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Spatiotemporal variations in atmospheric aerosols in East Asia: Identifying local pollutants and transported Asian aerosols in Osaka, Japan using DRAGON

Makiko Nakata[1], Itaru Sano[1], Sonoyo Mukai[2] and Brent N. Holben[3]

[1]{ Kinki University, Osaka, Japan }

[2]{ Kyoto College of Graduate Studies for Informatics, Kyoto, Japan }

[3]{Goddard Space Flight Center / NASA, MD, USA }

Correspondence to: M. Nakata (nakata@socio.kindai.ac.jp)

Abstract

In this work, we document the spatial and temporal variations of atmospheric aerosols in East Asia, specifically focusing on the NASA/AERONET-Osaka site in March 2012 during the AERONET "DRAGON-Japan" campaign. Air pollution has become a serious issue in East Asia in recent years, with particular problems caused by fine particles having diameters of up to 2.5 μm ($PM_{2.5}$). Emissions of anthropogenic aerosols are known to increase with economic growth, whereas natural dust concentrations show significant variation with season and changing wind patterns. We focus on variations in the mass concentrations of particulate matter (PM) gathered by a sampler (SPM-613D) at the NASA/AERONET-Osaka site in March 2012, and have compositionally analyzed individual PM types using a scanning electron microscope (SEM) coupled with an energy dispersive X-ray analyzer (EDX). Our data show that Asian aerosols derived from distal sources were unequivocally detected in Japan on 11th March 2012. Such pollutants can be carried by winds from continental China and subsequently merge with local emissions, thus accentuating air pollution problems.

1 Introduction

Since the early 1990s, the NASA/AERONET (National Aeronautics and Space Administration /Aerosol Robotics Network) project has provided the scientific community

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with a wide range of valuable data concerning the global distribution of aerosols (Holben et al. 1998). In recent years, 400 Cimel radiometers have begun collecting data worldwide, with six instruments located in Japan having been operational at the Shirahama, Noto, Osaka, Fukuoka, Sapporo, and Chiba AERONET sites since 2000, 2001, 2004, 2012, 2015, and 2015, respectively. Using these facilities, the NASA/AERONET/DRAGON (Distributed Regional Aerosol Gridded Observation Networks) campaign was initiated by Japan. Atmospheric aerosol distributions in East Asia are complex, being influenced by both natural phenomena and human activity (Kahn et al. 2004), with urban areas in particular being dominated by fine anthropogenic aerosols released from diesel-powered vehicles and industrial activity. Increased emissions of anthropogenic sulfuric, nitric, carbonaceous, and other aerosols associated with economic growth can lead to increased concentrations of hazardous air pollutants. In particular, large cities in East Asia have experienced numerous heavy haze episodes (dense concentrations of atmospheric aerosols) over the past thirty years. Furthermore, yellow dust storms or biomass burnings represent serious environmental hazards, yet their aerosol properties are poorly understood. Recent publications from our research group have documented the development of an efficient algorithm for aerosol retrieval from space in haze episodes around urban areas in China (Mukai et al. 2012, 2014, and 2015).

Air pollution in East Asia is severely influenced by heavy air pollutants and Asian dust transported from China to its neighboring countries throughout the year. Fine particulate matter with a diameter of up to 2.5 μm ($PM_{2.5}$) is particularly problematic. Emissions of fine anthropogenic aerosols are known to increase with economic growth. The mass concentrations of particulate matter (PM) have been measured by SPM-613D at our AERONET-Osaka site (Mukai et al. 2007). The SPM-613D sampler collects suspended PM on a Teflon filter and measures the weight of the particles by their absorbance of beta rays. The PM sampler can determine the individual concentrations of $PM_{2.5}$, PM_{10}, and optical black carbon (OBC) particles by measuring the proportions of fine particles ($PM_{2.5}$) and coarse particles (PM_C), and calculating the difference between PM_{10} and $PM_{2.5}$ concentration levels (i.e., $PM_C = PM_{10} - PM_{2.5}$). PM_C is generally a better indicator of dust events than $PM_{2.5}$ because soil-derived dust particles are typically large. The sampler measures PM concentrations every hour and data are eliminated if the PM_{10} concentration is smaller than that of $PM_{2.5}$. After this initial data screening, daily and monthly PM concentrations are calculated.

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Ueda et al. (2012) recently highlighted a correlation between health concerns and high levels of $PM_{2.5}$ in Japan and neighboring Asian countries. In particular, $PM_{2.5}$ increases the mortality rate of patients suffering from heart and/or lung diseases because these fine particles penetrate deeper into the lungs than coarser inhalable particles can, and so have a more severe impact on an individual's wellbeing. Temporal variations in aerosol properties have been investigated here by compositional analysis of individual PM types using a scanning electron microscope (SEM) coupled with an energy-dispersive X-ray analyzer (EDX).

