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Individual Particle Analysis of Atmospheric Aerosols at Nam Co, Tibetan Plateau

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ABSTRACT

Twelve filter samples were sampled monthly during a continuous 1-year campaign at a remote site, Nam Co in central Tibetan Plateau. The total aerosol concentrations ranged from 0.48 μg/m^3 to 36.11 μg/m^3 with the annual average of 6.74 μg/m^3, reflecting a typical background level. The composition and morphology of atmospheric aerosols with size ranging from 0.5 to 10 μm were investigated using a scanning electron microscope coupled with energy dispersive X-ray analysis. Then aerosol particles were classified into 7 groups: soot, tar ball, aluminosilicates/quartz, calcium sulfate, Ca/Mg carbonate, Fe/Ti oxide, and biological particle. Aerosol optical properties from the sun photometer and the results obtained by SEM-EDX could support and verify each other, providing complementary information on aerosol characteristics at Nam Co. Two distinct types of air masses arriving at Nam Co correspond to different aerosol constituent, showing that the summer monsoon circulation may bring considerable pollution from South Asia, while the westerly flow dominated in winter is relative clean.

Keywords: Aerosols; SEM-EDX; Nam Co; Tibetan Plateau.

INTRODUCTION

The Tibetan Plateau (TP) is the highest and most extensive plateau on Earth. As a unique geomorphic unit, the TP plays a key role in the Asian monsoon system and it greatly influences the climate of Asia. The atmosphere over TP remains pristine due to the sparse human population and minimal industrial activities (Wang *et al.*, 2006).

Recently, climate change has become a crucial issue receiving tremendous attention worldwide (IPCC, 2007; Li *et al.*, 2007). Especially, the TP has been regarded as a sensitive region to global climate change owing to its special landform, ecosystem, and monsoon circulation. For example, over TP, the climate changes have proximately caused a rise in temperature of up to 0.3°C a decade, which is approximately three times the global warming rate (Qiu, 2008). Therefore, an essential question now arises, i.e. what are the regional and global teleconnections and responses of TP climate to changes in external forcing (e.g. aerosols and greenhouse gases)? In this context, increasing attention has been devoted to the study of aerosol effects on regional climate change such as the slowing down of South Asian Monsoon system and the retreat of mountain glaciers (Jin, 2006; Lau *et al.*, 2006; Ramanathan *et al.*, 2007; Xia *et al.*, 2008). However large uncertainties still exist in the climate modeling and assessments due to the lack of comprehensive knowledge of aerosol properties over TP, especially considering the significant spatial and temporal variations of aerosols.

The majority of pervious studies from TP focused on bulk aerosol composition (Liu *et al.*, 1997; Wen *et al.*, 2001; Zhang *et al.*, 2001; Cong *et al.*, 2007). While little is known about the characteristics of individual aerosol particles, although those information are of crucial importance for understanding their source and formation, as well as assessing their scattering and absorption capabilities. Aerosols consist of many different types of particles (e.g. sea salt, mineral dust, fly ash and biogenic particles). The aerosol optical properties depend on which type they are. For example, the sulfate aerosols are considered to result in a negative radiative forcing that leads to a cooling of the Earth surface. By contrast, soot, or black carbon is an effective absorber of solar radiation and therefore has a warming effect (Mogo *et al.*, 2005).

In this study, as a part of comprehensive characterization of atmospheric aerosols in central TP, the aerosols from Nam Co were analyzed at the individual particle level using scanning electron microscopy coupled with energy dispersive X-ray analysis (SEM-EDX). Our intention is to illustrate the range of diversity of aerosol particles in central TP and to provide the insight of aerosol properties for better assessments of aerosol radiative forcing.

METHODOLOGY

Study Site and Sampling Description

Nam Co is the largest lake in Tibet as well as the highest great lake in the world. On account of the harsh climate, the local population in the Nam Co region is about one thousand. The surrounding areas are covered by grassland with Kobresi pygmae as the predominant vegetation form, which are widely distributed on the alpine meadow/peat soil. Therefore there is scarcely atmospheric pollutant emission in the vicinity. In 2005, Nam Co Monitoring and Research Station for Multispheric Interactions (N30°46.44′, E90°59.31′, 4730 m a.s.l.) was established.

According to the records of automatic weather station (AWS), the annual average of temperature at Nam Co is 0°C with a large seasonal and daily variability. The surface weather conditions of each sampling day are tabulated in Table 1. Generally, there are four distinct seasons, winter (December to February), spring

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Table 1. Summary of sampling date and surface weather conditions at Nam Co Station.

