Responses to the reviewers' comment

Anonymous Referee #1

Review of "Alteration of the microphysical properties of black carbon through transport in the boundary layer in East Asia" by Takuma Miyakawa et al. submitted to Atmospheric Chemistry and Physics.

We appreciate the reviewer's helpful and constructive comments on the manuscript entitled "Alteration of the microphysical properties of black carbon through transport in the boundary layer in East Asia". As the reviewers suggested, we have modified the manuscript. Major points for the revisions are listed as follows.

1) Title has been changed.

2) Supporting information (SI) has been prepared.

3) We have modified the discussion section.

*Note the reviewers' comments in **bold**.

The manuscript discusses ground-based measurements, with several instruments, of black carbon (BC) near an industrial source region and at a location removed from the source to study the effects of precipitation on the size distribution and properties of the BC-containing particles. The manuscript is well written and competently explains the study, but several of the arguments do not seem supported by the data. If the comments below are addressed I would recommend that the manuscript be accepted for publication. The title refers to "microphysical properties," which is true, but perhaps "size distribution and amount of associated non-BC material" would be more accurate, as the former term implies a host of properties that were not addressed.

>As the reviewer suggested, this study has investigated a part of the microphysical parameters of BC. Shape and chemical composition of BC-containing particles, which were not directly measured in this study, are important for considering the climatic impacts of BC-containing particles. However, chemical composition of non-refractory (non-BC) materials for both BC-free and -containing particles was measured using an Aerosol Chemical Speciation Monitor (ACSM). We addressed just simply the mixing state of BC-containing particles, and therefore revised the title slightly to "Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in East Asia".

Line 56: The sweeping statement that "washout cannot substantially affect the

lifetime of atmospheric BC-containing particles," even with a reference to Seinfeld and Pandis, seems difficult to justify. Do the authors mean that because most of the BC-containing particles have diameters of several hundred nanometers, their ability to be scavenged by falling precipitation is not very large? This would seem to depend on the intensity of precipitation.

>As the reviewer suggested, the accumulation mode aerosols including BC are not effectively removed by the falling rain droplets.   Washout process is dependent on the precipitation intensity (PI) and rain drop size as well as the particle size range.   In this study, the information of rain drop size is not available.   The average PIs along a backward trajectory were calculated for the rain period in 3d-backward time (PI > 0 mm h^{-1}).   They ranged from 0.1 to 2.5 mm h^{-1} (median = ~0.6 mm h^{-1}).   Using the PI value of 0.6 mm h^{-1}, the scavenging rates of accumulation mode particles were estimated to be 6E-3 h^{-1} (6E-5 h^{-1}) with the assumed rain drop diameter of 0.2 mm (2 mm).   The corresponding time constants are around 7 and 694 days.   These are longer than the typical transport time from the continent to the observation site.   The details are described in SI.

Line 148: Rather than "lower and upper boundaries" it would be preferable to state "outside the diameter range . . ." so that it is clear what size is being referred to.

>We have revised as suggested.

Lines 152-154: Some discussion of why the EC and rBC concentrations differ, and especially why the rBC concentration is less, seems to be necessary. Line 168: Some justification for the selection of 0.5 as the collection efficiency for sulfate in the ACSM is required.

>In this study, we compared rBC with effective BC (EBC) measured using a light absorption technique (COSMOS).   As we stated in the original manuscript, the difference between rBC and EBC is within the uncertainties related to both measurements.   One of the unclear uncertainties, which have not well been studied, is the detection sensitivity of SP2 to the ambient rBC particles (incandescence signal intensity per rBC particle mass, S_{LII}-m_{pp}) in a remote atmosphere.   It was found in previous studies (Moteki and Kondo, 2010; Miyakawa et al. 2016) that the S_{LII}-m_{pp} relationship of fullerene soot (FS) particles, which is used as a calibration standard for the SP2, is similar to that of ambient rBC particles in urban/industrial area.   We hence assume the same sensitivity of SP2 to the ambient rBC in a remote atmosphere as that

of FS particles and rBC particles in urban/industrial area.   I added some explanations on the related uncertainties to the section 2.1 in the revised manuscript.

The collection efficiency of ACSM-SO_4^{2-} was derived from Yoshino et al. (2016). This paper is included in the reference list of the revised manuscript.

Line 206: Some discussion of how sensitive the results are to different choices for the percentile (i.e., does the background value change if concentrations lower than the 10th percentile were averaged?) would be helpful, or better yet, a distribution of the CO concentrations should be shown.

>When we set 10th percentile of CO mixing ratio as the threshold value, the derived background CO mixing ratio was calculated to be 131 ppb, which is slightly higher than the original value (120 ppb).   We prepared SI including the descriptions on the determination of the background CO mixing ratio.   Please see SI for details.

Line 277: The statement that the ACSM-SO4 and the IC-SO4 "generally agreed well" is true, but from Fig. 5c there appears to be little variability in either at concurrent times when comparison could be made.

>The variability in IC-SO_4^{2-} mass concentration was ~9 µg m^{-3} at STP (min - max ~1 - ~10).   Wider range of concentrations (<~20 µg m^{-3}) were observed during an intercomparison experiment in Queens/New York (Drewnick et al., 2003).   To the best of our knowledge, the observed range was larger enough to discuss the intercomparison results.   For example, Takegawa et al. (2005) reported the intercomparison results of SO_4^{2-} mass concentration between Aerodyne AMS and PILS-IC.   The range given in their study (< ~7 µg m^{-3}) is smaller than ours.

Line 284: It is not clear why the positive correlation of SO4 and CO suggests that the SO4 was secondary and that SO4 contributed to the BC coatings; more explanation of these assumptions/conclusions is required.

>Growth of BC-containing particles should be explained separately from the formation. Besides our observation results, previous studies support the description of formation and structure of the coating of BC in the original manuscript.   As the reviewer suggested, we revised the related sentences and included more explanation in the revised manuscript.

Line 290: The authors note "the small variability of SO4/CO ratios," yet Figure 6b shows that these ratios vary considerably.

>As the reviewer suggested, this statement and Figure 6b seem to contradict each other. We removed this sentence for the clarity.

Lines 294, 297: The two "experiments," which consisted of two brief time periods out of a month of data, were used to justify conclusions regarding flow patterns. While the results are indeed consistent with the arguments made, it seems difficult to justify such conclusions on the basis of one comparison.

>As the reviewer suggested, the results shown in this study are based on the observation during not-so-long time periods. We agree that it is actually difficult to draw the general conclusions. However, we still believe that this paper shows the significance in the observational studies of the relationship between removal process and the changes in the BC microphysical properties, because the observed meteorological conditions in the spring of 2015 were not special and similar to those with an average year. We added the sentences "The migrating anticyclone and cyclone were observed during this period, which is typically dominant in spring over East Asia (Asai et al., 1988). We here only briefly describe the meteorological fields (wind flow and precipitation) in the following." behind the first sentence in section 3.1, and modified the last sentence in section 3.5 to "As the results from this study are based on observations during a limited length of time, it would be worthwhile to further investigate the possible connections of the variabilities in BC microphysical properties and meteorological conditions in this region to provide useful constraints on more accurate evaluations of climatic impacts of BC-containing particles (Matsui, 2016)". Please see the revised manuscript for details.

