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Soot particles and their impacts on the mass cycle in the Tibetan atmosphere

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Abstract

Atmospheric aerosol particles in urban and mountain areas around Lhasa city (29.65°N, 91.13°E) in the Tibetan Plateau were collected in the summers of 1998 and 1999. The particles were analyzed with electron microscopes and an energy dispersive X-ray spectrometer. Individual particle morphology, elemental composition and mixture of sulfate and nitrate were investigated. In the urban area, soot particles emitted from vegetation burning were dominant. These particles were characterized by chain or aggregate morphologies, and an elemental composition of potassium and sulfur. Such particles were frequently detected in mountain areas out of the city, where they formed droplets acting as condensation nuclei. Quantitative estimation indicated that sulfur was accumulated onto the soot particles during their dispersion from the urban area to mountain areas. Sulfate and nitrate detections indicated that soot particles collected in the urban area did not contain nitrate and BaCl₂-reactive sulfate, which revealed that the combination of sulfur and potassium in the particles was not K₂SO₄. In contrast, the particles dispersed to mountain areas contained BaCl₂-reactive sulfate and some contained nitrate, suggesting that soot particles emitted from the urban area could increase the buffering capacity of aerosol particles and enhance the formation of particulate sulfate through heterogeneous conversion in the Tibetan atmosphere. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Tibet; Vegetation burning; Soot; Transport

1. Introduction

Soot particles emitted from biomass burning have attracted more attention recently since they could impact the budget of solar radiation through the absorption of visible light (Penner et al., 1992; Myhre et al., 1998) and modify cloud processes through acting as condensation nuclei (Crutzen and Andreae, 1992). They also actively participate in the atmospheric mass cycle through direct reactions with other species or providing reaction sites, since the surface structure of soot particles make them efficient absorbers for various molecules (Sheridan et al., 1992; Parungo et al., 1994;

Yamasoe et al., 2000; Liu et al., 2000). Due to their small density, soot particles are easily dispersed to areas far away from their sources and this extends their effects over large regions. Soot particles found in the upper troposphere over Wyoming, USA, in spring 1991 were most likely from the oil fires in Kuwait (Sheridan et al., 1992) and those found in middle Atlantic were from the continental origin (Andreae, 1983).

In summer 1998, we collected aerosol particles around Lhasa, the capital city of the Tibet Autonomous Region of China. The subsequent analysis indicated that the aerosols at Lhasa were mainly soot particles emitted from the urban area and many such particles could be dispersed out of the city (Zhang et al., 2000). This observation was a part of a project that was performed to explore the mechanism of the “Tibet Ozone Valley”, a

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phenomenon of a region of low ozone concentration over the Tibetan Plateau in summer (Zhou and Luo, 1994; Zou, 1996), and we focused on the clarification of the category of aerosol particles, the evaluation of the contributions of different sources, and the assessment of the roles of aerosol particles in local and regional atmospheric mass cycles. Recent studies have indicated that ozone depletion in some cases might be due to soot particles in the free troposphere (Tsutsumi et al., 1999; Bhugwant et al., 2000), although some other works suggested that the ozone depletion associated with soot particles was negligible (Kamm et al., 1999; Jacob, 2000; Disselkamp et al., 2000). Soot particles possibly react with ozone directly or provide sites for ozone to react with other species leading to ozone destruction (Smith and Chughtai, 1996, 1997; Lary et al., 1997, 1999; Poschl et al., 2000). Therefore, it is important to clarify as to which are the physical and chemical properties of the soot particles found at Lhasa and what happens to them during their dispersion.

In order to further investigate the activities of the soot particles during their dispersion, we collected particles at two sites farther from the city besides the sites of 1998 and 1999. In this paper, results obtained during the observational campaigns of 1998 and 1999 are summarized. The origin and dispersion of the soot particles and their impacts on the Tibetan atmosphere are discussed.

2. Particle collection and analysis

In the urban area, particle collections were carried out on the roof of the Tibet Meteorological Observatory in the central area of Lhasa city. The observatory is about 18 m high above the ground. Out of the city, particles were collected at summits of different mountains in the south from the city. Fig. 1 illustrates the sampling sites' altitudes above sea level, the approximate distances of mountain sites from the city and the sampling dates. Particle

collections were carried out between 12:00 and 16:00 Beijing standard time (8 h prior to GMT) on daytime.

Aerosol particles were collected onto transmission electron microscope (TEM) copper grids (Maxtaform H7) using a two-stage impactor with jet diameters of 1 mm (stage-1) and 0.5 mm (stage-2). The pumping rate of air was 1.21 min^{-1} and the estimated 50% cut-off aerodynamic diameters were 1 and $0.3 \mu\text{m}$, respectively. Airflow passed through stage-1 first and then through stage-2. In August 1998, the largest monthly precipitation during the last 30 yr was recorded and the relative humidity was higher than the usual. For this reason, a diffusion dryer was applied to dry the inlet air for particle collection at that time. Tests at normal atmospheric conditions revealed that the diffusion dryer could dry the air of 80% relative humidity below to 30%. Sampling grids were covered with polyvinyl formvar [molecular formula: $(-\text{C}_5\text{H}_8\text{O}_2-)_n$] film sprayed by carbon. Samples of such grids were applied for morphological analysis and EDX analysis. In order to detect sulfate and nitrate in the individual particles, some grids were also coated with multiple film of nitron and barium chloride by vacuum deposition before particle collection. For one set of stage-1 and stage-2 grids, particle collection time was 5–10 min in the urban area and 10–20 min in mountain areas. Each grid was kept in a single plastic capsule, which was sealed into a box with silica gel right after particle collection and brought to laboratories for subsequent analysis.

