Aerosol mixing states over Kosa source regions : single particle analysis of free tropospheric particles collected with a balloon-borne sampler over Dunhuang, China

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氏　　名	山田　丸
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学位授与の題目	Aerosol mixing states over KOSA source regions: Single particle analysis of free tropospheric particles collected with a balloon-borne sampler over Dunhuang, China （黄砂発生源地域上空のエアロゾル混合状態：中国敦煌にて気球搭載型サンプラーにより採集した自由対流圏粒子個々の分析）
論文審査委員（主査）	岩坂　泰信（自然計測応用研究センター・教授）
論文審査委員（副査）	早川　和一（自然科学研究科・教授），大谷　吉生（自然科学研究科・教授）， 柏谷　健二（自然計測応用研究センター・教授）， 佐藤　　努（自然計測応用研究センター・助教授）

Abstract: Free tropospheric particles were collected using a balloon-borne aerosol sampler on August 29, 2002 and March 24, 2003 at Dunhuang (40°00'N, 94°30'E) in northwestern China, in order to investigate the mixing states of aerosol particles over KOSA source regions. The morphology and elemental composition of the individual particles were analyzed with a scanning electron microscope and an energy dispersive X-ray analyzer. The single-particle analysis revealed that mineral dust particles were major constituents in coarse mode range (d>1 μm), and sulfate particles were predominantly present in fine mode range (0.2<d<1 μm) in the free troposphere over the Taklamakan Desert both in spring and in summer. However, the elemental composition of the individual dust particles significantly differed between the results of August 2002 and March 2003. Si-rich particles were dominant in mineral dust on March 2003. On the other hand, evaporite particles such as Ca- and Na-rich dust particle stood out at the results of August 2002, although Si-rich particles were a main type of mineral dust as usual. Additionally, comparison of the elemental compositions of the particles collected over Dunhuang with those of particles collected over Japan strongly suggests that these particles were significantly modified by sulfur during their long-range transport.

1. Introduction

Asian dust particles (KOSA particles, literally mean "yellow sand" in Japanese) originated in arid and semi-arid regions in the Asian continent are frequently dispersed to eastern China, Korea peninsula, Japan, the North Pacific and even to North America. According to recent many investigations, KOSA particles make important contribution to regional and/or global climate and environment, because the particles can scatter and/or absorb solar radiation, act as chemical reaction site and play as cloud condensation nuclei or ice nuclei in the atmosphere. Moreover, it has been believed that KOSA particles will facilitate oceanic primary productivities, scince Fe contained in KOSA fall into ocean. Therefore the long-range transport of KOSA particles and chemical and physical modification of KOSA particles during their transport become matters of great concern, and

consequently various research campaigns have been made to clarify environmental and climate effects of KOSA.

However, there have been very few observations for behavior of aerosol particles in the free troposphere over KOSA source areas owing to technical difficulties. We therefore have strongly desired observations of the chemical and physical properties for free tropospheric particles and the particle number-size distribution in the free troposphere over the source areas.

2. Observation and analysis

Knowledge about aerosol mixture states in the free troposphere over KOSA source regions is essential to assess the impact on environment change caused by KOSA, because KOSA particles travel long distance mainly through the free troposphere. For that reason, we developed the balloon-borne sampler to collect free tropospheric particles, and we desired several advantage points to the sampler. That is (1) weight saving to allow launching the sampler by rubber balloon, (2) cutting off the rope between balloon and sampler by the rope cutter after finishing the particle collection, and (3) recovery of the sample on the basis of GPS signal. Figure 1 shows the balloon-train for particle collection in the free troposphere and the block diagram of the balloon-borne aerosol sampler.