Alongside causing air pollution, aerosols also affect the Earth's radiation balance through scattering, absorbing solar and thermal radiation, and by promoting chemical reactions in the atmosphere, thus altering its composition (Takemura et al. 2005). The study of aerosols is therefore important in order to understand their influence on the environment, the climate, and public health. As such, this work aims to use the NASA/AERONET/DRAGON campaign to draw attention to the importance of studying aerosols from a variety of perspectives.

The remainder of this paper is organized as follows: section 2 introduces the DRAGON-Japan campaign conducted in spring 2012, and sections 3 and 4 present and discuss the results obtained, respectively. Section 5 presents a brief summary of all of the work.

2 DRAGON-JAPAN campaign

The first aim of the DRAGON campaign in Asia was to investigate the optical properties of trans-boundary aerosols in East Asia, where air quality has recently declined due to heavy air pollution and Asian dust being transported from China to neighboring countries throughout the year (Lee and Kim 2010). These pollutants influence both the local atmosphere at their points of origin and also remote locations due to long-range transportation. The second aim of DRAGON was to measure aerosol properties in a megacity; thus, high-resolution measurements of the variation in atmospheric aerosols on both a spatial and temporal scale were desired for selected Asian urban population centers. Consequently, NASA/DRAGON-Asia was undertaken mainly at Osaka in Japan from 15th February 2012 to the end of May 2012. The DRAGON campaign in Asia in 2012 is alternatively called DRAGON-Japan or DRAGON-Osaka. During DRAGON-Japan, AERONET instruments were established from Fukue Island to Osaka (each ~200 km) alongside a NIES (National Institute for Environmental Science) 2ch LIDAR (Light Detection and Ranging) system and PM samplers, as shown in Fig. 1. Localities in Seoul and South Korea were also available for analysis. This

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campaign intended to establish a network composed of intensively distributed sites to constrain the variability of atmospheric aerosols, including those transported from continental China. The observation site discussed in this work was located in eastern Osaka (Fig. 1). Osaka city is at the center of the second largest metropolitan area of Japan. The greater Osaka metropolitan area covers 7,800 km^2, is located within 50–60 km of the city center, and contains over 10 million inhabitants, making it one of the biggest metropolitan areas in the world. Its industry is largely focused on manufacturing, with many small- and medium-sized enterprises, and suffers from severe air pollution caused by particles emitted from diesel-powered vehicles and industrial activities (Nakata et al. 2013, 2015). Airborne pollutants transported from continental China exacerbate the problem. As such, Osaka was viewed as a key location for the DRAGON-Japan campaign.

Our investigation aimed to document the spatial and temporal variations of atmospheric aerosols in East Asia, specifically focusing on the AERONET/Osaka site, thus concentrating on air pollution in a megacity (Osaka). Air pollution is a serious problem in megacities, as particulate matter containing high proportions of $PM_{2.5}$ is a severe health hazard. Anthropogenic sources of $PM_{2.5}$ include automobiles, factories, coal-burning power plants, and domestic heaters. In addition, the size of dust particles generally decreases as they are transported via westerly winds over long distances; therefore, dust storms may contain high concentrations of fine particles (M. Mukai et al. 2004). Accordingly, $PM_{2.5}$ concentration levels are influenced by both anthropogenic and natural factors.

3 Results of DRAGON-JAPAN

3.1 Fukue Island

During the DRAGON-Japan campaign period, three AERONET instruments were located on Fukue Island in the East China Sea (Fig. 1). Fukue Island has a generally warm and very wet climate, with hot summers and cool winters that lack snowfall owing to its southerly latitude. Typhoon activity throughout summer and autumn leads to high precipitation, although it is usually sunny during spring. The first measurement made by a standard AERONET processing system is that of aerosol optical thickness (AOT), which is an important aerosol parameter that can be derived from the transmittance measured by direct sun photometry. The resolution of AOT is better than 0.01 at all observational wavelengths and all data obtained

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are cloud-screened before aerosol retrieval (Eck et al. 1999, Smirnov et al. 2000, Dubovik et al. 2000, O'Neill 2003). As the basic parameter for describing atmospheric aerosols, AOT indicates the degree of opacity, and hence the atmospheric concentration of aerosol particles.

