

Long-term Measurements of Carbonaceous Aerosol at Cape Hedo, Okinawa, Japan: Effects of Changes in Emissions in East Asia

Kojiro Shimada^{1,2,9*†}, Akinori Takami³, Takatsugu Ishida², Yuta Taniguchi²,
Shuichi Hasegawa⁴, Chak K. Chan⁵, Yong Pyo Kim^{1,6,7}, Neng-Huei Lin^{1,8},
Shiro Hatakeyama^{1,2†}

¹Global Innovation Research Organization, Tokyo University of Agriculture and Technology, Tokyo, Japan

²Institute of Agriculture, Graduate School of Tokyo University of Agriculture and Technology, Tokyo, Japan

³National Institute for Environmental Studies, Onogawa, Tsukuba, Ibaraki, Japan

⁴Center for Environmental Science in Saitama, Kazo, Saitama, Japan

⁵School of Energy and Environment, City University of Hong Kong, Hong Kong, China

⁶Department of Chemical Engineering & Materials Science, Ewha Womans University, Seoul, Korea

⁷Department of Environmental Science & Engineering, Ewha Womans University, Seoul, Korea

⁸National Central University, Chung-Li, Taiwan

⁹School of Creative Science and Engineering, Waseda University, Tokyo, Japan

OPEN ACCESS 

Received: August 5, 2020

Revised: April 29, 2021

Accepted: May 6, 2021

* Corresponding Author:

kshimada@sci.u-ryukyuu.ac.jp;
kshimada@cs.u-ryukyuu.ac.jp

† Present address: Asia Center for Air Pollution Research, Niigata, Japan

‡ Present address: Department of Chemistry, Biology, and Marine Science, University of the Ryukyus, Okinawa 903-0213 Japan

Publisher:

Taiwan Association for Aerosol Research

ISSN: 1680-8584 print

ISSN: 2071-1409 online

 Copyright: The Author(s).

This is an open access article distributed under the terms of the [Creative Commons Attribution License \(CC BY 4.0\)](https://creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium, provided the original author and source are cited.

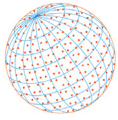
ABSTRACT

We investigated the changes in the carbonaceous-aerosol sources and their effects on the long-term elemental carbon (EC) and organic carbon (OC) concentration trends at the Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS) in Okinawa, Japan, during the period 2004–2013. We obtained the EC and OC concentrations by conducting semi-real-time measurements using a carbon monitor, and performing an offline thermal/optical filter analysis according to the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol. The annual average concentration of the EC remained constant between 2004 and 2013, but that of the OC decreased at a rate of $0.11 \mu\text{g C m}^{-3} \text{y}^{-1}$ ($\alpha > 0.05$). The secondary OC (SOC)/OC ratio showed an increasing trend from 2004 till 2011, which may have been caused by a reduction in primary emissions of OC and compositional changes in the organic compounds originating in China, from which air pollutants were frequently transported during spring and winter. Although the EC concentration did not change appreciably in either season, the OC concentration decreased at rates of $0.10 \mu\text{g C m}^{-3} \text{y}^{-1}$ and $0.11 \mu\text{g C m}^{-3} \text{y}^{-1}$ during spring and winter, respectively. We estimated the contributions from the various sources of carbonaceous aerosol, viz., biomass burning, fossil-fuel combustion, and air-pollutant transport from China, based on the OC/EC ratio, which decreased from 5.7 to 2.4 in terms of the annual average. The growing share from fossil-fuel combustion is responsible for the decline in biofuel-burning OC emissions transported from China to CHAAMS.

Keywords: Carbonaceous aerosol, CHAAMS, Fossil fuel combustion, Biomass burning, Long-term trend

1 INTRODUCTION

Elemental carbon (EC), or black carbon (BC), is known to cause both warming and degraded air quality. BC is the most critical light-absorbing component in atmospheric aerosols and a contributor to climate change although short-lived compared to other long-lived gases. Air



pollutants such as BC are called *short-lived climate pollutants* (SLCPs) (Shindell *et al.*, 2012). BC concentrations have been reported to show a downward trend in Europe (Bower *et al.*, 2009) and the United States (Kirchstetter *et al.*, 2008; Murphy *et al.*, 2011; Hand *et al.*, 2013) but an upward trend in Asia (Ohara *et al.*, 2007). For example, Bower *et al.* (2009) reported that the BC concentration decreased 1952–2005 in England, and Kirchstetter *et al.* (2008) reported a decrease 1967–2003 in the ambient BC concentration in San Francisco, California, United States, despite an increase in fuel consumption. These observations were attributed to the successful implementation of technologies controlling BC emissions. In the American and European Arctic region, BC has decreased at Zeppelin, Ny-Ålesund, Alert, Barrow, and Split (Eleftheriadis *et al.*, 2009; Hirdman *et al.*, 2010) due to a downward trend of BC concentrations in United States and Europe. On the other hand, the contribution of BC and sulfate concentrations derived from northern Eurasia including East Asia increase in 1989 to 2009 (Hirdman *et al.*, 2010).

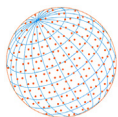
According to Kondo *et al.* (2011) and Matsui *et al.* (2011), in the Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission, the amount of BC and sulfate transported from Asia to the Arctic region was found to be higher than that transported from the North America to the Arctic. As one of the reasons, rapid economic development has caused increased energy consumption along with a higher rate of increase in anthropogenic air pollution in Asia (Ohara *et al.*, 2007; Zhang *et al.*, 2009). Estimates of the global and regional effects of air pollutants (such as BC) in Asia on climate change contain large uncertainties. Analyses of the long-term trend in BC in Asia based on global and regional monitoring are rather scarce.

At the Cape Hedo Atmosphere and Aerosol Monitoring Station (CHAAMS) in Okinawa, Japan, observations of transboundary air pollution have been performed since 2004. CHAAMS has been designated as a major atmosphere-monitoring station by the United Nations Environment Programme in the Asian region (Takami *et al.*, 2007; Nakajima *et al.*, 2008; Shimada *et al.*, 2015). To understand the impact of air pollution on the climate, ecosystems, and human health, we examined the formation of secondary aerosols, such as sulfate and nitrate (Takami *et al.*, 2005, 2007; Takiguchi *et al.*, 2008). In particular, in order to improve the three-dimensional regional model, we investigated the process of chemical transformations of these secondary aerosols during long-range transport. We previously studied seasonal variations and source areas of carbonaceous aerosols from 2004 till 2008 in air masses transported from East Asia to CHAAMS (Shimada *et al.*, 2011) as well as identified specific sources within the source area (Shimada *et al.*, 2015). Our long-term observations at CHAAMS from 2004 till 2009 showed that large-scale transboundary air-pollution (LSP) events frequently occur in winter (Shimada *et al.*, 2016). In addition, we have compared emission inventories with each of the aforementioned studies to understand carbonaceous-aerosol sources and emissions in China. Studying the long-term trends in OC and EC concentrations also supplies information for improving three-dimensional regional model in East Asia. Therefore, in this study, by investigating the long-term trends of OC and EC concentrations, we focus on the relationship between the long-term trends in OC and EC concentrations over CHAAMS and examine the relation between these trends and emission changes in China.

2 EXPERIMENTAL METHOD

CHAAMS (26.87°N, 128.25°E, 60 m above sea level) is located at the northern end of Okinawa Island in Japan. The site is approximately 100 km from Naha, which is the largest city on the island, and approximately 650 km from Shanghai in a straight line, one of the major cities in China. There is no major industrial or residential activity around the station. Air masses reaching CHAAMS are primarily transported from China, Korea, Japan, Southeast Asia, and the Pacific Ocean; therefore, this site is suitable for observing the long-range transport characteristics of air pollutants in East Asia (Takami *et al.*, 2007).

The OC and EC concentrations in aerosols were measured using an ambient-carbon monitor (Series 5400; Rupprecht & Patashnick Co., Inc. [R&P], Albany, New York, USA) at 3-h intervals from March 2004 till May 2011. OC concentration and total carbon (TC) concentration were analyzed with a non-dispersive infrared CO₂ sensor on the basis of desorption at 350°C and 750°C, respectively. EC concentration was calculated as the difference between TC concentration and OC concentration (Shimada *et al.*, 2016).