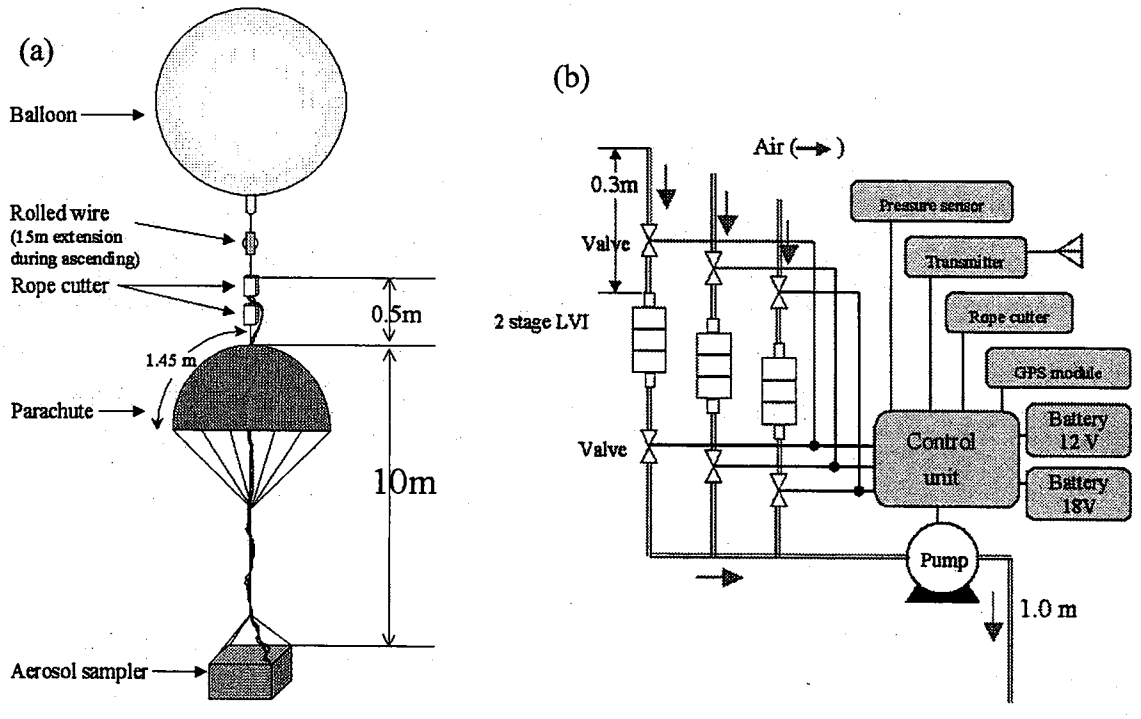

Figure 1. (a) Balloon-train for particle collection in the free troposphere, and (b) block diagram of the balloon-borne aerosol sampler.

We made electron microscopic analysis of the free tropospheric particles over Dunhuang, China (40°00'N, 94°30'E), which is located in the eastern part of the Taklamakan Desert and has been suggested to be an important source of KOSA particles. Their particles were collected in the three different layers—3–5 km, 5–7 km, and 7–9 km above sea level—with a balloon-borne aerosol sampler equipped three low-volume impactor on August 29, 2002 and on March 24, 2003 (Table.1). The recovery system worked very well at that time, and it took only several hours to recover the samples after landing, which seems to avoid the contamination from surrounding air. The distance between balloon launching and landing positions was about 12 km on August 2002, and 48 km on March 200. This kind of measurement is very effective to know the mixing state of free tropospheric aerosols and will bear good ripple effect in the atmospheric science field, although the handling need the technical skill.

Table 1. . Sampling time and weather conditions.

Date	Time (GMT)	Altitude (a.s.l.)	Sampling time	Wind		Weather
				Direction	Speed (m/s)	
29-Aug-02	3:31:12–3:37:12	3.2–5.2 km	6 min	NNE–WNW	5	Fine
	3:37:12–3:42:27	5.2–6.9 km	5.25 min	NW	9.2	
	3:42:27–3:45:27	6.9–8.0 km	3 min	NNW–WNW	14.3	
24-Mar-03	1:52:07–2:00:52	3–5 km	7.75 min	W	11.6	Fine
	2:00:52–2:07:07	5–7 km	6.25 min	WNW–W	16.8	
	2:07:07–2:13:37	7–9 km	6.5 min	WNW	18.6	

The particles collected onto carbon-coated collodion films supported by nickel grids, which were set on each stage of the low volume impactors, were analyzed with a scanning electron microscope and an energy dispersive X-ray analyzer, in order to see the particle morphology, size, and elemental composition. The diameter of each particle was calculated as the arithmetic mean of the longest width and its width orthogonal.

We simultaneously performed balloon-borne OPC(Optical Particle Counter) measurements, aerosol particle collection near the surface atmosphere, and lidar measurements in Dunhuang, China.

3. Result and summary

3.1. Existence of mineral dust particles both in the spring and the summer free troposphere

Electron microscopic experiments revealed that most of the coarse particles collected in the free troposphere over Dunhuang in spring and even in summer composed of mineral dust particles. Figure 2 shows the representative mineral dust particles collected using the balloon-borne sampler. The observation suggests that dust particles constantly exist in the free troposphere over Dunhuang, although the number concentrations are higher in spring than in summer. Balloon-borne OPC and lidar measurements also suggested the existence of dust particles in the free troposphere not only in spring but also in summer. Mineral types of the individual particles collected in the lower free troposphere showed a significant difference considerably between the cases of spring and the summer. The comparison with the previous results and the meteorological data suggested that the aerosol particles collected over Dunhuang

on March 22, 2003 exhibited the mixing state of aerosol particles under dusty condition. On the other hand, the particles collected on August 29, 2002 showed relatively high number fractions of Ca- and Na-rich particles, which are considered to be evaporite. It seems that the relative abundance of Ca- and Na-rich particles increases when the mineral dust concentration is low.