Additional measurements were made by portable sun photometers (MT-2) located on Fukue Island. These MT-2 were calibrated with a standard AERONET Cimel radiometer. Figure 2 shows the AOT values recorded by MT-2 on 11th March 2012, when a high AOT was detected in the morning. Observation by portable sun photometer was conducted at Fukue Island (Nakata et al. 2012). AOT was seen to increase throughout the morning, peaking at around 09.00 (Japan standard time: JST) and decreasing afterwards. These features were consistent with LIDAR measurements. Figure 3 presents an example of these measurements obtained by a Mie scattering LIDAR instrument located on Fukue Island. Notably, LIDAR provides the vertical distribution of atmospheric aerosols and the total depolarization ratio of perpendicular components to parallel components of the backscattering intensity. The measurements made on 11th March 2012 are shown by a black line on Fig. 3, where it can be seen that high concentrations of air pollutants were recorded in the morning.

Environmental standards for air quality in Japan are designed to reduce impacts on residents' health, and suspended particulate matter (SPM)—PM with a diameter up to 7 μm—is widely sampled across the country in order to monitor the air quality. Figure 4 shows mean monthly SPM concentrations measured by the Japanese Ministry of the Environment on Fukue Island over a five-year period from April 2009 to March 2014, which encompasses that of the DRAGON-Japan campaign. The error bars on Fig. 4 mark the ranges between the maximum and minimum monthly values for the entire five-year period. The concentration of SPM increased from February to May, decreased in June, and remained low during the winter months. The five-year averaged SPM concentration for March was 25 μg/m^3, although it exceeded 100 μg/m^3 on 11th March 2012, showing that a high concentration of air pollutants was registered over Fukue Island at that time.

3.2 Aerosols along a transportation path on 11th March 2012

Air pollutants were measured on 11th March 2012 in other locations in western Japan, with SPM concentrations at the Fukue Island, Saga, Yamaguchi, and Osaka sites (Fig. 1) shown in Fig. 5. Following measurement of an SPM concentration peak at 09:00 JST at Fukue Island, concentration spikes were recorded in Saga at 12.00 JST, in Yamaguchi at 14:00 JST, and in

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Osaka at 17:00 JST, although the actual SPM concentration decreased from west to east throughout the day. These features indicate that air pollutants were transported by the wind. Figure 6 shows sulfur dioxide (SO_2) concentrations at the same four sites on the same day, which also increased. SO_2 is derived from the combustion of sulfur-bearing fossil fuels and is a major air pollutant. Oxidation of SO_2 leads to the formation of sulfurous and sulfate aerosols. Anthropogenic sources of SO_2 include the use of sulfur-bearing fossil fuels for domestic heating, stationary power generation, and motor vehicles. In recent years the use of high-sulfur coal for domestic heating has declined in Japan, such that energy production and motor vehicles are now the predominant sources. Although this has led to a continued reduction in atmospheric SO_2 levels, its concentration remains at a relatively high level in China. GCM/SPRINTARS (Takemura et al. 2005) simulations shown in Fig. 7 suggest that sulfate aerosols were transported from continental China to western Japan on 11th March 2012. Thus, both ground measurements and simulation data imply that the high concentration of pollutants recorded above Fukue Island on the morning of 11th March 2012 originated from China.

3.3 Osaka site

The Osaka analysis site utilized all of the ground-measurement devices described in the previous section. NIES/LIDAR measurements in Osaka recorded high concentrations of air pollutants in the evening of 11th March 2012 (Fig. 8), whereas LIDAR measurements showed that similar such high concentrations occurred on Fukue Island on that morning, indicating that the pollutant-bearing air parcel travelled to Osaka from Fukue during the day.

PM sampler SPM-613D can measure the concentrations of various PMs (e.g., PM_{10} and $PM_{2.5}$) separately, alongside OBC. Figure 9 shows hourly $PM_{2.5}$ and PM_{10} concentration data obtained from the Osaka site, where it is clear that concentrations of both types increased in the afternoon and peaked at around 17:00 JST. In the morning of 11th March 2012, high concentrations of air pollutants were recorded at Fukue Island (Figs 2 and 3). The absence of an effective source of anthropogenic particles close to the island indicates that the airborne pollutant was transported from continental China to the East China Sea (Fig. 7). Large PM concentrations recorded in Osaka in the afternoon of the same day suggest that the air parcel responsible then migrated from the East China Sea to Osaka (cf. Figs 5 and 6).