In addition to the aforementioned continuous measurements, carbonaceous aerosol was collected on quartz filters (Shimada *et al.*, 2011) and analyzed using a thermal/optical carbon analyzer (Model 2001; Desert Research Institute [DRI], Reno, Nevada, USA), following the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol (Chow *et al.*, 2001). Aerosol particles for analyses of OC and EC in PM_{2.5} were collected on quartz-microfiber filters with a diameter of 47 mm (Pallflex Tissuquartz 2500QAT-UP; Pall Corp., East Hills, New York, USA) using an Federal Reference Method 2025 apparatus (Thermo Electron Corp., Franklin, Massachusetts, USA) operated at a flow rate of 16.7 L min⁻¹. None of the quartz fiber filters were baked before sampling because the amount of volatile organic compounds (VOCs) adsorbed onto unbaked quartz fiber filters is lower than the amount adsorbed onto baked (900°C) quartz fiber filters (Hasegawa, 2014). Three categories of blank filters were used: field (at the rate of 4 filters per month), travel (6 filters per month), and laboratory (4 filters per month) (Shimada *et al.*, 2015). The detection limits of OC and EC were 0.90 and 0.10 µg C m⁻³, respectively. The OC and EC samples were collected on a weekly basis from March 2010 till March 2014, but they were intensively sampled on a daily (24-h) basis during spring (March–May) in 2010 and 2011.

Because the Rupprecht and Patashnick [R&P] Series 5400 (RP5400) carbon monitors are known to have low collection efficiency and high time resolution (3 h), the concentrations measured by this thermal method were scaled using methods developed by U.S. EPA (2001), Green *et al.* (2007) and Quincey *et al.* (2009) with a semi-continuous EC and OC field analyzer (Sunset Laboratory Inc., Oregon, USA). The measurement principle of Sunset Laboratory instruments is similar to DRI instrument (thermal/optical method). Applying a thermal/optical method, both studies compared Sunset Laboratory instruments in the field with RP5400 instruments using the data observed at roadsides in England. Their conversion equation was $EC_{(RP5400)} = (0.22 \pm 0.02)EC_{(sunset)} + (0.88 \pm 0.26)$ ($N = 92$). In our case, the conversion coefficient was calculated by comparing the daily measured raw data obtained from the DRI instrument with the data acquired from the R&P instrument for the periods from March till May of 2010 and 2011 (Shimada *et al.*, 2016). The conversion equation for OC is:

$$Y_{(DRI)} = (2.3 \pm 0.23)X_{(RP5400)} + (0.11 \pm 0.10)$$

and that for TC is:

$$Y_{(DRI)} = (2.3 \pm 0.18) X_{(RP5400)} + (0.16 \pm 0.1)$$

where X is the concentration obtained using RP5400. As mentioned, EC was calculated as TC – OC (Shimada *et al.*, 2016).

Water-soluble organic carbon (WSOC) concentration in PM_{2.5} was analyzed with DRI instrument. The analysis of WSOC concentration was performed in the same manner as for OC and EC concentrations. Five punches of 0.5 cm² were taken from sample filters, and then, WSOC contained in these filters was extracted with ultra-pure water. The extracted solution was absorbed in a heat-treated 0.5-cm² filter. After the filter was dried by infrared light, WSOC concentration was analyzed by DRI instrument.

At CHAAMS, we also measured polycyclic aromatic hydrocarbons (PAHs) contained in filtered aerosol samples collected from 2004 till 2013. Concentrations of retene, one of the molecular markers of biomass burning, were measured. Methods for sampling and analyzing retene were described by Miura *et al.* (2019). We used the same data in Sato *et al.* (2007b), Sato *et al.* (2008), Yoshino *et al.* (2011), Ogawa *et al.* (2012) and Miura *et al.* (2019). The sampling periods and number of samples used in the current study are shown in Table S1. Furthermore, the durations of the seasons were considered to be as follows: March–May for spring, June–August for summer, September–November for autumn, and December–February for winter.

3 RESULTS AND DISCUSSION

3.1 Trends in OC and EC Concentrations from 2004 till 2013

Fig. 1 shows the monthly trends in OC and EC concentrations from 2004 till 2013. The

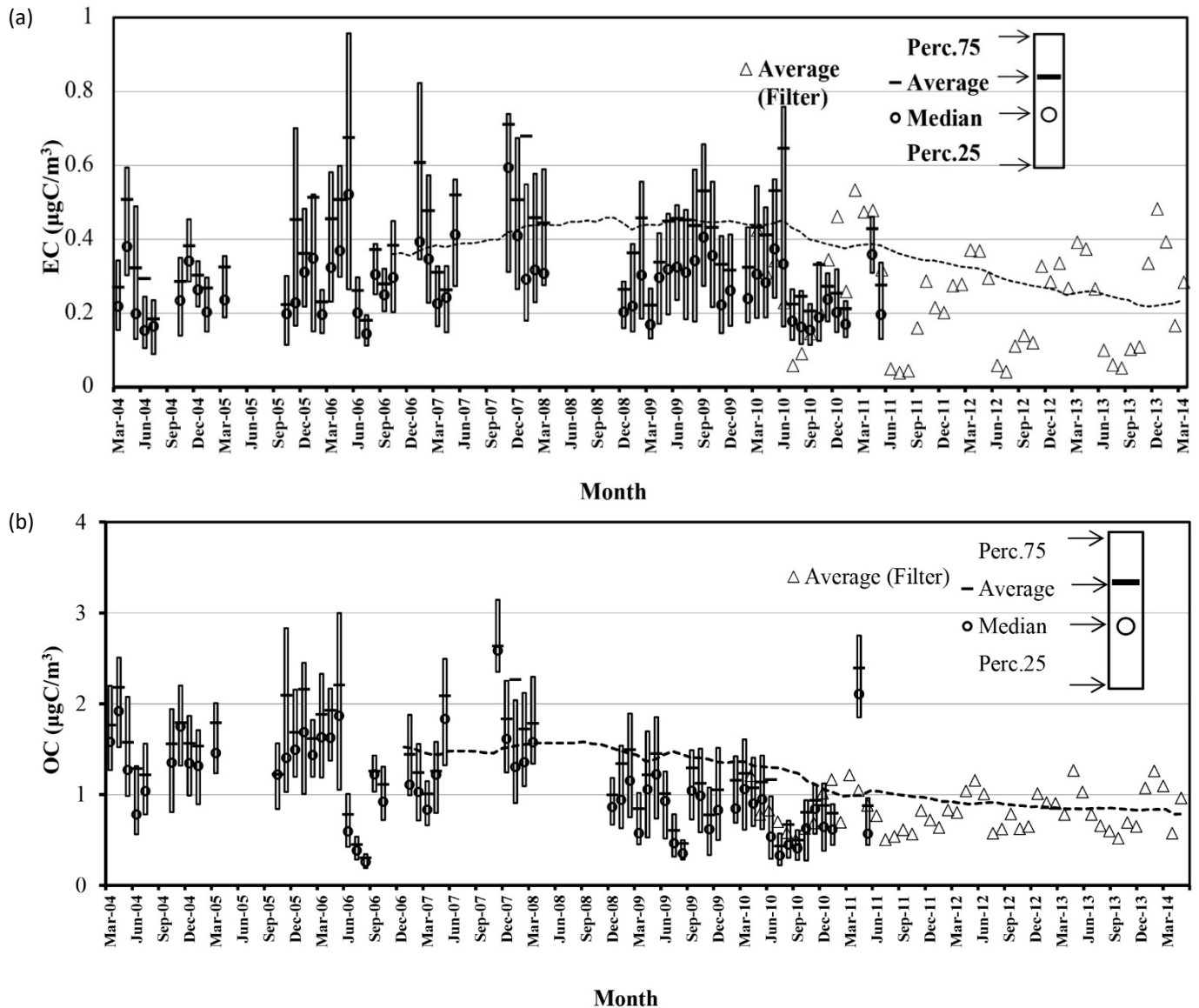
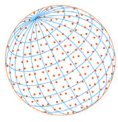


Fig. 1. The trends in monthly concentrations of (a) EC and (b) OC from 2004 till 2014. The dashed line represents a 3-y running mean of monthly averages. The solid bars and circles represent monthly means and median concentrations in box-and-whisker plot (same hereinafter).

concentrations were high in spring and winter, typically because of aerosols transported from China. In our previous studies, we reported high concentrations of $PM_{2.5}$, OC, EC, CO, O_3 , and major ionic components in rainwater during spring and winter and low concentrations during summer at CHAAMS (Shimada *et al.*, 2011). In addition, metallic and ionic species in $PM_{2.5}$ were all high during spring and winter but low during summer (Shimada *et al.*, 2015). The flow of air masses is governed by the wind field, created by pressure gradients as shown in Miura *et al.* (2019) in which the seasonal mean wind fields from 2008 to 2015 are shown. In winter, spring and fall, a high-pressure system is often located over northwestern China and a low-pressure over the western Pacific Ocean. On the other hand, in summer, a high-pressure system is often located over the western Pacific Ocean. Therefore, air pollutants were mainly transported from China during spring and winter. The OC and EC data for summer were insufficient for a detailed analysis to examine the relation between OC and EC trends and emission changes in China, and therefore, we excluded summer data.