3.2. The Taklamakan Desert as a possible source of background KOSA observed over Japan

Mineral dust particles were predominantly existed in coarse mode range in the free

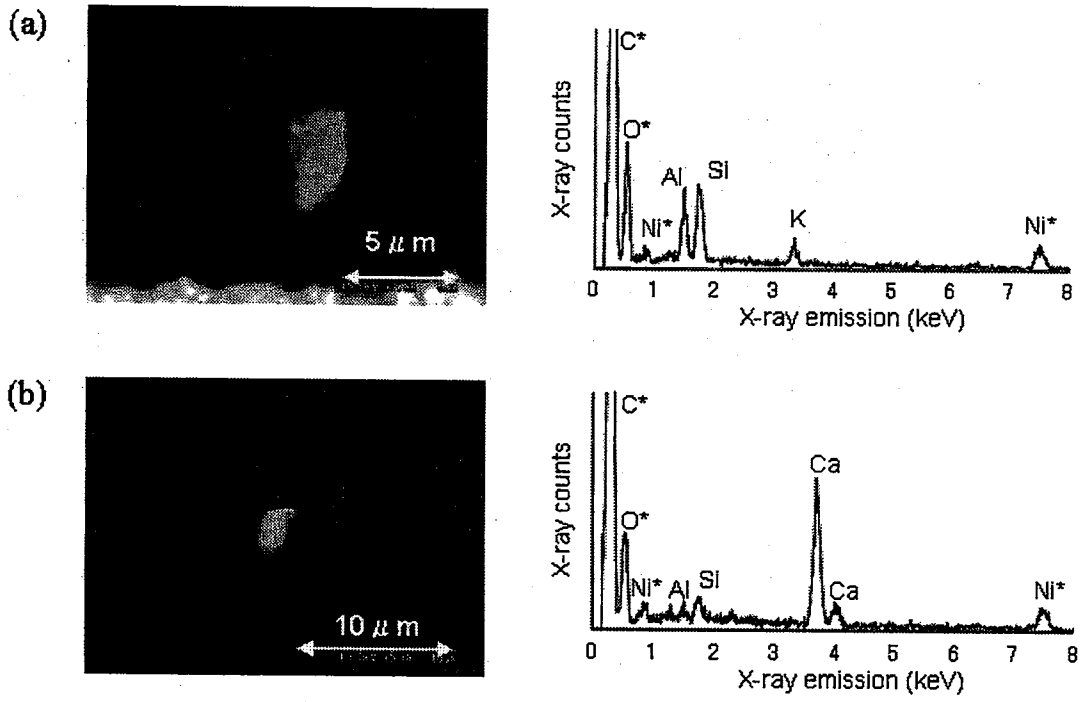

Figure 2. Scanning electron microscopic images (left) and X-ray spectra (right) on representative mineral dust particles collected in the free troposphere over Dunhuang. The particles of (a) and (b) are examples of aluminosilicate, and calcite ($CaCO_3$) containing a little aluminosilicate.

troposphere over Dunhuang in summer. According to air-mass forward trajectories, the air masses observed in the summer passed over Japan (Figure 3). It can be suggested that the Taklamakan Desert is a possible source region of KOSA particles which diffuse to downwind areas such as Japan islands through the free troposphere even in summer seasons. The results could support the previous research that discussed background KOSA on the basis of aircraft borne measurements made over Japan.

3.3. Mixing state of the fine particles over the KOSA source area:

We classified the fine aerosol particles ($0.2 < d < 1.0$ μm) collected in the free troposphere over Dunhuang into four types (sulfate particles, biomass burning particles, mineral dust particles, heavy metal particles), according to detected elements and size. Figure 3 shows Number fractions of particle types in each altitude on March 24, 2003 and on August 29, 2002. In the free

troposphere, sulfur particles were main particles in fine mode ranges (0.2<d<1.0 μm). In particular this tendency was more significant in diameter range of 0.2–0.3 μm, and the dominant number fractions of sulfur particles could be independent on atmospheric condition. On the other hand, the number fractions of sulfur particles in diameter range of 0.3–1.0 μm showed large variation in each sample. In this size range, the mixing state of aerosols is affected relatively strong by dust particles. Balloon-borne OPC measurements also supported the above results. Besides sulfur particles and mineral dust particles, we also found the particles derived from biomass burning and heavy metal particles in fine mode range in the free troposphere over Dunhuang.

