

Long-range transport of organic aerosol to Cape Hedo, Japan



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ABSTRACT

An investigation at Cape Hedo, Japan, from 2005 to 2006, focused on the long-range transport of organic aerosol (OA) from the Asian continent. An Aerodyne aerosol mass spectrometer was used to investigate the OA data collected over the study. OA concentrations were low from July to September and peaked during March and April. Based on air mass origins, four OA source regions were identified: northern China, southern China, Japan, and Korea. OA concentrations measured at Cape Hedo from the four sources did not exhibit large differences. Conversely, the frequencies of the air masses reaching Cape Hedo from the different regions varied considerably. Northern China was identified as the primary source of organic aerosols at Cape Hedo. Examination of variations in the ratio of m/z 44 to OA concentrations with transport time showed that OAs were partially oxidized during transport

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1. Introduction

Organic aerosol (OA) typically accounts for a large, sometimes even dominant, fraction of air particulate matter. OAs are well-known to affect climate, the biosphere, and human health. Despite recent advances, our understanding of the composition, physical and chemical properties, sources and transformation of OA are still limited, and the environmental effects of OA remain poorly understood.

Fine OA particles are similar in size to the wavelengths of light in the visible spectrum, indicating that they should have stronger climatic impacts than larger particles because they will interact more with incoming solar radiation. Fine OA particles can also be transported far from their source regions, delocalizing their climatic and environmental impacts relative to their emission sources.

East Asia is a rapidly developing region, with increased energy consumption producing considerable anthropogenic emissions of both gaseous species and particulate matter (Streets et al., 2003). Compositions of these pollutants in the atmosphere and their environmental impacts are the focus of considerable research.

Recently, the long-range transport of pollutants from continental areas to remote islands in the East China Sea has been studied using models and direct measurements. A deflation module was designed to model the long-range transport of yellow sand over East Asia (Wang, Ueda, & Huang, 2000). Zhang et al. (2004) studied the transport and chemical transformation of sulfur compounds over East Asia using the community multi-scale air quality (CMAQ) modeling system, using meteorological fields calculated from the regional atmospheric modeling system (RAMS). Numerous measurement campaigns have also been carried out. Kato, Pochanart, and Kajii (2001) measured ozone and non-methane hydrocarbons at Chichi-jima Island. Suthawaree et al. (2007) measured CO and O₃ at the Okinawa Islands to investigate the long-range transport of air pollution from highly polluted regions in East Asia. Takami, Miyoshi, Shimono, and Hatakeyama (2005) studied aerosols using an Aerodyne aerosol mass spectrometer (Q-AMS) at Fukue Island, with a primary focus on inorganic compounds. Many of these measurements and simulations covered short periods and continuous measurements over at least one year had never been performed before we began measuring aerosols in October 2004 (Lun, Takami, Miyoshi, & Hatakeyama, 2009).

Our measurement station was located at Cape Hedo, Okinawa, which is far from any large land mass. In this study, we present results from our continuous investigations of OA from October 2005 to August 2006. Contributions from different pollutant sources were identified and the oxidation of OA during transport was shown to occur.

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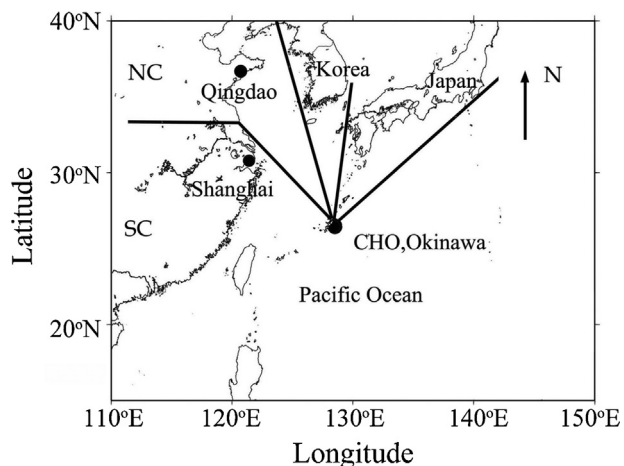


Fig. 1. Map of East Asia and the location of Okinawa and its surrounding islands. The four air mass regions in East Asia (northern China (NC), southern China (SC), Japan and Korea) are indicated by the black lines on the map.

2. Experimental

2.1. Site description

Cape Hedo (26.87° N, 128.25° E) is situated at the northern end of Okinawa Island. The location of Okinawa Island is shown in Fig. 1. The site is approximately 1800 km from Beijing and 700 km from Shanghai, which are the two largest cities in China. Approximately 600 km to the northeast is the large harbor city of Fukuoka and approximately 1000 km to the northwest is the coastline of Korea. There were no large industrial or residential areas near our measurement station. During the summer, the island is affected by maritime air masses from the Pacific Ocean that bring clean air. However, during the winter, Okinawa Island is exposed to strong westerly winds and polluted air from Chinese coastal areas is transported downwind to Okinawa. As such, this site is ideally positioned for investigating air pollution after long-range transport over East Asia.