Line 317: The authors refer to the SO4/CO ratio, but does this really refer to the deltaSO4/delta-CO ratio? It was unclear to me here and a number of places elsewhere in the test whether the CO and SO4 values referred to delta-CO and delta-SO4 values or not. For clarity, I would recommend using "delta-" values throughout.

>We clearly found the lower concentrations of SO_4^{2-} relative to CO for the data with the higher APT in Figure 6b of the original manuscript. Another reason not to include the $\Delta SO_4^{2-}/\Delta CO$ ratio is the uncertainty related to the variability in the background of SO_4^{2-} in East Asia. Although the use of the same data treatment would be clear for the readers, we did not quantitatively analyze the hourly ΔSO_4^{2-} and ΔCO values for considering the relative enhancements of SO_4^{2-} to CO in this study. We hence added the sentences to explain why we do not analyze Δ values in the revised manuscript in

section 2.2.

Lines 317-319: The difference in slopes shown in the inset to Figure 6b doesn't seem sufficiently large, given the scatter of the data, to be significantly different, and certainly not to justify the conclusion that the controlling process is rainout.
>The rainout lowered the transport efficiency of SO_4^{2-} as well as BC (to CO). However, the cloud process not associated with the precipitation can affect the relative increases of SO_4^{2-} concentration. The major purpose to include this figure is to elucidate the impact of the cloud process on the aqueous-phase formation of SO_4^{2-}, and is not to discuss the loss processes. Figure 6b is modified in the revised manuscript to clarify the data points with the APT of zero (no precipitation through the transport). These data points are highlighted by marking using cross markers. Please see the revised Figure 6b for details.

Line 343: Here and elsewhere the argument is made that aging leads to growth of BC particles, which is well accepted, but such aging can also lead to loss of larger particles through rainout, het size distributions in Figure 7 doesn't show much of a difference between size distributions for air masses with BC loss and those without, and certainly not more of a difference for larger BC particles than for smaller ones. This discrepancy requires explanation.
>All the size distributions shown in Figure 7 are normalized by the number or mass integrated for the measured size range, which is described in the caption of this figure. The "absolute" size distributions show more differences between with and without BC loss. We modified the size distributions from "normalized" to "absolute" and added a new figure (fig 7c of the revised manuscript) of the relationship between BC peak diameters and $\Delta BC/\Delta CO$ (i.e., degree of the removal of BC). This figure clarifies the significance of the observed changes in the peak diameter. Please see the revised figure for more details.

Line 345: The statement that "small BC-containing particles were scavenged by larger particles in the coagulation process" is a hypothesis, but stated as truth. It would seem that concentrations are too low for much coagulation over the brief period (a few days), especially for particles that are many tens of nanometers in diameter. Calculations or a simple model would be required to support this hypothesis. Line 353: It would be preferable, and less ambiguous, to rephrase "BC size of 0.2" to "BC diameter of 0.2".

>In the consideration of the washout process, the removal of small BC-containing particles through the washout is expected to be significant as well as the coagulation process.   We hence describe the possibility of both processes in the revised manuscript. We rephrased "BC size of 0.2" to "BC diameter of 0.2".

Line 368: The discussion focused on transport pathways of particles in the particular region of the study, but I was expecting more discussion on the results, what they mean, and so forth. There seemed to be little relevance to the second paragraph of the discussion.

>We reorganized the discussion part (section 3.5).   We merged and reorganized the first paragraph and the half of the second paragraph into one paragraph.   The latter half of the first paragraph of section 3.5 discusses the observed features and its relevance to the finding in previous studies.   We consider that the relationship between transport pathways (i.e., processes during transport) and its impact on the aerosol particles is a key and relevant to our observation results.   We hence modified the sentences of the third (second in the revised manuscript) paragraph.

Line 372: The decrease in the peak diameter of the mass size distribution is very small, and within uncertainty.

>The change in the peak diameter is small, however, significant.   Corresponded change in BC mass is ~1 fg/particle.   This difference can be resolved by the SP2 and beyond the uncertainty.   The variabilities of the peak diameters are summarized in Table 1 in the original and revised manuscript and are smaller than those measured. As we described in the above, we added a new figure to show the tendency of the BC particle diameter as a function of the degree of BC removal (Fig 7c of the revised manuscript).

Line 373: The statement that the evidence implies selective removal of large BC containing particles is not supported by Figure 7, which shows a very slight difference in the size distribution between "with BC loss" and "without BC loss" but not apparent selective decrease of larger particles. If there were selective removal, I would expect the size distribution to not be lognormal, but to have a deficit on the large side below what a lognormal would be. Figure 3a is very difficult to read; could it be made larger? Figure 3b requires units for q_v to accompany the scale. Figure 4a should be made larger also, if possible. Figure 5b: it is difficult to distinguish the COSMOS and SP2 BC values; perhaps make one

red and the other black? Figure 6a: do the axes refer to delta-CO and delta-BC? If so, they should be labeled as such. Figure 6b, inset: what does "all data" refer to? If this is to label the gray dot, then it is not clear.

>The activation of aerosol particles to cloud droplets has occurred during transport. We did not observe the aerosol particles below the convective cloud, because the migratory cyclone was the dominant process for the upward transport in spring in East Asia. We thus considered that SP2 detected BC-containing particles which have been aged (about a half ~ a day, typical transport time) since affected by the wet removal. The size distributions of BC-containing particles can change during transport again after the rainout process, and therefore do not always conserve the original shape.

We have corrected some figures as suggested. We enlarged all figures as large as possible as suggested. Units of all parameters in Fig 3 were clarified in the modified one. The color of SP2-BC in Fig 5 was changed to red. Axes of Fig 6a do not refer to delta (so we didn't change). Fig 6b was modified, because it was not clear. All the values in Figure 6 are absolute concentrations (not delta). Fig 7c was newly added (Please see the texts for details).

References

Asai, T., Y. Kodama, and J.-C. Zhu (1988), Long-term variations of cyclone activities in East Asia, Adv. Atmos. Sci., 5, 149−158.

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Matsui, H., Black carbon simulations using a size- and mixing-state-resolved three-dimensional model: 1. Radiative effects and their uncertainties (2016), J. Geophys. Res. Atmos., 121, 1793–1807, doi:10.1002/2015JD023998

Takami, A., et al. (2013), Structural analysis of aerosol particles by microscopic observation using a time-of-flight secondary ion mass spectrometer, J. Geophys. Res. Atmos., 118, 6726–6737, doi:10.1002/jgrd.50477.

Takegawa, N., Miyazaki, Y., Kondo, Y., Komazaki, Y., Miyakawa, T., Jimenez, J. L., Jayne, J. T., Worsnop, D. R., Allan, J. D., and Weber, R. J. (2005), Characterization of an Aerodyne Aerosol Mass Spectrometer (AMS): Intercomparison with Other Aerosol Instruments, Aerosol Sci. Technol., 39:760–770.