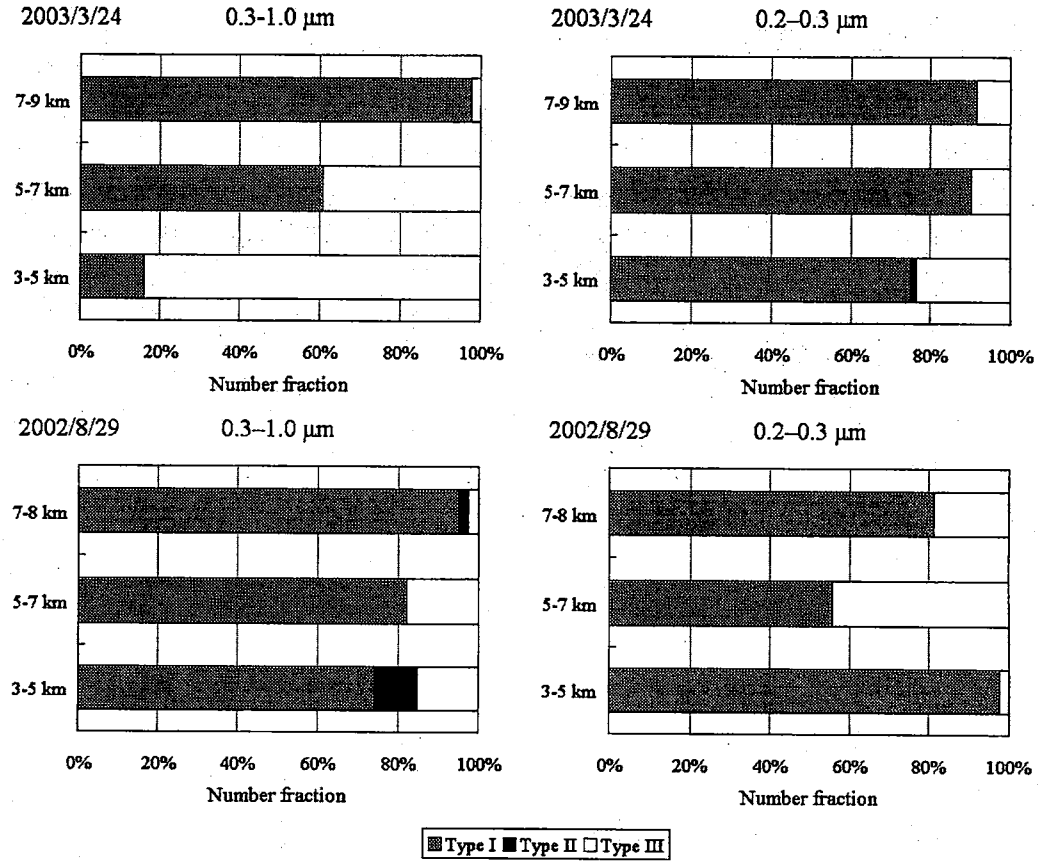

Figure 3. Number fractions of particle types in each altitude on March 24, 2003 and on August 29, 2002. Each type are represent sulfate particle (Type I), particle derived from biomass burning (Type II), and mineral dust particle (Type III).

3.4. Modification of KOSA particles by sulfur

Figure 4 shows the relative weight ratios of Al, S, and Ca for the individual mineral particles of six cases. The dashed line in the scatter-plot shows the weight ratio of S/Ca for gypsum ($CaSO_4 \cdot H_2O$) (=0.8). If there are many points plotted to the right of the dashed line, it can be assumed that considerable uptake of SOx on mineral particles hardly occurs. Most of the dust particles collected in the free troposphere and in the surface atmosphere in Dunhuang, from relative weight ratios of S to Ca in Fig. 4.11, indicated the value of lower than 0.8. The mineral dust particles collected in the free troposphere over KOSA source regions strongly suggest that

significant uptake of SOx by dust particles hardly occurred. Also the mineral dust particles in the surface atmosphere in Dunhuang did not relate with sulfur not only during dust storm but also non dust storm. In contrast about half of dust particles collected in the free troposphere over Japan was seemed to be dust particles significantly modified by sulfur. The fact is of considerable interest when discussing the modification during their long-range transportation. The results suggest that the significant modification of KOSA particles have not occurred at least in an initial stage of their long-range transport, and will be occur in the humid air along with high concentration of sulfur dioxide.

