Journal of Aerosol Science 100 (2016) 97–107

Contents lists available at ScienceDirect

Journal of Aerosol Science

journal homepage: www.elsevier.com/locate/jaerosci

Characteristics of carbonaceous aerosols in large-scale Asian wintertime outflows at Cape Hedo, Okinawa, Japan

K. Shimada [a,b,*], A. Takami [c], S. Kato [d], Y. Kajii [e], S. Hasegawa [f], A. Fushimi [c], A. Shimizu [c], N. Sugimoto [c], C.K. Chan [g], Y.P. Kim [a,h,i], N.H. Lin [a,j], S. Hatakeyama [a,b]

[a] Global Innovation Research Organization, Tokyo University of Agriculture and Technology, Tokyo, Japan
[b] Institute of Agriculture, Graduate School of Tokyo University of Agriculture and Technology, Tokyo, Japan
[c] National Institute for Environmental Studies, Onogawa, Tsukuba, Ibaraki, Japan
[d] Department of Applied Chemistry, Faculty of Urban Environmental Sciences, Tokyo Metropolitan University, Minami-oosawa, Hachioji, Tokyo, Japan
[e] Department of Natural Resources, Graduate School of Global Environmental Studies, Kyoto University, Yoshida-honmachi, Sakyoku, Kyoto, Kyoto, Japan
[f] Center for Environmental Science in Saitama, Kazo, Saitama, Japan
[g] School of Energy and Environment, City University of Hong Kong, Hong Kong, China
[h] Department of Chemical Engineering & Materials Science, Ewha Womans University, Seoul, Republic of Korea
[i] Department of Environmental Science & Engineering, Ewha Womans University, Seoul, Republic of Korea
[j] National Central University, Chung-Li, Taiwan

ARTICLE INFO

Article history:
Received 24 November 2015
Received in revised form
30 April 2016
Accepted 13 May 2016
Available online 29 June 2016

Keywords:
Carbonaceous aerosols
Large-scale Asian wintertime outflows
CHAAMS
Long-term observations

ABSTRACT

We have performed long-term measurements of organic carbon (OC) and elemental carbon (EC) in atmospheric aerosols at Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS). Large-scale East Asian continental outflows of aerosols have frequently been observed at CHAAMS in both winter and spring, but large-scale air pollution (LSP) events in winter have not yet been described in the literature. Examination of $PM_{2.5}$ in LSP events showed that it consisted mainly of carbonaceous aerosols and sulfate. We compared OC and EC concentrations measured at CHAAMS during LSP events with those measured at the same time in East Asian cities, and with concentrations measured at CHAAMS during an Asian dust event in spring 2006. The maximum OC and EC concentrations at Cape Hedo during LSP events were comparable to those at China Atmosphere Watch Network sites in China. The average OC concentration was also similar to concentrations measured at urban sites in Taipei (Taiwan), Seoul (Korea), and Tokyo (Japan); EC, however, was several times lower at Cape Hedo than EC concentrations at these urban sites. We identified three types of air masses associated with LSP events: Type 1, a polluted air mass composed of freshly emitted pollutants; Type 2, an air mass that underwent photochemical aging during transport from China to Okinawa; and Type 3, an air mass that was almost fully aged in China before being transported to Japan. Simulations of LSP events with a 3-D chemical transport model (Chemical Weather Forecasting System) indicated that Type 1 air masses were associated with “cone type” transport in a low-pressure system, and Type 2 air masses were associated with “giant puff type” transport by a migratory high-pressure system. Generally speaking, air pollutants are transported from China to CHAAMS in winter in association with the north or northwest monsoon. High levels of air pollutants in China in winter are due to higher emissions and poorer dispersion because of the low boundary layer height and low wind speeds. Our results suggest that LSP events occur when a cold front forms under accumulated air

* Corresponding author at: Global Innovation Research Organization, Tokyo University of Agriculture and Technology, Tokyo, Japan.
E-mail address: shimadai@cc.tuat.ac.jp (K. Shimada).

http://dx.doi.org/10.1016/j.jaerosci.2016.05.006
0021-8502/

pollutants. The mass contribution of carbonaceous materials to $PM_{2.5}$ during the LSP event in winter 2008 was 36%, much larger than the contribution of 19% during the spring 2006 dust event.