Yoshino, A., A. Takam, K. Sato, A. Shimizu, N. Kaneyasu, S. Hatakeyama, K. Hara, and M. Hayashi (2016), Influence of Trans-Boundary Air Pollution on the Urban

Referee #2 Dr. Gavin McMeeking
Review of Miyakawa et al.

We appreciate your helpful and constructive comments on the manuscript entitled "Alteration of the microphysical properties of black carbon through transport in the boundary layer in East Asia". As the two reviewers suggested, we have modified the manuscript. Major points for the revisions are listed as follows.

1) Title has been changed.
2) Supporting information (SI) has been prepared.
3) We have modified the discussion section.

*Note the reviewers' comments in **bold**.

The authors present a one month case study examining measurements of black carbon properties at a remote island site, using co-located measurements of CO and sub-micron aerosol composition and reanalysis data to evaluate precipitation impacts on the observed properties. The manuscript focuses on contrasting observed properties during periods with differing accumulated precipitation along backward trajectories. The paper is well prepared and well organized and the subject is well within the topic area for ACP. There are several areas where minor revisions are needed, however, before the paper can be recommended for publication. I agree with the points raised by Reviewer #1, so have tried to not repeat too much of what has been already raised. The comments should be addressed in a revised manuscript. In addition:

+ Given the focus of the manuscript, the introduction would benefit from a more thorough discussion of the various BC removal mechanisms, with more mechanistic details given as to why various processes may or may not be important in the study area. Distinction should be made between in-cloud processes (nucleation scavenging versus scavenging by pre-existing droplets), below-cloud (washout) and dry deposition.

We added the sentence describing the removal processes of BC to the second paragraph of section "Introduction". Relate to this, as the reviewer #1 suggested, we have modified the descriptions on the relative importance of the washout (to the rainout) (in section 3.3 of the original manuscript). A previous study (Kanaya et al., 2016) is referred for quantitatively elucidating the impacts of dry deposition. The timescale of the removal through the washout was evaluated in this study to be longer than the typical time scale of the transport (details in S.I. newly-prepared). Please see the

revised manuscript and SI for more details.

+ Two points regarding reported SP2-measured BC number/mass distributions. First, the manuscript needs to make it more clear when BC core versus shell diameters are being discussed, especially when linking the observations to theory. For example, while it is true we would expect larger particles to be removed in air masses heavily impacted by precipitation, the effects on BC core distributions will be confounded by other material mixed with the cores. Related to this, the diameter range for which the optical sizing of the BC particles should be provided in the methods section. Second, small changes in the detection efficiency of the SP2 at its lower limit due to changes in cavity laser power can look like changes in BC core number distribution. A short statement regarding any checks on cavity laser power or other approaches used to ensure consistent behavior at lower size limits for the instrument would be useful.

As the reviewer suggested, we added explanations on these SP2 working conditions in section2.1.   Please see the revised manuscript for more details.

+ Potentially useful additional information provided by the ACSM is being ignored by examining only sulfate. Is there a reason for this?

>We analyzed the concentration of SO_4^{2-} measured using the ACSM for the reasons, (1) "its precursor gas (sulfur dioxide) shares the emission sources and locations with CO", and (2) "its formation process in the aqueous phase reaction is useful for analyzing the effect of a possible cloud processing through air parcel transport".   We added more explanations especially on (2) to section 2.1.

References
Kanaya, Y., X. Pan, T. Miyakawa, Y. Komazaki, F. Taketani, I. Uno, and Y. Kondo (2016), Long-term observations of black carbon mass concentrations at Fukue Island, western Japan, during 2009-2015: Constraining wet removal rates and emission strengths from East Asia, Atmos. Phys. Chem., 16, 10689-10705, doi:10.5194/acp-16-10689-2016, 2016

Alteration of the ~~microphysical properties~~size distributions and mixing states of black carbon through transport in the boundary layer in East Asia

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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~~ we report on the temporal variations in the transport, size distributions, and mixing states of the BC-containing particles. These particles

were characterized by 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 aerosols through gas- and cloud-phase reactions 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 (ΔBC/ΔCO), which were used as an indicator of the transport efficiency of BC. The findings showed that ΔBC/ΔCO ratios were controlled mainly by the rainout process 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 rainout originated mainly from a region in Southern China (20º–35ºN) in the spring.   Selective removal of large and thickly-coated BC-containing particles was detected in the air masses that were substantially affected by the rainout 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 rainout process 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 types of effects, namely, direct (light absorption and scattering) effect 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 of BC-containing particles relative to other greenhouse gases and the variable physicochemical properties of ~~BC containing~~such particles. The BC itself is water-~~ ~~insoluble immediately after emission, but it subsequently ~~takes~~exhibits ~~on~~hygroscopicity (McMeeking et al., 2011) and cloud condensation nuclei (CCN) activity (Kuwata et al., 2007) ~~through~~following atmospheric transport and aging. Only small amounts of water-soluble materials on BC particles are needed for ~~to cause~~their activation, whereby they will ~~to~~ form cloud droplets under moderate supersaturation conditions (Kuwata et al., 2007; 2009). ~~It is considered that~~The BC-containing particles are thought to be removed from the atmosphere mainly by ~~the rainout~~wet deposition~~process. This is because other removal processes such as gravitational settling, dry deposition, and washout cannot substantially affect the lifetime of atmospheric BC containing particles~~ (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, their studies suggested that ~~T~~the removal processes of BC such as dry deposition, below-cloud (i.e., washout), and in-cloud (i.e., rainout) processes 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 process 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 ~~resulting~~results in significant variations in global direct radiative forcing values. The removal of BC has been considered to be an important issue for

the geochemical carbon cycle as well as for climate sciences. The BC-containing particles deposited onto the ocean surface can affect ocean surface particles, dissolved organic carbon (DOC), and microbial processes, by via absorbing DOC, stimulating particle aggregation, and changing the size distribution of suspended particles (Mari et al., 2014).

Previous modeling studies have dealt with the BC aging processes (condensational growth and coagulation) infor box and regional-scale models, and parameterized timescales for the conversion of BC-containing particles from water- insoluble to soluble for in global models (Oshima et al., 2009; Liu et al., 2011; Oshima and Koike, 2013). However, Quantitative quantitative knowledge of the variability of microphysical parameters of BC-containing particles and the timescale of their aging processes is still limited, and thus is more data are 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 to during the formation of cloud droplets, in air masses uplifting from the planetary boundary layer (PBL) to the free troposphere (FT) in East Asia in during the spring of 2009, during as part of the Aerosol Radiative Forcing in East Asia (A-FORCE) aircraft campaigns (Oshima et al., 2012). The A similar altitude dependence of the BC size distribution and similarity in the BC mixing state were was observed in other aircraft measurements conducted in East Asia in during the winter (Kondo et al., 2016). Selective removal of larger BC-containing particles though the cloud process, 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 control the global- and regional-scale spatial distributions of BC through their upward transport

from the PBL to the FT in ~~associated~~ association with rainout processes.   Despite the importance of the size distributions and mixing states of BC-containing particles in the PBL, the ~~continuous~~ 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 synthetically reported on the emission and removal of BC in East Asia by using these data sets.   ~~It was found i~~n their study, they found that wet removal through transport in the PBL substantially reduced the transport efficiency of BC aerosols.   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 taken at the same site in the spring of 2015 by using a single particle soot photometer (SP2) and an Aerosol Chemical Speciation Monitor (ACSM).   We first describe~~show~~ the meteorological characteristics of the East Asian region in the spring of 2015.   Then, ~~W~~we discuss the relative importance of ~~below cloud~~the washout and rainout~~in cloud~~ processes for the wet removal of BC as well as the transport patterns of the East Asian outflow air masses in spring.   ~~Then~~Lastly, the loss of BC-containing particles for that period is investigated by 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 ~~with~~to the transport pathways.

