



Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in east Asia

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Received: 1 July 2016 – Discussion started: 8 July 2016

Revised: 29 March 2017 – Accepted: 14 April 2017 – Published: 12 May 2017

Abstract. Ground-based measurements of black carbon (BC) were performed near an industrial source region in the early summer of 2014 and at a remote island in Japan in the spring of 2015. Here, we report the temporal variations in the transport, size distributions, and mixing states of the BC-containing particles. These particles were characterized using a continuous soot monitoring system, a single particle soot photometer, and an aerosol chemical speciation monitor. The effects of aging on the growth of BC-containing particles were examined by comparing the ground-based observations between the near-source and remote island sites. Secondary formation of sulfate and organic aerosols strongly affected the increases in BC coating (i.e., enhancement of cloud condensation nuclei activity) with air mass aging from the source to the outflow regions. The effects of wet removal on BC microphysics were elucidated by classifying the continental outflow air masses depending on the enhancement ratios of BC to CO ($\Delta\text{BC} / \Delta\text{CO}$), which were used as an indicator of the transport efficiency of BC. It was found that $\Delta\text{BC} / \Delta\text{CO}$ ratios were controlled mainly by the wet removal during transport in the planetary boundary layer (PBL) on the timescale of 1–2 days. The meteorological conditions and backward trajectory analyses suggested that air masses strongly affected by wet removal originated mainly from a region in southern China (20–35° N) in the spring of 2015.

Removal of large and thickly coated BC-containing particles was detected in the air masses that were substantially affected by the wet removal in the PBL, as predicted by Köhler theory. The size and water solubility of BC-containing particles in the PBL can be altered by the wet removal as well as the condensation of non-BC materials.

1 Introduction

Black carbon (BC)-containing particles in the atmosphere can significantly affect the radiative budget of the Earth through two effects: direct (light absorption and scattering) and indirect (aerosol–cloud interactions) effects (Bond et al., 2013; references therein). The difficulty in the estimation of these effects in the atmosphere results from both the short lifetime relative to other greenhouse gases and the variable physicochemical properties of BC-containing particles. The BC itself is water-insoluble immediately after emission, but it subsequently exhibits increased hygroscopicity (McMeeking et al., 2011) and cloud condensation nuclei (CCN) activity (Kuwata et al., 2007) through atmospheric transport and aging. Only small amounts of water-soluble materials on BC particles are needed to cause their activation to form cloud droplets under moderate supersaturation conditions (Kuwata

et al., 2007, 2009). It is considered that BC-containing particles are removed from the atmosphere mainly by wet deposition (Seinfeld and Pandis, 2006).

The horizontal and vertical distributions of aerosols can be substantially altered by their atmospheric lifetimes (e.g., Lawrence et al., 2007). Moreover, Lawrence et al. (2007) suggested that the removal processes of BC such as dry deposition, below-cloud processes (i.e., washout), and in-cloud processes (i.e., rainout) can greatly change the atmospheric lifetimes. The in-cloud processes include nucleation scavenging and scavenging by the preexisting cloud droplets. Precipitation followed by in-cloud processes leads to the irreversible removal of BC-containing particles. Samset et al. (2014), using multiple global model data sets constrained by aircraft observations, suggested that the atmospheric lifetime of BC largely affects its distribution, especially in the Northern Hemisphere, and this results in significant variations in global direct radiative forcing values. The removal of BC has been considered an important issue for the geochemical carbon cycle as well as for climate science. The BC-containing particles deposited onto the ocean surface can affect ocean surface particles, dissolved organic carbon (DOC), and microbial processes by absorbing DOC, stimulating particle aggregation, and changing the size distribution of suspended particles (Mari et al., 2014).

Previous modeling studies have dealt with BC aging processes (condensational growth and coagulation) in box and regional-scale models and parameterized timescales for the conversion of BC-containing particles from water-insoluble to -soluble in global models (Oshima et al., 2009; Liu et al., 2011; Oshima and Koike, 2013). However, quantitative knowledge of the variability in microphysical parameters of BC-containing particles and the timescale of their aging processes is still limited, and thus more investigation is needed for near-source and remote regions (Samset et al., 2014). Moteki et al. (2012) reported the first observational evidence of the size-dependent activation of BC in air masses uplifting from the planetary boundary layer (PBL) to the free troposphere (FT) in east Asia in the spring of 2009, as the part of the Aerosol Radiative Forcing in east Asia (A-FORCE) aircraft campaigns (Oshima et al., 2012). A similar altitude dependence of the BC size distribution and similarity in the BC mixing state were observed in other aircraft measurements conducted in east Asia in winter (Kondo et al., 2016). Selective removal of larger BC-containing particles through cloud processing, which is predicted by Köhler theory, was qualitatively observed in the atmosphere. This observational evidence indicates that the size distributions and mixing states of BC-containing particles have a large impact on the global- and regional-scale spatial distributions of BC through their upward transport from the PBL to the FT associated with cloud processes. Despite the importance of the size distributions and mixing states of BC-containing particles in the PBL, the measurements of their microphysical properties are still limited around the source regions in east Asia.

Kanaya et al. (2016) have conducted long-term measurements of BC for 6 years (2009–2015) at Fukue Island, and they reported the emission and removal of BC in east Asia using these data sets. This study determined that the transport efficiency of BC aerosol particles through the PBL was substantially reduced by wet removal. Here, we examine the effects of aging and wet removal during transport on the changes in BC size distributions and mixing state, as well as concentrations, based on ground-based measurements conducted at the same site in the spring of 2015 using a single particle soot photometer (SP2) and an aerosol chemical speciation monitor (ACSM). We first describe the meteorological characteristics of the east Asian region in the spring of 2015. Then, we discuss the relative importance of the below-cloud (i.e., washout) and in-cloud scavenging (i.e., rainout) processes for the removal of BC as well as the transport patterns of the east Asian outflow air masses in spring. The loss of BC-containing particles for that period is investigated using a similar approach to that used by Kanaya et al. (2016), and this is performed in connection with the associated changes in BC microphysics and their relevance to the transport pathways.

2 Experimental and data analysis

2.1 Atmospheric observations

Continuous measurements of PM_{2.5} and BC aerosols have been conducted at a remote island, Fukue Island, since February 2009 (Kanaya et al., 2013; Ikeda et al., 2014). The observation site is located at the Fukue Island Atmospheric Environment Monitoring Station (32.75° N, 128.68° E; Fig. 1). The site is located in the northwest portion of Fukue Island, approximately 20 km from the main residential area in the southeast. The PM_{2.5} aerosols sampled at the site are mostly transported from areas beyond the island. The enhanced concentrations of BC aerosols in Fukue Island can be mainly attributed to long-range transport from the Asian continent, according to a previous study (Shiraiwa et al., 2008) and an emission inventory work (Fig. 1; REAS ver. 2.1; Kurokawa et al., 2013).