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4 Discussion

4.1 Analysis of PM recorded from 11th to 12th March 2012

Changes in aerosol properties over time were investigated by performing individual analyses on PM populations using an SEM coupled with an EDX analytical system. We analyzed $PM_{2.5}$ populations collected by SPM-613D in Osaka at the peak of its measured concentration on 11th March 2012 (17:00 JST), one hour before (16:00 JST) and after (18:00 JST) this peak, in the morning of the same day (08:00 JST), and in the late evening (02:00 JST). The pie charts in Fig. 10 show the contribution of each component to the mean mass concentration obtained for ~100 particles for each time period. The proportion of sulfur, which possibly indicates an anthropogenic source, becomes dominant at the peak of the observed PM mass concentration (17:00 JST) and remains high afterwards. Nakata et al. (2011) showed that the proportion of sulfate increases when a parcel of air that contained anthropogenic aerosols reaches Osaka from China. It is clear, therefore, that the concentration of sulfate, which was possibly derived from the combustion of fossil fuels, became dominant at the peak hour of the recorded air pollution event, and remained high afterwards.

4.2 Normal air conditions in Osaka

Insight into the severity of the air pollution event recorded on 11th March 2012 at Osaka can be obtained via comparison with ground measurements taken at 16:00 JST on 14th March 2012, which are representative of normal air conditions. Such AERONET data show small AOT values and low PM mass concentrations (Fig. 11). The air pollution environmental quality standard in Japan for $PM_{2.5}$ is a daily average of 35 $\mu g/m^3$ of air. All $PM_{2.5}$ data for 14th and 15th March 2012 are less than this daily quality standard; in particular, the concentration is lowest at 16:00 JST on 14th March 2012, and hence the air over Osaka is interpreted to have been clear at that time. Figure 12 presents the compositional analysis of this matter, where it can be seen that although sulfur is one of the major components, its concentration is lower than that in PM collected during the period of severe pollution on 11th March (Fig. 10).

5 Summary

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In this study, we have analyzed only one day's worth of data (11th March 2012), despite the DRAGON-Japan project running for more than three months. However, it has been shown herein that airborne pollutants can influence both the local atmosphere near to their source and relatively remote locations due to long-range transportation. The following results have been derived from this work:

1. High concentrations of airborne pollutants were recorded on Fukue Island in the East China Sea on the morning of 11th March 2012, which was during the period of operation of the DRAGON-Japan campaign. In the afternoon on the same day, large PM mass concentrations were recorded in Osaka. Component analysis of this particulate matter using SEM/EDX showed that the averaged proportion of sulfur in the total mass concentration of ~100 particles clearly increased above levels typical for normal days, and remained high until the late evening.

2. Examination of component proportions in the air on normal days showed that the amount of sulfur was significantly larger on 11th March 2012, especially at the time of the pollutant's peak concentration. Very clear atmosphere in Osaka is rare, as it is usually polluted with particles in suspension derived from diesel-powered vehicles and local industries. As such, sulfur derived from anthropogenic sources is one of the major components of aerosols over Osaka; nonetheless, there was an unusually high ratio of sulfur within the measured PM in the afternoon and evening of 11th March 2012, indicating contamination from an additional source.

3. We speculate that the parcel of air that carried these anthropogenic aerosols reached Fukue Island on the morning of 11th March 2012 from China, and migrated to Osaka on the same afternoon.

In this work, we focus on concentrations of $PM_{2.5}$ during spring when its concentration can be attributed to both anthropogenic production and natural dust aerosols. In order to investigate the change in aerosol properties, individual analysis of PM types was performed using an SEM coupled with an EDX analytical system. The proportion of sulfate was seen to increase during air pollution spikes. However, it is clear that silicon, which is possibly derived from soil particles, becomes the dominant element in large particles during dust events. Sulfur (in the form of sulfate) becomes the dominant element in fine particles for air pollution reaching Osaka alongside dust. In this study, we have focused on the analysis of $PM_{2.5}$ during periods when winds carry pollution from China to Japan. Air pollution has become a serious issue in

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China, with $PM_{2.5}$ being of particular concern. This problem has also become particularly acute in neighboring parts of Asia, including Japan, given airborne transmission. The Ministry of Environmental Protection in China has stated that up to a quarter of the country was covered with thick fog in January 2013. High concentrations of $PM_{2.5}$ were commonly observed in Osaka during this time period, despite $PM_{2.5}$ concentrations rarely being so high at that time of year. This indicates that seasonal winds carry pollution from China to Japan, exacerbating local pollution levels. Recently, the Chinese government declared a red alert due to very high $PM_{2.5}$ concentrations around Beijing city in December 2015. As the air quality worsens in urban areas in Asia, continued high-resolution measurements of atmospheric aerosols at different spatial, temporal, and spectral scales are necessary. Alongside such global observations, we will continue to investigate the effect of local air pollution at Osaka and other sites.

