general, when the  $\text{EF}_X$  of a sample approaches unity, the dominant source for that element is treated as crustal; if  $\text{EF}_X > 10$ , a significant fraction of the element is considered to be from anthropogenic sources. As shown in [Fig. 2](#), only the EFs of As, Pb, and Zn exceeded 10, and high correlations were observed among the three elements in both  $\text{PM}_{2.5}$  ( $R = 0.80\text{--}0.97, P < 0.01$ ) and TSP ( $R = 0.75\text{--}0.95, P < 0.01$ ) samples, indicating that the elements were influenced by anthropogenic sources and should have the same source or transport process.

### 3.3. Carbonaceous composition

The average concentrations of OCs and ECs and OC/EC ratios in the  $\text{PM}_{2.5}$  and TSP samples at Qinghai Lake are listed in [Table 3](#). Selected data from other rural and urban sites are also listed in [Table 3](#) for comparison. The concentration of OCs in  $\text{PM}_{2.5}$  and TSP was  $1.49 \pm 0.63$  and  $2.30 \pm 0.95 \mu\text{g m}^{-3}$ , respectively, whereas the concentration of ECs in  $\text{PM}_{2.5}$  and TSP was  $0.33 \pm 0.17$  and  $0.47 \pm 0.28 \mu\text{g m}^{-3}$ . Both OC and EC concentrations in the  $\text{PM}_{2.5}$  or TSP samples in Qinghai Lake were lower than the concentrations in the urban sites listed in [Table 3](#) and were lower compared with those in Mt. Hua ([Li et al., 2011](#)) and Daihai ([Han et al., 2006](#)). However, these concentrations were close to those in Zhuzhuang and Akdala ([Qu et al., 2009](#)), which are both regarded as remote rural sites far from human activities. However, these concentrations were significantly greater compared with those collected at Muztagh Ata peak ([Cao et al., 2009a](#)), which is a remote high-altitude site. By contrast, EC concentration in the

**Table 2**  
Concentration of elements ( $\text{ng/m}^3$ ) at Qinghai Lake, and a comparison with other sites.

	Qinghai Lake		Chengdu <sup>a</sup>	Mt. Gongga <sup>b</sup>	Daihai <sup>c</sup>		Mt. Yulong <sup>d</sup>	Nam Co <sup>e</sup>	Waliguan <sup>f</sup>
	$\text{PM}_{2.5}$	TSP	$\text{PM}_{2.5}$	$\text{PM}_{2.5}$	$\text{PM}_{2.5}$	TSP	TSP	TSP	TSP
K	275.79	670.16	2802.1	498.2	98	599.6		82	1190–2200
Ca	384.06	1639.7	1306.9	372.8	618.4	2757.5	4814.84	251	2010–4280
Ti	25.31	88.74	121.6		11.1	95	149.94	10	106–220
V	0.59	2.07		0.7	6.8	6.7	1.76	0.06	3.02–5.88
Cr	2.31	5.75	7.2		6.1	6.6	3.16	1	
Mn	5.2	28.76	74.8		63.9	72	36.03	3.7	27.5–52.9
Fe	273.68	1111.16	1425.9	224	385.8	1824	1258.31	94	1720–3910
Ni	0.62	1.56	3.3	0.9	7	16.5	2.95	0.95	
Zn	38.49	57.31	323.9	154.6	27.8	12.4	71.75	1.8	9.5–26.3
As	3.04	3.7	0.4	4.3	7.4	10.3	3.61	0.04	0.70–3.37
Ba	13.33	28	27	6			12.67		21.3–45.3
Pb	20.03	24.46	132.9	39.4	20.4	44.3	12.37		

<sup>a</sup> Wang et al. (2010).

<sup>b</sup> Yang et al. (2009).

<sup>c</sup> Han et al. (2006).

<sup>d</sup> Zhang et al. (2012).

<sup>e</sup> Cong et al. (2007).

<sup>f</sup> Wen et al. (2001).