We deployed an SP2 (Droplet Measurement Technologies, Inc., USA) for the analysis of microphysical parameters of refractory BC (rBC; Petzold et al., 2013) from 26 March 2015 to 14 April 2015. The SP2 was calibrated before starting the ambient measurements. The calibration protocol for our SP2 is described in Miyakawa et al. (2016). Fullerene soot (FS; stock 40971, lot L20W054, Alfa Aesar, USA) particles were used as a calibration standard for the SP2. A differential mobility analyzer (DMA; Model 3081, TSI Inc., USA) was used for preparing the monodisperse FS particles. The analysis of the calibration results suggests that the full width at half maximum (FWHM) was typically 30 % of the modal incandescence signal intensity (S_{LI}) for the diameter range

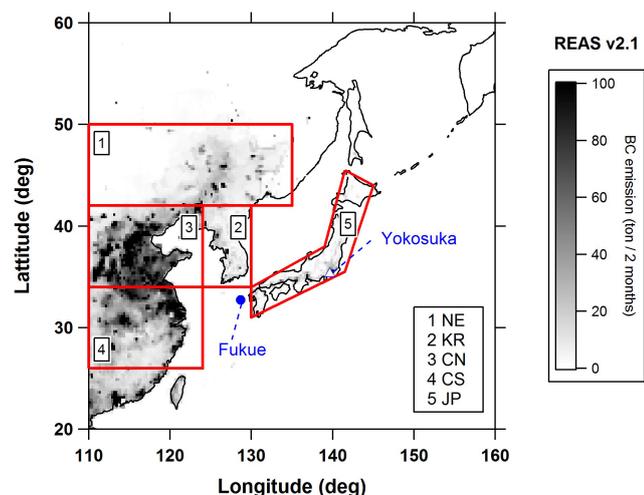


Figure 1. Map of the investigated region with two observation sites (Yokosuka – open triangle; Fukue Island – closed circle) and five defined areas (1 – Northeast China; 2 – Korea; 3 – Central North China; 4 – Central South China; 5 – Japan). The bimonthly mean BC emission rate (March–April) in 2008 is overlaid on the map (REAS ver. 2.1, Kurokawa et al., 2013).

studied. Note that the FWHM can be regarded as an upper limit to describe the resolving power of rBC mass per particle using our SP2 because the combination of polydisperse size distribution of FS particles and the transfer function of the DMA can broaden the distributions of S_{LII} for the prepared FS particles. The variations in the laser power were within $\pm 3\%$ during the observation period, thus indicating that the fluctuations in laser power did not largely affect the lower limit of the detectable rBC size using the SP2. The mass equivalent diameter (D_{core}) of an rBC particle was derived from the rBC mass per particle (m_{pp}) with an assumed particle density for BC (1800 kg m^{-3} ; Bond and Bergstrom, 2006). A large diameter Nafion dryer (MD-700, Perma Pure, Inc., USA) was placed in front of the SP2 for drying the sample air without significant loss of the aerosol particles greater than 50 nm. The dry air for MD-700 was generated by a heatless dryer (HD-2000, Perma Pure, Inc., USA) and a compressor (2AH-23-M222X, MFG Corp., USA). The relative humidity of the sample air was less than 20% during the observation period. The hourly number or mass size distributions and hourly median values of shell (D_S) to D_{core} ratios (D_S/D_{core}) for the selected D_{core} ranges were calculated. The retrievals of D_S from the light-scattering signals measured by an avalanche photodiode and a position sensitive detector (Gao et al., 2007) were performed using a time-resolved scattering cross section method given by Laborde et al. (2012). In this study, we quantified the D_S/D_{core} ratios with a D_{core} range between 0.15 and 0.35 μm . The upper limit of the estimation of D_S/D_{core} ratios is 4 in this study. Maximum levels of D_S/D_{core} ratios retrieved were ~ 2.5 at D_{core} of 0.2 μm . We also analyzed the microphysical param-

eters of rBC particles measured using the SP2 in the early summer of 2014 at Yokosuka (35.32° N , 139.65° E ; Fig. 1), located near industrial sources along Tokyo Bay (Miyakawa et al., 2016). These data sets were used as a reference for the BC-containing particles in air masses strongly affected by combustion sources.

Equivalent BC (EBC; Petzold et al., 2013) mass concentrations are continuously measured at Fukue Island using two instruments: a continuous soot-monitoring system (COSMOS; model 3130, Kanomax, Japan) and a multi-angle absorption photometer (MAAP; MAAP5012, Thermo Scientific, Inc., USA). The details of the air sampling and inter-comparisons for EBC measurements at Fukue Island have been described elsewhere (Kanaya et al., 2013, 2016). In this study, mass concentrations of EBC measured using the COSMOS were evaluated by comparison with those of SP2-derived rBC. The intercomparison between SP2 and COSMOS will be briefly discussed below.

Figure 2 depicts the correlation between COSMOS–EBC and SP2–rBC hourly mass concentrations. The unmeasured fraction of the rBC mass was corrected by the extrapolation of the lognormal fit for the measured mass size distributions outside the measurable D_{core} range (0.08–0.5 μm). Note that the unmeasured fraction of rBC mass was minor ($< 5\%$) in this study. The linear regression slope of the correlation between EBC and rBC was 0.88 (± 0.03). Uncertainty with respect to the calibration was examined in an industrial region and found to be within around 3% (Miyakawa et al., 2016). The average discrepancy between EBC and rBC was beyond the uncertainty of the calibration and was comparable to the uncertainty of COSMOS (10%) as evaluated by Kondo et al. (2009). While the validity of the calibration standard, FS particles, has been evaluated only near source regions (Moteki and Kondo, 2011; Miyakawa et al., 2016), the discrepancy may be partly attributed to the differences in physicochemical properties between ambient BC in remote air and FS particles. Onsite calibration of the SP2 using ambient BC particles prepared by a thermal denuder and particle mass classifier, such as an aerosol particle mass analyzer (APM), is desirable for better quantification of the rBC mass based on the laser-induced incandescence technique in remote areas. Although we need to make further attempts to evaluate SP2 in remote areas, this study indicated that SP2–rBC mass concentrations agreed well with COSMOS–EBC. Therefore we simply use “BC”, instead of EBC and rBC defined depending upon the measurement techniques. We analyzed the COSMOS data for the BC mass concentrations, and the SP2 data for the BC microphysics.

The chemical composition of non-refractory submicron aerosols was measured using an Aerodyne ACSM (Aerodyne, Inc., USA.) placed in an observatory container at Fukue Island during the observation period. The details of the ACSM at Fukue Island have been described in Irei et al. (2014). The collection efficiency (CE) of the ACSM was assumed to be 0.5 for this period (Yoshino et al., 2016). We

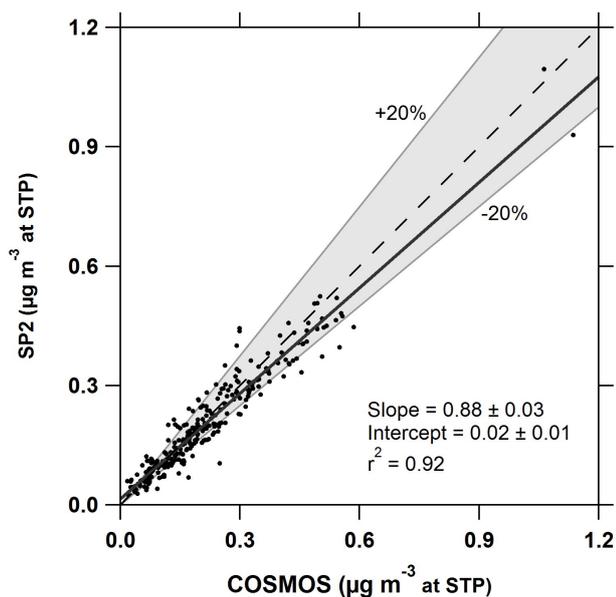


Figure 2. Correlation plot of SP2-rBC and COSMOS-EBC mass concentrations (at standard temperature and pressure). The shaded region corresponds to within $\pm 20\%$ from the 1 : 1 line (the dashed line). The bold line depicts the linear regression.

considered sulfate (SO_4^{2-}) ions to be a major non-BC material and one of the most important secondary aerosols in east Asia (Takami et al., 2007) for the data interpretation. The fact that SO_4^{2-} is produced in the cloud phase as well as in the gas phase is beneficial for interpreting temporal changes in SO_4^{2-} concentration associated with the wet-removal processes. We also analyzed other non-refractory components such as nitrate (NO_3^-), ammonium (NH_4^+), and organic matter (OM). During the period 1–7 April 2015, the critical orifice of the inlet assembly of the ACSM became clogged. ACSM-derived SO_4^{2-} , NO_3^- , NH_4^+ , and OM (ACSM- SO_4^{2-} , ACSM- NO_3^- , ACSM- NH_4^+ , and ACSM-OM) for this period were not used in the analysis.

