Available online at www.sciencedirect.com
ScienceDirect
Atmospheric Environment 41 (2007) 717–729
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Vertical profiles of ultrafine to supermicron particles measured by aircraft over Osaka metropolitan area in Japan

Shuichi Hasegawa[a,*], Shinji Wakamatsu[b], Toshimasa Ohara[a], Yasuyuki Itano[c], Katsumi Saitoh[d], Masamitsu Hayasaki[a,e], Shinji Kobayashi[a]

[a]*National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan*
[b]*Ehime University, 3-5-7 Tarumi, Matsuyama, Ehime 790-8566, Japan*
[c]*Osaka City Institute of Public Health and Environmental Sciences, 8-34 Tojo-cho, Tennoji-ku, Osaka 543-0026, Japan*
[d]*Akita Prefectural Research Center for Public Health and Environment, 6-6 Senshu-Kubota, Akita 010-0874, Japan*
[e]*JST Cooperative System for Supporting Priority Research, Kawaguchi, Saitama, Japan*

Received 6 February 2006; received in revised form 4 September 2006; accepted 8 September 2006

Abstract

Intensive aircraft- and ground-based measurements of ultrafine to supermicron particles in the Osaka metropolitan area, Japan, were carried out on 17–19 March 2003, in order to investigate vertical profiles of size-resolved particles in the urban atmosphere. Differently sized particles were observed at different altitudes on 19 March. Relatively higher concentrations of ultrafine particles (31 nm) and submicron particles (0.3–0.5 μm) were measured (100–200 cm^{-3}) at altitudes of 300 and 600 m, whereas supermicron particles (2–5 μm) were present (300–600 cm^{-3}) at higher altitudes (1300 m in the morning and 2200 m in the afternoon). The chemical composition analysis showed that supermicron particles evidently comprised mainly soil particles mixed internally with anthropogenic species such as carbonaceous components and sulfate. Numerical simulation using the Chemical weather FORecasting System (CFORS) suggested the long-range transport of soil dust and black carbon from the Asian continent. Total number concentrations of particles sized 10–875 nm ranged from 4.8×10^3 to 3.0×10^4 cm^{-3} at an altitude of 300 m and from 7.3×10^2 to 4.8×10^3 cm^{-3} at an altitude of 1300 m. Total number concentrations of particles sized 10–875 nm correlated very well with NO_X concentrations, and, therefore, ultrafine and submicron particles were likely emitted from urban activities such as car traffic and vertically transported. Number size distributions at lower altitudes obtained by aircraft measurements were similar to those obtained by ground measurements, with modal diameters of 20–30 nm on 18 March and about 50 nm on 19 March.

Keywords: Ultrafine particles; Submicron particles; Supermicron particles; Chemical composition; Long-range transport

1. Introduction

To understand the behavior of particles in the atmosphere, it is important to know their spatial and temporal variations, and various kinds of physical and chemical properties of particles as well as related gaseous matter concentrations. Aircraft

*Corresponding author. Tel.: +81 29 850 2901; fax: +81 29 850 2580.
E-mail address: hasegawa.shuichi@nies.go.jp (S. Hasegawa).

1352-2310/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2006.09.031

measurement is an effective way to observe particles in the atmosphere for these purposes. Therefore, many particle measurements have been collected by aircraft, particularly during major observation projects such as ACE-1, INDOEX, ACE-Asia, and TRACE-P (Bates et al., 1998; de Gouw et al., 2001; Huebert et al., 2003; Jacob et al., 2003).

Weber et al. (1998) observed nanoparticles (2.7–10 nm) at altitudes of ~1200 m over Macquarie Island (between Antarctica and Tasmania) during ACE-1 and discussed the process of new particle formation. de Reus et al. (2001) measured number concentrations and size distributions from the surface to the upper troposphere over the northern Indian Ocean during INDOEX and discussed particle formation in relation to nucleation mode in the free troposphere. Bahreini et al. (2003) measured vertical profiles of mass concentrations and size distributions of sulfate, ammonium, and organics using an aerosol mass spectrometer (AMS) at altitudes of ~3700 m over western Japan during ACE-Asia. Lee et al. (2003) measured inorganic anions and cations of submicron particles with high sensitivity and time resolution using a particle-into-liquid sampler coupled with ion chromatography (PILS-IC) during ACE-Asia and TRACE-P. Moreover, Hatakeyama et al. (2004, 2005) measured inorganic anions and cations over the East China Sea and Bo Hai, respectively. Zaizen et al. (1996) and Schröder and Ström (1997) observed vertical profiles of number concentrations and size distributions in the troposphere over the western Pacific Ocean and western Europe, respectively.

The aircraft measurements mentioned above have been carried out over oceans or coastal areas; however, the measurements have also been carried out over urbanized land. During the Southern California Air Quality Study (SCAQS) (Lawson, 1990), gaseous matters (such as SO_2, NO_X, O_3, and hydrocarbons) and particle physical and chemical properties (such as light scattering, size distribution, and concentrations of carbonaceous and ionic species) were measured by aircraft. Collins et al. (2000) surveyed the physical and chemical properties of particles by aircraft over the Los Angeles Basin, United States, during the Southern California Ozone Study (SCOS). They estimated the three-dimensional distribution of particles within the basin from data obtained during several spiral flights and discussed spatial and temporal variations.

Urban areas in Japan such as the Tokyo and Osaka metropolitan areas not only have mobile and stationary emission sources but are influenced by long-range transport from the Asian continent. Some observation campaigns have been made in these areas. Wakamatsu et al. (1983, 1990) and Uno et al. (1985) observed ozone, NO_X, and non-methane hydrocarbons (NMHC) by aircraft or airship over the Tokyo metropolitan area, and analyzed their behavior in conjunction with meteorological conditions. Wakamatsu et al. (1998a) conducted aircraft measurements of NO_X and ozone in spring over the Osaka metropolitan area, and observed high concentration episodes. These observation data were used for validation of a numerical simulation model (Wakamatsu et al., 1998b; Ohara et al., 2001). However, particles have not been focused in these studies; it is a matter of course that particles are also investigated. Therefore, we carried out intensive aircraft- and ground-based measurements of gaseous and particulate matters in the Osaka metropolitan area in spring 2001 and 2003, and Itano et al. (2005) presented vertical profiles of ozone and discussed local and regional contributions to ozone.

In this paper, we present the vertical profiles of ultrafine to supermicron particles and their chemical compositions, obtained in the Osaka metropolitan area in spring 2003. Comprehensive size-resolved particle measurements from aircraft and chemical analysis of major components, such as carbonaceous, water-soluble, and elemental species, in the urban atmosphere were carried out. Such measurements have hardly been performed previously. We discuss the behavior of particles in the urban atmosphere based on such measurements.

2. Instrumentation and measurement methods

2.1. Aircraft measurements

The aircraft measurements were carried out from 17 to 19 March 2003 over the Osaka metropolitan area. Table 1 shows the information for aircraft-measurement flights. Although three aircraft-measurement flights (ME0301–ME0303) were carried out on 17 March, those data are not presented in this paper. The location of the Osaka area and the typical flight route are shown in Fig. 1. Osaka City, with a population of 2.6 million, is the prefecture capital and is located in the center of the Osaka Plain facing Osaka Bay. The city is a major source