Particles were viewed and photographed using the HS-9 Hitachi TEM of the Institute of Hydrospheric–Atmospheric Sciences of Nagoya University of Japan. Sulfuric acid and partially neutralized sulfate particles, if existed, could be identified on the basis of their distinctive morphology: a central spherical cap surrounded by one or more rings of small satellite droplets (Frank and Lodge, 1967; Bigg et al., 1970). Before the investigation of the particles collected on nitron–barium chloride film, the grids were exposed to octanol vapor for 18 h at room temperature to promote the reactions of

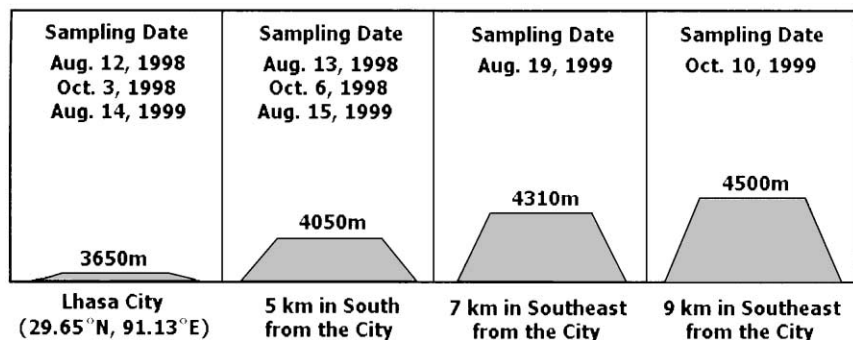


Fig. 1. Illustration of the sampling sites, heights above sea level and sampling dates.

sulfate and/or nitrate with the reagent film. Particles containing sulfate would react with barium chloride and produce barium-sulfate Liesegang rings, and particles containing nitrate would react with nitron and produce nitron–nitrate bundles during the exposure. Reactions of particles containing both sulfate and nitrate with the film would produce both Liesegang rings and bundles. Whether the particles contained sulfate and nitrate could be specified through the morphologies of reaction products around particles. According to the laboratory experiments of Isawa and Ono (1979) and Qian et al. (1991), the detection limits of the reagent film for sulfate and nitrate contained in the individual particles were 10^{-17} and 10^{-14} g, respectively.

Analysis was also carried out by using the Hitachi scanning electron microscope attached with a Horiba energy dispersive X-ray spectrometer (SEMEDX) of Solar Terrestrial Environmental Laboratory of Nagoya University of Japan. The information about particle morphology and elemental composition was obtained. To avoid artificial damage and distortion of particles, we did not coat anything on the samples after particle collection. Consequently, we did not obtain clear SEM pictures of particles smaller than $1\ \mu\text{m}$ diameter although particles smaller than $0.2\ \mu\text{m}$ could be viewed and measured using these instruments. The EDX spectrometer was able to quantify elements with atomic numbers $Z \geq 5$ (nitrogen was undetectable). Carbon in spectra was distorted by the formvar film and, sometimes greatly, by the carbon pedestal for SEM analysis, oxygen was distorted by the formvar film and sodium was disturbed by copper grid (Cu-L_α) slightly. For these reasons, carbon and oxygen will not be mentioned and discussed. The accelerating voltage of the SEM was 20 keV and the filament current was about $112\ \mu\text{A}$. X-ray spectra were generated from squares covering particles and were collected for 100 live seconds with probe current of 0.3 nA. After the spectrum was collected, the detected elements were automatically specified by the EDX equipments. The software Horiba Smedx was applied and the atom number fractions and weight fractions of different elements in individual particles were calculated by means of standard-less method with ZAF correction.

3. Results

Lhasa city is located in a narrow west–east valley in the southern part of the Tibetan Plateau. Its geographic position is (29.65°N , 91.13°E) and its altitude is 3650 m asl. August is the rainy season in Tibet and October is the dry season. Precipitation in August 1998 was much larger than the average level according to historical records. Except in August 1998, the relative humidity at the urban area was around 35%.

The average pressure was 652 hPa. Wind was weak and wind directions were usually eastward along the valley where the city is located. Particle collection was carried out in the urban and mountain areas when the weather was clear.

3.1. Elemental composition

A small number of particles were collected on stage-1 grids in the urban area. Most of them were soil particles characterized by Al, Si with/without Ca, Fe and Mg peaks in their X-ray spectra. Some of the particles were abundant in calcium. Besides, there were a few of particles that contained only potassium and sulfur. As could be expected, soil particles were rarely found in the mountain areas out of the city.

Compared to the particles of stage-1 samples, particles collected on stage-2 grids were much different. A great number was collected at both urban and mountain areas. They were classified into five types according to elemental composition. Type I was K–S particles, characterized with clear potassium and sulfur peaks in their X-ray spectra and no other elements were detected (Fig. 2a). Type II was the particles of potassium, sulfur and silicon characterized with peaks of potassium, sulfur and silicon in their X-ray spectrum (Fig. 2b). Compared to soil particles in which silicon was the main component, potassium and sulfur were main components in Type II particles. It is worth pointing out that the element contained in most Type II particles except S and K was only silicon and a few of the particles contained calcium whereas none contained aluminum. Type III was S particles characterized by sulfur content and no other elements were detected in them (Fig. 2c). Type IV was the particles in which no elements were detected (Fig. 2d). Type V was that which could not be specified as any of the above four types. The number fractions of the five types of particles at different sites and the numbers of the analyzed particles observed in summer 1999 are shown in Fig. 3. Potassium-containing particles were dispersed in large regions from the urban area to mountain areas. The fraction of Type II particles decreased from urban to mountain areas, while the fractions of Type I particles were not much different at different sites compared to Type II particles, suggesting that Type I particles were more easily dispersed in wide ranges. Few Type III particles were detected in the urban atmosphere, and it indicated that the concentration of Type III particles was very small compared to other kinds of particles there but the fractions increased remarkably with the increase of distance from the city. Similarly, the fraction of Type IV particles in the urban area was pretty smaller than those in the mountain areas.