Acknowledgements

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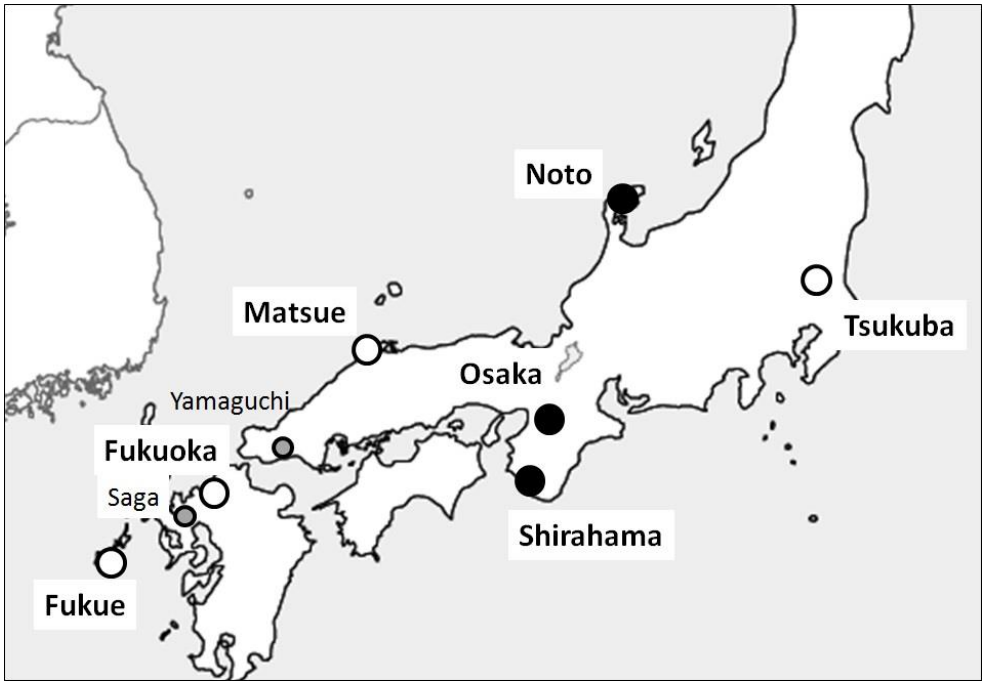

Figure 1. DRAGON-Japan site distribution. Black and open circles indicate permanent AERONET sites and temporary monitoring locations during the DRAGON campaign period, respectively. Grey circles indicate sites measuring SPM concentrations.

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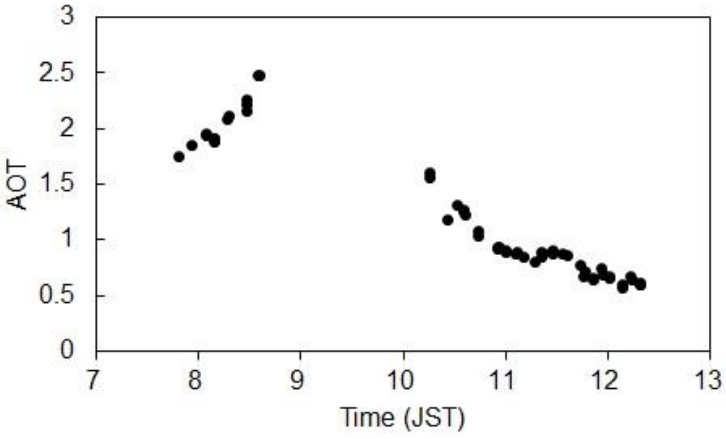

Figure 2. AOT at a wavelength of 440 nm at three points on Fukue Island on 11th March 2012.

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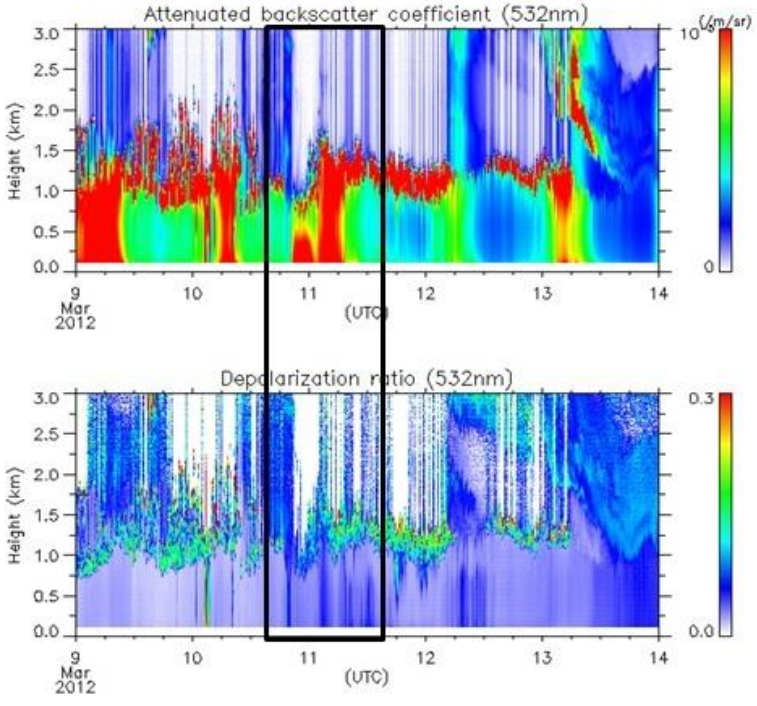