Two high-volume air samplers (HV500F, Sibata Scientific Technology Ltd., Japan) were deployed on the rooftop of the observatory container. The sampling flow rate for both samplers was 500 liters per minute (L min^{-1}). Air sampling was carried out for 21 h (from 10:00 to 07:00) on a 110 mm pre-combusted (900°C for 3 h) quartz filter (QR-100, Advantec Toyo Kaisha Ltd., Japan). Both samplers have a $\text{PM}_{2.5}$ impactor for classifying the particle size. One impaction plate was coated with vacuum grease (HIVAC-G, Shin-Etsu Chemical Co., Ltd., Japan) to minimize the impact of coarse-mode particles on the chemical analysis of fine mode particles such as radiocarbon analysis, and a pre-combusted quartz fiber filter with slits was set on another impaction plate to collect the coarse particles. Water soluble ions were analyzed using ion chromatography (IC; Dionex ICS1000, Thermo Fisher Scientific K.K., Japan). The results from the chemical analysis of filter samples are not discussed

in this study in detail. We only used the mass concentration of SO_4^{2-} (IC- SO_4^{2-}) in this study to evaluate the uncertainty in relation to CE of the ACSM and to analyze the temporal variations during the period when the ACSM- SO_4^{2-} data were not available (1–7 April 2015).

The carbon monoxide (CO) mixing ratio was also continuously measured using a nondispersive infrared (NDIR) CO monitor (model 48C, Thermo Scientific, Inc., USA). Details of the CO measurements including the long-term variations in sensitivity and zero level are discussed elsewhere (Kanaya et al., 2016).

2.2 Enhancement ratio of BC and SO_4^{2-} to CO as an indicator of the transport and transformation of aerosol particles

In order to quantify the extent of the removal of BC, we calculated the hourly enhancement ratio of BC mass concentrations to CO mixing ratios ($\Delta\text{BC} / \Delta\text{CO}$) against the east Asian background air concentrations as follows:

$$\frac{\Delta\text{BC}}{\Delta\text{CO}} = \frac{[\text{BC}] - [\text{BC}]_{\text{bg}}}{[\text{CO}] - [\text{CO}]_{\text{bg}}}, \quad (1)$$

where $[\text{BC}]$ and $[\text{CO}]$ are measured hourly concentrations of BC and CO, respectively, and $[\text{BC}]_{\text{bg}}$ and $[\text{CO}]_{\text{bg}}$ are their estimated background concentrations. Here, we assumed that $[\text{BC}]_{\text{bg}}$ is 0 (Oshima et al., 2012). The background concentration of CO during the analysis period (11 March–14 April 2015) was calculated by averaging the concentrations lower than the fifth percentile (120 ppb). The validity of this value is discussed in the Supplement.

Relative changes in SO_4^{2-} to CO were also analyzed using the linear regression slopes of their correlation in this study. We did not calculate their hourly values because it was difficult to determine the background concentration of SO_4^{2-} . The use of CO as a tracer of sulfur compounds in east Asia was validated by Koike et al. (2003). Although sulfur dioxide (SO_2), which is a major precursor of anthropogenic SO_4^{2-} , does not always share the emission sources with CO, the spatial distributions of SO_2 emissions are similar to those of CO emissions in east Asia (Koike et al., 2003; Kurokawa et al., 2013). Analyzing the increase or decrease in the slopes of the SO_4^{2-} –CO correlation is beneficial to the investigation of the formation and removal processes for SO_4^{2-} . Especially, the aqueous-phase reaction of SO_4^{2-} in clouds is discussed using this parameter.

2.3 Meteorological field analysis

We used the 6-hourly meteorological data, with a resolution of 1° in terms of the latitude and longitude, from the National Centers for Environmental Prediction (NCEP) Final (FNL) operational global analysis, and daily precipitation data, with a resolution of 1° in terms of the latitude and longitude, from the Global Precipitation Climatology Project (GPCP) data set

(Huffman et al., 2001). We analyzed these data sets to investigate the general features of the meteorological field in east Asia during the observation period.

2.4 Backward trajectory analysis

We calculated backward trajectories from the observation site to elucidate the impact of the Asian outflow. Three-day backward trajectories from the observation site (the starting altitude was 0.5 km) were calculated every hour using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory model (Draxler and Hess, 1997; Draxler, 1999; Stein et al., 2015) with the meteorological data sets (NCEP's Global Data Assimilation system, GDAS). In this study, the residence time over specific source regions was used as an indicator of their impacts on the observed air masses. We defined five domains for assessing the impact over the Asian continent: Northeast China (NE), Korea (KR), Central North China (CN), Central South China (CS), and Japan (JP) (Fig. 1). The period when air masses passed over the domains NE, KR, CN, and CS at least for 1 h is defined as that of "continental outflow". The impacts of precipitation on the observed air masses were assessed by a parameter referred to as the "accumulated precipitation along trajectory" (APT; Oshima et al., 2012). In this study, we calculated the APT values by integrating the amount of hourly precipitation in the Lagrangian sense along each 3-day back trajectory of the sampled air masses. The hourly variations in APT were merged into the observed gas and aerosol data sets.

3 Results and discussion

3.1 The meteorological field in the spring of 2015

The mean meteorological field during the observation period (11 March–13 April 2015) is discussed for the purpose of characterizing the general features of the wind flow and precipitation in this region. The migrating anticyclone and cyclone passed alternately over east Asia during this period; this pattern is typically dominant in spring over east Asia (Asai et al., 1988). Figure 3a shows the mean sea level pressure (SLP) and mean horizontal winds at the 850 hPa level in east Asia during the observation period. The mean equivalent potential temperature (θ_e) and the meridional moisture transport at the 850 hPa level during the same period are also shown in Fig. 3b. The midlatitude region (35–50° N, 120–140° E) was under the influence of a modest monsoonal northwesterly flow, which advected cold, dry air from the continent to the observation area. The subtropical region (20–30° N, 110–130° E) was under the influence of a persistent southwesterly flow, part of which was converging into the observation area (30–35° N), and this flow was confluent with the northwesterlies from the continent. The low-level

southerly flow advected warm, moist air into the observation area to sustain a large amount of precipitation (Fig. 4a).

Figure 3c shows the temporal variations in surface pressure and precipitable water at the observation site. The surface pressure is anticorrelated well with the precipitable water. During the observation period, migratory cyclones and anticyclones occurred occasionally (3 times each). The occurrence of migratory cyclones advected moist air, which could have contributed to the wet removal of BC during transport in the PBL. In contrast, the occurrence of anticyclones advected dry air, which could have contributed to the efficient transport of BC from the source regions.