Sampling date	Temperature (°C)	Relative Humidity (%)	Wind speed (m/s)	Air pressure (hPa)
1 Aug. 2006	8.49	63.53	2.23	575.7
9 Sep. 2006	6.16	75.84	1.90	575.4
8 Oct. 2006	-2.47	56.06	2.16	576.8
9 Nov. 2006	-10.83	70.56	1.81	576.1
1 Dec. 2006	-13.64	53.63	1.46	567.9
6 Jan. 2007	-4.70	42.34	2.83	567.7
3 Feb. 2007	-10.78	50.06	1.30	569.5
5 Mar. 2007	-3.68	46.78	3.14	565.8
7 Apr. 2007	2.76	41.86	3.63	570.2
1 May 2007	1.94	36.51	2.28	576.8
8 Jun. 2007	7.30	49.96	1.32	573.6
3 Jul. 2007	8.77	73.18	2.70	571.7

(March to May), summer (June to September) and autumn (October to November). In spring, the dust episode occasionally impacts Nam Co from the upwind arid and semi-arid regions such as the upper reach of Yalu Zangbu River. In summer, Nam Co is under the influence of the Indian Monsoon. Therefore, it is characterized by relatively higher temperature and humid weather and accounts for about 70% of annual precipitation (You *et al.*, 2007). Autumn is generally characterized by cloudless sky and lower precipitation. Winter is very cold and the surrounding area is covered by snow.

From August 2006 to July 2007, total 12 total suspended particle (TSP) samples were collected on polycarbonate filters (Nuclepore, Costar Corp. 0.4 μm pore size) in the beginning of each month. The sampling was performed on the flat roof of the monitoring house at Nam Co Station, about 4 m above ground level. The filter holder was protected with a rain cover and connected to a vacuum pump at an average flow rate of 16.7 L/min. The electric current to run the pump was supplied by a solar panel system. The pump was equipped with an automatic air mass flow meter (Taihe Automation Control and Instrument Corp.), calibrated with an accuracy of 1.5%. The duration of each TSP sampling was set to 24 hours. The air volume was converted into standard condition according to the ambient conditions in the Nam Co region. TSP masses on the filers were determined gravimetrically (AUW220D, Shimadzu), before and after the sample collection. And then the TSP mass concentrations were calculated according to mass weighed and total air volume.

Experiment

Individual aerosol particles were analyzed manually using scanning electron microscopy coupled with energy dispersive X-ray analysis (PHILIPS XL 30 FEG, Hitachi S-2700). X-ray analysis was carried out with an energy-dispersive Si (Li) detector Model Saphire, SUTW (super ultrathin window), allowing chemical elemental analysis ($Z > 11$). Prior to the analysis, one eighth of total filter was cut and Au/Pd coated. Approximately 150 particles were analyzed on each sample to examine particle morphology, size (equivalent projected area diameter) and elemental composition. Number of particles analyzed per sample was similar to other studies where authors used manual mode (Mogo *et al.*, 2005). The EDX spectrum was obtained by the irradiation of electron beams at the center of individual particles with accelerating voltage 10 kV and a counting time 60-100 s. The intensities of the characteristic X-ray lines were converted to the corresponding elemental concentration by the standardless ZAF correction method.

The optical properties of aerosol such as aerosol optical depth (AOD) and Angstrom parameters (α) were measured by CIMEL Sun photometer following the procedure of ANRONET (http://aeronet.gsfc.nasa.gov/). The sun photometer makes direct solar radiation measurements with a 1.28 full field of view every 15 min at 340, 380, 440, 500, 675, 870, 940, and 1020 nm. These direct solar extinction measurements are used to compute AOD at each wavelength. The instrument calibration and derivation of AOD and α are discussed in detail by Holben *et al.* (2001).