Figure 3. LIDAR observations at Fukue Island in universal time (UTC) from 9th to 13th March 2012.

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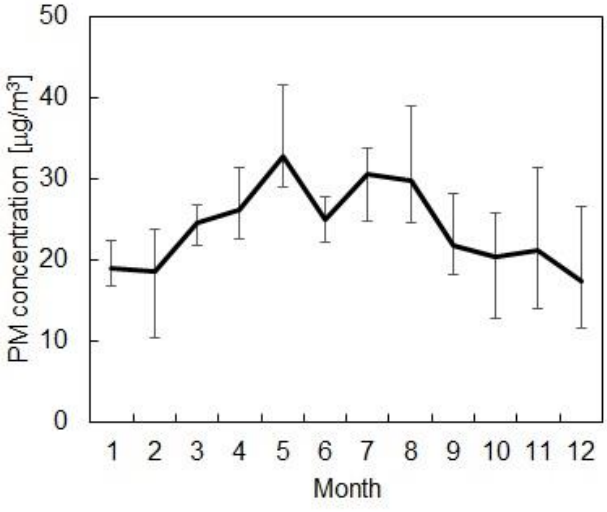

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3 Figure 4. Monthly mean SPM concentration on Fukue Island averaged from April 2009 to
4 March 2014. Error bars represent the ranges between the maximum and minimum monthly
5 values recorded over the entire five-year period.

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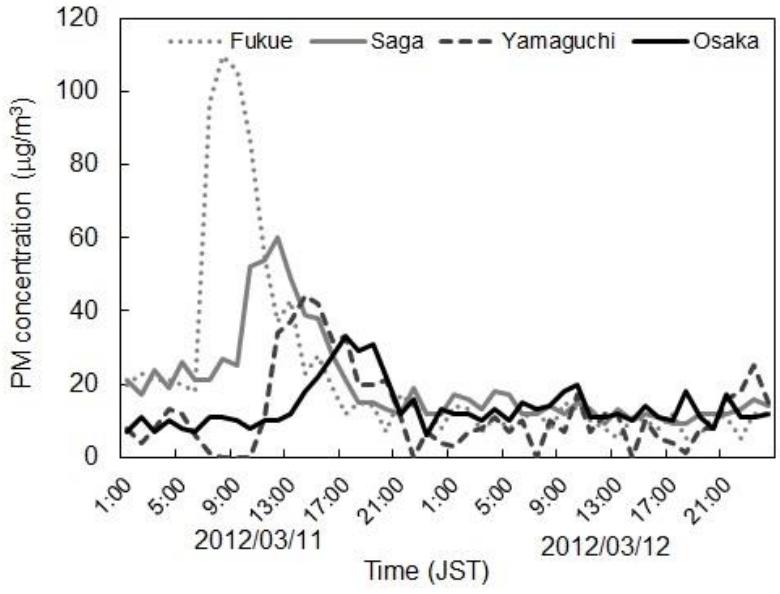

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3 Figure 5. SPM concentrations at Fukue Island, Saga, Yamaguchi, and Osaka from 11th to 12th
4 March 2012. Times are shown in Japan standard time (JST), which is nine hours ahead of
5 universal time (i.e., UT + 09:00).

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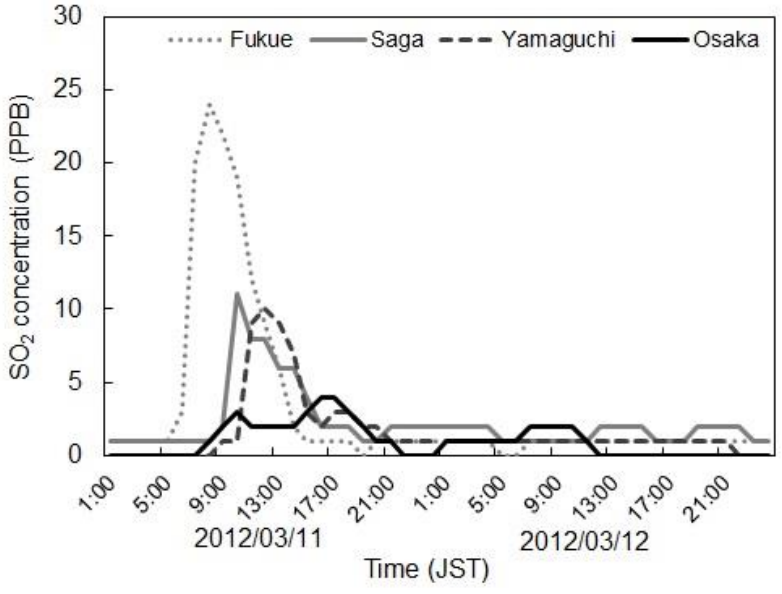

