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Aerial observations of air masses transported from East Asia to the Western Pacific: Vertical structure of polluted air masses

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HIGHLIGHTS

- We conducted aerial observations in December 2010 over the northern East China Sea.
- On Dec. 11 Ca^{2+}, SO_4^{2-}, Al, Ca, Fe, and Zn in aerosol were high with Asian dust.
- Level of pollutants was low in the boundary layer on Dec. 12 in oceanic air.
- Layered structures were characterized by spiral-like vertical observation flights.

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ABSTRACT

There has been only limited information about the vertical chemical structure of the atmosphere, so far. We conducted aerial observations on 11, 12, and 14 December 2010 over the northern part of the East China Sea to analyze the spatial distribution of atmospheric pollutants from East Asia and to elucidate transformation processes of air pollutants during the long-range transport. On 11 December, a day on which Asian dust created hazy conditions, the average PM_{10} concentration was 40.69 μg m^{-3}, and we observed high concentrations of chemical components such as Ca^{2+}, NO_3^-, SO_4^{2-}, Al, Ca, Fe, and Zn. The height of the boundary layer was about 1200 m, and most species of pollutants (except for dust particles and SO_2) had accumulated within the boundary layer. In contrast, concentrations of pollutants were low in the boundary layer (up to 1000 m) on 12 December because clean Pacific air from the southeast had diluted the haze. However, we observed natural chemical components (Na^+, Cl^-, Al, Ca, and Fe) at 3000 m, the indication being that dust particles, including halite, were present in the lower free troposphere. On 14 December, peak concentrations of SO_2 and black carbon were measured within the boundary layer (up to 700 m) and at 2300 m. The concentrations of anthropogenic chemical components such as NO_3^-, NH_4^+, and Zn were highest at 500 m, and concentrations of both anthropogenic and natural chemical components (SO_4^{2-}, Pb, Ca^{2+}, Ca, Al, and Fe) were highest at 2000 m. Thus, it was clearly indicated that the air above the East China Sea had a well-defined, layered structure below 3000 m.

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1. Introduction

Rapid economic growth in East Asia has caused emissions of atmospheric pollutants to increase. At the end of the 20th century, East Asia became the area responsible for the largest emissions of sulfurdioxide (SO_2) and nitrogen oxides ($NO_x = NO + NO_2$) in the world, exceeding those of the United States and Europe (Akimoto, 2003; Ohara et al., 2007). Atmospheric pollutants emitted in East Asia not only affect the emission source area but also are transported long distances downwind to areas such as Japan (Liang et al., 2004; Liu et al., 2008; Aikawa et al., 2010; Li et al., 2010; He et al., 2012). In the context of effects on climate and human health, atmospheric pollutants in aerosols are very important. In September 2009, an annual average of less than 15 μg m^{-3} and a daily average of less than 35 μg m^{-3} were established as the environmental standard for $PM_{2.5}$ (Particulate Matter with diameter of 2.5 μm or less) in Japan.

Concentrations of $PM_{2.5}$ exceeding this environmental standard are often observed in Fukuoka (a large city in northern Kyushu in Japan), in Fukue (a remote, small island in the East China Sea), and at Cape Hedo, Okinawa (a rural area). The observed concentrations and the temporal variations of $PM_{2.5}$ are similar at these three sites, the suggestion being that the entire area is sometimes covered by a large, polluted air mass (Kaneyasu et al., 2011).

To understand the characteristics and effects of long-range transboundary transport of atmospheric pollutants in an air mass, the three-dimensional structure of the air mass must be understood; two-dimensional ground-based observations are not enough. To achieve this understanding, several aerial observation campaigns have been conducted in the Western Pacific, including PEM/WEST (The Pacific Exploratory Mission-West) Phase A and B, ACE-Asia (Asian Aerosol Characterization Experiments), and TRACE-P (Transport and Chemical Evolution over the Pacific) (Hoell et al., 1997; Huebert, 2003; Jacob et al., 2003). We have also conducted aerial observations of atmospheric pollutants over the East China Sea and mainland China (Hatakeyama et al., 2001, 2004, 2005, 2011; Wang et al., 2006, 2008). Here, we report on experiments carried out in December 2010.

2. Experiments

Aerial sampling was carried out from Fukue Airport (32°45′N, 128°41′E) (Nagasaki Prefecture), which is the site of a ground-based observation station for atmospheric research. Aerial observations were made between Fukue and Point A (31°21′N, 126°32′E, ~200 km south of Jeju Island, Korea) on 11, 12, and 14 December 2010 (Fig. 1). Two 40-min round-trip flights (four level flights at altitudes of 500, 1000, 2000, and 3000 m above sea level) were made between Fukue and Point A on each of these days. In addition, the vertical distributions of gases and aerosols above Fukue Island were observed by making 20-min, circular flights at 500, 1000, 2000, and 3000 m above the island on each sampling day.

