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Modification of Asian-dust particles transported by different routes – A case study

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HIGHLIGHTS

- Different mixing states were evident between Asian dusts from same origin.
- The modifications of mineral dust by sulfate and sea-salt were size-dependent.
- S-absorption did not depend on mineralogy for submicron particles in a polluted case.

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ABSTRACT

Two separate Asian dust events occurred before and after the passage of a cold front over Japan on 21 March 2010. According to back trajectories and a model simulation, the two dusty air-masses originated from the same region in Mongoria or northern China and were transported over different routes to Japan. Samples of aerosol particles from both airmasses were collected at Tsukuba and Mt. Haruna and examined by single-particle analysis using a transmission electron microscope and an energy dispersive X-ray analyzer. The mixing properties of mineral aerosol were quite different in the two airmasses and size ranges. In the prefrontal airmass, which were associated with pollution, most of fine (<1 μm) mineral aerosol was internally mixed with sulfate. On the contrary, mineral aerosols in the postfront airmass, which were relatively natural, were mostly externally mixed. In the latter case, the internal mixing was associated with Ca, however in the former case, mixing processes not concerning mineralogy was suggested.

1. Introduction

Asian dust events in Japan occur frequently in spring and fall (Sugimoto et al., 2003; Shimizu et al., 2004). Mineral dust particles influence the radiation budget (Tegan et al., 1996; Yoshioka et al., 2005), cloud formation through their role as cloud condensation nuclei (Twohy et al., 2009; Koehler et al., 2009), and ice nucleation (DeMott et al., 2003; Klein et al., 2010), among other atmospheric physicochemical processes. They also have documented effects on marine phyto-plankton (Uematsu et al., 1983; Duce et al., 1991). Furthermore, they are adversely effective to human health, for example, increasing asthma (Yang et al., 2005; Kanatani et al., 2010).

Asian dust particles are frequently transported along with anthropogenic aerosols from industrial sources in China. A considerable fraction of Asian dust particles have been found to be internally mixed with sulfate or nitrate as a result of chemical reactions with gaseous sulfuric or nitric acid and coagulation with particulate sulfate or nitrate (Yaacov and Judith, 1989; Okada et al., 1990; Dentener et al., 1996; Kojima et al., 2006). Asian dust particles were also frequently internally mixed with seasalt (Niimura et al., 1998, Zhang et al., 2003). These modifications of Asian dust particles may be relevant to climate change through cloud processes, such as water cloud formation (Karydis et al., 2011), ice cloud formation (Falkovich et al., 2004), and rain-fall (Wurzler et al., 2000).

The size of mineral particles in Asian dust may be important in modeling and assessing chemical modifications, because these processes have a dependence on particle size. Mineralogy, too, may differ in different particle size classes (Okada and Kai, 1995). However, few studies have analyzed modifications of Asian dust in terms of particle size (Osada, 2013).

Submicron mineral particles account for a small fraction of Asian dust in terms of mass (Zaizen et al., 1995; Chun et al., 2001), and have not been considered separately in studies of bulk chemical analysis. But they have considerable numbers and surface areas, giving them an influence on radiation balances and cloud formation

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(D'Almeida and Schütz, 1983; Sviridenkov et al., 1993; Okada and Kai, 1995).

Asian dust is frequently observed in Japan in the spring and fall seasons, when frequent extratropical cyclones pass over Japan. Therefore, the transport route of Asian dust is responsible to the wind system of these cyclones. Transport routes are typically differentiated by their relationship to fronts into cold and warm sectors.

Asian dust is commonly observed in the cold sector after the passage of a cold front (Bates et al., 2004; Zhang et al., 2005; Inomata et al., 2011). Increased particle concentrations, especially fine particles, are also observed in the warm sector when southerly winds precede cold fronts (Uematsu et al., 2002; Matsumoto et al., 2003; Bates et al., 2004; Zhang et al., 2005; Takahashi et al., 2010; Inomata et al., 2011). Warm-sector increases have been attributed to anthropogenic pollution transported from China (Inomata et al., 2011), and Zhang et al. (2005) have reported that anthropogenic influences are clearly different between dusty airmasses observed before and after the passage of a cold front at Qingdao, China.

On 21 March 2010, Asian dust was observed over Japan both before and after the passage of a cold front. In this study, aerosol samples were collected from both warm and cold airmasses, and elemental compositions and mixing properties of individual particles were examined using an electron microscope. The aim of this study was to clarify the differences between the samples and to obtain information about the particle modification processes in the different airmasses.

This study used single-particle analysis with a transmission electron microscope (TEM). The method is reliable for determinations of aerosol composition, mixing properties and morphologies (Iwasaka et al., 1988; Okada et al., 1990; Vester et al., 2007; Pósfai et al., 2013).