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3 Figure 6. SO_2 concentrations at Fukue Island, Saga, Yamaguchi, and Osaka from 11th to 12th
4 March 2012. Times are shown in Japan standard time (JST), which is nine hours ahead of
5 universal time (i.e., UT + 09:00).

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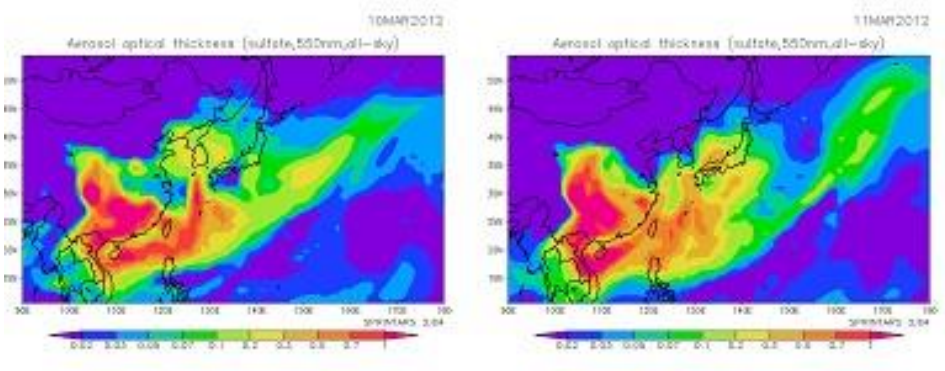

Figure 7. AOT (sulfate) at a wavelength of 550 nm simulated by SPRINTARS for 10th and 11th March 2012.

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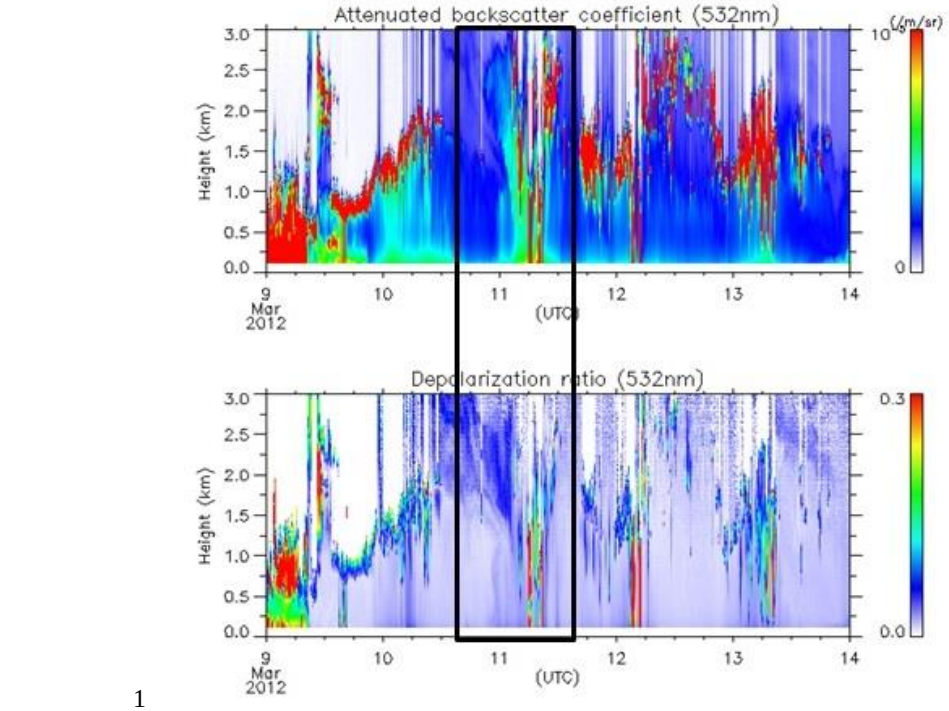

Figure 8. LIDAR observations at Osaka in universal time (UTC) from 9th to 13th March 2012.