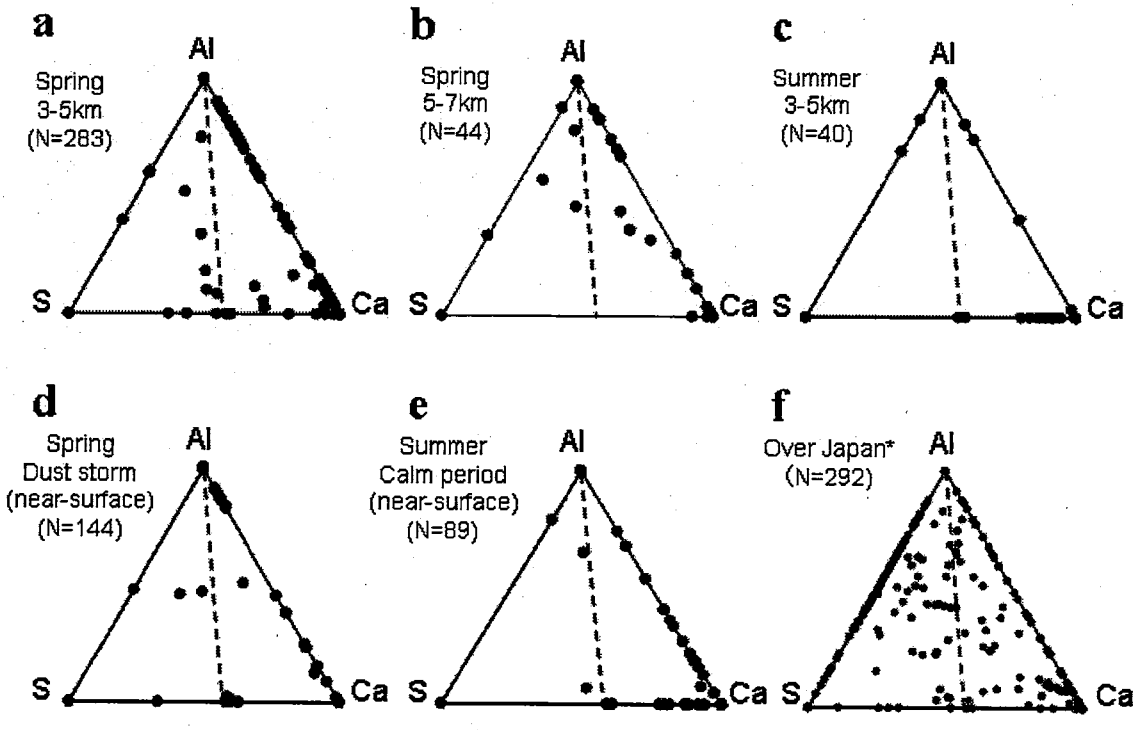

Figure 4. Relative weight ratios of Al, S and Ca for mineral particles collected (a) 3-5 km above sea level over Dunhuang on March 24, 2003, (b) 5-7 km above sea level over Dunhuang on March 24, 2003, (c) 3-5 km above sea level over Dunhuang on August 29, (c) near the surface atmosphere at Dunhuang on April 29, 2002, (e) near the surface atmosphere at Dunhuang, and (f) in the free troposphere over Japan.

学位論文審査結果の要旨

平成18年1月26日の第1回審査委員会で予備的な審査を行い審査方針を決定した。同年2月2日に最終発表を行い、引き続き第2回審査委員会を開いて以下の結論を得た。

本論文は、タクラマカン砂漠上空（自由大気圏高度）の大気エアロゾルの混合状態を明らかにし、それに基づき「黄砂粒子への硫黄化合物の沈着が長距離輸送途中で生じている」こと、及び「タクラマカン砂漠がバックグランド黄砂の有力な発生源である」ことを示した。

地球環境問題が社会的にも学問的にも大きな関心事となって以来、黄砂の変質過程の解明、東アジア西太平洋域での硫黄や窒素の生物地球化学的循環の解明、地球温暖化予測の精度向上などのために、「黄砂発生源の上空における黄砂粒子の混合状態」に関する知見は必須のものとみなされてきた。しかし、黄砂発生源地域の上空での観測は、技術的難度が高く、長い間、実証的研究が皆無であった。申請者は、気球搭載型の大気エアロゾル採集装置を開発（気球切り離し装置部分を担当）し、その成果をもとに中国の敦煌などで現地観測を行った。現地の自由大気圏で採集した試料の分析に基づく上記の結果は、関係分野で長い間待たれていたものであり、気象学や大気環境学の発展に大きな貢献をした。したがって、論文は博士（理学）を授与するに充分値するものと認められた。