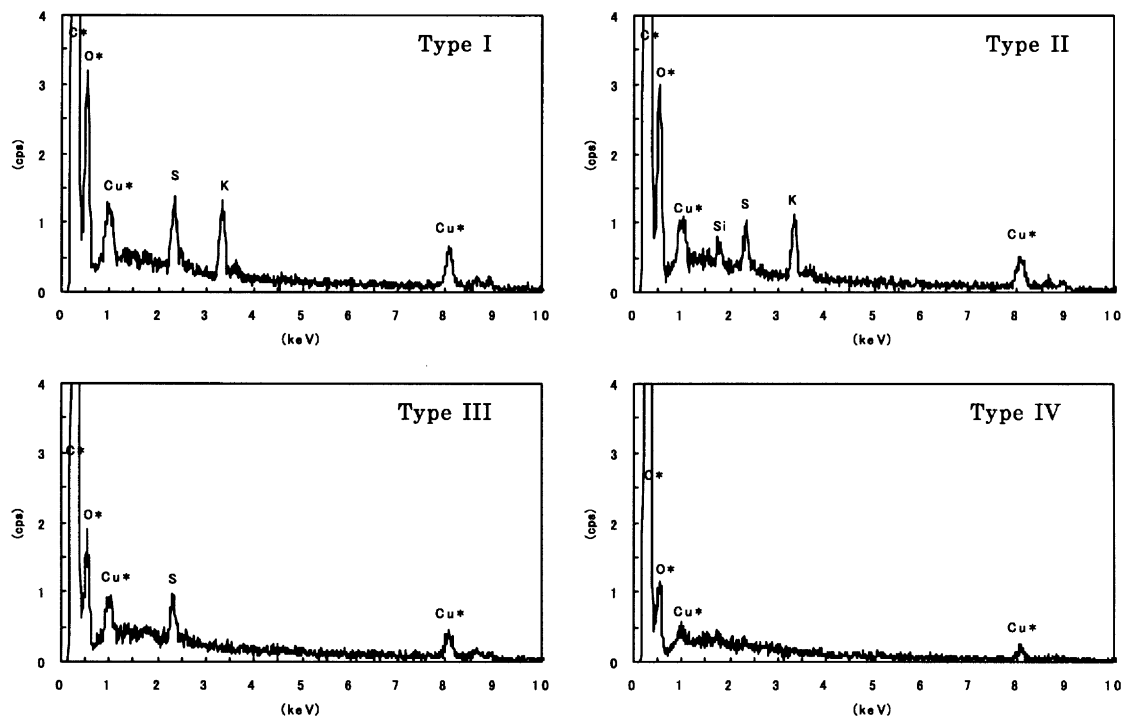


Fig. 2. X-ray spectra of different types of particles.

3.2. Particle morphologies

As is well known, soil particles have irregular shapes and are electron-dense in TEM pictures. In Lhasa, such particles were mainly found on stage-1 grid samples collected in the urban atmosphere and therefore they should not be important in the mass cycle between the urban and mountain atmosphere. Hereafter, we will focus on the particles collected on stage-2 grids.

Fig. 4 shows typical particles collected by stage-2 grids in the urban atmosphere. The predominant particles were aggregates of fragments and chains of small spheres or their mixture. Diameters of the small spheres were from 0.05 to 2 μm . Some particles contained electron-dense large cores. It has been known that such particles were soot particles or fly ashes that were usually generated from biomass burning. For the convenience of description, we will not distinguish soot particles and fly ashes, but apply soot particles to represent the particles of aggregates and chains. Elemental analysis of the particles revealed that most of them were Type I and II particles. Chains and aggregates without electron-dense cores were mainly Type I particles and a few chains were Type IV particles. Those with electron-dense cores were mainly Type II particles. It was found that almost no particles showed the images of former presence of a liquid layer on their

surface and only one particle showed the morphology of partially neutralized sulfuric acid droplet in the urban samples (figure omitted).

Particles collected at the site farthest from the city (4500 m mountain) showed much different morphologies from those of urban samples. Most of the particles were small droplets (Fig. 5a). Some of them had obvious cores. Further magnification of the cores revealed that they were aggregates of chains or small spheres. This indicated that soot particles from anthropogenic sources in the urban area could reach the mountain areas and act as condensation nuclei to form droplets. Elemental analysis of the small droplets indicated that they were mainly Type I or IV particles. In addition, a few particles showed satellite morphologies suggesting that they contained sulfuric acid. However, it seems that they contained a lot of water although there was no indication that they were neutralized. Fig. 5b shows examples of such particles.