The airplane employed and the sampling inlet set-ups were the same as those described previously (Hatakeyama et al., 2011). Gaseous pollutants, including O_3 (Model 49i, Thermo Fisher Scientific), SO_2 (Model 43i-TLE, Thermo Fisher Scientific), total reactive nitrogen oxides (NO_y) (Model 42i-TL, Thermo Fisher Scientific, modified for NO_y measurements; Yuba et al., 2010), and CO (Model 48i, Thermo Environmental Instrument); particle number concentrations (sampling flow rate, 2.83 L min^{-1}; duration, 2 min; Sibata GT-521); and particle mass concentrations (Sibata GT-331) were measured on board by methods reported previously (Hatakeyama et al., 2011). Total suspended particles (TSP) were collected on board with a high-volume tape sampler (Kimoto Model AS-191) on a Teflon roll filter (Sumitomo Electric Industry, Poreflon membrane

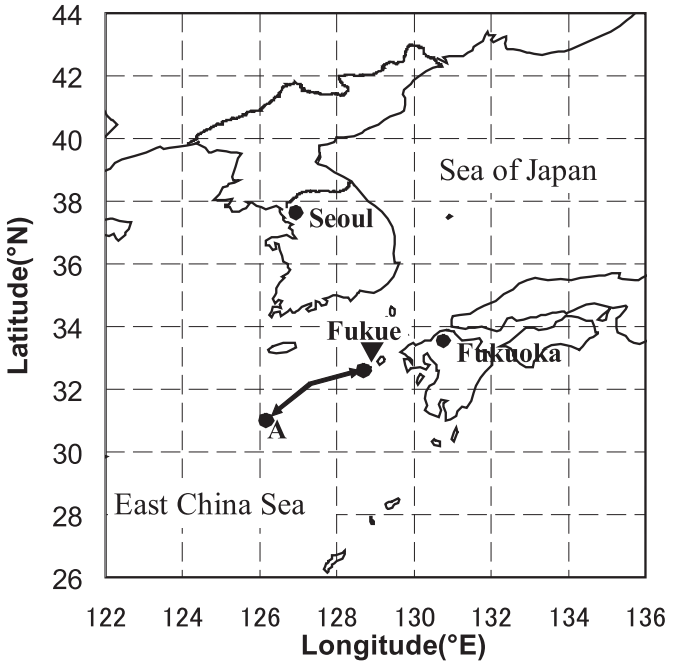

Fig. 1. Map of the observation area. The arrow shows the flight track for the December 2010 aerial observation experiment.

filter 100 mm × 10 m, pore size 5.00 μm). Ionic species (NH_4^+, Ca^{2+}, K^+, Na^+, Mg^{2+}, SO_4^{2-}, NO_3^-, Cl^-, and F^-) and metallic elements (Li, Na, Mg, Al, K, Ca, V, Cr, Mn, Fe, Co, Cu, Zn, Ga, As, Se, Rb, Sr, Ag, Cd, In, Sb, Cs, Ba, Hg, Tl, and Pb) on the filter samples were analyzed in a laboratory on land by ion chromatography (Shimadzu IC-C4 without suppressor for cations, and Shimadzu IC-SA2 with suppressor for anions) and inductively coupled plasma mass spectrometry (Agilent 7500 ICP-MS).

The CFORS (Chemical weather FORecast System) model was developed by Uno et al. (2003). CFORS simulates the transport and distribution of pollutants such as SO_2, SO_4^{2-}, and dust. Back-trajectory analyses were carried out with the HYSPLIT4 model from the U.S. National Oceanic and Atmospheric Administration (NOAA) (Draxler and Rolph, 2003). The initial altitude and calculation time were set to the observation altitude and 72 h or 96 h, respectively.

3. Results and discussion

3.1. Distributions of air pollutants over the northern East China Sea

Fig. 2 shows the variation of ionic species and the variation of metallic elements in aerosols collected on board the aircraft on 11, 12, and 14 December.

3.1.1. 11 December

Average concentrations of SO_2, NO_y, HNO_3, CO, and black carbon (BC) were relatively high at lower altitudes (500 and 1000 m). The concentrations at 500 m (1000 m) were 3.60 (5.32) ppb, 7.48 (6.33) ppb, 2.20 (1.83) ppb, 372 (314) ppb, and 1.29 (0.96) μg m^{-3}, respectively. At an altitude of 2000 m (3000 m) the concentrations of these components were lower: 0.99 (0.21) ppb, 1.91 (1.63) ppb, 1.17 (1.14) ppb, 126 (115) ppb, and 0.13 (0.06) μg m^{-3}, respectively.

As shown in Fig. 2b, concentrations of Ca^{2+} and metallic elements such as Al, Ca, Fe, and Zn were high during the early sampling of 11 December. The Ca^{2+} concentrations in the samples that were obtained at lower altitudes were the highest that we