As exhibited in Fig. 2, the annual average and median concentrations of EC did not show any significant increase in the period 2004–2014. By contrast, the annual average and median

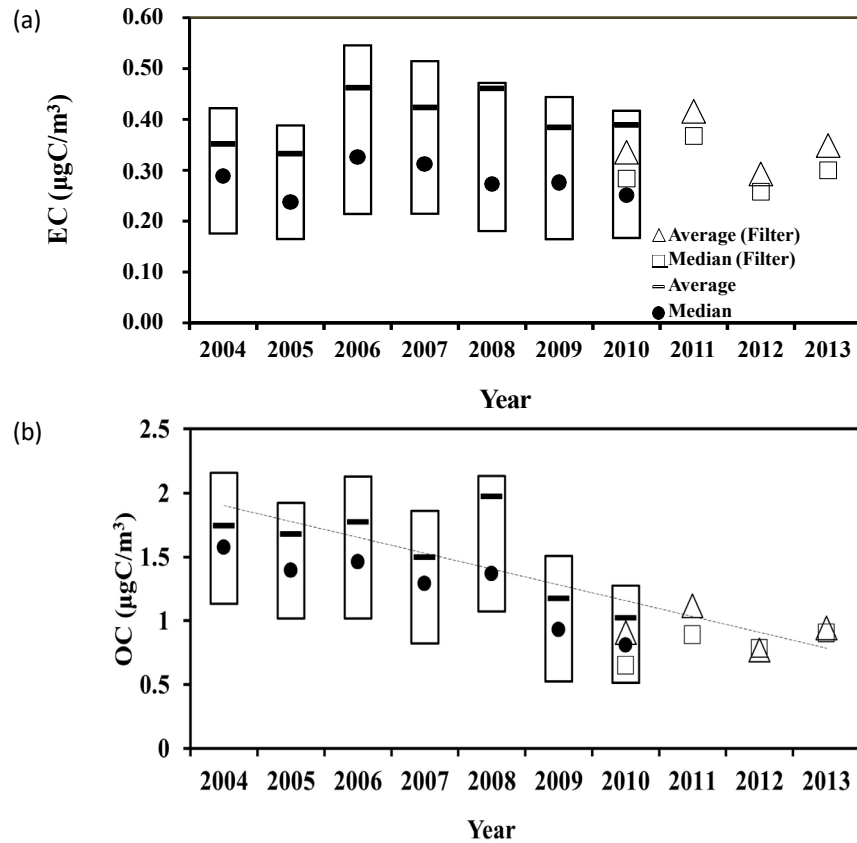
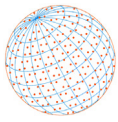


Fig. 2. The annual trends of (a) EC and (b) OC concentrations from 2004 till 2013, excluding summer data. $OC_{(average)}$: $Y = -0.1X + 1.7$ ($\alpha > 0.05$, $R = 0.64$); $OC_{(median)}$: $Y = -0.1X + 2.0$ ($\alpha > 0.05$, $R = 0.84$).

concentrations of OC clearly decreased at the rate of $0.12 \mu\text{g C m}^{-3} \text{y}^{-1}$ ($7\% \text{y}^{-1}$; $R = 0.73$, $\alpha > 0.05$ when 2004 is considered the base year) and $0.10 \mu\text{g C m}^{-3} \text{y}^{-1}$ ($7\% \text{y}^{-1}$; $R = 0.83$), respectively.

The average EC concentrations are typically higher than the median concentrations. Furthermore, the difference between the average and the median concentrations increases at higher concentrations. Several major episodes increased the average EC concentrations in spring and winter. We observed LSP events in winter. The OC and EC concentrations were highest in month of the year when LSP persisted over the CHAAMS 4–6 days in winter (Shimada *et al.*, 2016). Although these observations indicate the importance of episodic events to EC, the difference between average and median values is less pronounced than that for OC (Fig. 1), indicating that the aforementioned episodes affect EC concentration more than OC concentration, even though the latter is higher. To investigate the influence of episodic events on average and median EC concentrations, we analyzed the increase–decrease rates of the annual trends in OC and EC concentrations. Fig. S2 shows the frequency distributions of the quantity (average – median)/median for EC concentration and OC concentration, and Fig. 2 shows the annual trends in OC and EC concentrations from 2004 till 2013. The frequency of EC concentration is predominantly higher than that of OC concentration, and the quantity (average – median)/median for EC concentration varies between 20% and 70%. It was found that the above episodes affect EC concentration rather than OC concentration. Although LSP might influence the annual trend in EC, it does not affect the annual trend in OC.

Fig. 3 shows the annual trends in EC and OC in spring and winter, when air pollutants were frequently transported from China to Japan. The average and median EC concentrations did not show appreciable changes (Figs. 3 and S1), whereas the average and median OC concentrations showed a considerable decrease in both winter and spring. Episodes such as those of LSP did not affect the average and median OC concentrations (Fig. S2), which decreased at the rate of $0.10 \mu\text{g C m}^{-3} \text{y}^{-1}$ ($5\% \text{y}^{-1}$) in spring and $0.11 \mu\text{g C m}^{-3} \text{y}^{-1}$ ($7\% \text{y}^{-1}$) in winter (Figs. 3 and S1). We

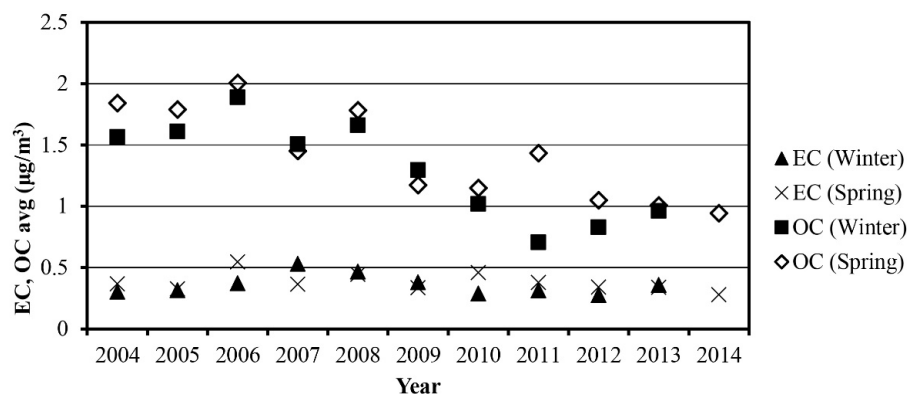
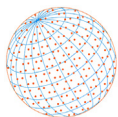


Fig. 3. The annual trends of average (a) EC and (b) OC concentrations in spring and winter from 2004 till 2014. $OC_{(spring)}: Y = -0.1X + 1.9$ ($\alpha > 0.05$, $R = 0.64$); $OC_{(winter)}: Y = -0.1X + 1.9$ ($\alpha > 0.05$, $R = 0.76$).

checked for trends by applying the Mann–Kendall test, which is widely used to verify the significance of tendencies in data sets (Miura *et al.*, 2019). No significant increasing or decreasing trends in EC ($P > 0.05$) were found in any given season over the study period. On the other hand, decreasing trends in OC ($P < 0.05$) were found in spring and winter. The decreasing trend from winter through spring observed at CHAAMS may indicate a reduction in OC emissions in China from the past decade.