2.   Experimental and data analysis
2.1. Atmospheric ~~Observations~~observations
~~The c~~Continuous measurements of PM$_{2.5}$ and BC aerosols ~~has~~ have been conducted at a remote island, Fukue Island, since February 2009 (Kanaya et al., 2013; Ikeda et al.,

122 2014).   The observation site is located at the Fukue Island Atmospheric Environment

123 Monitoring Station (32.75°N, 128.68°E, **Fig. 1**).), and The the site is located situated

124 in the northwest portion of Fukue Island, approximately 20 km from the main

125 residential area in the southeast.   The fine mode aerosols sampled at the site are

126 mostly transported from areas beyond the island.   The enhanced concentrations of BC

127 aerosol detected at in Fukue Island are can be mainly attributed to long-range transport

128 from the Asian continent, according to a previous study (Shiraiwa et al., 2008) and an

129 emission inventory work (**Fig. 1**, REAS ver. 2.1, Kurokawa et al., 2013).

130   We deployed an SP2 (Droplet Measurement Technologies, Inc., USA) for the

131 analysis of microphysical parameters of refractory BC (rBC, Petzold et al., 2013) from

132 March 26, 2015 to April 14, 2015.   The SP2 was calibrated before starting the

133 ambient measurements.   The calibration protocol for our SP2 is described in

134 Miyakawa et al. (2016).   Fullerene soot (FS, stock 40971, lot L20W054, Alfa Aesar,

135 USA) particles were used as a calibration standard for the SP2.   The variations in the

136 laser power were within ±3% during the observation period, thus indicating that the

137 fluctuations of laser power did not largely affect the lower limit of the detectable BC

138 size of the SP2.   The Mass mass equivalent diameter (MED) was derived from the

139 rBC mass per particle (m_{pp}) with the an assumed particle density of for BC (1800 kg

140 m^{-3}, Bond and Bergstrom, 2006).   A large diameter Nafion dryer (MD-700, Perma

141 Pure, Inc., USA) was placed in front of the SP2 for to drying the sample air without

142 significant loss of the aerosol particles greater than 50 nm.   The dry air for MD-700

143 was generated by a heatless dryer (HD-2000, Perma Pure, Inc., USA) and a

144 compressor (2AH-23-M222X, MFG Corp., USA).   The relative humidity of the

145 sample air was less than 20% during the observation period.   The hourly

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number/mass size distributions and hourly median values of shell (D_S) to rBC diameter (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 by using the time-resolved scattering cross section method given by Laborde et al. (2012). In this study, we analyzed the D_S of BC-containing particles with a D_{core} range between 0.15 and 0.35 µm. We also analyzed the microphysical parameters of rBC particles that were measured by using the SP2 in the early summer of 2014 at Yokosuka (35.32°N, 139.65°E, **Fig. 1**), which is 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 were continuously measured at Fukue Island by 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 intercomparison for EBC measurements at Fukue Island have been described elsewhere (Kanaya et al., 2013; 2016). In this study, mass concentrations of EBC measured by using the COSMOS were evaluated by comparison with those of SP2-derived rBC. The intercomparison between SP2 and COSMOS is 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 extrapolation of the lognormal fit for the measured mass size distributions, to the outsides of the measurable D_{core} range (0.08 and 0.5 µm,

170 respectively). Note that the uncertainty with respect to the unmeasured fraction of

171 rBC mass was minor (<5%) in this study.   The linear regression slope of the

172 correlation between EBC and rBC was 0.88 (±0.03).   Uncertainty with respect to the

173 calibration was examined in an industrial region and found to be within around 3%

174 (Miyakawa et al., 2016).   The average discrepancy between EBC and rBC was

175 beyond the uncertainty of the calibration and was comparable to the uncertainty of

176 COSMOS (10%) as evaluated by Kondo et al. (2009).   AsWhile the validity of the

177 calibration standard, FS particles, has been evaluated only near source regions (Moteki

178 and Kondo, 2011; Miyakawa et al., 2016), the discrepancy can be partly attributed to

179 the differences in physicochemical properties between ambient BC in remote air and

180 FS particles.   Onsite calibration of the SP2 using ambient BC particles prepared by a

181 thermal denuder and particle mass classifier, such as an aerosol particle mass analyzer

182 (APM), is desirable for the better quantification of the rBC mass based on the

183 laser-induced incandescence technique in a remote areas.   Although we need to make

184 further attempts to evaluate SP2 in remote areas, the data derived from this study

185 suggested indicated that SP2-rBC mass concentrations agreed well with

186 COSMOS-EBC values within the uncertainty uncertaintiesof COSMOS.

187 ThereforeThus, wWe simply use "BC", here instead of the EBC and rBC defined

188 depending upon the measurement techniques.   We analyzed the COSMOS data for

189 the BC mass concentrations, and the SP2 data for the BC microphysics.

190   The chemical composition of non-refractory submicron aerosols was measured by

191 using an Aerodyne Aerosol Chemical Speciation Monitor (ACSM, Aerodyne, Inc.,

192 USA.) placed in an observatory container at Fukue Island during the observation

193 period.   The details of the ACSM at Fukue Island have been described in Irei et al.

194 (2014).   The collection efficiency (CE) of the ACSM was assumed to be 0.5 for this

195 period (Yoshino et al., 2016).   We considered sulfate ~~(SO_4^{2-})~~ ions (SO_4^{2-}) ~~as~~ to be the~~a~~

196 major non-BC material and ~~the~~ most important secondary aerosols in East Asia

197 (Takami et al., 2007) for the data interpretation.   The fact that SO_4^{2-} is produced in the

198 cloud phase as well as the gas phase is ~~a benefical~~ ~~to~~for interpreting temporal changes

199 in SO_4^{2-} concentration associated with the wet removal process.   During the period

200 April 1—7, 2015, the critical orifice of the inlet assembly of the ACSM ~~was~~ became

201 clogged.   Therefore, the ACSM-derived SO_4^{2-} (ACSM- SO_4^{2-}) for this period wa~~i~~s not

202 used in the analysis.