RESULTS AND DISCUSSION

Aerosol Mass Concentration

The mass concentration of aerosol is a key criteria for the assessment of air quality. The range of TSP concentrations measured at Nam Co was 0.48-36.11 μg/m^3 with the annual average of 6.74 μg/m^3, reflecting a typical background level. These values are generally comparable to those observed at other remote sites (Zhang et *al.*, 2008). For example, Mazzera *et al.* (2001) reported the average PM_{10} mass concentration at McMurdo (Hut Point), Antarctic during 1995/96 and 1996/97 austral summers was 3.4 μg/m^3 with the maximum of 7.5 μg/m^3. In our previous work, TSP concentrations at an extremely high elevation site (6520 m) on Mt. Qomolangma ranged from 2.02 μg/m^3 to 6.34 μg/m^3 (Cong *et al.*, 2009). Fig. 1 shows the temporal variation of TSP concentration at Nam Co. The most striking is that TSP values in spring (especially April and May) are significantly higher than other seasons. In spring, the snow layer covering on TP begins to melt, and more ground surface is exposed to the air accompanied more windy days (Table 1), resulting in considerable amount of dust emitted into the atmosphere.

Major Aerosol Particle Types

On the base of the morphology and chemical composition determined by SEM-EDX, totally 1800 particles on 12 filters were classified into 7 groups (Table 2). The size of particle analyzed in this study ranges from 0.5 to 10 μm. Then their characteristics and possible sources were discussed.

Soot

Soot, also known as black carbon, is clearly distinguishable from other aerosol types due toits unique morphology (aggregates of 20-50 nm spherules, Fig. 2(a)). Soot, as an important particle pollutant in air, is primarily emitted from biomass burning and incomplete fossil fuel combustion (Li *et al.*, 2003; Posfai *et al.*, 2003; Cao *et al.*, 2007). The morphology of this kind of carbonaceous particle varied from short chains to complex clusters, which depend on fuels, burning conditions and atmospheric processing. At Nam Co, all soot particles present in fine mode (< 2.5 μm, Table 3). The elemental composition of soot was predominant carbon with minor sulfur.

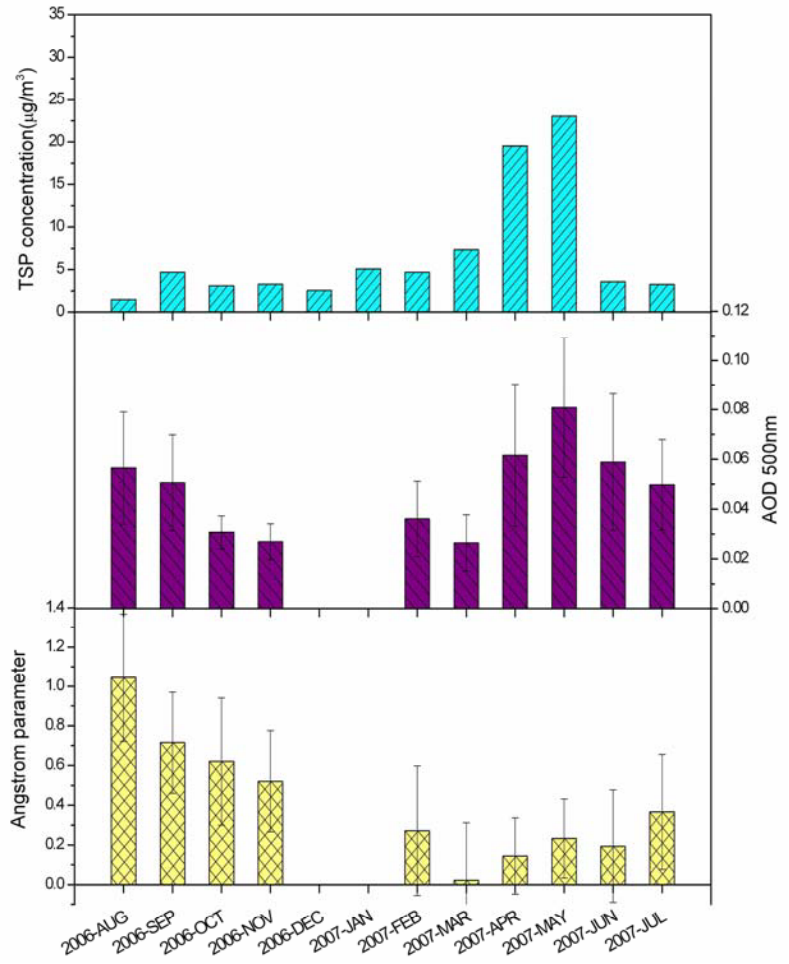


Fig. 1. Temporal variation of total suspended particle concentrations, aerosol optical depth (AOD) and Angstrom parameter.

Table 2. Description of the 7 aerosol groups used in the data analysis and the major selection criteria.