Particles collected at the 4050 and 4310 m sites showed intermediate features between those of the urban and 4500 m sites. Soot particles of chains and aggregates of small spheres without a liquid layer were frequently detected and droplets with soot aggregates as their condensation nuclei also appeared on the sampling grids. Moreover, a few of partial neutralized sulfuric acid particles were detected and some of them contained

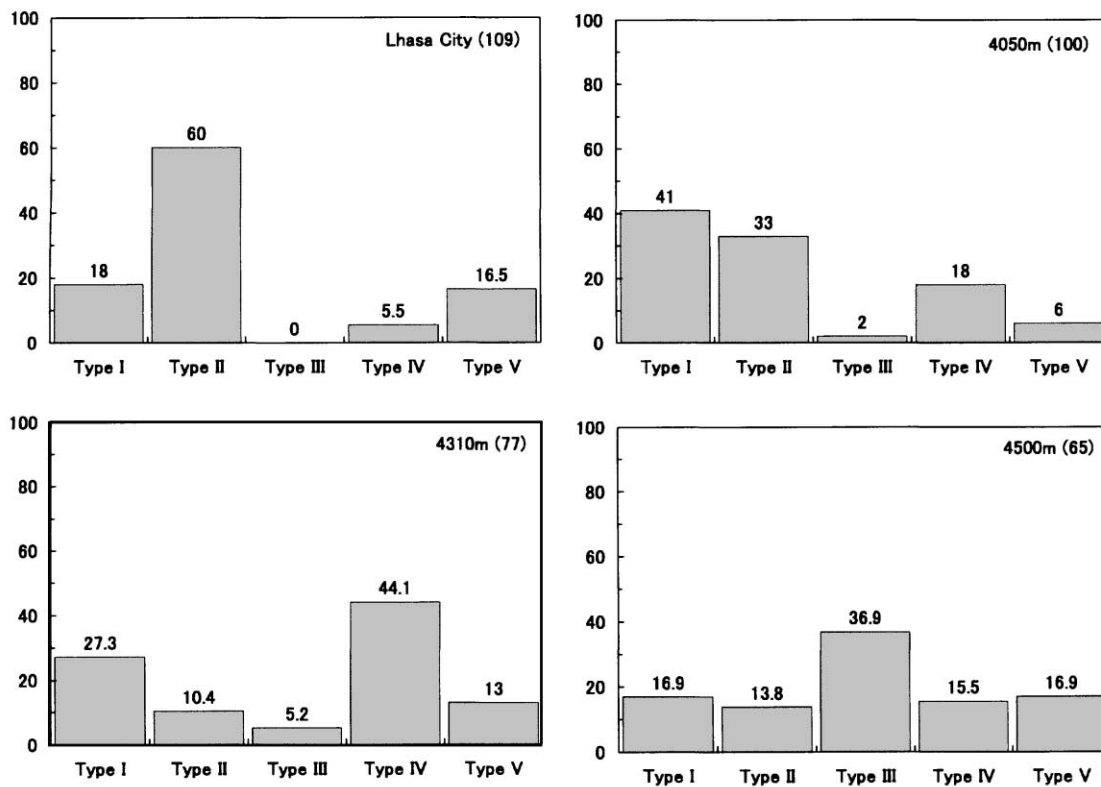


Fig. 3. Number fractions of different types of particles at different sampling sites. Numbers of counted particles are listed in parentheses.

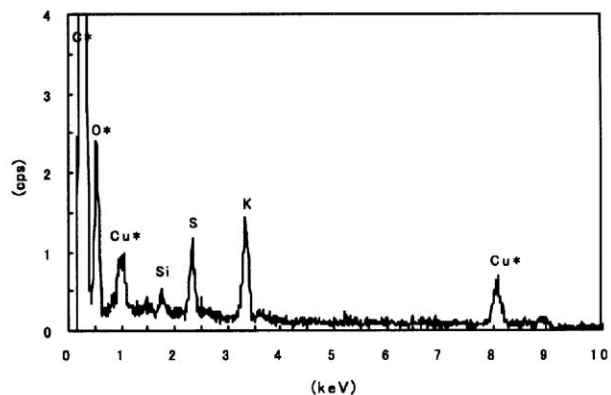
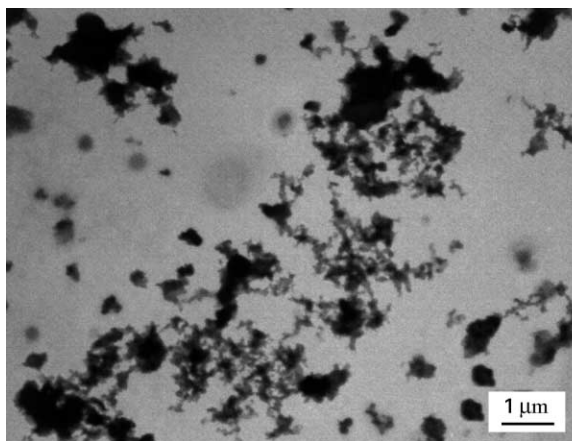


Fig. 4. A TEM picture of particles collected on carbon film by stage-2 grids at the urban area on August 12, 1998, and an X-ray spectrum of a typical particle with diameter smaller than $0.3\mu\text{m}$.

soot particles (Fig. 6). This indicated that sulfuric acid had been formed or absorbed on the surface of the soot particles and these processes could transfer soot particles into those with acidic surface.