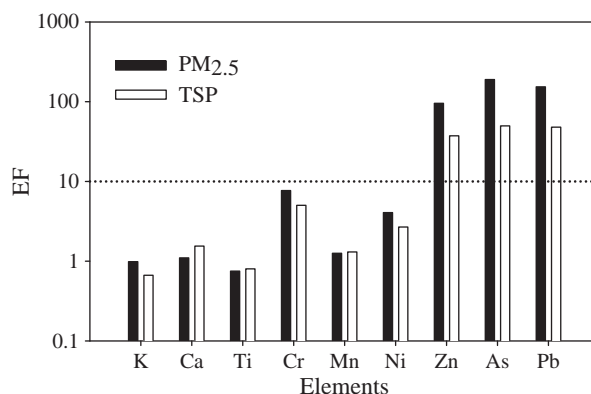


Fig. 2. EFs of detected elements.

TSP samples in Qinghai Lake was significantly greater than that monitored at remote sites, such as Mt. Waliguan ( $0.05 \mu\text{g m}^{-3}$  to  $0.13 \mu\text{g m}^{-3}$ ) (Tang et al., 1999), Nam Co ( $0.082 \mu\text{g m}^{-3}$ ) (Ming et al., 2010) and Qilian Shan ( $0.02 \mu\text{g m}^{-3}$  to  $0.07 \mu\text{g m}^{-3}$ ) (Zhao et al., 2012). These comparisons suggest that the concentration of carbonaceous aerosol was also influenced by human activities at Qinghai Lake.

According to previous studies (Castro et al., 1999; Cao et al., 2003), primary organic carbon (POC) and secondary organic carbon (SOC) can be estimated as follows:

$$\text{POC} = \text{EC} * (\text{OC}/\text{EC})_{\min}$$

$$\text{SOC} = \text{OC} - \text{POC}$$

where  $(\text{OC}/\text{EC})_{\min}$  is the minimum ratio observed. The results show that the estimated POC was  $0.93 \pm 0.48$  and  $1.75 \pm 0.98 \mu\text{g m}^{-3}$  and the estimated SOC was  $0.60 \pm 0.40$  and  $0.59 \pm 0.45 \mu\text{g m}^{-3}$  in the PM<sub>2.5</sub> and TSP samples, respectively. The estimated SOC/POC ratio was  $0.48 \pm 0.38$  in the TSP samples and  $0.73 \pm 0.43$  in the PM<sub>2.5</sub> samples, suggesting that carbonaceous aerosols in fine particles were more oxidized.

### 3.4. Relationship between OCs and ECs

EC comes from primary anthropogenic sources, and OC is not only emitted directly from primary particles but is also formed by atmospheric chemical reactions involving gaseous hydrocarbon precursors. The ratio of OC to EC concentrations is used to identify the presence of secondary organic aerosols when the OC/EC ratios exceed 2.0. However, in rural sites,

Table 3  
Concentration of OCs and ECs ( $\mu\text{g}/\text{m}^3$ ) at Qinghai Lake, and a comparison with other sites.

Sites	Size fraction	Sampling period	OCs	ECs	OC/EC	Reference
Qinghai Lake	TSP	June–September 2010	2.30	0.48	5.23	This study
Rural site with altitude of 3200 m	PM <sub>2.5</sub>		1.49	0.33	4.77	
<i>Rural sites</i>						
Daihai	TSP	2005–2007	12.40	2.01	6.17	Han et al. (2008)
Muztagh Ata	TSP	Dec. 2003–Feb. 2005	0.48	0.06	8	Cao et al. (2009a)
Remote mountain with the altitude of 4500 m						
Zhuzhuang	PM <sub>10</sub>	Summer 2004–spring 2005	$3.1 \pm 0.91$	$0.34 \pm 0.16$	9.11	Qu et al. (2009)
Rural background site with the altitude of 3583 m						
Akdala	PM <sub>10</sub>	Summer 2004–spring 2006	$2.9 \pm 1.6$	$0.35 \pm 0.31$	8.29	Qu et al. (2009)
Rural background site with the altitude of 562 m						
Mt. Hua	TSP	January 10–22, 2009	$6.9 \pm 2.9$	$0.9 \pm 0.6$	$8.2 \pm 3.0$	Li et al. (2011)
Rural mountain site with the altitude of 2160 m						
<i>Urban sites</i>						
Xi'an	PM <sub>10</sub>	Winter 2003–2004	93.00	22.70	4.20	Cao et al. (2005)
Hangzhou	PM <sub>10</sub>	September 2001–August 2002	$21.41 \pm 18.03$	$4.06 \pm 4.13$	5.27	Cao et al. (2009b)
Guangzhou	PM <sub>10</sub>	August 2–September 10, 2004	37.93	13.83	2.00–3.77	Wang et al. (2006)
Bangkok	PM <sub>2.5</sub>	May–October 2007	5.3	3.0	1.9	Sahu et al. (2011)