203   Two high volume air samplers (HV500F, Sibata Scientific Technology, Ltd., Japan)

204 were deployed on the rooftop of the observatory container.   The sampling flow rate

205 for both samplers was 500 liters per minute (lpm).   Air sampling was carried out for

206 21 h (from 10:00 AM to 7:00 AM) on a 110-mm pre-combusted (900°C for 3 h) quartz

207 filter (QR-100, Advantec Toyo Kaisha Ltd., Japan).   Both have a $PM_{2.5}$ impactor for

208 classifying the particle size.   One impaction plate was coated with vacuum grease

209 (HIVAC-G, Shin-Etsu Chemical Co., Ltd., Japan) to minimize the impact of coarse

210 mode particles on the chemical analysis of fine mode particles, such as during

211 radiocarbon analysis, and a pre-combusted quartz fiber filter with slits was set on

212 another impaction plate to collect the coarse particles.   Water soluble ions were

213 analyzed by using ion chromatography (IC, Dionex ICS1000, Thermo Fisher Scientific

214 K.K., Japan).   The results from the chemical analysis of filter samples are not

215 ~~included~~ discussed in this study in detail.   We only used the mass concentration of

216 sulfate ions (IC-SO_4^{2-}) in this study to evaluate the uncertainty in relation to CE of the

217 ACSM, and to analyze the temporal variations during the period when the

218 ACSM-SO$_4^{2-}$ data weare not available (April 1–7, 2015).

219 The carbon monoxide (CO) mixing ratio wais also continuously measured by using

220 a nondispersive infrared (NDIR) CO monitor (model 48C, Thermo Scientific, Inc.,

221 USA). Details of the CO measurements including the long-term variations in

222 sensitivity and zero level are discussed elsewhere (Kanaya et al., 2016).

223

224 **2.2. Enhancement ratio of BC and SO$_4^{2-}$ to CO as an indicator of the ~~BC~~**

225 **~~removal~~transport and transformation of aerosol particles**

226 In order to quantify the extent of the removal of BC, we calculated the hourly

227 enhancement ratio of BC mass concentrations to CO mixing ratios (ΔBC/ΔCO) against

228 the East Asian background air concentrations as follows:

229

230
$$\frac{\Delta BC}{\Delta CO} = \frac{[BC] - [BC]_{bg}}{[CO] - [CO]_{bg}},$$
(1)

231

232 where [BC] and [CO] are measured hourly concentrations of the BC and CO,

233 respectively, and [BC]$_{bg}$ and [CO]$_{bg}$ are their estimated background concentrations.

234 Here, we assumed that [BC]$_{bg}$ is zero (Oshima et al., 2012). The background

235 concentration of CO during the analysis period (March 11–April 14, 2015) was

236 calculated by averaging the concentrations lower than the 5th percentile (120 ppb).

237 The validity of this value wais discussed in the supporting information (S.I.).

238 Relative changes in SO$_4^{2-}$ to CO were also analyzed by using the linear regression

239 slopes of their correlation in this study. We havedid not calculated the hourly

240 ΔSO$_4^{2-}$/ΔCO values, because it iwas difficult to determine the background

10

concentration of SO_4^{2-}. Analyzing the slope of the SO_4^{2-}–CO correlation is beneficial to the investigation of the formation processes as well as the removal processes offor SO_4^{2-}. Especially, the aqueous-phase formation of SO_4^{2-} in clouds is discussed by 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 along with 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. 3Three-day backward trajectories from the observation site (the starting altitude was 0.5 km) were calculated every hour by using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory model (Draxler and Rolph, 2012; Rolph, 2012) with and 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

11

265 China (CN), Central South China (CS), and Japan (JP) (**Fig. 1**).   The period when air

266 masses passed over the domains NE, KR, CN, and CS at least for one hour is defined

267 as that of "continental outflow".   The impacts of precipitation on the observed air

268 masses were assessed by a parameter referred to as the "Accumulated Precipitation

269 along Trajectory" (APT, Oshima et al., 2012).   In this study, we calculated the APT

270 values by integrating the amount of hourly precipitation in the Lagrangian sense along

271 each 3-day back trajectory of the sampled air masses.   The hourly variations of APT

272 were merged into the observed gas and aerosol data sets.

273

274 **3.   Results and discussion**

275 **3.1.   The meteorological field in the spring of 2015**

276   The mean meteorological field during the observation period (March 11–April 14,

277 2015) is discussed for the purpose of characterizing the general features of the wind

278 flow and precipitation in this region.   The migrating anticyclone and cyclone were

279 observed during this period, which is typically dominant in spring over East Asia (Asai

280 et al., 1988).   The meteorological conditions over East Asia during the observation

281 period were generally usual.   We here only briefly describe the meteorological fields

282 (wind flow and precipitation) in the following.   **Figure 3a** shows the mean sea level

283 pressure (SLP) and mean horizontal winds at the 850 hPa level in East Asia during the

284 observation period.   The mean equivalent potential temperature (θe) and the

285 meridional moisture transport at the 850 hPa level during the same period are also

286 shown in **Figure 3b**.   The mid-latitude region (35–50°N, 120–140°E) was under the

287 influence of a modest monsoonal northwesterly flow, which advected cold, dry air

288 from the continent to the observation area.   The subtropical region (20°–30°N, 110°–

289 –130°E) was under the influence of a persistent southwesterly flow, part of which was

12

conversing into the observation area (30°–35°N), and this flow wasbeing confluent with the northwesterlies from the continent.   The low-level southerly flow advected warm, moist air into the observation area, which produced 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 was well anti-correlated 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 south to Northnorth), and these results suggesting 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 backwardly from the time of -24 h (Lat_{ORIG}) (**Fig. 4b**), which and these data can be interpreted as an indicator for 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°–40ºN).   The data points are colored according to the maximum relative humidity (RH) values along each backward trajectory (RH_{max}).   The lower RH_{max} were observed in the air masses with low APT values and that originated from the northern regions (30°–50ºN).   These air mass

13

314 characteristics ~~are~~ were consistent with the mean precipitation field (**Fig. 4a**).   Some

315 of the data points showed high values of RH$_{max}$ (~100%) when their APT was almost

316 zero.   These data ~~would~~ probably correspond to the air masses that experienced cloud

317 process~~es~~ not associated with precipitation.   Possible effects of cloud process~~es~~

318 without precipitation on the removal of aerosol particles during transport will be

319 discussed by using these data points in the following section.

320

3.2. Temporal variations in BC, SO$_4$$^{2-}$, and CO

322   Temporal variations in the concentrations of BC (measured by using COSMOS and

323 SP2), SO$_4$$^{2-}$ (measured by using ACSM and IC), and CO are shown in **Figure 5**

324 ~~(middle and bottom panels respectively)~~.   ACSM-SO$_4$$^{2-}$ generally agreed well with

325 IC-SO4, thus indicating that the assumed CE (0.5) was valid for the observation period.

326 In general, BC and SO$_4$$^{2-}$ were positively correlated with CO at Fukue Island, and these

327 results illustrate~~showing~~ the impact of continental outflow affected by incomplete

328 combustion sources ~~for~~ on aerosol mass concentrations.   **Figure 5** also includes the

329 temporal variation~~s~~ in the fractional residence time over the selected region defined in

330 section 2.4 (top panel).   The CO concentrations were typically enhanced for the

331 period with the higher contributions of CN and CS.   ~~A previous study~~Sahu et al.