Particle group	Elemental associations	Typical morphology
soot	Abundant C, often with minor S	Chain aggregations of carbonaceous spherules
tar ball	Abundant C	Individual carbonaceous spherules
alumosilicates/Quartz	Al + Si > 60%, with minor Na, Mg, K, Ca, or Fe	Irregular shape
calcium sulfate	Ca > 30% and S > 30%	Irregular shape
Ca/Mg carbonate	Ca > 60% or (Ca + Mg) > 60%	Irregular shape
Fe/Ti oxide	Fe > 70% or Ti > 70%	Irregular shape
biological particle	Abundant C with minor elements (Ca, Na, Mg, P or K)	Variable morphology

Tar ball

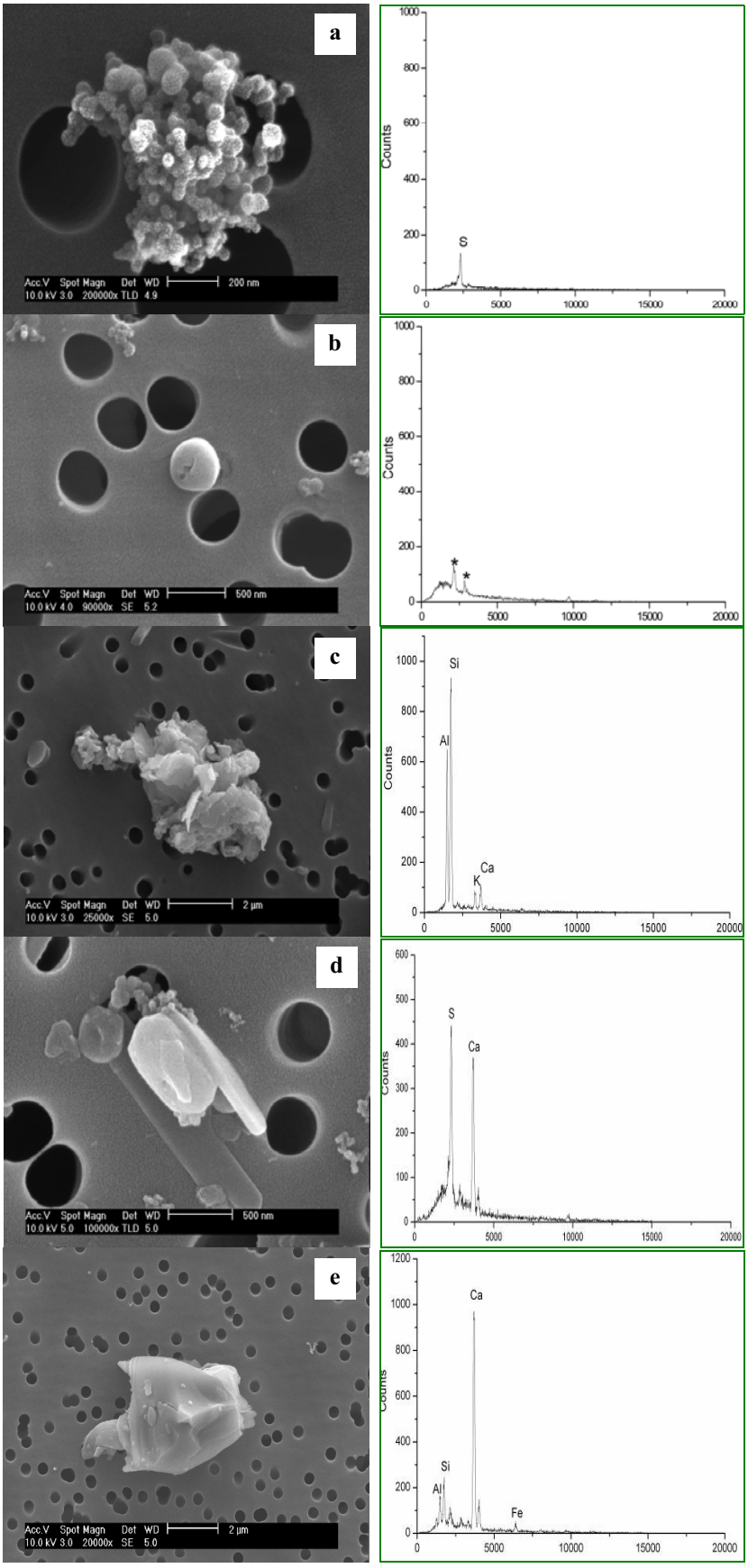
Tar ball is a distinct carbonaceous particle type from soot (Fig. 2(b)). It is spherical, amorphous, and typically not aggregated with other particles (Posfai *et al.*, 2004). The composition of the tar ball observed in our study was dominant by C and O. The size of tar ball generally ranged from 200 to 600 nm (Table 3). Tar ball is produced from biomass and biofuel burning. The further investigation shows tar ball can scatter and absorb light efficiently and then may play an important role in climate forcing (Hand *et al.*, 2005).