3.3. Sulfate and nitrate in individual particles

Tests of nitron–barium chloride multiple film of urban aerosols revealed that only a few particles reacted with

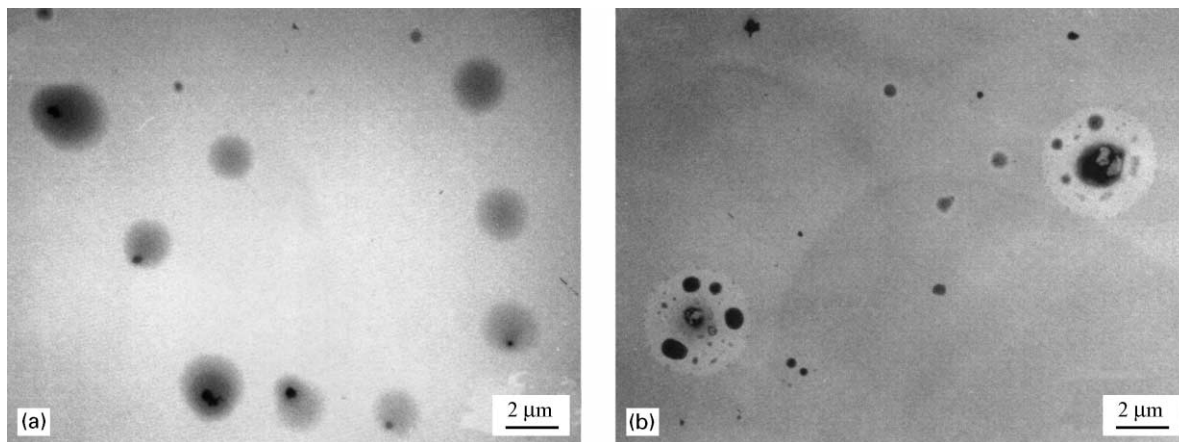


Fig. 5. TEM pictures of particles collected on carbon film at 4500 m on 10 October 1999.

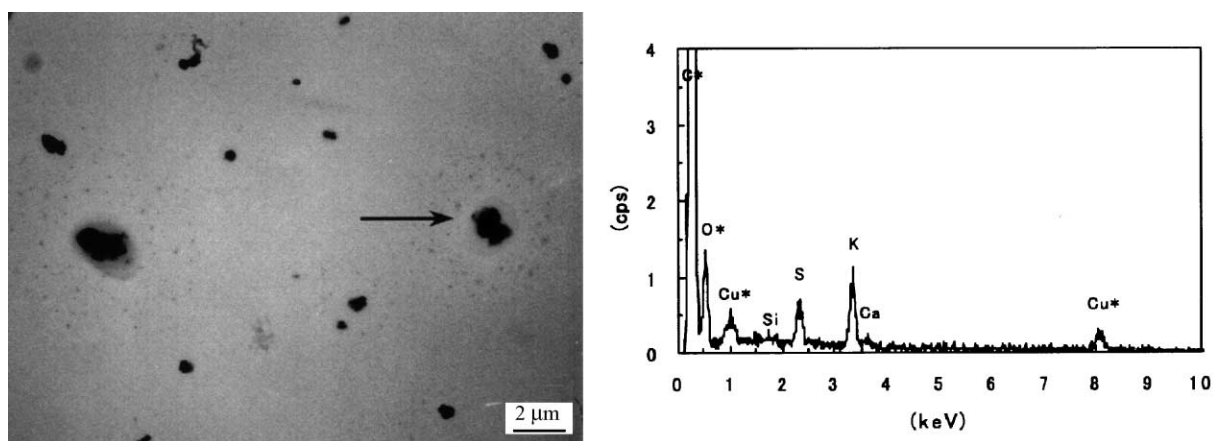


Fig. 6. A TEM picture of partial neutralized sulfuric acid particles on carbon film and the X-ray spectrum of the arrow-marked particle. The particle was collected at 4310 m on 19 August 1999.

the film. Most chains and aggregates were not much different from those on carbon film. Fig. 7a shows the typical image of non-reactive particles on a reagent film. Since about 78% of particles collected on the stage-2 grids contained sulfur (Fig. 2), it was concluded that Type I and II particles in the urban atmosphere of Lhasa did not contain BaCl₂-reactive sulfate on their surface. Therefore, sulfur in Type I and II particles was not from sulfur deposition in the atmosphere. It was in the particles when the particles were generated and did not exist as potassium sulfate (K₂SO₄). Nitrate was rarely detected except some particles larger than 1 μm diameter (figure omitted).

The mixture of sulfate and nitrate particles collected at 4050 and 4310 m sites showed similar characteristics. Figs. 7b and c show particles on the reagent film at the 4050 m site in 1998 and 1999. Many particles had

reacted with barium chloride. Some reactions left clear Liesegang rings while smaller ones left only reacting vestiges. It is noted that some reactions produced needle-like products besides Liesegang rings, suggesting that those particles contained both sulfate and nitrate. Such reaction spots in samples of 1998 were clearer than 1999.

Particles collected at the 4500 m site showed different characteristics from other sites (Fig. 7d). Most of the particles produced rings on the film. However, the rings looked different from Liesegang rings and were more likely to be formed by the coagulation of droplets after colliding with the film. For this reason, we could not confirm if these particles contained sulfate, although some other particles seemed to have reacted with the film slightly (particles marked by arrows in the picture). Estimation of number fraction indicated that the droplet particles occupied more than 50% of total particles.