3.2 Annual Trends in POC and SOC from 2004 till 2013

Primary emissions of OC are expected to have decreased over the past decade. Here, we examined the secondary formation of OC using the EC-tracer method. According to Turpin and Huntzicker (1991), the contributions of primary organic carbon (POC) and secondary organic carbon (SOC) concentrations can be estimated as:

$$C_{POC} = EC \times (OC/EC)_{min} \quad (1)$$

$$C_{SOC} = OC - C_{POC} \quad (2)$$

where $(OC/EC)_{min}$ is the observed minimum OC/EC ratio. The $(OC/EC)_{min}$ values were determined by using the RP5400 data for spring and winter and taking the average of the 10 minimum OC/EC ratios (Turpin and Huntzicker, 1995) for the period 2004–2010. The obtained values were 2.91 ($R^2 = 0.99$), 2.95 ($R^2 = 0.85$), 1.78 ($R^2 = 0.99$), 0.98 ($R^2 = 0.84$), 1.57 ($R^2 = 0.84$), 0.64 ($R^2 = 0.74$), and 0.67 ($R^2 = 0.90$) for 2004, 2005, 2006, 2007, 2008, 2009, and 2010, respectively. Moreover, for the filtered samples collected in 2010 and 2011, $(OC/EC)_{min}$ was determined as the average value of the two lowest OC/EC ratios in these years. We used OC and EC data from the spring of 2010 and 2011, when intensive daily sampling was conducted throughout the season. The $(OC/EC)_{min}$ values for 2010 and 2011 were 0.95 ($R^2 = 0.83$) and 1.17 ($R^2 = 0.98$), respectively. Because the samples with the minimum OC/EC ratios at CHAAMS might contain SOC, the contribution of SOC could be underestimated by the EC-tracer method. Zhang *et al.* (2008) reported that $(OC/EC)_{min}$ ranged from 0.9 to 2.4 at 18 stations of the China Atmosphere Watch Network (CAWNET) in China in 2006. In our study, the SOC/OC ratio increased from 2004 till 2007 and thereafter stabilized (Fig. 4). It is noteworthy that the use of yearly $(OC/EC)_{min}$ from 2010 to 2013 instead of average $(OC/EC)_{min}$ would only lead to a larger increase in the SOC/OC ratio. This is because SOC/OC data from filter sampling were used for spring of 2010 and 2013. The annual trends of the SOC/OC ratio from 2004 till 2010, excluding summer data from 2004 till 2010, was used from the RP5400 data. For reference, we showed SOC/OC ratios from 2010 till 2013 using the calculated average $(OC/EC)_{min}$ values (1.06) based on 2010 and 2011 data. These ratios were consistently higher (~75%) in rural areas due to reduced contributions of primary OC concentrations.

Moreover, in the current study, the relationship between SOC and WSOC concentrations was found to be $Y = 0.70X + 0.32$ ($R = 0.62$) (Fig. S3). Here, X is the estimated SOC concentration, and

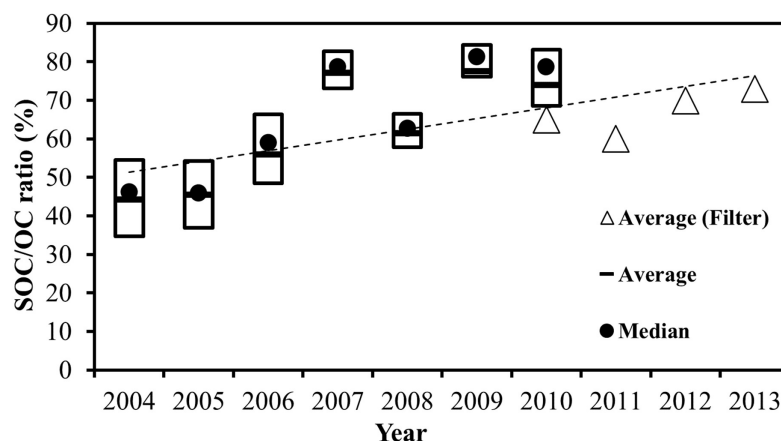
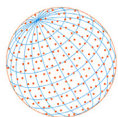


Fig. 4. The annual trends of the SOC/OC ratio from 2004 till 2010, excluding summer data from 2004 till 2010, from RP5400 measurements. SOC/OC data from filter sampling were used for spring of 2010 and 2013. SOC/OC: $Y = 2.8X + 48.7$ ($\alpha > 0.05$, $R = 0.67$).

Y is the measured WSOC concentration. When SOC is produced by a photochemical oxidation reaction as shown below, the main component of SOC is water-soluble. Therefore, SOC and WSOC are highly correlated (Miyazaki *et al.*, 2006). SOC concentration is known to be formed from gas-phase photo-oxidation of VOCs, such as isoprene (Kroll *et al.*, 2006) and monoterpenes (Hatakeyama *et al.*, 1989), and aromatic compounds (Sato *et al.*, 2007a) as well as carbonyl and semi-volatile organic species from anthropogenic emissions (Hatakeyama *et al.*, 1994; Robinson *et al.*, 2007) formed through oxidation by ozone or radicals such as OH and NO₃.

The trend in ozone concentration at CHAAMS from 2001 till 2013 did not show either upward or downward trends (Kato *et al.*, 2014), and therefore, the high SOC content in OC was not due to an increase in the ozone concentration. Huang *et al.* (2015) reported that based on the OC/EC ratios in literature, SOC is increasing in urban Shanghai. Thence, they discussed the contribution of VOC emissions, such as those with high aromatic contents, to secondary organic aerosol (SOA) formation and the relative importance of vehicular emissions. They suggested that gasoline exhaust is an important factor in SOA formation based on the relative contributions of gasoline and diesel vehicles to vehicle-origin organic aerosol. Kurokawa *et al.* (2013) reported that emissions from solvent and paint increased rapidly to 38% in 2008, exceeding those from road transport (35%). Zheng *et al.* (2017) also reported that solvent emissions are estimated to have increased by 52% since 2010. Toluene and C₈ aromatics were the most abundant species, accounting for 76% of the total VOCs emitted from paint applications (Yuan *et al.*, 2010). De Smedt *et al.* (2010) reported that an upward trend of formaldehyde-column abundance was observed over northeastern China (4% y⁻¹) based on data retrieved from the Global Ozone Monitoring Experiment (GOME) and Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instruments between 1997 and 2009. Formaldehyde is one of the important intermediate products from the oxidation of VOCs. They also reported that the detected trend in formaldehyde-column abundance could be explained by changes in the emissions of corresponding anthropogenic non-methane hydrocarbon (NMHC), which were reported by Ohara *et al.* (2007). The trend of formaldehyde can mainly be used as an indicator of tropospheric VOC emissions. It seems that the SOC/OC ratio is increasing at the rate of 2.7% y⁻¹ due to the reduction of OC emission and the change in composition of OC in China.

3.3 OC/EC Ratio and Retene as a Molecular Marker of Biomass Burning

Fig. 5 shows the annual trend in the OC/EC ratio from 2004 till 2013. This ratio is useful for examining the relative importance of sources of carbonaceous aerosols, namely biomass burning and fossil-fuel combustion. The OC/EC ratios of the products of coal combustion, oil combustion, and vehicle emissions, and biomass burning have been suggested to be 3.0 and 12.3, respectively (Bond *et al.*, 2004; Cao *et al.*, 2005), indicating the greater importance of biomass burning. The annual average OC/EC ratio decreased from 5.7 to 2.4, at the rate of 0.28 y⁻¹ (5% y⁻¹; $R = 0.66$,

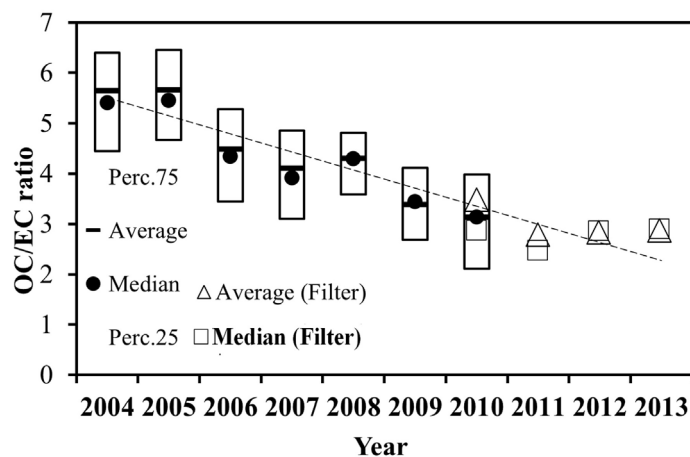
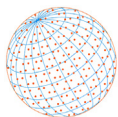


Fig. 5. The annual trends of OC/EC ratios from 2004 till 2013, excluding summer data. OC/EC: $Y = -0.28X + 5.5$ ($\alpha > 0.05$, $R = 0.66$).