Measurements were conducted at this station since October 2005 and are ongoing. Some data over this time period were lost because of instrument failure and poor weather. Meteorological conditions including wind direction, wind velocity, temperature, pressure, relative humidity, and precipitation were measured using a weather transmitter (Vaisala, WXT510). Aerosols were sampled through a half-inch copper tube projecting from the roof of the station. The sampling height was approximately 4 m from the ground.

2.2. Instruments

The sub-micron, non-refractory chemical composition of OA was analyzed using a quadrupole aerosol mass spectrometer (Q-AMS) produced by Aerodyne Research Inc (America). A detailed description of this device can be found in a number of other publications (Allan, Alfarra, et al., 2003; Allan, Jimenez, et al., 2003; DeCarlo, Slowik, Worsnop, Davidovits, & Jimenez, 2004; Jayne et al., 2000; Jimenez, Bahreini, et al., 2003; Jimenez, Jayne, et al., 2003; Takami et al., 2005, 2007).

Briefly, ambient air is introduced into the Q-AMS through a half-inch copper pipe. Particles larger than 2.5 μm are cut off using a cyclone. The AMS draws particles (with aerodynamic diameters of less than 1 μm), using high vacuum, through an “aerodynamic lens” that contracts and expands the sampled air stream through

Table 1

Concentration ($\mu\text{g}/\text{m}^3$) of organics in PM₁ at Cape Hedo from Oct 2005 to Aug 2006.

	Average	STDEV	Max	Min
Oct	1.12	0.94	4.27	Below detection limit
Nov	1.21	0.95	7.06	Below detection limit
Dec	1.99	1.34	10.93	Below detection limit
Jan	2.05	1.62	7.04	Below detection limit
Feb	2.02	1.49	7.57	Below detection limit
Mar	2.72	2.12	19.76	Below detection limit
Apr	2.36	1.67	14.63	Below detection limit
May	1.64	1.52	7.64	Below detection limit
Jun	0.49	0.47	3.94	Below detection limit
Jul	0.66	0.57	5.17	Below detection limit
Aug	0.89	0.83	4.66	Below detection limit

an orifice. The aerosols are then vaporized at 600 °C. The vaporized molecules are then ionized by standard 70-eV electron impact ionization and the positive ions are analyzed using a quadrupole mass spectrometer that provides a mass spectrum of the aerosol components. The aerosol size distribution is then calculated from the aerosol flight time (DeCarlo et al., 2004; Jimenez, Bahreini, et al., 2003), which is defined as the time taken to travel from the chopper to the detector.

AMS data were analyzed using an Igor-based software package described by Allan, Jimenez, et al. (2003) and Allan et al. (2004). Briefly, the ion current detected by the quadrupole mass spectrometer was converted into a mass concentration using the measured ionization efficiency (IE) of nitrate, which was calibrated with respect to ammonium nitrate aerosol (Jayne et al., 2000). For all other compounds, the relative ionization efficiency (RIE) was used as calibrated in our laboratory because each compound has its own unique ionization efficiency (Alfarra et al., 2004; Allan et al., 2004; Jimenez, Jayne, et al., 2003). The RIE values for ammonium, nitrate, sulfate, and organics used in our study were 3.5, 1.1, 1.2, and 1.4, respectively. The particle collection efficiency (CE) in the AMS chamber was also taken into account. A CE value equal to 0.5 is commonly used for the analysis of Q-AMS data, based on comparisons between particle-into-liquid sampler combined with an Ion Chromatography analyzer (PILS-IC) and Q-AMS measurements (Takegawa et al., 2005). To verify the CE value, we compared sulfate concentrations measured using Q-AMS (Q-AMS-sulfate) with those measured using a low-pressure impactor (LPI-sulfate) during the spring of 2006. The results showed that the Q-AMS-sulfate analyzed using a CE of 0.5 was about twice that of the LPI-sulfate. A CE value of 1 was identified as suitable for analyzing the Q-AMS data obtained at Cape Hedo 2006 (Takami et al., 2007). A CE of 1 was used for all species because the mass distributions were similar for ammonium, sulfate, and organics, suggesting that each of the species could be found on the same particle. Using another method, we found similar results. Here we give a brief introduction of the method. We suggest that ammonium bisulfate was a major component because the molar ratio of ammonium to sulfate was commonly 1. Ammonium bisulfate was considered to be in the liquid phase because the deliquescence point of ammonium bisulfate is 25 °C at a relative humidity (RH) of 40% and the RH of the inlet of the Q-AMS ranged from 20% to 60%. Aerosols in the liquid phase are known to have a CE value of 1 (Allan et al., 2004).

3. Results and discussion

3.1. Concentration of organic aerosol

Average monthly OA concentrations are presented in Table 1. The lowest average monthly concentration of $(0.49 \pm 0.47) \mu\text{g}/\text{m}^3$ occurred when air masses generally originated from the Pacific

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