332 (2009) suggested that the majority of SO$_4$$^{2-}$ aerosols ~~was~~ formed ~~by~~in less than around

333 1.5 days after the air masses left~~ave~~ the Chinese continent ~~(Sahu et al., 2009)~~.   The

334 positive correlation of SO$_4$$^{2-}$ and CO suggests that the secondary formation of SO$_4$$^{2-}$

335 through transport was significant during the observation period~~, and that SO$_4$$^{2-}$~~

336 ~~contributed to the coating of BC containing particles.   A previous study suggested~~

337 ~~that the majority of SO$_4$$^{2-}$ aerosol was formed by less than around 1.5 days after the air~~

14

338 ~~masses left the Chinese continent (Sahu et al., 2009).~~ Kanaya et al. (2016) showed

339 that the typical transport time of continental outflow air masses at Fukue Island was

340 around 1–2 days in spring. ~~The small variability of SO_4^{2-}/CO ratios is consistent~~

341 ~~with these facts.~~ The structure and composition of submicron aerosols in East Asian

342 outflow ~~have been~~were analyzed by using a secondary ion mass spectrometer in a

343 previous study (Takami et al., 2013). The~~y~~ose findings suggest that SO_4^{2-} and

344 organics ~~compose~~are constituents in ~~a~~the coating of almost all ~~of~~ BC-containing

345 particles. Hence ~~W~~we ~~hence~~ have concluded that the formation of SO_4^{2-} during

346 transport can contribute to the growth of BC-containing particles.~~–~~ The period with

347 the APT > 3 mm is highlighted by light blue in **Figure 5** to show the impacts of wet

348 removal on the transport of BC and SO_4^{2-} aerosols. The maximum concentrations of

349 BC, SO_4^{2-}, and CO were observed on the morning of March 22 (Ep. 1) under the

350 influence of the anticyclone (corresponding to the trajectories colored red in **Fig. 4a**)

351 when the APT values were almost zero. In contrast, BC and SO_4^{2-} concentrations did

352 not increase with CO in the period from the evening of April 5 to the morning of April

353 6 (Ep. 2) under the influence of the migratory cyclone (corresponding to the

354 trajectories colored black in **Fig. 4a**), when the APT was greater than 10 mm.

355

356 **3.3. Correlation of BC, SO_4^{2-}, and CO as an indicator of the removal of aerosols**

357 **Figures 6a** and **6b** show scatter plots of CO with BC and SO_4^{2-}, respectively.

358 Positive correlation of BC and SO_4^{2-} with CO was clearly found in air masses with low

359 APT values. It is evident from these scatter plots that the ~~correlations~~ relative

360 enhancements of BC~~/CO~~ and SO_4^{2-}~~/~~ to CO ~~are~~ were mainly ~~affected~~ reduced by the

361 APT. The cloud process~~es~~ of aerosol particles not associated with precipitation can

15

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 zero was found during the observation period (data not shown).   Kanaya et al. (2016) found that the estimated emission ratios of BC to CO over the East Asian continent ranged from 5.3 ($\pm$2.1) to 6.9 ($\pm$1.2) ng m^{-3} ppb^{-1}, and slightly ~~depending~~ depended on the origin of the air masses (this range is overlaid on **Fig. 6a**). ΔBC/ΔCO observed in the PBL over the Yellow Sea ~~in~~ during the same season was 6.2 ng m^{-3} ppb^{-1} (Kondo et al., 2016).   The data points with ΔBC/ΔCO in these ranges ~~show~~ displayed low APT values (less than or ~1 mm).   Wet removal (rainout) was one of the most important controlling factors on the transport efficiency of BC in this region during the observation period.   This work demonstrates that ~~Tt~~he use of ~~the~~ ΔBC/ΔCO ratios is feasible for examining the wet removal of BC during the observation period.

~~The SO_4^{2-}/CO slope slightly increased with RH_{max} increasing when the APT was zero, as indicated in the subset of~~ **Figure 6b**, ~~suggesting 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 are controlled largely by the rainout process and weakly by aqueous phase formation during transport.~~In this study, the other removal processes including dry deposition and washout ~~wea~~re considered to be minor.   The dry deposition in this region has been evaluated by Kanaya et al. (2016).   The removal through the washout process is dependent on the precipitation intensity and rain drop size as well as the particle size range.   We quantitatively considered the relative importance of rainout to washout. The removal timescale of submicron accumulation particles through the washout was estimated to be around a week or longer, which is much longer than the typical

transport time of air masses exported from the East Asian continent.   ~~The m~~More details ~~of~~about the estimation are given in the S.I.   Note that the Aitken and coarse mode particles, which were not measured by the SP2, can be significantly affected.   ~~The more details of the estimation are given in S.I.~~

The SO_4^{2-}/CO slopes with the APT values of zero (i.e., non-precipitation) were analyzed as a function of RH_{max} (**Figure 6b**).   The SO_4^{2-}/CO slopes without precipitation varied f~~o~~rom 30.7 to 44.1 ng m^{-3} ppb^{-1} ~~in~~under the conditions ~~between~~ without (RH_{max} ~~>8~~<50%) and with~~out~~ (RH_{max} ~~<5~~>80%) cloud impacts, respectively. The SO_4^{2-}/CO slope increased with increases in RH_{max} ~~increasing~~ when the APT was zero, thus suggesting 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 we~~a~~re controlled largely by the rainout process and weakly by aqueous-phase formation during transport.

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

Number- and mass -size distributions of BC classified by the values of ΔBC/ΔCO are shown in **Figures 7a** and **7b**, respectively.   When ΔBC/ΔCO values in continental outflow air masses we~~a~~re greater than 3 ng m^{-3} 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 ΔBC/ΔCO values of continental outflow air masses are less than 1 ng m^{-3} ppb^{-1}, the air masses we~~a~~re defined as "outflow with BC loss".   Considering the typical emission ratios of BC to CO (6_-7 ng m^{-3} ppb^{-1}; Kanaya et al., 2016), transport efficiency for the "outflow with BC loss" air masses can be estimated to be less than

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411 ~17%.   These air masses originated mainly in CS.   The low and high APT values for

412 "outflow without BC loss" and "outflow with BC loss" air masses, respectively, ~~give~~

413 gave us confidence in the validity of our classification as discussed in the previous

414 section.   As a reference for emission sources ("source"), the average size distributions

415 of BC in a Japanese industrial area (see section 2.1, Miyakawa et al., 2016) are shown

416 in **Figure 7**.   The statistics of the size distributions a~~we~~re summarized in **Table 1**.