$\alpha > 0.05$, with 2004 as the base year). The yearly trend in OC/EC ratios for spring and winter from 2004 till 2013 showed an appreciable decrease (Fig. S4) at the rate of 0.26 y^{-1} ($4\% \text{ y}^{-1}$) in spring and 0.28 y^{-1} ($5\% \text{ y}^{-1}$) in winter. According to the Mann–Kendall test, decreasing trends in OC/EC ratio ($P < 0.05$) were found in spring and winter over the study period, suggesting that the contribution of fossil-fuel combustion increase more than biomass burning to carbonaceous aerosols. Our results agree with the results of Kim *et al.* (2011), who reported a decrease in the OC/EC ratio at Gosan, Korea, from 1992 till 2008. Handa *et al.* (2010) reported that $^{14}\text{C}/^{12}\text{C}$ analysis of the EC indicated a greater contribution from fossil-fuel combustion ($\sim 60\%$) than biomass burning ($\sim 40\%$). When low EC concentrations were observed, $^{14}\text{C}/^{12}\text{C}$ analysis of OC indicated that the contribution of fossil-fuel combustion ($\sim 40\%$) was less than that of biomass burning ($\sim 60\%$). The contribution of OC probably does not reflect biomass burning; rather, this increase may have been caused by SOA derived from biogenic VOCs (Handa *et al.*, 2010).

Moreover, the OC/EC ratios in the air masses that originated in North China and South China were 1.98 ± 0.05 and 2.83 ± 0.03 , respectively, using the same data from 2004 to 2008 (Shimada *et al.*, 2011). OC/EC ratios estimated for North China and South China based on the Asian emission inventory data set were 1–2 and 2–3, respectively (Shimada *et al.*, 2011). Thus, our observational data are in accordance with the emission inventory data. The low OC/EC ratios (< 3) in both North China and South China indicate that the sources of the EC and OC were anthropogenic (Shimada *et al.*, 2011). In order to identify specific source categories for EC and OC using the same data in the air masses from China and Korea, we used Positive Matrix Factorization–Potential Source Contribution Function (PMF–PSCF) analysis results (Shimada *et al.*, 2015), which provided information on air-mass transport. While oil combustion occurred in South China, coal combustion occurred in North China, with high contributions from the former in spring and the latter in winter (Shimada *et al.*, 2015).

The yearly trend in the OC/EC ratio indicates that the contribution to carbonaceous aerosols from biomass burning has decreased at CHAAMS. We also compare the yearly trend in the OC/EC ratio with emission inventories in Section 3.4.

The website of the Acid Deposition Monitoring Network in East Asia (EANET; led by the Ministry of the Environment, Japan) reported the monthly trends of potassium concentration in PM_{10} during spring and winter at Cape Hedo from 2004 till 2012 (<http://www.eanet.asia/>). The main sources of potassium are biomass burning and sea salt. The non-sea-salt potassium (nss-potassium) concentration, obtained from the Na^+ concentration, first increased from 2004 and then decreased, showing a slight overall decrease during the aforementioned period (Fig. 6). At Gosan, it slightly decreased overall from 1992 till 2008 (Kim *et al.*, 2011). The trend at CHAAMS from 2004 till 2008 is in agreement with the observations made at Gosan. Overall, the trend in the potassium concentration supports the observed decrease in the contribution of biomass burning to ambient aerosols. However, due to low potassium concentration, PMF analysis did not show the contribution from biomass burning (Shimada *et al.*, 2015).

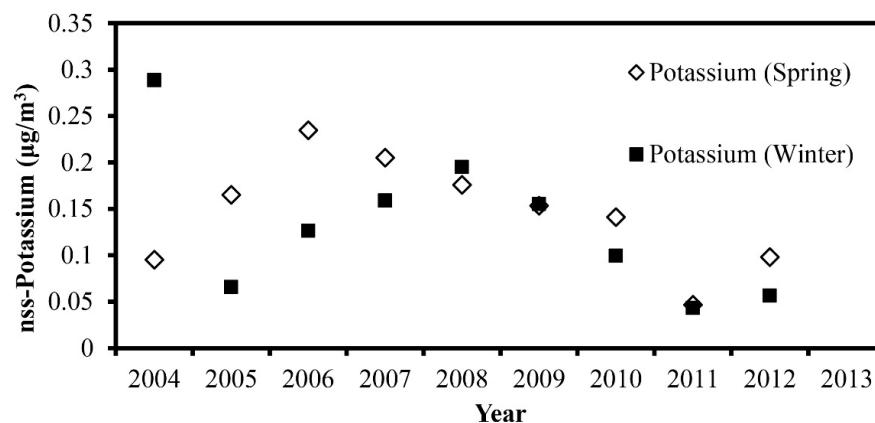
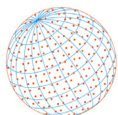


Fig. 6. The annual trends of non-sea-salt potassium concentrations in spring and winter from 2004 till 2013.

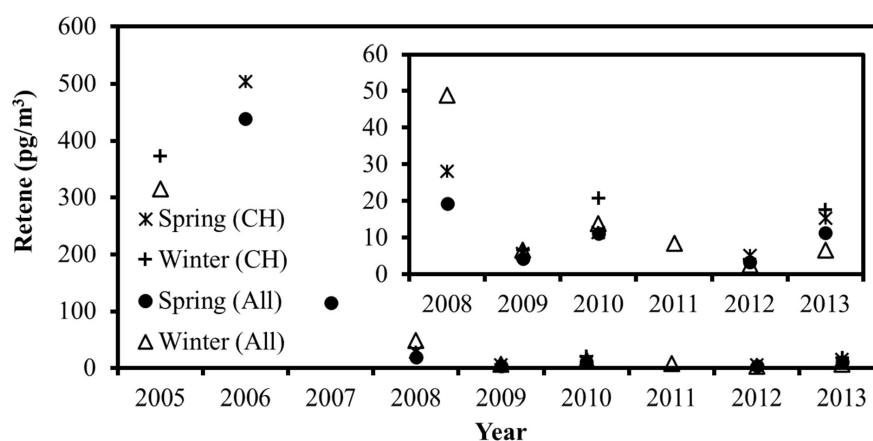


Fig. 7. The annual trends of retene concentrations in spring and winter from 2004 till 2013. *CH* means the fraction of air-mass origins from the area including part of China.

Levoglucosan has often been used as a marker of biomass burning (Simoneit *et al.*, 2002). However, recent findings have reported that its lifetime is rather short, up to 12 h, because of rapid photochemical oxidation in the droplet phase (Teraji *et al.*, 2010) and that it may not be suitable for use as a marker in humid environments. Retene concentration is another marker of biomass burning, especially softwood burning (Ramdahl, 1983; McDonald *et al.*, 2000; Fine *et al.*, 2001; Simoneit, 2002). We monitored retene, a persistent organic pollutant, to examine changes in the biomass-burning source strength. Fig. 7 shows the annual average concentrations of retene in spring and winter from 2004 till 2013. Fig. 7 also shows the annual average concentrations of retene transported from China classified by back-trajectory analysis. Table S2 shows sampling periods and sample number from 2005 to 2013. PAH concentrations at CHAAMS in each season were dominated by PAHs transported from China. PAH concentrations were highest in winter and lowest in spring (Miura *et al.*, 2019; Shimada *et al.*, 2020). We used the data classified as of Chinese origin by back trajectory in these references (Sato *et al.*, 2007b, 2008; Yoshino *et al.*, 2011; Ogawa *et al.*, 2012; Miura *et al.*, 2019). Retene concentrations transported from China dominate the long-term trend of Cape Hedo. In other words, the retene concentrations transported from China determine the long-term trends of retene.

During this period, the retene concentration based on all data sets decreased from 315 pg m^{-3} to 7 pg m^{-3} (in winter) and from 439 pg m^{-3} to 11 pg m^{-3} (in spring), indicating a decrease in the contribution from biomass burning to carbonaceous aerosols.

Examining the OC/EC ratio, nss-potassium concentration, and retene concentration, we found that the contribution from biomass burning to carbonaceous aerosols has decreased at CHAAMS.

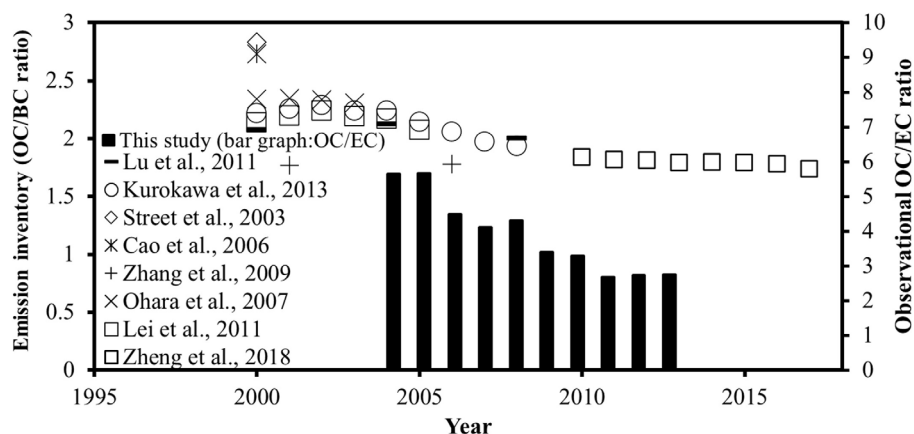
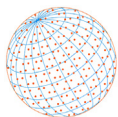


Fig. 8. Comparison of OC/BC emission inventories from China based on references (left axis) with observed OC/EC concentrations (bar graph) at CHAAMS in this study (right axis).