Figure 4a depicts the mean precipitation over east Asia during the observation period. Mean precipitation showed a latitudinal gradient over eastern China and the Yellow Sea and East China Sea region (i.e., increasing precipitation from south to north), and these results suggest that transport pathways can greatly affect the wet removal of aerosols. The APT was compared with the averaged latitude of each trajectory for 48 h backwards from the time of -24 h (Lat_{ORIG}) (Fig. 4b), which can be interpreted as an indicator of the latitudinal origin of the air masses arriving at Fukue Island. The high APT values corresponded to the air masses that originated from the southern regions (20–40° N). The data points are colored according to the maximum RH values along each backward trajectory (RH_{max}). Lower relative humidity (RH_{max}) was observed in the air masses with low APT values that originated from northern regions (30–50° N). These air mass characteristics were consistent with the mean precipitation field (Fig. 4a). Some of the data points showed high values of RH_{max} ($\sim 100\%$) when their APT was almost 0. These data probably correspond to the air masses that experienced cloud processes not associated with precipitation. Possible effects of cloud processes without precipitation on the removal of aerosol particles during transport will be discussed using these data points in the following section.

3.2 Removal processes of fine-aerosol particles

In this study, the removal processes including dry deposition and below-cloud scavenging were considered to be minor. The dry deposition in this region has already been evaluated by Kanaya et al. (2016), who found a minimal decrease in $\Delta BC / \Delta CO$ for air masses not affected by wet removal but with different transport times. The below-cloud scavenging is dependent on the precipitation intensity and rain drop size as well as the particle size range. The removal rates of submicron accumulation mode particles through the washout (Λ_{accum}) were estimated to be $\sim 1 \times 10^{-3} \text{ h}^{-1}$ ($0.5\text{--}2 \times 10^{-3} \text{ h}^{-1}$) using a parameterization given by Wang et al. (2014) and the average precipitation intensity along the trajectories ($0.78 \pm 0.6 \text{ mm h}^{-1}$) as an input to the parameterization. The values of Λ_{accum} can be underestimated by 1 order of magnitude by using the parameterization (Wang et al., 2014), which is, however, overly pessimistic. The tem-

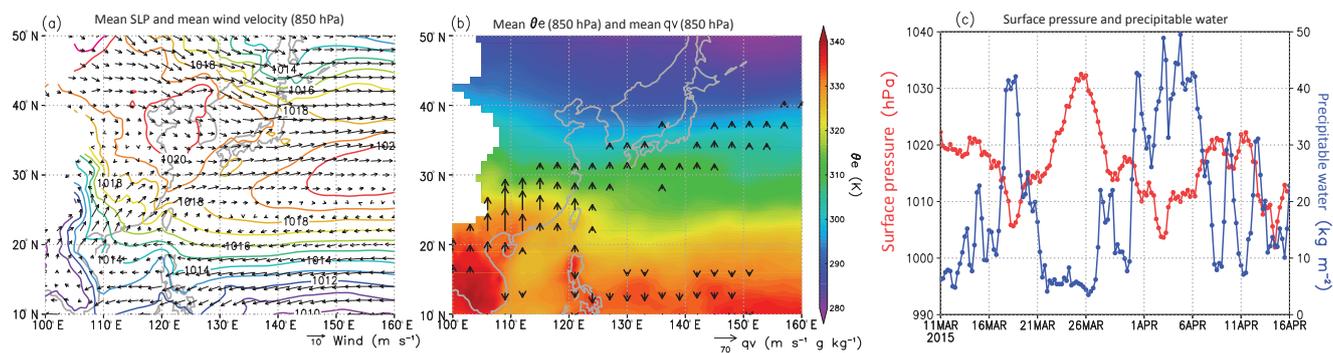


Figure 3. Meteorological fields in east Asia during the observation period (11 March–14 April 2015) based on NCEP FNL data. **(a)** Mean SLP (hPa, contours) and mean horizontal wind velocity at the 850 hPa level (m s^{-1}). Regions without data correspond to those of high-altitude mountains. **(b)** Mean θ_e (K) and total meridional moisture transport (qv values) at the 850 hPa level ($\text{m s}^{-1} \text{ g kg}^{-1}$). Only qv vectors with magnitudes greater than $10 \text{ m s}^{-1} \text{ g kg}^{-1}$ were plotted. **(c)** Temporal variations in the surface pressure (hPa; red line and markers, left axis) and precipitable water (kg m^{-2} ; blue line and markers, right axis) at the Fukue observation site (32.75° N , 128.68° E).

poral duration of rain along trajectories for air masses with an APT greater than 0 mm was $10 (\pm 8)$ h on average. The average fraction of submicron aerosols removed was 1% ($+2.59\%/-0.9\%$). Even though we took into account the uncertainties for estimating Λ_{accum} , it was found that below-cloud scavenging did not play a major role in the removal of BC in east Asian outflow.

3.3 Temporal variations in aerosols and CO

Temporal variations in the concentrations of BC (measured using COSMOS and SP2), SO_4^{2-} (measured using ACSM and IC), NO_3^- , OM, and CO are shown in Figure 5. ACSM- SO_4^{2-} generally agreed well with IC- SO_4 , thus indicating that the assumed CE (0.5) was valid for the observation period. As NO_3^- and SO_4^{2-} were almost fully neutralized by NH_4^+ , we assumed their chemical forms were ammonium salts. In general, BC, SO_4^{2-} , and OM were positively correlated with CO at Fukue Island, and these results illustrate the impact of continental outflow affected by incomplete combustion sources on aerosol mass concentrations. The mean chemical composition of fine aerosols during the observation period was listed in Table 1. Ammonium sulfate and OM were abundant components. Figure 5 also includes the temporal variations in the fractional residence time over the selected region defined in Sect. 2.4 (top panel). The CO concentrations were typically enhanced for the period with the higher contributions of CN and CS. A previous study suggested that the majority of SO_4^{2-} aerosols were formed in less than around 1.5 days after the air masses left the Chinese continent (Sahu et al., 2009). Kanaya et al. (2016) showed that the typical transport time of continental outflow air masses at Fukue Island was around 1–2 days in spring. The positive correlation of SO_4^{2-} and CO suggests that the secondary formation of SO_4^{2-} through transport was significant during the observation period. The structure and composition of fine

aerosols in east Asian outflow were analyzed by using a secondary ion mass spectrometer in a previous study (Takami et al., 2013). Takami et al. (2013) suggest that SO_4^{2-} and OM are constituents in the coating of almost all BC-containing particles. Hence, we concluded that ammonium sulfate and OM contributed to the growth of BC-containing particles. The period with an APT > 3 mm is highlighted by light blue in Fig. 5 to show the impact of wet removal on the transport of BC and SO_4^{2-} aerosols. The maximum concentrations of aerosols and CO were observed on the morning of 22 March (Ep. 1) under the influence of the anticyclone (corresponding to the trajectories colored red in Fig. 4a) when the APT values were almost 0. In contrast, aerosol concentrations did not increase with CO in the period from the evening of 5 April to the morning of 6 April (Ep. 2) under the influence of the migratory cyclone (corresponding to the trajectories colored black in Fig. 4a), when the APT was greater than 10 mm.

3.4 Correlation of BC, SO_4^{2-} , and CO

Figure 6a and b show scatterplots of CO with BC and SO_4^{2-} , respectively. Positive correlation of BC and SO_4^{2-} with CO was clearly found in air masses with low APT values. Linear regression was performed for the data points with an APT higher than 15 mm for BC–CO and SO_4^{2-} –CO. Note that the linear regression slope for BC–CO was determined by forcing through the background concentrations of BC ($0 \mu\text{g m}^{-3}$) and CO (120 ppb). The slopes of the fitted lines were $1.4 (\pm 0.06)$ and $9.8 (\pm 2.7) \text{ ng m}^{-3} \text{ ppb}^{-1}$ for BC–CO and SO_4^{2-} –CO, respectively. They were close to the lower envelopes of the correlations. It is evident from these scatterplots that the relative enhancements of BC and SO_4^{2-} to CO were mainly affected by the APT. Kanaya et al. (2016) found that the estimated emission ratios of BC to CO over the east Asian continent varied slightly depending on the origin of the air masses (this range is overlaid on Fig. 6a). In their

Table 1. Mean chemical composition of fine aerosols during the observation period.