2. Methods

Aerosol samples were collected using a two-stage impactor with nozzle diameters of 1 and 0.5 mm at a flow rate of 2 L/min. The 50% cutoff diameters for the two stages were 0.66 and 0.14 μm, respectively. The collection surface was a carbon-coated nitrocellulose (collodion) film on a reference copper grid (Maxtaform H7, Graticules Ltd.). The sampling sites were the Meteorological Research Institute (MRI) in Tsukuba (36.08°N, 140.12°E, 40 m altitude) and Mt. Haruna (36.48°N, 138.87°E, 1390 m altitude) (Fig. 1). On 21 March 2010, aerosol particles were collected every 3 h from 0:00 to 21:00 Japan Standard Time (JST) at Tsukuba and every 2 h from 7:00 to 21:00 JST at Mt. Haruna. The sampling duration was 30 min for each sample. Three of these samples were selected for this study: one from 3:00–3:30 JST at Tsukuba before passage of the front (PRE), one from 12:00–12:30 JST at Tsukuba after passage of the front (POST), and one from 9:00–9:30 JST at Mt. Haruna after passage of the front (HARUNA).

Particles were examined with a JEM-1400 TEM using accelerating voltage of 120 kV. Elemental compositions of individual particles were determined by an energy dispersive X-ray (EDX) analyzer (Oxford MAX-80) coupled to the TEM, which was operated in scanning TEM mode with live times of 20 s and beam current of 70 μA. Relative weight of 18 elements in individual particles was calculated by the thin-film method (Cliff and Lorimer, 1975).

In order to ensure the representativeness of particle selection, following method was adopted. We used TEM grids with 200 meshes. From the meshes, 2–10 meshes (depending number of collected particles) were selected. The selection was made randomly, but the location was limited to near the center of grids where particles were collected effectively. From the each selected mesh, a rectangle area at the center of the mesh was defined as analysis area. For the each area, all particles detected by image analysis were analyzed. The size of individual particles was determined by image analysis as area-equivalent diameters.

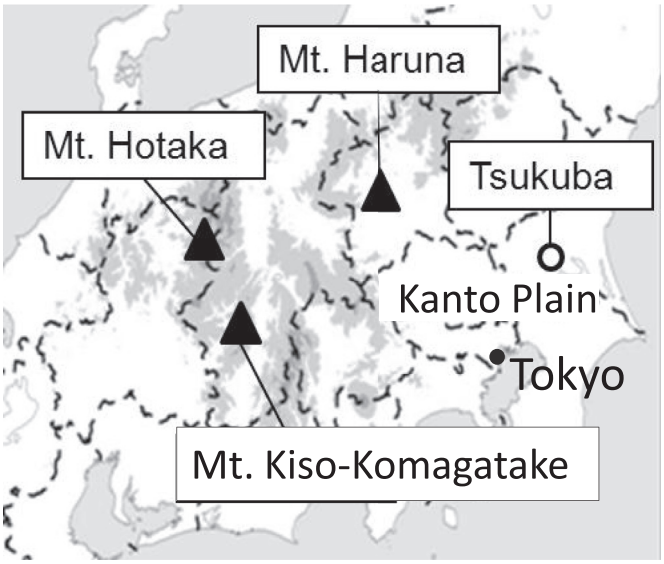

Fig. 1. Locations of observation sites in central Japan. Aerosol samples were collected at Tsukuba and Mt. Haruna. Aerosol concentrations were measured at Tsukuba and the three mountain sites.

In the EDX analysis, sulfur is possibly underestimated due to vaporization of ammonium sulfate. Empirically, the error was considerable for the particles smaller than 0.3 μm. It is factor 2 for the weight ratio of sulfur in the particles of 0.1 μm diameter.

Size distributions of aerosol particles were measured with an optical particle counter (OPC) (Sigmatec, TD-100) and a scanning mobility particle sizer (SMPS) (TSI, 3071A and 3010) at Tsukuba. Aerosol size distributions were also measured at Mt. Haruna, Mt. Hotaka (36.27°N, 137.60°E, 2165 m altitude), and Mt. Kiso-Komagatake (35.77°N, 137.80°E, 2610 m altitude) (Fig. 1) with laser OPCs (Yamanashi Gijutu Kobo Co.).

A global aerosol chemical transport model coupled with the MRI/JMA98 General Circulation Model (MASINGAR; Tanaka et al., 2003) was used to simulate the dust events. The model used a grid of 192 × 96 horizontal cells and 30 vertical layers. The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 2004) was used to calculate the back trajectories of air parcels.

3. Results and discussions

3.1. Two dusty airmasses observed at Tsukuba and mountain sites

According to meteorological data from the Japan Meteorological Agency (JMA) Aerological Observatory at Tsukuba, a southerly wind began to blow at Tsukuba at 14:00 JST on 20 March. The wind direction changed from southwest to northwest at 8:00 JST on 21 March, at which time the air temperature decreased rapidly from 17 °C to 14 °C, indicating the passage of a cold front. After the passage of the front, the JMA observed an Asian dust (Kosa) event over Japan's main island, Honshu. The event was evident in the lidar network of the National Institute for Environmental Studies (NIES) and in MODIS satellite images, which confirmed the presence of a belt of dusty air along the northern side of the cold front.