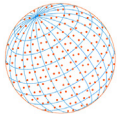
3.4 Comparison of the Annual Trends in OC and EC with Emission Inventories

The annual trends in the OC and EC concentrations at CHAAMS from 2004 till 2008 and 2010 to 2017 were also quite consistent with changes in the emission inventories in China reported by Kurokawa *et al.* (2013) and Zheng *et al.* (2018). The annual trend in the OC/BC ratio in emissions in China decreased during this period, consistent with the decrease in the OC/EC ratio at CHAAMS (Fig. 8). Carbonaceous aerosol emissions in China originate mainly from residential biofuel and coal combustion. According to the Regional Emission Inventory in Asia (REAS; version 2.1; Kurokawa *et al.*, 2013) and Multi-resolution Emission Inventory for China (MEIC; Zheng *et al.*, 2018), due to a reduction in biofuel consumption in China, BC emissions have increased since 2000, and OC emissions have decreased since 2006. Kurokawa *et al.* (2013) and Zheng *et al.* (2018) reported that more than half of the total BC emissions in China during 2000 were from the domestic sector (e.g., biofuel consumption). BC emissions from this sector have decreased since 2006, and industrial emissions reached levels equal to domestic emissions in 2008 (with each category contributing approximately 45% to the total BC emissions). The road-transport sector has become more important, with a share of 11% (Kurokawa *et al.*, 2013). Kurokawa *et al.* (2013) reported that OC emissions decreased by 16% between 2006 and 2008.

It is noteworthy that Kurokawa *et al.* (2013) and Zheng *et al.* (2018) considered biofuel consumption such as wood burning but not open biomass burning in the emission inventory. Lu *et al.* (2010) described the overall trend in the annual change in emissions from open biomass burning in China from 2004 till 2010 on the basis of the Global Fire Emissions Database (GFED; version 3.1), reporting that OC emissions from open biomass burning of forests and savannas vary from year to year. The emissions are largely dependent on the amount of precipitation and soil moisture. Although the emission factor is highly uncertain, OC emissions from this source type account for 2–11% of the total emissions in China, depending on the year. Li *et al.* (2009) reported that the OC emission factor varies in the range 0.85–3.97 g kg⁻¹ among different types of biofuels. OC emission also depends on the moisture content of biomass fuels (Venkataraman *et al.*, 2005). In a laboratory experiment, OC emission factors of fuelwood were observed to be approximately 0.4 g kg⁻¹ at low burn rates and as high as 2.7 g kg⁻¹ at high burn rates (Venkataraman *et al.*, 2005). In general, the contribution of open burning to OC emissions is considered low. Therefore, we believe that a decrease in biofuel consumption in China caused the decreasing trend in OC concentration observed at CHAAMS. The annual trend in the OC/EC ratio at CHAAMS, and the trend in the OC/BC ratio in emissions in the study by Kurokawa *et al.* (2013) and Zheng *et al.* (2018) (Fig. 8) consistently show the large contribution of fossil-fuel combustion to carbonaceous aerosols at CHAAMS.

4 CONCLUSION

This study assessed the levels of the carbonaceous aerosols at Cape Hedo from 2004 till 2014.



Using an ambient-carbon-particulate monitor, we measured the OC and EC concentrations in these aerosols every 3 h between March 2004 and May 2011. Additionally, following the IMPROVE protocol, we collected samples of the carbonaceous aerosols on quartz filters between March 2010 and March 2014 and quantified the carbon content with a DRI Model 2001 thermal/optical carbon analyzer.

The annual EC concentration exhibited a stable trend from 2004 till 2014. However, both the annual average and median OC concentrations decreased at a rate of $0.10 \mu\text{g C m}^{-3} \text{y}^{-1}$ during this period owing to the reduction in OC emissions in China over the past decade. Specifically, the levels of OC dropped at rates of $0.10 \mu\text{g C m}^{-3} \text{y}^{-1}$ and $0.11 \mu\text{g C m}^{-3} \text{y}^{-1}$ in spring and winter, respectively, during which air pollution from China frequently arrived in Japan.

The annual average of the OC/EC ratio, which can be used to estimate the contributions of biomass burning and fossil-fuel combustion to carbonaceous aerosols, decreased from 5.7 in 2004 to 2.4 in 2014, or at a rate of 0.28y^{-1} , indicating a reduction in the relative contribution of biomass burning. Comparing the BC to the OC emission-inventory data in REAS v.2.1 and MEIC, we found that the declining OC concentrations observed at CHAAMS can be attributed to the remarkable decrease in biofuel consumption in China.

ACKNOWLEDGMENTS

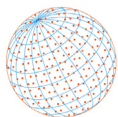
This study was partly supported by a Grant-in-Aid for Scientific Research in Innovative Areas (No. 4003, Impacts of Aerosols in East Asia on Plants and Human Health) from the Ministry of Education, Culture, Sports, Science and Technology, Japan, by a Sasagawa Scientific Research Grant from the Japan Science Society, and by the Global Environment Research Fund of the Japanese Ministry of the Environment (2-1403).

SUPPLEMENTARY MATERIAL

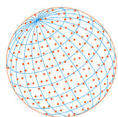
Supplementary material for this article can be found in the online version at <https://doi.org/10.4209/aaqr.200505>

REFERENCES

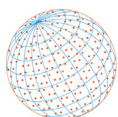
- Bond, T.C., Streets, D.G., Yarber, K.F., Nelson, S.M., Woo, J.H., Klimont, Z. (2004). A technology-based global inventory of black and organic carbon emissions from combustion. *J. Geophys. Res.* 109, D14203 <https://doi.org/10.1029/2003JD003697>
- Bower, S.A., Susan, M.L., Stefan, F.G., Claus, W.B. (2009). Interior pathways of the North Atlantic meridional overturning circulation. *Nature* 459, 243–247. <https://doi.org/10.1038/nature07979>
- Cao, G.L., Zhang, X.Y., Zheng, F.C. (2006). Inventory of black carbon and organic carbon emissions from China. *Atmos. Environ.* 40, 6516–6527. <https://doi.org/10.1016/j.atmosenv.2006.05.070>
- Chow, J.C., Watson, J.G., Crow, D., Lowenthal, D.H., Merrifield, T. (2001). Comparison of IMPROVE and NIOSH carbon measurements. *Aerosol Sci. Technol.* 34, 23–34. <https://doi.org/10.1080/02786820119073>
- De Smedt, I., Stavrakou, T., Müller, J.F., van der A, R.J., Van Roozendaal, M. (2010). Trend detection in satellite observations of formaldehyde tropospheric columns. *Geophys. Res. Lett.* 37, L18808. <https://doi.org/10.1029/2010GL044245>
- Eleftheriadis, K., Vratolis, S., Nyeki, S. (2009). Aerosol black carbon in the European Arctic: Measurements at Zeppelin station, Ny-Ålesund, Svalbard from 1998-2007. *Geophys. Res. Lett.* 36, L02809. <https://doi.org/10.1029/2008GL035741>
- Fine, P.M., Cass, G.R., Simoneit, B.R.T. (2001). Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the northeastern United States. *Environ. Sci. Technol.* 35, 2665. <https://doi.org/10.1021/es001466k>
- Green, D., Alexander, J., Fuller, G., Quincey, P., Butterfield, D. (2007). Marylebone Road Aethalometer Trial Report. UK-AIR: Air Information. https://uk-air.defra.gov.uk/assets/documents/reports/cat13/0711261353_Marylebone_Road_Aethalometer_Trial.pdf