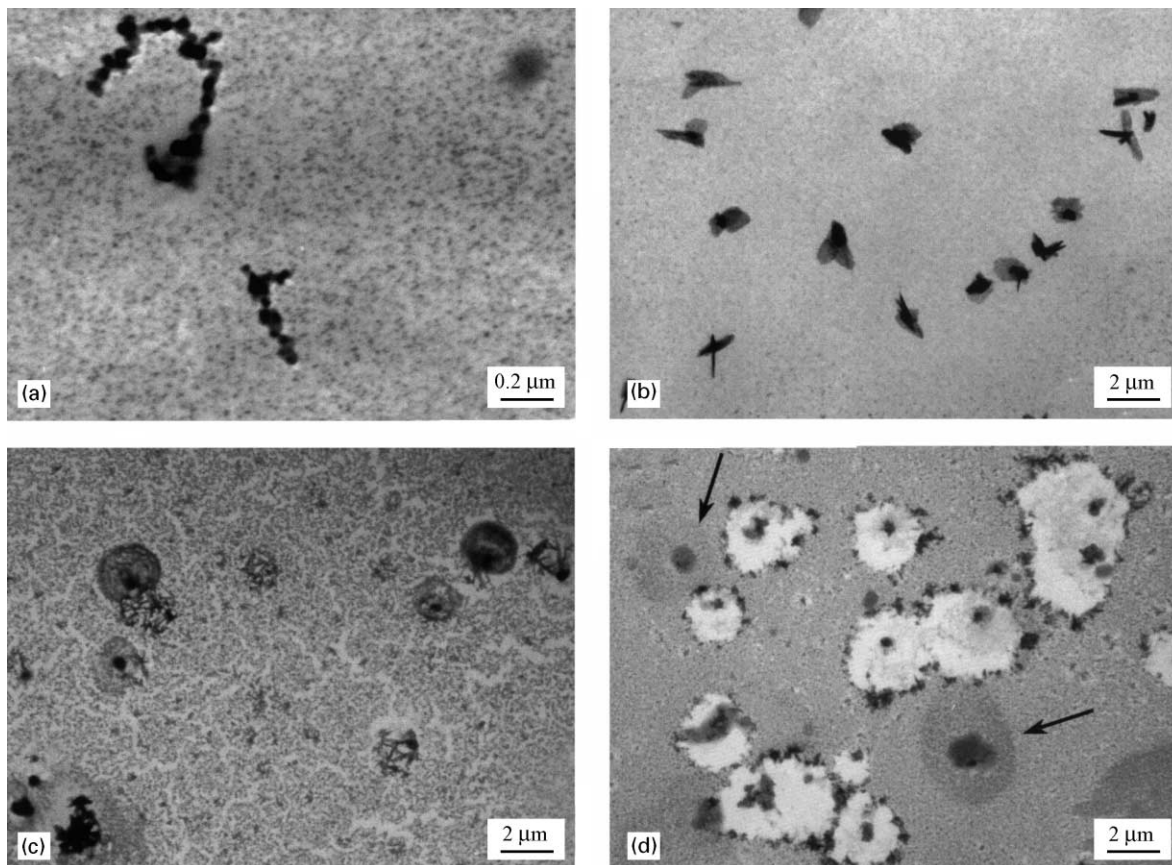


Fig. 7. TEM pictures of the typical images of particles on multiple film at different sites: (a) the urban area in 1999; (b) 4050 m in 1998; (c) 4050 m in 1999; and (d) 4500 m in 1999.

In order to investigate the deposition of sulfate onto Type I particles during their dispersion, atom number ratios of sulfur to potassium in the particles were estimated through element analysis. Fig. 8 shows the ratios in total measured Type I particles at different sites. The ratio at the urban area was almost the same as that of 4050 m site and those at the 4310 and 4500 m sites were a little larger. Although the numbers of the analyzed particles were not adequate for undoubted confidence, these results indicated that sulfur was deposited onto particles during their transport from the urban area to mountain areas. But the majority of sulfur in the particles originated from vegetation burning.

4. Discussion

4.1. Generation of different types of particles

Soot particles of small sphere chains and aggregates were usually generated by vegetation burning or fossil

fuel combustion. However, particles of vegetation burning are different from those of fossil fuel combustion. Measurements of the plumes of vegetation fires in Africa, Amazonia in Brazil and the United States indicated that the enrichment of potassium was the unique indicator for soot particles generated from vegetation burning and this indicator could be applied to differentiate the particles from those of fossil fuel combustion (Cachier et al., 1995; Andreae et al., 1988; Cofer et al., 1988). Lhasa's population is 200,000–300,000 and most of the inhabitants are Tibetans living on agriculture and stock raising. Besides using vegetation for cooking and heating, Tibetans burn a kind of flower at street crosses and temples as religious activities every morning. Since there were no other sources of potassium and sulfur besides the vegetation burning around Lhasa city, the particles abundant in potassium and sulfur found in this study should be associated with the burning activities. To examine the particles generated by the burning processes, particle collection was carried out at the roof of Daji Hotel (15 m high from the ground) adjacent to a cross with the inlet air from the

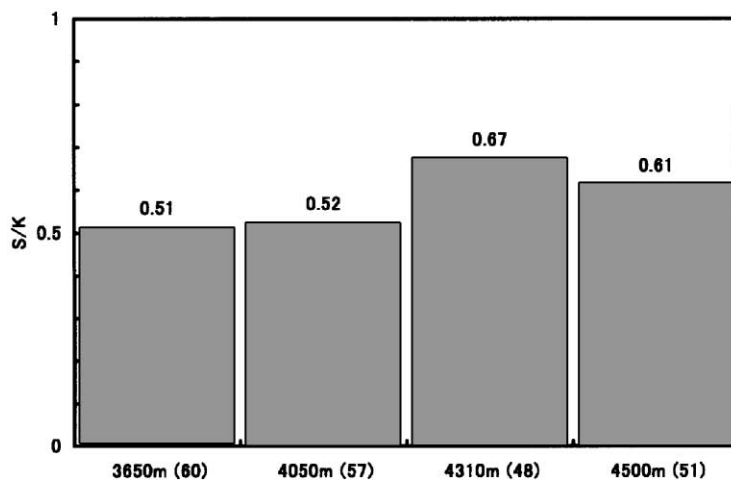


Fig. 8. S/K atom ratios in total detected Type I particles collected at different sites. Numbers of counted particles are listed in parentheses