Components	Period average	APT			
		0 mm	0 mm RH _{max} < 50 %	0 mm RH _{max} > 80 %	> 15 mm
Ammonium sulfate	44.9 %	41.8 %	34.0 %	48.9 %	50.4 %
Ammonium nitrate	11.7 %	15.7 %	10.7 %	8.0 %	5.0 %
OM	40.9 %	40.1 %	52.0 %	40.4 %	42.0 %
BC	2.5 %	2.4 %	3.2 %	2.6 %	2.5 %

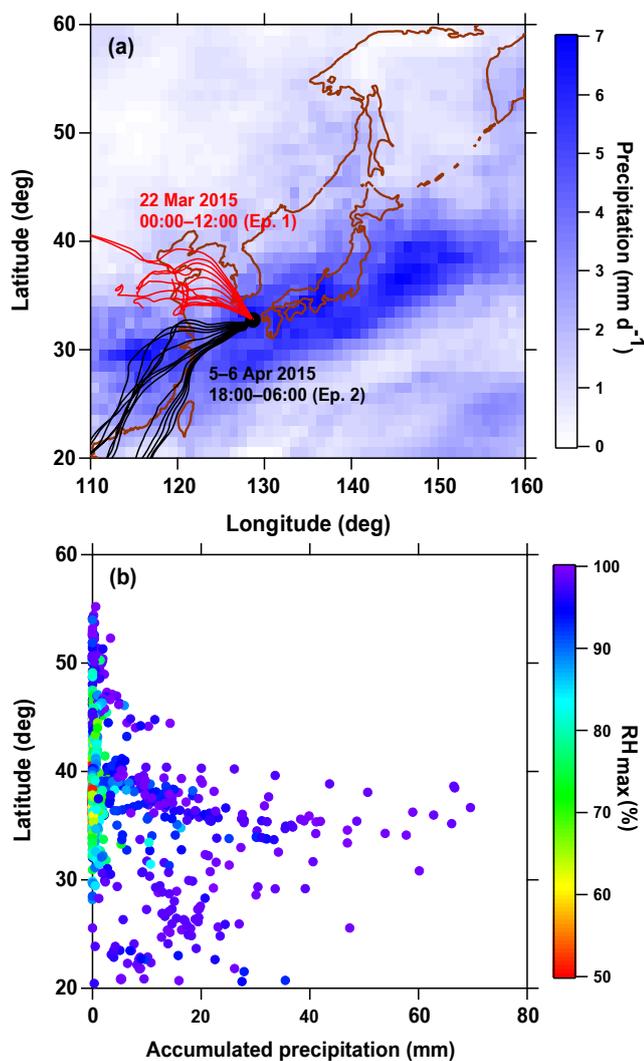


Figure 4. (a) Mean precipitation derived from GPCP during the observation period (11 March–14 April 2015). Three-day backward trajectories for selected periods are overlaid (red lines; 00:00–12:00 LT, 22 March 2015, Ep. 1; black lines; 08:00 LT 5 April–06:00 LT, 6 April 2015, Ep. 2). (b) The relationship between APT and Lat_{ORIG} (see text for details) colored by the maximum RH along the backward trajectories.

study, the $\Delta BC / \Delta CO$ ratios for Central North and Central South China regions were estimated to be $5.3 (\pm 2.1)$ and $6.9 (\pm 1.2) \text{ ng m}^{-3} \text{ ppb}^{-1}$, respectively. $\Delta BC / \Delta CO$ observed in the PBL over the Yellow Sea during the same season was $6.2 \text{ ng m}^{-3} \text{ ppb}^{-1}$ (Kondo et al., 2016). The data points with $\Delta BC / \Delta CO$ in these ranges show low APT values (less than or ~ 1 mm). Wet removal (in-cloud scavenging) was one of the most important controlling factors for the transport efficiency of BC in this region during the observation period.

The cloud processes of aerosol particles not associated with precipitation can also reduce the slope of their correlation. However, no decreasing tendency of BC–CO and SO_4^{2-} –CO slopes against RH_{max} when APT was 0 was found during the observation period (data not shown). The SO_4^{2-} –CO slopes with the APT values of 0 were analyzed as a function RH_{max} (Fig. 6b) and varied from $30.7 (\pm 1.8)$ to $44.1 (\pm 13.4) \text{ ng m}^{-3} \text{ ppb}^{-1}$ under the conditions without (RH_{max} < 50 %) and with (RH_{max} > 80 %) cloud impacts, respectively. The difference in the slope without and with cloud impacts is small but significant (based on the analysis of covariance to these data sets). The fact that the SO_4^{2-} –CO slope increased with RH_{max} when the APT was 0 suggests that aqueous-phase formation and subsequent droplet evaporation partly contributed to the mass concentrations of SO_4^{2-} observed at Fukue Island. Therefore, the changes in the SO_4^{2-} –CO correlation were controlled largely by in-cloud scavenging and weakly by aqueous-phase formation during transport.

3.5 Changes in fine-aerosol compositions

Chemical compositions of fine aerosols were investigated in terms of the APT and RH_{max}. Four cases are selected here, namely (1) APT of 0 (no precipitation), (2) APT of 0 with RH_{max} < 50 % (no precipitation without cloud impacts), (3) APT of 0 with RH_{max} > 80 % (no precipitation with cloud impacts), and (4) APT > 15 mm (heavily affected by wet removal). The comparison between cases (3) or (4) and (2) is useful to elucidate the effect of cloud processing. The results are summarized in Table 1. Ammonium sulfate and OM were dominant in all cases. The relative changes in chemical compositions of fine-aerosol particles were within around 10 %. As all components of fine aerosols were removed through

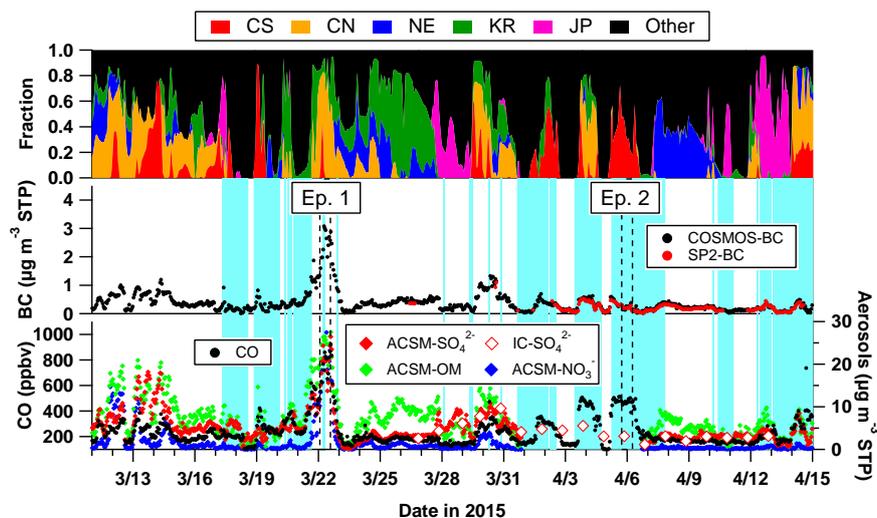


Figure 5. Temporal variations in air mass origin and concentration of trace species. Top panel: fractional residence time of air masses passing over selected area (red – Central South China; orange – Central North China; blue – Northeast China; green – Korea; pink – Japan; black – other regions such as the ocean). Middle panel: mass concentrations of BC measured using COSMOS (black markers) and SP2 (red markers). Bottom panel: concentrations of CO (black markers), SO_4^{2-} (red closed and open markers for ACSM and IC, respectively), ACSM- NO_3^- (blue markers), and ACSM-OM (light green markers). The periods with an APT > 3 mm are highlighted in light blue in the middle and bottom panels. The periods denoted as Eps. 1 and 2 (see the text for details) are enclosed by dashed lines.