- Hand, J.L., Schichtel, B.A., Malm, W.C., Frank, N.H. (2013). Spatial and temporal trends in PM_{2.5} organic and elemental carbon across the United States. *Adv. Meteorol.* 2013, 367674. <https://doi.org/10.1155/2013/367674>
- Hatakeyama, S., Akimoto, H. (1994). Reactions of Criegee intermediates in the gas phase. *Res. Chem. Intermed.* 20, 503–524. <https://doi.org/10.1163/156856794X00432>
- Hatakeyama, S., Izumi, K., Fukuyama, T., Akimoto, H. (1989). Reactions of ozone with α -pinene and ϵ pinene in air: Yields of gaseous and particulate products. *J. Geophys. Res.* 94, 13013–13024. <https://doi.org/10.1029/JD094iD10p13013>
- Hirdman, D., Burkhardt, F.J., Sodemann, H., Eckhardt, S., Jefferson, A., Quinn, K.P., Sharma, S., Strom, J., Stohl, A. (2010). Long-term trends of black carbon and sulphate aerosol in the Arctic: Changes in atmospheric transport and source region emissions. *Atmos. Chem. Phys.* 10, 12133–12184. <https://doi.org/10.5194/acp-10-9351-2010>
- Huang, C., Wang, H.L., Li, L., Wang, Q., Lu, Q., de Gouw, J.A., Zhou, M., Jing, S.A., Lu, J., Chen, C.H. (2015). VOC species and emission inventory from vehicles and their SOA formation potentials estimation in Shanghai, China. *Atmos. Chem. Phys.* 15, 11081–11096. <https://doi.org/10.5194/acp-15-11081-2015>
- Kato, S., Jeeranut, S., Kajii, Y., Shiroma, T., Iwasaki, A., Kadena, H., Tomoyose, N., Toguchi, T. (2014). Long term observation of CO, O₃, VOC at cape Hedo, Okinawa. 55th Annual meeting of the Japan Society for Atmospheric Environment, Japan. (in Japanese)
- Kim, N.K., Kim, P.Y., Kang, C.H. (2011). Long-term trend of aerosol composition and direct radiative forcing due to aerosols over Gosan: TSP, PM₁₀, and PM_{2.5} data between 1992 and 2008. *Atmos. Environ.* 45, 6107–6115. <https://doi.org/10.1016/j.atmosenv.2011.08.051>
- Kirchstetter, T.W., Aguiar, J., Tonse, S., Fairley, D., Novakov, T. (2008). Black carbon concentrations and diesel vehicle emission factors derived from coefficient of haze measurements in California: 1967–2003. *Atmos. Environ.* 42, 480–491. <https://doi.org/10.1016/j.atmosenv.2007.09.063>
- Kondo, Y., Matsui, H., Moteki, N., Sahu, L., Takegawa, N., Kajino, M., Zhao, Y., Cubison, J.M., Jimenez, L.J., Vay, S., Diskin, S.G., Anderson, B., Wisthaler, A., Mikoviny, T., Fuelberg, E.H., Blake, R.D., Huey, G., Weinheimer, J.A., Knapp, J.D., Brune, H.W. (2011). Emissions of black carbon, organic, and inorganic aerosols from biomass burning in North America and Asia in 2008. *J. Geophys. Res.* 116, D08204. <https://doi.org/10.1029/2010JD015152>
- Kroll, J.H., Ng, N.L., Murphy, S.M., Flagan, R.C., Seinfeld, J.H. (2006). Secondary organic aerosol formation from isoprene photooxidation. *Environ. Sci. Technol.* 40, 1869–1877. <https://doi.org/10.1021/es0524301>
- Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., Akimoto, H. (2013). Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2. *Atmos. Chem. Phys.* 13, 11019–11058. <https://doi.org/10.5194/acp-13-11019-2013>
- Lei, Y., Zhang, Q., He, K.B., Streets, D.G. (2011). Primary anthropogenic aerosol emission trends for China, 1990–2005. *Atmos. Chem. Phys.* 11, 931–954. <https://doi.org/10.5194/acp-11-931-2011>
- Lu, Z., Streets, G.D., Zhang, Q., Wang, S., Carmichael, R.G., Cheng, F.Y., Wei, C., Chin, M., Diehl, T., Tan, Q. (2010). Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. *Atmos. Chem. Phys.* 10, 6311–6331. <https://doi.org/10.5194/acp-10-6311-2010>
- Lu, Z., Zhang, Q., Streets, D.G. (2011). Sulfur dioxide and primary carbonaceous aerosol emissions in China and India, 1996–2010. *Atmos. Chem. Phys.* 11, 9839–9864. <https://doi.org/10.5194/acp-11-9839-2011>
- Matsui, H., Kondo, Y., Moteki, N., Takegawa, N., Sahu, K.L., Zhao, Y., Fuelberg, E., Sessions, R.W., Diskin, G., Blake, R.D., Wisthaler, A., Koike, M. (2011). Seasonal variation of the transport of black carbon aerosol from the Asian continent to the Arctic during the ARCTAS aircraft campaign. *J. Geophys. Res.* 116, D05202. <https://doi.org/10.1029/2010JD015067>
- McDonald, J.D., Zielinska, B., Fujita, E.M., Sagebiel, J.C., Chow, J.C., Watson, J.G., (2000). Fine particle and gaseous emission rates from residential wood combustion. *Environ. Sci. Technol.* 34, 2080–2091. <https://doi.org/10.1021/es9909632>
- Miura, K., Shimada, K., Sugiyama, T., Sato, K., Takami, A., Chan, C.K., Kim, I.S., Kim, Y.P., Lin, N.H., Hatakeyama, S., (2019). Seasonal and annual changes in PAH concentrations in a remote site in the Pacific Ocean. *Sci. Rep.* 9, 12591. <https://doi.org/10.1038/s41598-019-47409-9>
- Miyazaki, Y., Kondo, Y., Takegawa, N., Komazaki, Y., Fukuda, M., Kawamura, K., Mochida M.,