Aluminosilicates/Quartz

The aluminosilicates are characterized by high contents of Si and Al with varying Na, K, Fe, S and/or Ca. Particles containing predominant silicon are classified as quartz. Most particles in this group showed irregular shapes (Fig. 2(c)). From mineralogy, aluminosilicates are mainly made up of kaolinite, illinite, montmorillonite and feldspars, which are typical terrigenous minerals (Shao *et al.*, 2007). Aluminosilicates and silicates account for more than 25% of the mineral species. The major types of chemical compounds in the earth's crust consist approximately 75% of silicates and aluminosilicates in terms of weight (van Malderen *et al.*, 1996). Therefore, the most evident sources for this particle type would be soil related.

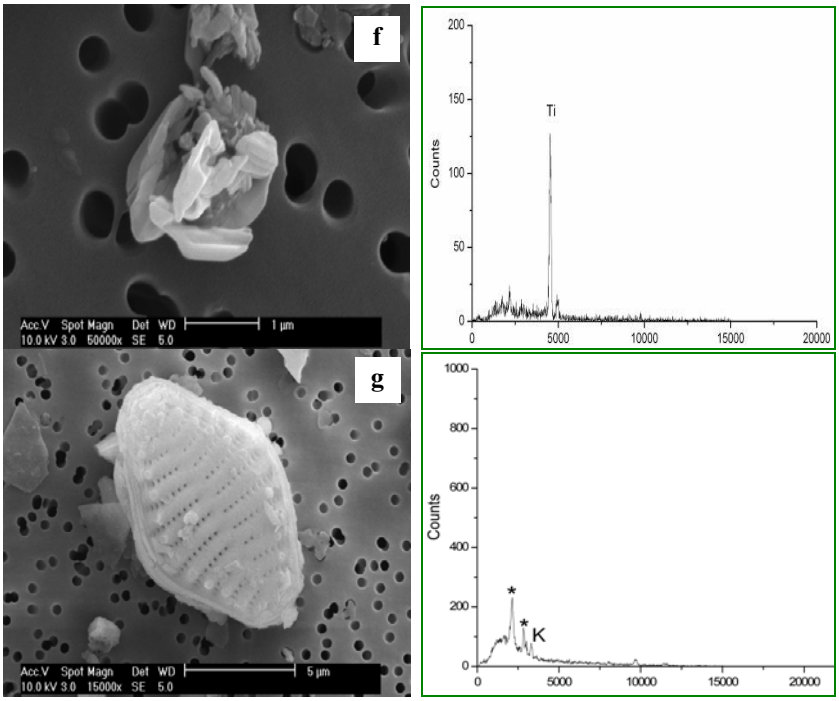
Calcium Sulfate

Particles composed of high and approximately equal abundance of Ca and S fall into this group. In this study, calcium sulfates mostly have columnar appearance (Fig. 2(d)). Gypsum originates generally from diverse sources (Zhang and Iwasaka, 1999). The



Note: * means the background signal from Au/Pd coating.

Fig. 2. Typical morphology and EDX spectra of seven particle groups.



Note: * means the background signal from Au/Pd coating.

Fig. 2. *(continued).*

Table 3. Size-resolved number of aerosol particles in different groups

Seasons	Spring			Summer			Autumn			Winter		
diameter (μm)	0.5-1	1-2.5	2.5-10	0.5-1	1-2.5	2.5-10	0.5-1	1-2.5	2.5-10	0.5-1	1-2.5	2.5-10
soot	31	42	0	36	78	0	24	66	0	18	30	0
tar ball	29	0	0	162	0	0	78	0	0	36	0	0
aluminosilicates/quartz	36	156	174	30	108	54	42	114	156	42	144	192
calcium sulfate	6	18	6	18	18	6	12	24	6	12	12	24
Ca/Mg carbonate	0	18	30	0	6	24	12	6	18	6	18	18
Fe/Ti oxide	0	24	12	0	6	12	0	6	0	0	24	6
biological particle	0	0	6	0	6	42	0	6	36	0	6	6
Sum	102	258	228	246	222	138	168	222	216	114	234	246

Note: the ultra-fine particles with diameter less than 0.5 μm are not analyzed.

primary source is crustal material because gypsum is a typical mineral in the earth crust (Hoornaert *et al.*, 1996). The other source is conversion of calcite to gypsum by chemical reactions with sulfuric acid or ammonium sulfate during transport (Liu *et al.*, 2005).