the in-cloud scavenging (Fig. 10 of Kanaya et al., 2016), it is expected that the relative abundance does not vary greatly with the in-cloud scavenging. The relative contributions of fine aerosols in cases (3) and (4) increased from case (2), indicating that cloud processes affected their relative abundances. Ammonium sulfate contribution slightly increased with the in-cloud scavenging (based on the comparison between cases (2) and (3) or (4)), while the relative contributions of ammonium nitrate, OM, and BC slightly decreased. The contributions of OM in case (2) increased from the average. The formation of secondary OM can be significant under dry conditions during transport. Detailed mass spectral analyses of OM, secondary formation of OM, and cloud-phase formation of OM in east Asia are beyond the scope of this study, and they are not discussed in this study. The first two issues have been investigated in previous studies (e.g., Irei et al., 2014; Yoshino et al., 2016).

3.6 Changes in microphysical parameters of BC-containing particles associated with wet removal

Number and mass size (D_{core}) distributions of BC classified by the values of $\Delta\text{BC}/\Delta\text{CO}$ are shown in Fig. 7a and b, respectively. When $\Delta\text{BC}/\Delta\text{CO}$ values in continental outflow air masses were greater than $3\text{ ng m}^{-3}\text{ ppb}^{-1}$ (within the range of the BC/CO emission ratios given by Kanaya et al., 2016), these air masses are defined as “outflow without BC loss”. These air masses originated mainly from CN via KR and NE. When $\Delta\text{BC}/\Delta\text{CO}$ values of continental outflow air masses were less than $1\text{ ng m}^{-3}\text{ ppb}^{-1}$, the air masses were

defined as “outflow with BC loss”. Considering the typical emission ratios of BC to CO ($6\text{--}7\text{ ng m}^{-3}\text{ ppb}^{-1}$; Kanaya et al., 2016), transport efficiency for the outflow with BC loss air masses can be estimated to be less than $\sim 17\%$. These air masses originated mainly in CS. The low and high APT values for outflow without BC loss and outflow with BC loss air masses, respectively, (Table 2) gave us confidence in the validity of our classification as discussed in the previous section. As a reference for emission sources (“source”), the average size distributions of BC in a Japanese industrial area (see Sect. 2.1, Miyakawa et al., 2016) are shown in Fig. 7. The statistics of the size distributions are summarized in Table 2. Observed differences in the size distributions between source and outflow were generally consistent with previous studies (Schwarz et al., 2010). Air mass aging leads to the growth of BC-containing particles. Number–size distributions of BC largely varied in the size range less than $0.1\text{ }\mu\text{m}$ (Fig. 7a). In outflow air masses, such small BC-containing particles would be scavenged by larger particles in the coagulation process during transport. Below-cloud scavenging can also affect the BC-containing particles in the smaller size range ($<0.1\text{ }\mu\text{m}$) when the air masses are affected by the precipitation. The peak D_{core} of mass (number) size distributions of BC became larger – from 0.16 (0.06 , which is estimated by the mass size distribution) μm to $0.18\text{--}0.2$ ($0.09\text{--}0.1$) μm – between source and outflow. The BC-containing particles have systematically different size distributions in outflow air masses with and without BC loss, indicating that the BC loss process also affected the size distributions. The peak D_{core} of BC number and mass size distributions in outflow

Table 2. Summaries of BC microphysical parameters measured at Yokosuka and Fukue Island.

Site	Air mass type	Averaging time* (h)	$\Delta\text{BC} / \Delta\text{CO}$ ($\text{ng m}^{-3} \text{ppb}^{-1}$)	APT (mm)	Lognormal fit parameters avg. (1σ)		1 h median D_S/D_{core} for selected D_{core} avg. (1σ)			
					MMD (μm)	σ_g	0.15–0.2	0.2–0.25	0.25–0.3	0.3–0.35 μm
Yokosuka	Source	184	–	–	0.160 (0.019)	1.84 (0.08)	1.18 (0.07)	1.15 (0.06)	1.10 (0.04)	1.07 (0.04)
Fukue	Outflow	87	> 3	1.2	0.195 (0.005)	1.57 (0.05)	1.37 (0.05)	1.32 (0.03)	1.21 (0.03)	1.17 (0.03)
Fukue	Outflow	51	< 1	19.9	0.182 (0.011)	1.62 (0.09)	1.25 (0.05)	1.24 (0.04)	1.16 (0.02)	1.12 (0.03)

* Time used for calculating averaged statistics of the microphysical properties of BC-containing particles. MMD: mass median diameter.

air masses with BC loss was slightly lower than that for air masses without BC loss. The changes in the peak diameter of the core and total (i.e., core and shell) size distributions of BC-containing particles as a function of $\Delta\text{BC} / \Delta\text{CO}$ ratios are shown in Fig. 7c. The peak values of D_{core} and D_S (with the D_{core} range of 0.15–0.35 μm) were determined by fitting the lognormal function to the hourly BC mass– D_{core} and BC number– D_S distributions of BC-containing particles, respectively. The reason why we did not analyze the peak values of D_{core} for BC number–size distributions is that they were mostly smaller than 0.08 μm (outside the measurable range). The observed decreases in the diameters or BC mass per particle were clear and were beyond the uncertainties of SP2 (see Sect. 2.1). The changes in the peak D_{core} and D_S from the highest to lowest bins of $\Delta\text{BC} / \Delta\text{CO}$ ratios were 0.02 μm (2–2.5 fg) and 0.05 μm , respectively, which are statistically significant ($p < 0.01$).

Figure 8 depicts the probability density of the D_S/D_{core} ratio for the BC size of 0.2 (± 0.02) μm for source and outflow air masses. The modal values of the D_S/D_{core} ratio were systematically changed with air mass aging and BC loss (in-cloud scavenging). The condensation of inorganic and organic vapors on BC-containing particles during transport can account for the increase in the D_S/D_{core} ratio, as discussed in previous studies (e.g., Shiraiwa et al., 2008; Subramanian et al., 2010). As discussed earlier, the results of this study suggested that SO_4^{2-} and OM substantially contributed to the increase in the D_S/D_{core} ratio. In outflow air masses with BC loss, modal values of the D_S/D_{core} ratio were clearly lower than those in outflow without BC loss. Furthermore, it is indicated that the wet-removal process also affected the coating thickness distributions for the BC sizes in the range 0.15–0.35 μm (Table 2). It should be noted that the coating of BC-containing particles is not always thick in remote regions and that the D_S/D_{core} ratio distributions, as well as size distributions, can be affected by the wet-removal process during transport in the PBL.