OCs can be produced by biogenic sources and formed during long-distance transport by various chemical reactions (Cao et al., 2003, 2009a). The OC/EC ratios were greater in remote sites compared with those in urban sites and almost exceeded 6.0 (Table 3). Comparing the OC/EC ratios in the PM<sub>2.5</sub> samples, Cao et al. (2003) found OC/EC ratios that were between 1.0 and 4.0 for most urban sites around the world. The OC/EC ratio at Qinghai Lake was greater than 4.0, however, the OC/EC ratios in the PM<sub>2.5</sub> (4.77 ± 1.32) and TSP (5.23 ± 1.39) samples in Qinghai Lake were both lower than those in rural sites, such as the ratio of 6.17 in the TSP samples in Daihai and 9.11 and 8.29 in the PM<sub>10</sub> samples in Zhuzhuang and Akdala, respectively. The value of the OC/EC ratio at Qinghai Lake fell between the values of the urban and remote sites, indicating that the OC and EC concentrations were influenced by anthropogenic sources at Qinghai Lake. These results are in accordance with the phenomenon that the OC/EC ratio was low in the polluted samples at Zhuzhuang and Akdala (Qu et al., 2009). Furthermore, a low OC/EC ratio in the PM<sub>2.5</sub> samples implies that anthropogenic sources are important in fine particles. The anthropogenic sources influenced not only the OC/EC ratio but also the relationship between OCs and ECs. The correlation between OCs and ECs in the PM<sub>2.5</sub> samples ( $R^2 = 0.59$ ) was lower than that in the TSP samples ( $R^2 = 0.78$ ), as shown in Fig. 3.

EC has various sources, including diesel engine emissions, fossil fuel combustion at low burning efficiency and biomass burning. Thus, the ratios of EC/TC and K<sup>+</sup>/EC in the PM<sub>2.5</sub> samples were calculated to identify the source types of ECs. Previous studies have pointed out that EC/TC ratios range from 0.6 to 0.7 in fuel combustion emissions and from 0.1 to 0.2 in biomass combustion emissions (Salam et al., 2003). The EC/TC ratio in our PM<sub>2.5</sub> samples was 0.18 ± 0.04, suggesting that EC mainly comes from biomass burning. The elevated fine K<sup>+</sup> is known as an indicator of biomass burning, and results of previous studies indicate that K<sup>+</sup>/EC ratios range from 0.20 to 0.69 in biomass burning aerosols (Andreae, 1983; Ram and Sarin, 2010). The K<sup>+</sup>/EC ratio in our PM<sub>2.5</sub> samples was 0.38 ± 0.17, suggesting that the concentration of ECs was more influenced by biomass burning emissions in the PM<sub>2.5</sub> than in

the TSP samples. This result can be attributed to the fact that yak dung is commonly used for cooking and heating in the traditional Tibetan pastoralist society in the Qinghai Lake region. Various vehicles were used in Qinghai Lake during our sampling periods because of the tourism rush season. Therefore, traffic emissions should comprise another source of EC that makes OC/EC ratios lower than those in some remote rural sites.

### 3.5. Sources and transport

#### 3.5.1. Local sources

Mass concentration distribution of aerosol species can provide clues on the sources or processes of species. Fig. 4 shows the average PM<sub>2.5</sub>/TSP ratios and associated standard deviations. As shown in the figure, common crustal components, such as Mg<sup>2+</sup>, Ca<sup>2+</sup>, Ca, Ti, Fe and K, were mainly in the coarse size fraction. Their correlation coefficients were all greater than 0.89 ( $P < 0.01$ ), suggesting that they were from crustal sources. For potassium, K<sup>+</sup> (water soluble potassium) was mainly in fine fraction contrast with the K (elemental potassium) dominant in coarse fraction (Fig. 4). The average K<sup>+</sup>/K ratio was 0.49 in PM<sub>2.5</sub> and 0.25 in TSP, suggesting that the water-soluble part of potassium had sources different from those of the insoluble part of potassium, and that most potassium was concentrated in the insoluble fraction in the Qinghai Lake aerosol samples. These results are in accordance with the minerals at Qinghai Lake, which mainly comprise illite and are rich in insoluble potassium (Xu et al., 1989).