417 Observed differences in the size distributions between source and outflow were

418 generally consistent with previous studies (Schwarz et al., 2010).   Air mass aging

419 leads to the growth of BC-containing particles.   Number-size distributions of BC

420 largely varied in the size range less than 0.1 μm (**Fig. 7a**).   In outflow air masses,

421 such small BC-containing particles were scavenged by larger particles in the

422 coagulation process during transport.   The washout process can also affect the

423 BC-containing particles in the smaller size range (<0.1 μm) as indicated in the S.I.

424 The peak diameter of mass (number) size distributions of BC became larger, i.e., from

425 0.16 (0.06) μm to 0.18–0.2 (0.09–0.1) μm, between source and outflow.   The

426 BC-containing particles ~~have~~ had systematically different size distributions in outflow

427 air masses with and without BC loss, thus indicating that the BC loss process also

428 affected the size distributions.   The peak diameter of BC number and mass size

429 distributions in outflow air masses with BC loss was slightly lower than that for air

430 masses without BC loss.   The changes in the peak diameter as a function of

431 ΔBC/ΔCO values are shown in **Figure 7c**.   The observed changes in the diameter or

432 mass per particle a~~were more~~very clear~~ly found~~ and a~~were beyond the uncertainties.

433   **Figure 8** depicts the probability density of the D_S/D_{core} ratio for the BC size of 0.2

434 (±0.02) μm, for source and outflow air masses.   The modal values of the D_S/D_{core}

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435 ratio were systematically changed with air mass aging and BC loss (wet removal).

436 The condensation of inorganic and organic vapors on BC-containing particles during

437 transport can account for the increase in the D_S/D_{core} ratio, as discussed in previous

438 studies (e.g., Shiraiwa et al., 2008; Subramanian et al. 2010; Shiraiwa et al., 2008).

439 As discussed earlier, the results of this study suggested that SO_4^{2-} substantially

440 contributed substantially to the increase in the D_S/D_{core} ratio. In outflow air masses

441 with BC loss, modal values of the D_S/D_{core} ratio were clearly lower than those in

442 outflow without BC loss. Furthermore, It it is indicated that the wet removal process

443 also affected the coating thickness distributions for the BC sizes in the range 0.15–

444 –0.35 μm (**Table 1**). It should be noted that the coating of BC-containing particles is

445 not always thick in remote regions, and that the D_S/D_{core} ratio distributions, as well as

446 size distributions, can be affected by the wet removal process during transport in the

447 PBL.

448

449 **3.5. Discussion**

450   Not only in-cloud scavenging of BC-containing particles but also subsequent

451 precipitation (i.e., the rainout process) can account for the changes in the

452 microphysical parameters of BC detected in this study. Our results showed the a

453 decrease of both the peak diameter of the BC mass size distribution, and the modal

454 value of the D_S/D_{core} ratios in relation to the rainout. The observed evidence implies

455 the that there is selective removal of large and water -soluble BC-containing particles

456 during transport in the PBL. The Köhler theory suggests that a lower super saturation

457 is needed for the large and highly water soluble particles,soluble particles, and this can

458 qualitatively accounts for the observed changes in the BC microphysics.–

459   Note that the magnitude of the change in the BC size distributions in the PBL (0.01–

460   –0.02 μm (~1 fg)) shown in this study is smaller than that observed in air masses

461   uplifted from the PBL to the FT, in association with wet removal (~0.04 μm (~3 fg),

462   Fig 2 of Moteki et al., 2012) at a similar level of transport efficiency (<~20%). The

463   deployment of a highly -sensitive instrument, SP2, enabled us to quantify such small

464   changes in the BC microphysics in the PBL.   Air masses sampled at the ground level

465   would be affected by turbulent mixing of those near the clouds around the top of the

466   PBL and those in cloud-free conditions at below-cloud levels.   On the other hands,

467   most air masses sampled by the aircraft measurements in the FT would experience the

468   cloud processes during upward transport from the PBL.   Mixing of air masses in the

469   PBL suggests that theytheir partially experience of the in-cloud scavenging processes

470   and therefore the suppression of the changes in the microphysical properties of

471   BC-containing particles during transport in the PBL.

472   The transport pathways of the continental outflow air masses are horizontally and

473   vertically variable in spring in East Asia because of the frequent alternateive

474   cyclone/anticyclone activities in spring (Asai et al., 1988).   Oshima et al. (2013) have

475   examined the three-dimensional transport pathways of BC over East Asia in spring and

476   showed that the PBL outflow through which BC originating from China was advected

477   by the low-level westerlies without uplifting out of the PBL was one of the most major

478   pathways for BC export from continental East Asia to the Pacific, thus supporting the

479   general features of microphysical properties of BC in continental outflow obtained by

480   this study.   Mori et al. (2014) measured the seasonal variations in BC wet deposition

481   fluxes at another remote island in Japan (Okinawa, ~500 km south of Fukue Island),

482   and revealed their maxima in spring, which is were consistent with the seasonal

483 variations in the cyclone frequencies. It ~~is~~ has been suggested that BC-containing

484 particles were efficiently activated to form cloud droplets in the continental outflow air

485 masses, especially from the CS region, and can affect the cloud physicochemical

486 properties in spring in East Asia, as indicated by Koike et al. (2012). As the results

487 from this study are based on ~~the~~ observations during ~~the~~a limited length of ~~the~~

488 ~~periods~~ime, it would be worthwhile ~~is needed~~ to further investigate the possible

489 connections of the variabilities in BC microphysical properties and meteorological

490 conditions in this region ~~for~~to provid~~ing~~e useful constraints on more accurate

491 evaluations of climatic impacts of BC-containing particles (Matsui, 2016)~~To further~~

492 ~~understand the possible connections of the variabilities in BC microphysical properties~~

493 ~~and meteorological conditions in this region can provide useful constraints on the~~

494 ~~better prediction of climatic impacts of BC-containing particles (Matsui, 2016).~~

495

4. Conclusions

497   Ground-based measurements of BC were performed near an industrial source region

498 and at a remote island in Japan. We have reported on the temporal variations in the

499 transport and the microphysics of the BC-containing particles that were, measured by

500 using ~~the~~ COSMOS, SP2, and ACSM. The impacts of air mass aging upon the

501 growth of BC-containing particles were examined by comparing the ground-based

502 observations between the near-source and remote island sites. $\Delta BC/\Delta CO$ was used as

503 an indicator of the transport efficiency of BC, because it was controlled mainly by

504 rainout during transport in the PBL. The BC size and coating increased during

505 transport from the near-source to the outflow regions on the timescale of 1–2 days

506 when the rainout during transport was negligible. SO_4^{2-} aerosol was secondarily

507 formed both in the gas- and cloud- phases during transport, and it contributed to the

508 significant increase in the coating materials of BC (i.e., it enhanced the whole size and

509 water -solubility of BC-containing particles).   Decreases in the peak diameter of mass

510 size distributions (~0.01 μm) and modal D_S/D_{core} ratios (~0.4 for BC with a diameter of

511 0.2 μm) of BC-containing particles were observed in air masses substantially affected

512 by rainout.   The observed evidence~~s~~, for the selective removal of large and

513 water-soluble BC-containing particles, was ~~are~~ qualitatively consistent with the Köhler

514 theory; however ~~the~~they values ~~are~~ were not as large as those found in air masses

515 uplifted from the PBL to the FT in East Asia associated with precipitation.   The

516 mixing of below-cloud and in-cloud air masses in the PBL would result in suppression

517 of the degree of changes in BC microphysical parameters by cloud processes.   This

518 study indicates (1) that the changes (sign and degree) in BC microphysics can be

519 affected by how the air masses are transported and~~,~~ (2) that the observed selective

520 removal of large and water-soluble BC-containing particles in East Asia ~~are~~ can be

521 expected to be significant in the PBL as well as in the FT in East Asia.