3.7 Discussion

Not only in-cloud scavenging of BC-containing particles but also subsequent precipitation (i.e., the rainout process) can account for the changes in the microphysical parameters of BC detected in this study. Our results show a decrease in both

the peak diameters, D_S and D_{core} , of the BC number and mass size distributions, respectively, and the modal value of the D_S/D_{core} ratios in relation to the rainout. The observed evidence implies that there can be removal of large and water-soluble BC-containing particles during transport in the PBL. The Köhler theory suggests that a lower supersaturation is needed for the large and highly water-soluble particles, and this can qualitatively account for the observed changes in the BC microphysics. The D_S/D_{core} ratios (with D_{core} of 0.2 μm) in Fig. 8 were converted to the critical supersaturation (SS_C) of BC-containing particles, which is estimated using the observed chemical composition of non-BC materials at Fukue Island. Sets of hygroscopicity parameters (“CCN-derived” κ ; Petters and Kreidenweis, 2007) and material densities used for the estimation are (0.67, 1.73 g cm^{-3}), (0.61, 1.77 g cm^{-3}), (0, 1.8 g cm^{-3}), and (0.1, 1.2 g cm^{-3}) for ammonium nitrate, ammonium sulfate, BC, and OM, respectively. The estimation includes another assumption, namely that all components are internally mixed with BC. As chemical compositions of non-BC materials did not vary greatly during the observation period (Sect. 3.5), the average value of κ was calculated to be 0.35 using the averaged chemical compositions (Table 1) and was used for the calculation of SS_C . The estimated SS_C decreases with the increases in the D_S/D_{core} ratios. The observed changes in the D_S/D_{core} ratios indicated that the BC-containing particles with lower SS_C were removed through the wet removal.

Note that the magnitude of the change in the peak D_{core} of the BC size distributions in the PBL (~ 0.02 μm , ~ 2 –2.5 fg) shown in Fig. 7c is smaller than that observed in air masses uplifted from the PBL to the FT, in association with wet removal (~ 0.04 μm , ~ 3 fg; Fig. 2 of Moteki et al., 2012) at a similar level of transport efficiency ($< \sim 20\%$). Although the shape of mass size distributions soon after the rainout processes can be distorted by the droplet activation of larger aerosol particles, the observed mass size distributions were well fitted by a lognormal function (Fig. 7b). Figure 8 showed the existence of BC-containing particles with D_S/D_{core} ratios higher than 1.2 even in outflow air masses with BC loss that are expected to readily act as CCN. Air masses sampled at the ground level would be affected by turbulent mixing of those near the clouds around the top of the PBL and those in cloud-free conditions at below-cloud

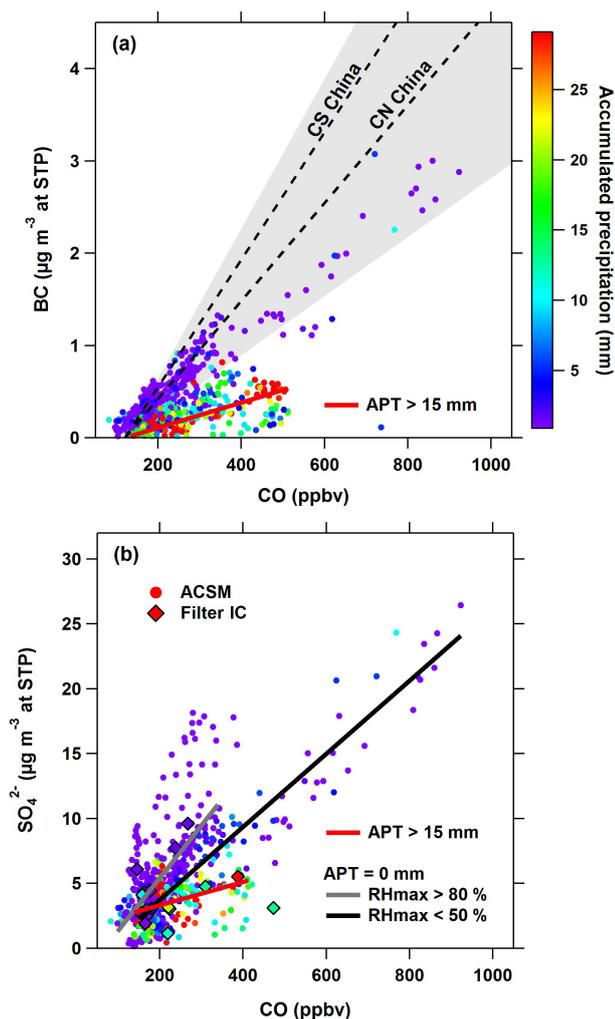


Figure 6. Correlation between aerosol mass concentrations and CO mixing ratio colored according to the APT. (a) BC measured by COSMOS and (b) SO_4^{2-} measured by ACSM and IC (circles and diamond markers, respectively). Dashed lines and shaded area in panel (a) represent the emission ratios of BC to CO over Central North and South (CN and CS) China and their variation ranges, respectively (Kanaya et al., 2016). The bold lines shown in panels (a) and (b) are the linear fitting to the BC–CO and ACSM– SO_4^{2-} –CO correlations for the selected data points, i.e., those with the APT > 15 mm for BC and SO_4^{2-} (red lines), those with the APT of 0 and $\text{RH}_{\text{max}} < 50\%$ for SO_4^{2-} (black line), and those with the APT of 0 and $\text{RH}_{\text{max}} > 80\%$ (shaded line).

levels. On the other hand, most air masses sampled by aircraft measurements in the FT would experience cloud processes during upward transport from the PBL. Mixing of air masses in the PBL suggests that they partially experience in-cloud scavenging processes. The aging (e.g., coagulation) of aerosols particles through the transport (i.e., around ~ 1 day) after the wet-removal events may also lead to the further modification of the shape of the particle size distributions and the mixing state distributions which have been