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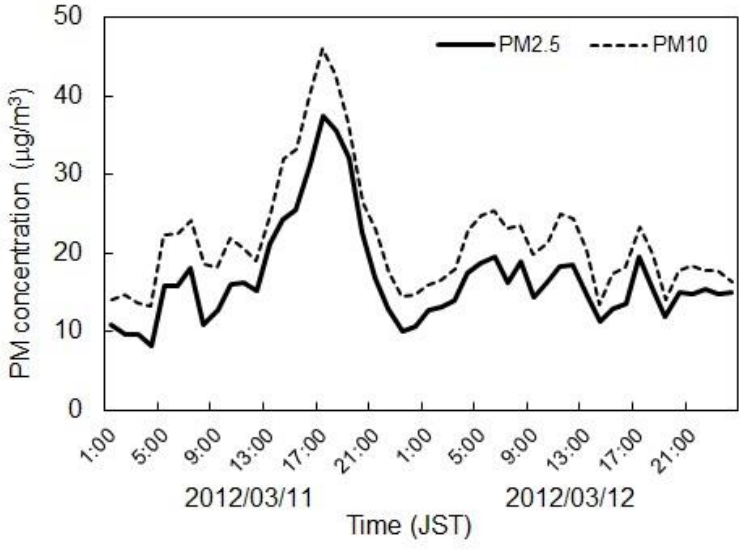

Figure 9. $PM_{2.5}$ and PM_{10} concentrations in Osaka from 11th to 12th March 2012. Times are shown in Japan standard time (JST), which is nine hours ahead of universal time (i.e., UT + 09:00).

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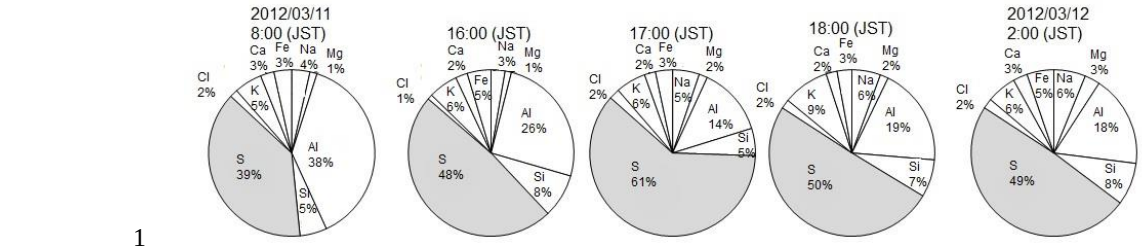

Figure 10. Mass concentration (%) of each component of $PM_{2.5}$ collected on 11th and 12th March, 2012, determined by a scanning electron microscope with an energy-dispersive X-ray analytical system (SEM/EDX). All times are given as JST

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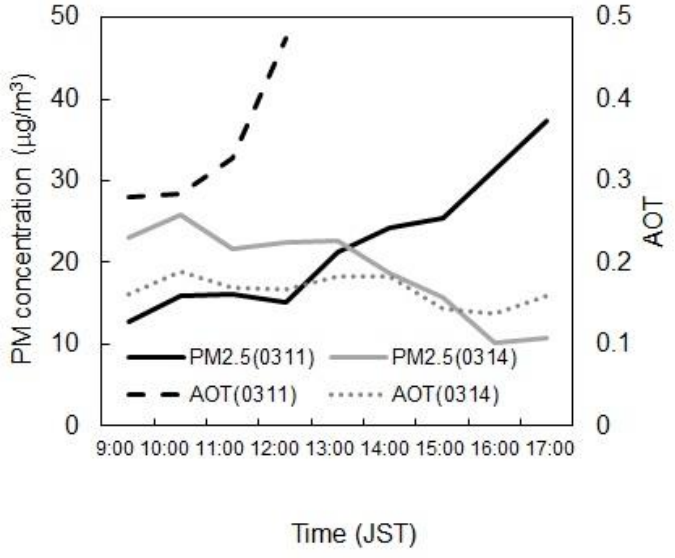

Figure 11. $PM_{2.5}$ concentrations and AERONET AOT at a wavelength of 675 nm in Osaka on 11th and 14th March in 2012, denoted by "0311" and "0314", respectively.

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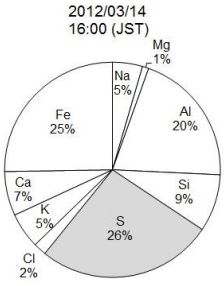

Figure 12. Mass concentration (%) of each component of $PM_{2.5}$ collected at 16:00 JST on 14th March 2012, determined by a scanning electron microscope with an energy-dispersive X-ray analytical system (SEM/EDX).