In our aerosol samples, Cl<sup>-</sup> and Na<sup>+</sup> were mainly distributed in coarse mode, indicating that they were from local sources. The correlation coefficient between them was 0.79 ( $P < 0.01$ ) and 0.94 ( $P < 0.01$ ) in the PM<sub>2.5</sub> and TSP, respectively, indicating that they were from the same source or process. As a saline water lake, the water chemical composition of Qinghai Lake is close to that of seawater (Hou et al., 2009). Moreover, the Cl<sup>-</sup>/Na<sup>+</sup> mass ratio of the lake water was approximately 1.5, which is slightly lower than the ratio of seawater (1.8). Chloride depletion of sea salt has been reported in previous studies conducted at coastal sites. The formation of

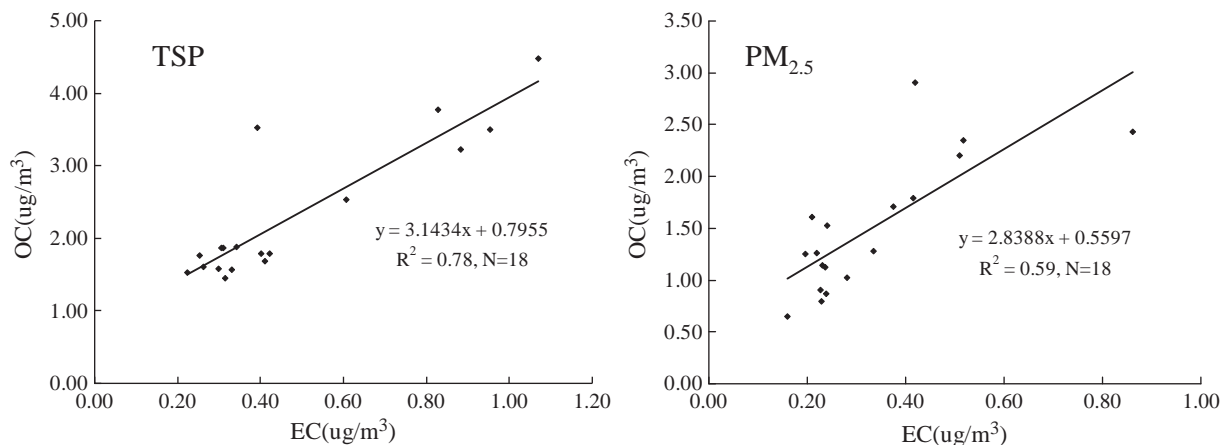


Fig. 3. Relationship between OCs and ECs in TSP and PM<sub>2.5</sub> samples.

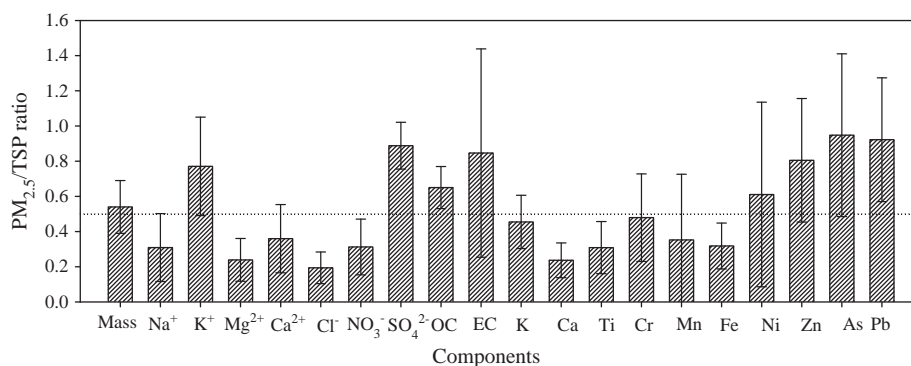


Fig. 4. PM<sub>2.5</sub>/TSP ratios of species.