- Okuzawa K., Weber, R.J. (2006). Time-resolved measurements of water-soluble organic carbon in Tokyo. *J. Geophys. Res.*, 111, D23206. <https://doi.org/10.1029/2006JD007125>
- Murphy, D.M., Chow, J.C., Leibensperger, E.M., Malm, W.C., Pitchford, M., Schichtel, B.A., Watson, J.G., White, W.H. (2011). Decreases in elemental carbon and fine particle mass in the United States. *Atmos. Chem. Phys.* 11, 4679–4686. <https://doi.org/10.5194/acp-11-4679-2011>
- Nakajima, T., Yoon, C.S., Ramanathan, V., Shi, Y.G., Takemura, T., Higurashi, A., Takamura, T., Aoki, K., Sohn, J.B., Kim, W.S., Tsuruta, H., Sugimoto, N., Shimizu, A., Tanimoto, H., Sawa, Y., Lin, H.N., Lee, T.C., Goto, D., Schutgens, N. (2007). Overview of the Atmospheric Brown Cloud East Asian Regional Experiment 2005 and a study of the aerosol direct radiative forcing in east Asia. *J. Geophys. Res.*, 112, D24S91. <https://doi.org/10.1029/2007JD009009>
- Ogawa, Y., Kaneyasu, N., Sato, K., Takami, A., Hayashi, M., Hara, K., Hatakeyama, S. (2012). PAHs and n-Alkanes transported long-range. *J. Jpn. Soc. Atmos. Environ.* 47, 18–25. <https://doi.org/10.11298/taiki.47.18>
- Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T. (2007). An Asian emission inventory of anthropogenic emission sources for the period of 1980–2020. *Atmos. Chem. Phys.* 7, 4419–4444. <https://doi.org/10.5194/acp-7-4419-2007>
- Quincey, P., Butterfield, D., Green, D., Coyle, M., Cape, N.J. (2009). An evaluation of measurement methods for organic, elemental and black carbon in ambient air monitoring sites. *Atmos. Environ.* 43, 5085–5091. <https://doi.org/10.1016/j.atmosenv.2009.06.041>
- Ramana, M.V., Ramanathan, V., Feng, Y., Yoon, C.S., Kim, W.S., Carmichael, R.G., Schauer, J.J. (2010). Warming influenced by the ratio of black carbon to sulphate and the black-carbon source. *Nat. Geosci.* 3, 542–545. <https://doi.org/10.1038/ngeo918>
- Ramdahl, T. (1983). Retene - A molecular marker of wood combustion in ambient air. *Nature* 306, 580–582. <https://doi.org/10.1038/306580a0>
- Robinson, A.L., Donahue, N.M., Shrivastava, M.K., Weitkamp, E.A., Sage, A.M., Grieshop, A.P., Lane, T.E., Pierce, J.R., Pandis, S.N. (2007). Rethinking organic aerosols: Semivolatile emissions and photochemical aging. *Science* 315, 1259–1262. <https://doi.org/10.1126/science.1133061>
- Sato, K., Hatakeyama, S., Imamura, T. (2007a). Secondary organic aerosol formation during the photooxidation of toluene: NO_x dependence of chemical composition. *J. Phys. Chem. A* 111, 9796–9808. <https://doi.org/10.1021/jp071419f>
- Sato, K., Tanaka, Y., Li, H., Ogawa, S., Hatakeyama, S. (2007b). Distributions and seasonal changes of organic aerosols at Cape Hedo, Okinawa: Polycyclic aromatic hydrocarbons observed during 2005–2006. *Chikyukagaku* 41, 145–153. <https://doi.org/10.14934/chikyukagaku.41.145> (in Japanese)
- Sato, K., Li, H., Tanaka, Y., Ogawa, S., Iwasaki, Y., Takami, A., Hatakeyama, S. (2008). Long-range transport of particulate polycyclic aromatic hydrocarbons at Cape Hedo remote island site in the East China Sea between 2005 and 2008. *J. Atmos. Chem.* 61, 243–257. <https://doi.org/10.1007/s10874-009-9135-4>
- Shimada, K., Takami, A., Kato, S., Yoshizumi, K., Hatakeyama, S. (2011). Variation of carbonaceous aerosol in polluted air mass transported from East Asia and evaluation of their source origin. *J. Jpn. Soc. Atmos. Environ.* 46, 1–9. <https://doi.org/10.11298/taiki.46.1> (in Japanese)
- Shimada, K., Shimada, M., Takami, A., Hasegawa, S., Fushimi, A., Arakaki, T., Watanabe, I., Hatakeyama, S. (2015). Mode and place of origin of carbonaceous aerosols transported from east Asia to Cape Hedo, Okinawa, Japan. *Aero. Air Quality Res.* 15, 799–813. <https://doi.org/10.4209/aaqr.2014.09.0190>
- Shimada, K., Takami, A., Kato, S., Yoshizumi, K., Hasegawa, S., Fushimi, A., Shimizu, A., Sugimoto, N., Chan, K.C., Kim, P.Y., Lin, H.N., Hatakeyama, S. (2016). Characteristics of carbonaceous aerosols in large-scale Asian wintertime outflows at Cape Hedo, Okinawa, Japan. *J. Aerosol Sci.* 100, 97–107. <https://doi.org/10.1016/j.jaerosci.2016.05.006>
- Shimada, K., Nohchi, M., Yang, X., Sugiyama, T., Miura, K., Takami, A., Sato, K., Chen, X., Kato, S., Kajii, Y., Meng, F., Hatakeyama, S. (2020). Degradation of PAHs during long range transport based on simultaneous measurements at Tuoji Island, China, and at Fukue Island and Cape Hedo, Japan. *Environ. Pollut.* 260, 113906. <https://doi.org/10.1016/j.envpol.2019.113906>
- Shindell, D., Kuylensstierna, J.C., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z., Anenberg, S.C., Muller, N., Janssens-Maenhout, G., Raes, F., Schwartz, J., Faluvegi, G., Pozzoli, L., Kupiainen, K., Hoglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Hicks, K., Oanh, N.T., Milly,



- G., *et al.* (2012). Simultaneously mitigating near-term climate change and improving human health and food security. *Science* 335, 183–189. <https://doi.org/10.1126/science.1210026>
- Simoneit, B.R.T. (2002). Biomass burning-A review of organic tracers for smoke from incomplete combustion. *Appl. Geochem.* 17, 129–162. [https://doi.org/10.1016/S0883-2927\(01\)00061-0](https://doi.org/10.1016/S0883-2927(01)00061-0)
- Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S.D., Fu, Q., He, D., Klimont, Z., Nelson, S.M., Tsai, N.Y., Wang, M.Q., Woo, J.H., Yarber, K.F. (2003). An inventory of 20 gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res.* 108, 8809. <https://doi.org/10.1029/2002JD003093>
- Takami, A., Miyoshi, T., Shimono, A., Hatakeyama, S. (2005). Chemical composition of fine aerosol measured by AMS at Fukue Island, Japan during APEX priod. *Atmos. Environ.* 39, 4913–4924. <https://doi.org/10.1016/j.atmosenv.2005.04.038>
- Takami, A., Miyoshi, T., Shimono, A., Kaneyasu, N., Kato, S., Kajii, Y., Hatakeyama, S. (2007) Transport of anthropogenic aerosols from Asia and subsequent chemical transformation. *J. Geophys. Res.* 112, D22S31. <https://doi.org/10.1029/2006JD008120>
- Takiguchi, Y., Takami, A., Sadanaga, Y., Lun, X., Shimizu, A., Matsui, I., Sugimoto, N., Wang, W., Bandow, H., Hatakeyama, S. (2008). Transport and transformation of total reactive nitrogen over the East China Sea. *J. Geophys. Res.* 113, D10306. <https://doi.org/10.1029/2007JD009462>
- Teraji, T., Arakaki, T. (2010). Bimolecular rate constants between levoglucosan and hydroxyl radical: Effects of pH and temperature. *Chem. Lett.* 39, 900–901. <https://doi.org/10.1246/cl.2010.900>
- Turpin, B.J., Huntzicker, J.J. (1995). Identification of secondary organic aerosol episodes and quantitation of primary and secondary organic aerosol concentrations during SCAQS. *Atmos. Environ.* 29, 3527–3544. [https://doi.org/10.1016/1352-2310\(94\)00276-Q](https://doi.org/10.1016/1352-2310(94)00276-Q)
- U.S. Environmental Protection Agency (U.S. EPA) (2001). Environmental Technology Verification Report – ETV Advanced Monitoring Systems Center – Rupprecht & Patashnick, Co. Series 5400 Ambient Particulate Carbon Monitor. U.S. Environmental Protection Agency. https://archive.epa.gov/nrmrl/archive-etv/web/pdf/01_vr_rupp_5400c.pdf
- Venkataraman, C., Habib, G., Eiguren-Fernandez, A., Miguel, A.H., Fiedlander, S.K. (2005). Residential biofuels in South Asia: Carbonaceous aerosols emissions and climate impacts. *Science* 307, 1454–1456. <https://doi.org/10.1126/science.1104359>
- Verma, R.L., Kondo, Y., Oshima, N., Matsui, H., Kita, K., Sahu, L.K., Kato, S., Kajii, Y., Takami, A., Miyakawa, T. (2011). Seasonal variations of the transport of black carbon and carbon monoxide from the Asian continent to the western Pacific in the boundary layer. *J. Geophys. Res.* 116, D21307. <https://doi.org/10.1029/2011JD015830>
- Weber, R.J., Sullivan, A.P., Peltier, R.E., Russell, A., Yan, B., Zheng, M., de Gouw, J., Warneke, C., Brock, C., Holloway, J.S., Atlas, E.L., Edgerton, E. (2007). A study of secondary organic aerosol formation in the anthropogenic influenced southeastern United States. *J. Geophys. Res.* 112, D1330. <https://doi.org/10.1029/2007JD008408>
- Yoshino, A., Nakayama, H., Ogawa, Y., Sato, K., Takami, A., Hatakeyama, S. (2011). Long-Range transport of polycyclic aromatic hydrocarbons from east Asia at Cape Hedo in 2010. *Eurozoru Kenkyu* 26, 307–314. <https://doi.org/10.11203/jar.26.307> (in Japanese)
- Yuan, B., Shao, M., Lu, S.H., Wang, B. (2010). Source profiles of volatile organic compounds associated with solvent use in Beijing, China. *Atmos. Environ.* 44, 1919–1926. <https://doi.org/10.1016/j.atmosenv.2010.02.014>
- Zhang, X.Y., Wang, Y.Q., Zhang, X.C., Guo, W., Gong, S.L. (2008). Carbonaceous aerosol composition over various regions of China during 2006. *J. Geophys. Res.* 113, D14111. <https://doi.org/10.1029/2007JD009525>
- Zhang, X., Streets, G.D., Carmichael, R.G., He, B.K., Huo, H., Kannari, A., Klimont, Z., Park, S.I., Reddy, S., Fu, S.J., Chen, D., Duan, L., Lei, Y., Wang, T.L., Yao, L.Z. (2009). Asian emissions in 2006 for the NASA INTEX-B mission. *Atmos. Chem. Phys.* 9, 5131–5153. <https://doi.org/10.5194/acp-9-5131-2009>
- Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L., Zhang, Y., Zhao, H., Zheng, Y., He, K., Zhang, Q. (2018). Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions. *Atmos. Chem. Phys.* 18, 14095–14111. <https://doi.org/10.5194/acp-18-14095-2018>