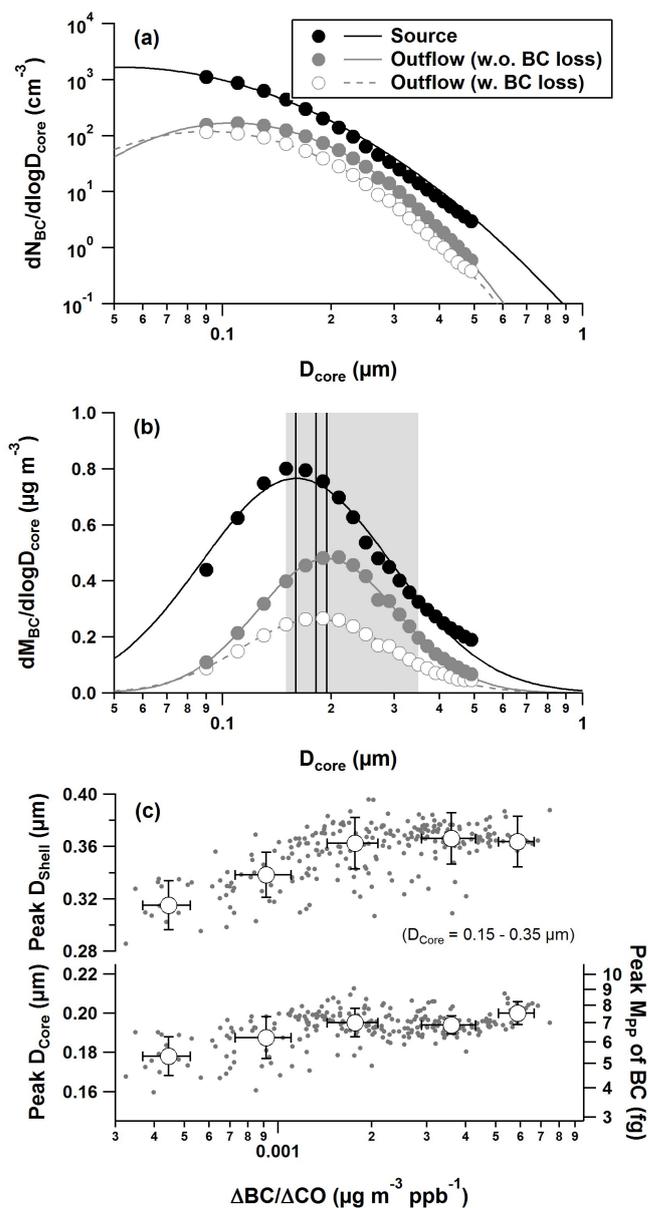


Figure 7. The (a) number and (b) mass size distributions of BC measured at Yokosuka (black markers) and at Fukue Island (gray markers). (c) The evolution of the peak D_S and D_{core} as a function of the degree of removal of BC. The size distributions at Fukue Island include the data for the outflow air masses with (open markers) and without (closed markers) BC loss. Lines in panels (a) and (b) are the lognormal fitting results. The shaded band in panel (b) corresponds to the size range analyzed to estimate D_S/D_{core} ratios. Vertical lines in panel (b) represent the peak D_{core} of the lognormal fit for each of three mass size distributions. Note that the peak D_{core} of lognormal fit for the BC number–size distributions at Yokosuka was estimated from the peak D_{core} of its mass size distribution (Table 2). The peak values of D_S and D_{core} shown in panel (c) were determined by fitting the lognormal function to the hourly number and mass size distributions of BC-containing particles, respectively.

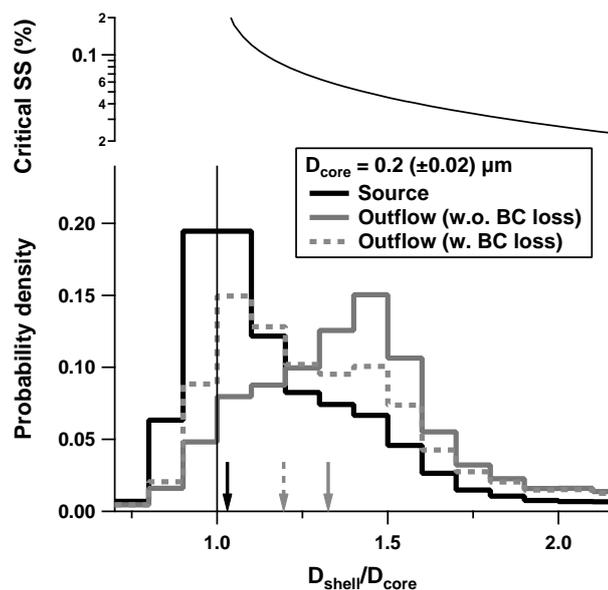


Figure 8. Probability density function of the estimated D_S/D_{core} ratios for BC-containing particles with the size $0.2 (\pm 0.02) \mu\text{m}$ at Yokosuka (black line) and in the air masses of continental outflow with (gray dashed line) and without (gray solid line) BC loss. Vertical arrows indicate the median values of D_S/D_{core} ratios for three different air masses. The estimated critical supersaturation of BC-containing particles detected at Fukue Island was also shown as a function of the D_S/D_{core} ratios (see the text for details).

affected by cloud processes. This factor is actually expected to be minor because the particle concentrations are too low to have high coagulation coefficients to accelerate this effect. The suppression of changes in the microphysical properties of BC-containing particles during transport in the PBL can be related to these factors. More quantitative assessments of the impacts of these factors on the observed features should be performed using a model which has a function to resolve the mixing state of aerosol particles (e.g., Matsui et al., 2013).

The transport pathways of the continental outflow air masses are horizontally and vertically variable in spring in east Asia because of the frequent alternate cyclone–anticyclone activities in spring (Asai et al., 1988). Oshima et al. (2013) examined the three-dimensional transport pathways of BC over east Asia in spring and showed that the PBL outflow through which BC originating from China was advected by the low-level westerlies without uplifting out of the PBL was one of the major pathways for BC export from continental east Asia to the Pacific, thus supporting the general features of microphysical properties of BC in continental outflow obtained by this study. Mori et al. (2014) measured the seasonal variations in BC wet deposition fluxes at another remote island in Japan (Okinawa, ~ 500 km south of Fukue Island) and revealed their maxima in spring, which were consistent with the seasonal variations in the cyclone frequencies. It has been suggested that BC-containing parti-

cles were efficiently activated to form cloud droplets in the continental outflow air masses, especially from the CS region, and this can affect the cloud physicochemical properties in spring in east Asia, as indicated by Koike et al. (2012). As the results from this study are based on observations over a limited length of time, it would be worthwhile to further investigate the possible connections of the variabilities in BC microphysical properties with meteorological conditions to provide useful constraints on more accurate evaluations of the climatic impacts of BC-containing particles in this region (Matsui, 2016).

4 Conclusions

Ground-based measurements of BC were performed near an industrial source region and at a remote island in Japan. We have reported the temporal variations in the transport and the microphysics of the BC-containing particles, measured using COSMOS, SP2, and ACSM. The impacts of air mass aging upon the growth of BC-containing particles were examined by comparing the ground-based observations from the near-source and remote island sites. $\Delta\text{BC} / \Delta\text{CO}$ was used as an indicator of the transport efficiency of BC because it was controlled mainly by rainout during transport in the PBL. The BC size and coating increased during transport from the near-source to the outflow regions on the timescale of 1–2 days when the rainout during transport was negligible. SO_4^{2-} and organic aerosols contributed to the significant increase in the coating materials of BC (i.e., it enhanced the whole size and water solubility of BC-containing particles). Decreases in the peak D_{core} and D_S of mass and number–size distributions (~ 0.02 and $0.05 \mu\text{m}$), respectively, and modal D_S/D_{core} ratios (~ 0.4 for BC of $0.2 \mu\text{m}$) of BC-containing particles were observed in air masses substantially affected by in-cloud scavenging. The observed evidence for the removal of large and water-soluble BC-containing particles was qualitatively consistent with the Köhler theory; however, the values were not as large as those found in air masses uplifted from the PBL to the FT in east Asia associated with precipitation. The mixing of below-cloud and in-cloud air masses in the PBL would result in the suppression of the degree of changes in BC microphysical parameters by cloud processes. This study indicates (1) that the changes (sign and degree) in BC microphysics can be affected by how the air masses are transported and (2) that the observed removal of large and water-soluble BC-containing particles through in-cloud scavenging in east Asia can be expected to be significant in the PBL as well as in the FT in east Asia.

Data availability. The data given in this study will be available on request to the corresponding author (miyakawat@jamstec.go.jp).

The Supplement related to this article is available online at doi:10.5194/acp-17-5851-2017-supplement.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This study was supported by the Environment Research and Technology Development Fund (S7, S12, and 2-1403) of the Ministry of Environment, Japan, and the Japan Society for the Promotion of Science (JSPS), KAKENHI Grant numbers JP26550021, JP26701004, JP26241003, JP16H01772, and JP16H01770, and was partially carried out in the context of the Arctic Challenge for Sustainability (ArCS) Project. The authors would like to thank N. Moteki at the University of Tokyo for assistance with the SP2 calibrations. M. Kubo, T. Takamura, and H. Irie (Chiba University) are also acknowledged for their support at the Fukue Island Atmospheric Environment Monitoring Station. The authors are grateful to Gavin McMeeking and an anonymous reviewer for constructive comments to improve this paper.

Edited by: R. Sullivan

Reviewed by: G. R. McMeeking and one anonymous referee

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