nitrate and sulfate has also been observed along with the chloride depletion (Savoie and Prospero, 1982; Zhuang et al., 1999), leading to coarse mode distribution of most nitrates and a small fraction of sulfate (Zhuang et al., 1999). In our aerosol samples, the mean value of Cl<sup>-</sup>/Na<sup>+</sup> ratio was 0.48 and 0.81 in the PM<sub>2.5</sub> and TSP samples, respectively, displaying the serious Cl deficit in our aerosol samples. Moreover, nitrates and sulfate were found to be mainly distributed in coarse and fine modes, respectively (Fig. 4), proving that Cl<sup>-</sup> and Na<sup>+</sup> can be produced by the salty lake, just like the sea salt particles emitted from seawater. Due to this phenomenon, the concentrations of Cl<sup>-</sup> and Na<sup>+</sup> in our TSP samples were higher than those in other sites around the Tibetan Plateau (Table 1). Moreover, significant inverse correlation between SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>/Na<sup>+</sup> ratios was found in both PM<sub>2.5</sub> and TSP samples because of the Cl deficit along with the formation of sulfate (Fig. 5), indicating that the depletion of Cl<sup>-</sup> becomes more serious with increasing SO<sub>4</sub><sup>2-</sup> concentration.

The existence of a saline lake can influence the regional concentration of sulfate via the reactions between salt particles and acid gases. Generally, sulfate has an important role in climate cooling by strongly scattering sunlight and reflecting solar radiation. According to statistical results, lakes with areas greater than 1.0 km<sup>2</sup> account for approximately 44993.3 km<sup>2</sup> over the whole Tibetan Plateau, and most of these lakes are

saline. Thus, emissions of salt particles from these saline lakes may affect local climate. However, the cooling effect of aerosols is reduced by its chemical composition changes, particularly the presence of black carbon (BC, which is equated to EC). Chýlek et al. (1995) pointed out that a BC-to-sulfate mass mixing ratio of 5% will reduce the local sulfate cooling effect, which is also accompanied by a change in the vertical distribution of solar heating and lapse rate. In our aerosol samples, the BC-to-sulfate mass ratio was 7.4% and 9.5% in PM<sub>2.5</sub> and TSP, respectively, suggesting that the cooling effect of sulfate can be affected by the BC emissions at the Qinghai Lake region. Therefore, the roles of a saline lake in regional aerosol chemistry and the chemical composition of aerosol should be considered in the evaluation of regional climate changes.

### 3.5.2. Long-distance transport

Aerosol particles in coarse mode can be deposited by gravity and have a short resident time in the atmosphere. Therefore, species discussed for long-distance transport are mainly focused on PM<sub>2.5</sub> samples. Backward trajectories were computed using the hybrid single-particle Lagrangian integrated trajectory (HYSPPLIT-4) model (<http://www.arl.noaa.gov/ready/hysplit4.html>) to identify the potential source regions and transport pathways. Backward trajectories setting the end location at our sampling sites were run for 6 days with heights of 500 and

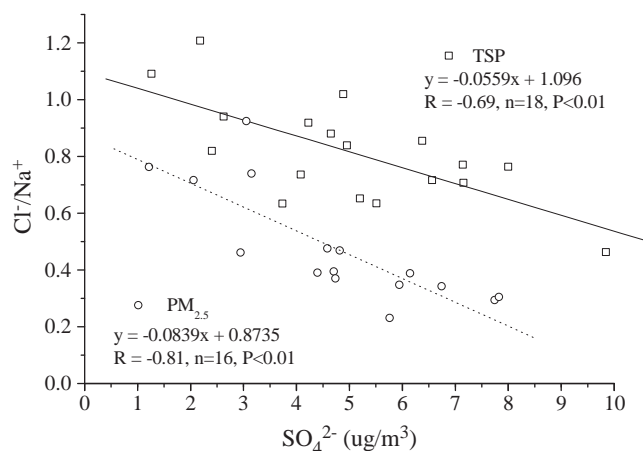


Fig. 5. Relationship between SO<sub>4</sub><sup>2-</sup> and Cl<sup>-</sup>/Na<sup>+</sup> ratios in TSP and PM<sub>2.5</sub> samples.