Ca/Mg Carbonate

This type of aerosol is characterized by high relative X-ray intensities for Ca and/or Mg (Fig. 2(e)). The size of Ca/Mg carbonate particle varied, ranging from 0.5 to 10 μm (Table 3). According to the irregular shape and coarse grain size, those Ca/Mg carbonate particles were assumed to be soil-related. Carbonate minerals including calcite ($CaCO_3$) and dolomite ($CaMg(CO_3)_2$) are common constituent of soils, and often observed in the individual aerosol particle analysis (Shao *et al.*, 2008).

Fe/Ti Oxide

Particles abundant in Fe or Ti element are classified into this group (Fig. 2(f)). Fe or Ti oxide particles have natural and anthropogenic origins. However, those metal oxide particles emitted from metallurgic industry usually present as spherical shape. In our study, according to their irregular morphology, Fe and Ti oxides from crustal materials (hematite, magnetite, goethite, rutile, etc) were most likely.

Biological Particle

Biological particles are recognized from their characteristic morphology (Fig 2(g)) with the size larger than 5 μm. Usually, such type particles have minor amounts of elements (Na, Mg, K, Ca, and P) which are essential tracers present in plants. Such biological particles were attributed to the likely occurrence of pollen, spores, bacteria, algae, parotozoa, fungi, fragments of leaves, as well as the excrements and fragments of insect.

Seasonality of Number Abundances for Particle Groups

The relative abundances of each particle group in different size fractions are shown in Fig. 3. According to the definition of different season, the samples used for spring, summer, autumn and winter are 3, 3, 4 and 2, respectively. Clearly,

aluminosilicates was predominant in all seasons, indicating the crustal sources were always the most important contributor. In the previous study, the elemental composition of aerosols in Nam Co also mainly consisted of Si, Ca, Fe, Al and K which originated from crust material (Cong *et al.*, 2007). For summer, the most remarkable characteristic was the significant increase of soot and tar ball's relative abundances (the sum up to 46%). The autumn took the second place in terms of soot and tar ball content, while the winter and spring exhibited the lowest. The seasonal variations of soot and tar ball content suggested the Nam Co area possibly received more anthropogenic influence in summer than other seasons. Moreover, the biological particles in the summer and autumn (6-8%) were slightly more than winter and spring, reflecting the increased contribution of vegetation on the land surface. While the number abundances of other aerosol type (i.e. calcium sulfate, Ca/Mg carbonate and Fe/Ti-rich particle) generally kept constant through the whole year. Furthermore, considering the large amount of soot and tar ball particles in summer, which could lead to considerable local climatic warming, more attention should be devoted to their potential contribution to the glacier retreat in Nam Co region.

Comparison with Sun Photometer Measurements

Aerosol optical depth and Angstrom parameter retrieved from the sun photometer (CIMEL 318) are shown in Fig. 1. The mean value of AOD for the whole year (0.05 at 500 nm) is comparable to other background sites such as Mauna Loa (Holben *et al.*, 2001). Very low aerosol optical values in Nam Co were consistent with the result of mass concentration of TSP. Actually, AOD and mass concentration revealed the clear atmospheric environment (low loading of aerosols, Fig. 1) of Nam Co from different aspects.

The seasonal variation of the monthly average AOD showed maximum values in the spring (April and May) and remained high values in summer monsoon season (June, July, August). This seasonal peak in spring was thought to be substantial dust particles. The high values of AOD in summer were possibly caused by the enhanced anthropogenic emissions.