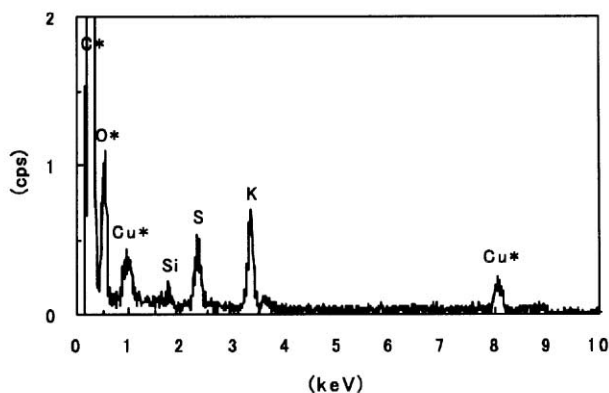
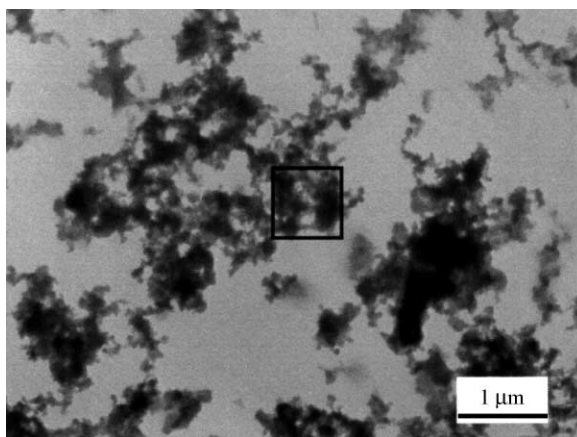


Fig. 9. A TEM picture of particles generated by flower burning and the X-ray spectrum generated from the square area marked in the picture. The particles were collected during 9:25–9:30 a.m. (BST) on 4 October 1998.

direction of the cross when large amount of flowers were burning in several cans during the observation campaign of October 1998. Fig. 9 shows the collected particles and an example of their X-ray spectra. The morphology and composition of these particles were consistent with those of Type I and II particles. Therefore, potassium and sulfur in Type I and II particles in the urban area were due to the resuspension of previous deposition in vegetations and their combination was determined by their state in the vegetations and the burning process. In plant cells, potassium usually exists as K^+ , whereas sulfur exists as organic sulfur compounds. Consequently, the combination of sulfur and potassium in the particles should not be K_2SO_4 although the atom ratio of sulfur to potassium at the urban area was

approximately $\frac{1}{2}$ (Fig. 8). This is consistent with the result of the reagent film detection that the compound of sulfur in soot particles in urban atmosphere could not react with barium chloride.

Type III particles mainly appeared at the 4500 m site and occupied about 37% of the total particles there (Fig. 3). It was noted that more than 50% of particles at the same site were droplets. Therefore, Type III particles, at least most of them, should be small droplets containing sulfur. Since no other elements were detected except sulfur, it was expected that sulfur in the particles existed as SO_4^{2-} , HSO_4^- and/or H_2SO_4 with or without ammonium. SO_2 from natural and anthropogenic sources is always being oxidized into sulfuric acid and sulfuric acid gas, in turn, accumulates to form con-

denensation nuclei by which water droplets are easily formed in the free atmosphere (Seinfeld and Pandis, 1998). Since Type I and II particles of anthropogenic sources were predominant in urban atmosphere and Type III particles originated from the natural source, the number fractions of Type III particles increased with the distance from the urban area (Fig. 3). It could be expected that Type III particles should comprise a significant fraction of aerosol in areas where there are less anthropogenic impacts.

Among Type IV particles, the droplets with soot cores were obviously formed through condensation on soot particles. The formation of those without obvious cores could not be specified. It was unclear if they contained cores that were too small to be detected by the electron microscopes.

4.2. Transport of soot particles generated from the urban area

The result that the number fraction of Type I particles did not decrease with the increase of distance from the urban area suggested that such particles could be frequently transported out of the city. Since soot particles have smaller density compared to soil particles, their lifetime in the atmosphere is longer and they are more easily transported far away from their source areas. Observations in the Atlantic and the Pacific indicated that soot particles originated from continents could be advected to remote marine atmosphere (Andreae, 1983; Parungo et al., 1994; Singh et al., 2000).

Morphology analysis indicated that soot particles which dispersed from the urban area to mountain areas had two main variations. One was that a number of them formed droplets. This is not surprising because soot particles could form droplets as condensational nuclei (Parungo et al., 1994; Lary et al., 1999). Another was that sulfuric acid or partially neutralized sulfuric acid was formed on the particles. Heterogeneous conversion of SO_2 to SO_4^{2-} could occur on soot particle surfaces and the conversion would become more efficient when the particles absorbed water vapor. Such mechanism was also reported by Clarke et al. (1997) and Posfai et al. (1999), who found that soot particles provided effective condensation sites for sulfate under conditions that ranged from extremely clean to heavily polluted marine troposphere. Analysis of soot particles of biomass burning in Africa suggested the deposition of excess sulfur, which was attributed to the deposition of sulfate onto particle surface through heterogeneous reactions (Gaudichet et al., 1995; Liu et al., 2000).