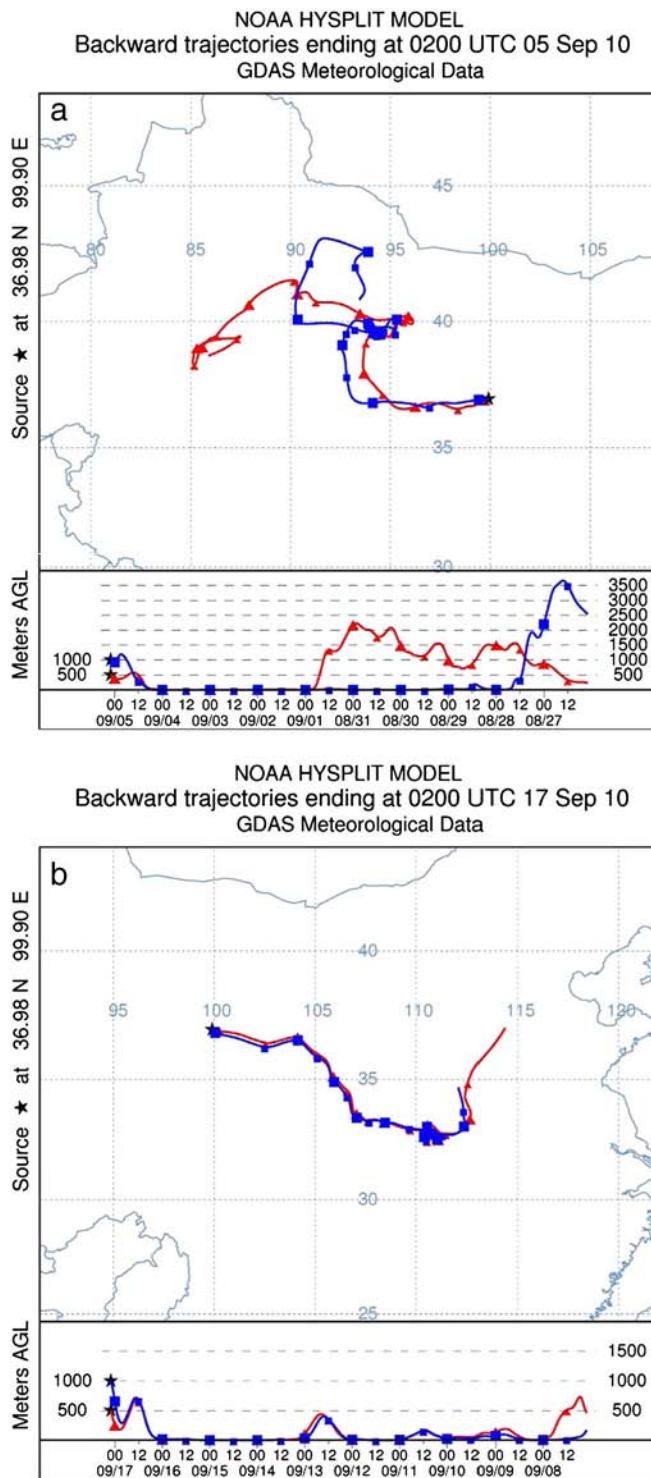


Fig. 6. Compared 6 day backward trajectories of lowest (a) and biggest (b) values of  $SO_4^{2-}$ .

1000 m above ground level. The ending time was set at the end of each sample (around 10 a.m.). Fig. 6(a) is a typical backward trajectory which associated with the lowest concentration of  $SO_4^{2-}$  during sampling time, showing that the air mass arrived at Qinghai Lake passed through the western area of the lake, which

have few residents and less anthropogenic pollutions. On the contrary, air mass transported from eastern China (Fig. 6b), which agrees well with East Asian monsoon pathway, is typical with the highest concentration of  $SO_4^{2-}$ . This indicates that the pollutants from eastern China seemed to have been transported



to Qinghai Lake. A similar result was found in the variation of OCs, and a significant correlation between  $\text{SO}_4^{2-}$  and OCs was found ( $R = 0.80$ ,  $P < 0.01$ ), which was greater than that between OCs and ECs ( $R = 0.77$ ,  $P < 0.01$ ). This result suggests that parts of  $\text{SO}_4^{2-}$  and OCs were formed by the oxidizing reaction during long-distance transport.