The high aerosol loading in spring accompanied with low α indicated that dust dominated the aerosol regime associated with loose ground surface and subsequent more soil dust in the aerosphere. In contrast, high AOD associated with high α in summer could be attributed to larger proportion of fine anthropogenic aerosols. The SEM-EDX results showed the dominant aerosol particles were aluminosilicates in all seasons, which are typical crustal minerals and mainly exist in coarse mode (Table 3). Especially in spring, the relative number concentration of aluminosilicates was about 61%. While insummer, the percentage of aluminosilicates decreased to 29% with increased amount of soot and tar ball (19% and 27%). Soot and tar ball are produced mainly from anthropogenic activities like fossil fuel and biomass burning and usually present in fine mode. Therefore, the result of aerosol optical properties and SEM-EDX support and verify each other, providing a reliable understanding of aerosol characteristics at Nam Co.

Back Trajectories Analysis

To analyze the source and transport pathways of the air masses arriving at Nam Co, the back trajectory analysis was done for the period of measurements using the HYSPLIT model, developed by NOAA/ARL (Draxler and Rolph, 2003). In this study, five-day back trajectories were calculated at a height of 500 m above ground level at 08:00 UTC for all sampling days. The trajectories were generated using GDAS (Global Data Assimilation

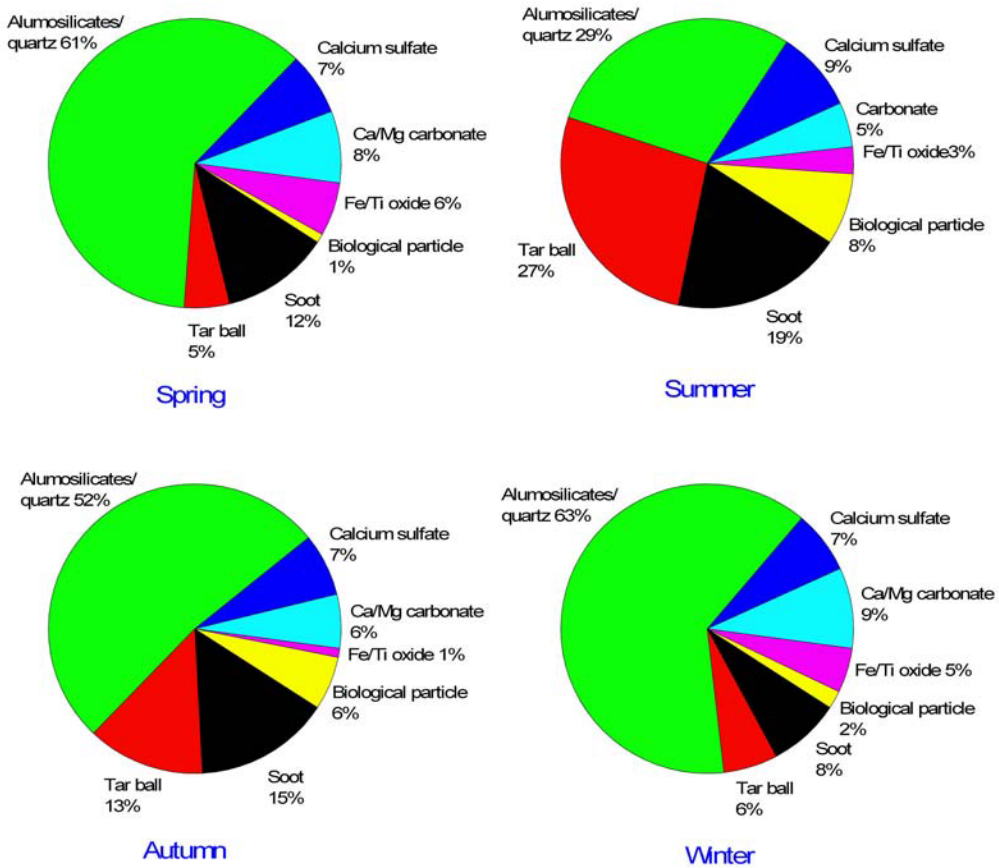


Fig. 3. Seasonality of number abundances for particle groups.

System) meteorological archive from NCEP (National Center for Environmental Prediction). The error accompanying HYSPLIT-generated trajectories were estimated to be anywhere from 15% to 30% of the travel distance, and uncertainty increases with the distance of transport (Stohl, 1998).

The trajectories (Fig. 4) reveal two different types of air mass arriving at the sampling site. In summer (June, July and August), slow-moving air masses come from Bangladesh and Northeast India at low altitude. In other seasons, most of air masses come from the west with a rapid moving speed. Such two kinds of air mass pathway generally correspond to the summer monsoon and westerly system over TP. Namely, in summer monsoon period, the low pressure over TP induces warm air from the Indian Oceans (especially Bay of Bengal) transport to the hinterland of the plateau. While in other time, the large scale atmospheric circulation patterns over TP were mainly dominated by westerlies.