The atom ratios of sulfur to potassium at the urban area and at the 4050 m site were 0.51 and 0.52, respectively. Analysis of integrated aerosol samples in Lhasa city during non-rain periods indicated that the ratio of sulfur atoms to potassium atoms was 0.48 (Tang

et al., 1999), which was consistent with our result. If we assumed 0.51 to be the baseline S/K ratio in soot particles immediately generated from the flower burning and the average ratio in Type I particles at the 4310 and 4500 m sites to be 0.64 (Fig. 8), for every one potassium atom in soot particles 0.13 sulfur atoms could accumulate onto the particles through heterogeneous conversions.

Reagent tests revealed that nitrate was formed on some particles smaller than $1\ \mu\text{m}$ at 4050 m (Fig. 7). Usually, nitrate is hardly formed on small particles due to its volatility (Bassett and Seinfeld, 1984). It was expected that the homogeneous formation of nitrate was impossible in Lhasa areas because acidic sulfate droplets were detected, while ammonia concentration was to be small. A possible explanation of the nitrate in small particles was that it was formed on soot particles through heterogeneous conversions of nitrogen oxides. Partition of large fraction of total nitrate into aerosols was found in Amazonia haze layers (Andreae et al., 1988). However, the major product of NO_2 deposition on soot particles was nitrite and the minor one was nitrate (Santis and Allegrini, 1992; Ammann et al., 1998; Gereck et al., 1998), suggesting that the soot particles might contain more nitrite than nitrate. Unfortunately, we could not learn any information about nitrite in the particles by our present methods.

4.3. Impacts of the soot particles on the Tibetan atmosphere

Our observation results indicated that soot particles generated due to the religious activities in the urban area of Lhasa could be transported out of the city and particulate sulfate and nitrate were formed during their dispersion. It is noted that such religious activities occur not only in Lhasa but also in other cities in the Tibetan Plateau. After analyzing aerosol particles collected around five cities in the Tibetan Plateau, Okada et al. (1999) reported that 20–30% of sulfur-containing particles contained potassium, suggesting that soot particles were emitted into the atmosphere in other cities as in Lhasa. In other words, soot particles of vegetation burning are constantly emitted into the atmosphere from the residence areas of Tibetans.

The Tibetan Plateau is the highest and largest plateau in the world. Asian monsoons occur from May to October every year and the upward flow over the plateau becomes strongest in midsummer (Hahn and Manabe, 1975; Endo et al., 1994; Ueda and Yasunari, 1998). Part of the upward air possibly arrives at 16 km from low level and penetrates the tropopause to enter the low stratosphere (Hahn and Manabe, 1975). These results suggest that the large-scale circulation associated with the Asian monsoon could bring surface air up to the free troposphere and even to the low stratosphere

over the Tibetan Plateau in summer. As a result, soot particles characterized by small density in the surface atmosphere were possibly brought up to high level and even to the low stratosphere. Backward trajectory analysis indicated that soot particles collected in the upper free troposphere in eastern China by balloon observations possibly came from the Tibetan Plateau (K. Okada, MRI of Japan, pers. Comm.). If this is true, then the soot particles generated from biomass burning at the Tibetan Plateau could influence the atmospheric mass cycle not only in the surface air but also in the upper level atmosphere.

Besides the interference in sulfur and nitrogen cycles and the formation of droplets, the soot particles might disturb the ozone distribution in the Tibetan atmosphere. Since the flower was burned in cans in open air, the burning was incomplete and smoldering, which would have emitted particles with large fractions of organic substances (Torok et al., 2000). Analysis of integrated samples revealed high concentrations of poly aromatic hydrocarbon in the urban atmosphere in Lhasa (Tang et al., 1999). The poly aromatic hydrocarbon in soot particles could react actively with ozone (Poschl et al., 2000). Therefore, ozone might be destroyed through the reactions with organic compounds on the soot particles. A similar mechanism was suggested to explain the anti-correlation between ozone mixing ratio and aerosol surface area density observed at the United States (Disselkamp et al., 2000). Recent observations about biomass burning smoke over Indonesia and Australia indicated that the decrease of ozone concentration was closely related to the increase of aerosol particle surface associated with biomass burning in the free troposphere (Tsutsumi et al., 1999). However, many other studies suggested that the decreasing level of ozone associated with soot particles was negligible (Kamm et al., 1999; Jacob, 2000; Disselkamp et al., 2000). Although the vertical distributions of aerosol number concentration obtained by balloon-borne particle counters showed large concentrations of particles right above the tropopause and lidar observations, and radio sondes suggested possible effects of upwelling air containing surface particulate matter in the Tibetan atmosphere during the periods of our observation (Iwasaka et al., 2000a,b), there is no evidence for clarifying if the large concentrations had relations with the soot particles, yet.

5. Summary

Through the analysis of individual aerosol particles around Lhasa city in Tibet, the following conclusions were obtained.

- Due to the religious activities in the urban area, a large amount of soot particles were emitted from vegetation burning at the Tibetan Plateau. These particles were abundant in potassium and sulfur, but the potassium and sulfur did not appear as potassium sulfate. Such particles could be dispersed in wide ranges away from their sources.
- During their wide-range dispersion, soot particles could enhance the formation of particulate sulfate and act as condensation nuclei to form droplets, leading to the increase of sulfate concentration wherever they arrive. The particles also interfere with the mass cycle of nitrogen compounds through the formation of particulate nitrate.

It appears that soot particles from biomass burning in the residence areas could impact the atmospheric mass cycle in wide ranges in the Tibetan Plateau. More details are needed to quantitatively evaluate their roles and carefully investigate their relations with the ozone decrease in the Tibetan atmosphere in summer. Particularly, did the soot particles contribute to the aerosols of high concentration above the tropopause? The answer to this question might provide the explanation of the appearance of the Tibet Ozone Valley.

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