1. Introduction

Black carbon (BC), brown carbon, and mineral dust are the most important light-absorbing aerosols that affect global climate. The impact of these substances on the global warming of atmospheric temperatures has garnered much attention in recent years (Ramana et al., 2010; Andreae & Gelencsér, 2006; Shapiro et al., 2009; Sareen, Schwier, Shapiro, Mitroo, & McNeill, 2010; Jacob et al., 2010; UNEP & WMO; 2011; Park, Kim, Jeong, Youn, & Kim, 2010). Moreover, these aerosols may include toxic substances and thus can negatively affect human health.

However, BC is usually internally mixed with sulfate, amplifying its role in solar absorption. Most of the inferred climate forcing by BC ($\sim$1 W m^{-2}, whether internally or externally mixed) is probably anthropogenic (Sato et al., 2003). BC warming is regulated by the presence of sulfate, which is derived mainly from sulfur dioxide (SO_2) emissions. Sulfate strongly reflects solar radiation, whereas BC strongly absorbs solar radiation; thus, the net radiation force is determined by the relative amounts of BC and sulfate (Jacobson, 2001). Brown carbon from combustion processes is that part of organic carbon aerosols that has been found to absorb sunlight (Andreae & Gelencsér, 2006).

The Atmospheric Brown Cloud (ABC) project of the United Nations Environment Programme (UNEP), currently in its second phase, is investigating black carbon emissions and air pollution in East Asia and their effect on climate change. Ohara et al. (2007) and Zhang et al. (2009) have reported increasing black carbon emissions in Asia, and according to Matsui et al. (2011), Kondo et al. (2011), and Wang et al. (2011b), BC and sulfate transported from Asia are affecting the North American Arctic.

Since 2004, we have conducted observations at the Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS) in Okinawa, Japan, which was a supersite by UNEP during a regional experiment conducted as part of the ABC first phase (Takami et al., 2007; Nakajima et al., 2007; Shimada et al., 2015). Moreover, for over 9 years we have been measuring secondary inorganic aerosols such as sulfate and nitrate, in addition to carbonaceous aerosols, to understand the effects of air pollution on climate change in East Asia.

Many studies have reported on the occurrence of transboundary air pollution by Asian dust in spring (Tsai et al., 2012; Wang et al., 2011a; Hatakeyama et al., 2011; Sawa et al., 2007), but most transboundary air pollution events occurring in spring are of shorter duration than wintertime events. In this paper, we report on large-scale transboundary air pollution events occurring in winter; such events have not been reported before in East Asia. Here, we define a large-scale plume with broad elemental carbon (EC) and organic carbon (OC) peaks lasting 4–7 days as a large-scale air pollution (LSP) event. Our long-term observations at CHAAMS from 2004 to 2009 showed that LSP events happen frequently in winter. In this study, we compared OC and EC concentrations in large-scale plumes at CHAAMS with concentrations measured in East Asian cities and with similar measurements made during the Asian dust event of spring 2006, a typical example of a springtime LSP event.

2. Experimental methods

CHAAMS (26.87 °N, 128.25 °E, 60 m above sea level) is located at the northern end of the Okinawa Island, Japan. The site is located about 100 km from Naha, the largest city on the island, and about 650 km from Shanghai, China. There is no large industrial or residential development around the station. Because air masses are transported to CHAAMS from China, Korea, Japan, Southeast Asia, and the Pacific Ocean, this site is suitable for observing characteristics of the long-range transport of air pollutants in East Asia.

Concentrations of total carbon (TC) as TC=OC+EC and OC in aerosols were measured by an ambient particulate carbon monitor (Model 5400; Rupprecht & Patashnick Co. Inc., Albany, NY, USA) at 3 h intervals from March 2004 to December 2009. OC and TC were analyzed by NDIR CO_2 sensor at a desorption temperature at 350 °C and 750 °C, respectively. EC was calculated as the difference of TC and OC.

However, the collection efficiency of the Model 5400 is known to be lower than filter sampling. Using the method described by Chow, Watson, Crow, Lowenthal, and Merrifield (2001), we compared raw data obtained simultaneously by a DRI instrument (Desert Research Institute, Reno, Nevada, USA) and the Model 5400 operated at CHAAMS (N=79) from March to May in 2010 and 2011.We also developed the following conversion equations:

OC=$(2.3 \pm 0.23)X+(0.11 \pm 0.10)$, and.

TC=$(2.3 \pm 0.18)X+(0.16 \pm 0.1)$ (Fig. S1),

where X is the concentration measured by the Model 5400. We then calculated EC by subtraction of TC from OC (EC=TC−OC). Aerosol particles for analyses of OC and EC in $PM_{2.5}$ were collected on quartz microfiber filters with a