Previous researches have suggested that the summer monsoon circulation can bring considerable pollution from South Asia. While air masses belonging to westerly flow do not carry large quantities of pollutants because they originate and travel far away from the industrialized and heavily populated regions (Duan *et al.*, 2007). The individual particle analysis in this study provides more data on this assumption. Obviously higher proportions of soot and tar ball, typical indicators of anthropogenic influence, were found in summer. Therefore, it could be deduced that the air mass from South Asia in summer may carry such anthropogenic particles. Atmospheric pollution is serious in South Asia (Salam *et al.*, 2003; Salam *et al.*, 2008). Model simulation has pointed out that those atmospheric pollution from South Asia could be lifted and transported to TP (Li *et al.*, 2005). Our previous analysis on elemental composition of aerosols from Nam Co also suggested that several heavy metals may be transported long distances from South Asia (Cong *et al.*, 2007). On the other hand, the possibility that some pollutants from areas inside the TP also exist. For example, the previous studies concerning emissions in Lhasa have suggested that soot particles generated due to the religious activities in the urban area could be transported out of the city, which may impact the atmospheric mass cycle in wide ranges (Zhang *et al.*, 2001).

SUMMARY AND CONCLUSIONS

Aerosols at Nam Co, a remote station in central Tibetan Plateau were characterized at the individual particle level. According to the morphology and chemical composition, aerosol particles could be generally classified into 7 groups: soot, tar ball, aluminosilicates/ quartz, calcium sulfate, Ca/Mg carbonate, Fe/Ti oxide, and biological particle.

The seasonal variation of the abundances of the different particle types was further investigated. In all of seasons, the aluminosilicates/quartz was the dominate aerosol type, showing that crustal material was the primary contributor of the aerosols. The relative abundances of soot and tar ball that all occur in fine mode increased significantly in summer, reflecting Nam Co received more influence of anthropogenic activities at that time.

By comparison of SEM-EDX results to aerosol optical properties (such as AOD and Angstrom parameter) obtained by sun photometer, it was clearly demonstrated that the individual particle analysis was meaningful to interpret the temporal variation of aerosol optical properties, providing complementary information on aerosol characteristics at Nam Co.

The backward trajectories reveal two different types of air masses arriving at Nam Co in the whole year. Furthermore, the individual particle analysis result shows that such two types of air masses correspond to different aerosol composition. The summer monsoon circulation may bring considerable pollution from South Asia, while the westerly flow dominated in winter is relative clean. More work is needed to figure out the definite source of those aerosols.

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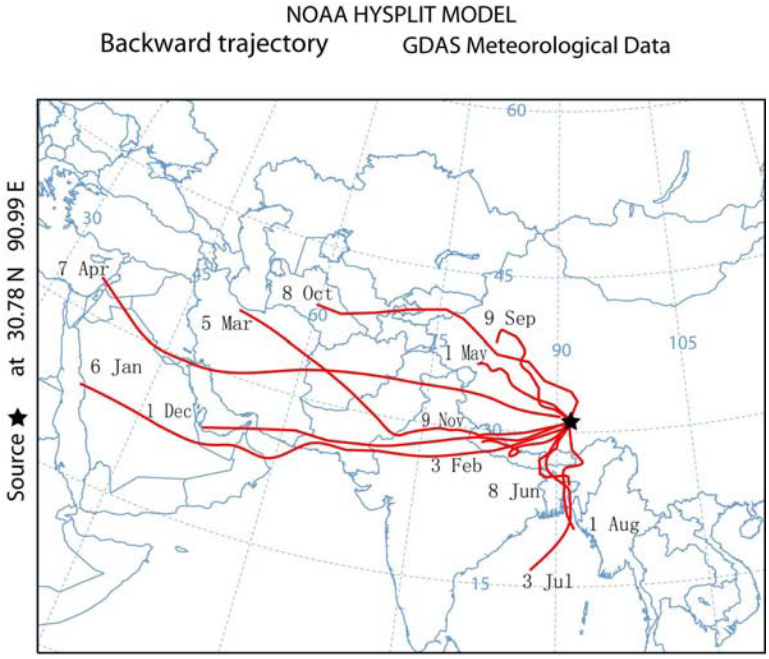

Fig. 4. Backward trajectories arriving at Nam Co in central TP for 12 sampling cases.

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