The elements whose EF value exceeded 10 were found to have good correlation with  $\text{SO}_4^{2-}$  and OCs in the  $\text{PM}_{2.5}$  samples. The correlation coefficients between  $\text{SO}_4^{2-}$  and Zn, Pb, and As, were 0.86 ( $P < 0.01$ ), 0.84 ( $P < 0.01$ ), and 0.69 ( $P < 0.01$ ), respectively. The correlation coefficients between OCs and Zn, Pb, and As were 0.96 ( $P < 0.01$ ), 0.91 ( $P < 0.01$ ), and 0.77 ( $P < 0.01$ ), respectively. The correlation coefficients among Zn, Pb, and As ranged from 0.80 to 0.97 ( $P < 0.01$ ), indicating that the three elements were from the same source and were influenced by monsoon circulation, as well as by  $\text{SO}_4^{2-}$  and OCs.

#### 4. Conclusions

In this study, the particulate mass, water soluble ions, 10 elements, OCs, and ECs of  $\text{PM}_{2.5}$  and TSP samples were determined during the summer season of 2010 at the rural site of Qinghai Lake. The mass concentrations of  $\text{PM}_{2.5}$  and TSP were  $21.27 \pm 10.70 \mu\text{g m}^{-3}$  and  $41.47 \pm 20.25 \mu\text{g m}^{-3}$ , respectively, which indicate lower mass loading of aerosols at Qinghai Lake than in other sites.  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  were the greatest anion and cation for both  $\text{PM}_{2.5}$  and TSP samples, and Ca, Fe, and K were the main elements. The concentration levels of the detected ions and elements indicate that they were influenced by anthropogenic sources at Qinghai Lake, and EF values of As, Pb, and Zn were greater than 10.

EC concentrations were  $0.33 \pm 0.17 \mu\text{g m}^{-3}$  and  $0.47 \pm 0.28 \mu\text{g m}^{-3}$  in  $\text{PM}_{2.5}$  and TSP, whereas OC concentrations in  $\text{PM}_{2.5}$  and TSP were  $1.49 \pm 0.63 \mu\text{g m}^{-3}$  and  $2.30 \pm 0.95 \mu\text{g m}^{-3}$ , respectively. The ratios of OC/EC were  $4.77 \pm 1.32 \mu\text{g m}^{-3}$  and  $5.23 \pm 1.39$  in  $\text{PM}_{2.5}$  and TSP, respectively. Comparing the ratio values of  $\text{K}^+/\text{TC}$  and  $\text{K}^+/\text{EC}$  in our aerosol samples with those in biomass burning and fuel combustion aerosol samples, the concentration of ECs was more influenced by biomass burning emissions in the  $\text{PM}_{2.5}$  samples.

As expected, components such as  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ , K, Ca, Ti, Mn, and Fe were mainly in the coarse size fraction. The correlation coefficients among  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ , Ca, Ti, Fe, and K were all greater than 0.89 ( $P < 0.01$ ), suggesting that they were both from crustal sources.  $\text{Cl}^-$  and  $\text{Na}^+$  can be produced by Qinghai Lake, and the reactions between salt particles and acid gases in aerosol led to the mean value of  $\text{Cl}^-/\text{Na}^+$  ratio being 0.48 and 0.81 in  $\text{PM}_{2.5}$  and TSP samples, displaying serious Cl deficit in our aerosol samples. A significant inverse correlation between  $\text{SO}_4^{2-}$  and  $\text{Cl}^-/\text{Na}^+$  ratios was found both in  $\text{PM}_{2.5}$  and in TSP samples. Backward trajectories and correlation coefficients were computed, along with components including  $\text{SO}_4^{2-}$ , OCs, Zn, As, and Pb associated with long-distance transport from eastern China.

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