522

523

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534

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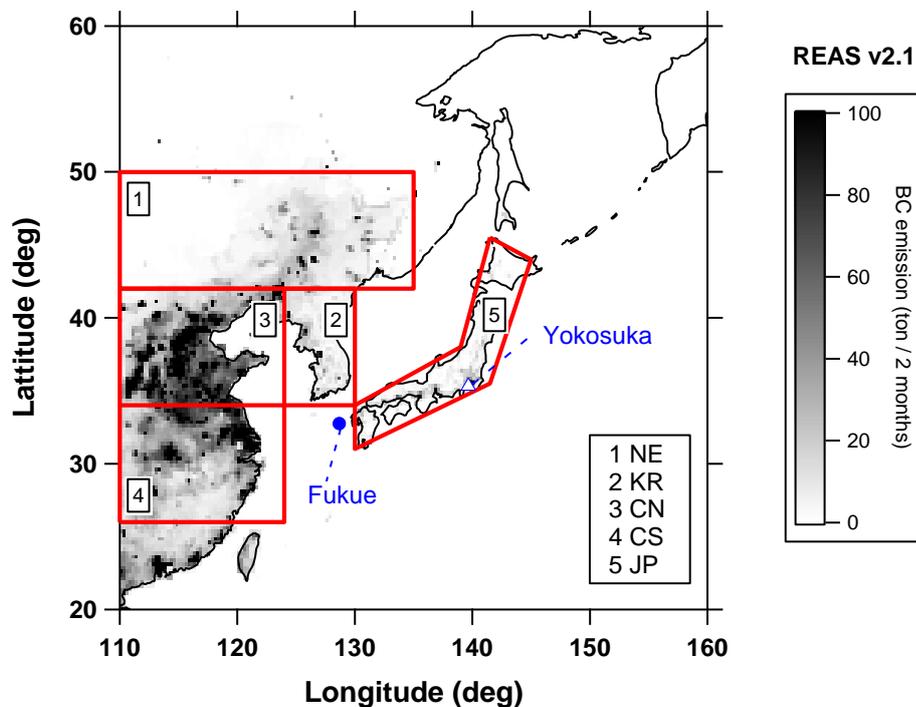
Figures

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).

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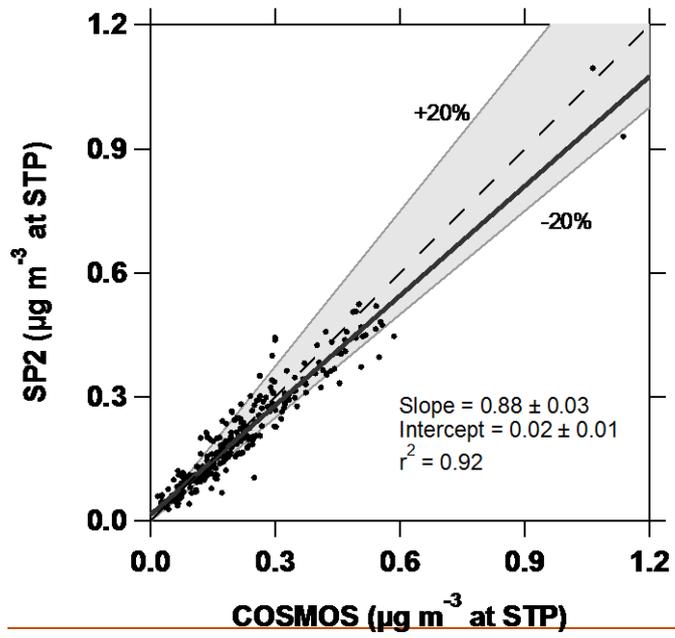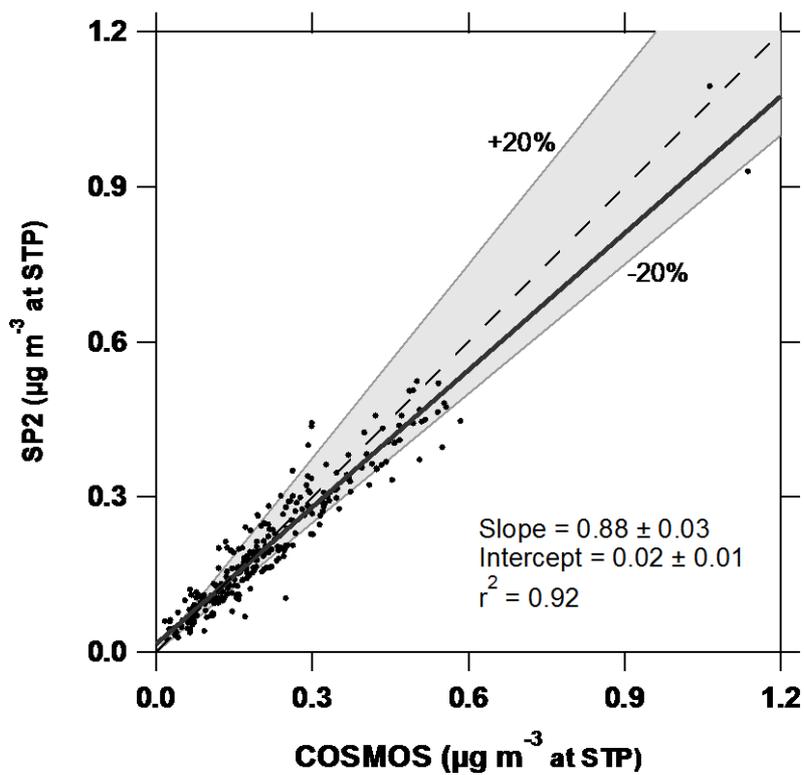
Figure 2. Correlation plot of SP2-rBC and COSMOS-EBC mass concentrations (at standard temperature and pressure~~STP~~). The shaded region corresponds to the levels within ±20%.

(a) Mean SLP and Mean Wind Velocity (850 hPa)

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(a) Mean SLP and Mean Wind Velocity (850 hPa)
(b) Mean θe (850 hPa) and Mean qv (850 hPa)

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(b) Mean θe (850 hPa) and Mean qv (850 hPa)

θe (K)

$\xrightarrow{70}$ qv (m s^{-1} g kg^{-1})

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(c) Surface Pressure and Precipitable Water

(c) Surface Pressure and Precipitable Water
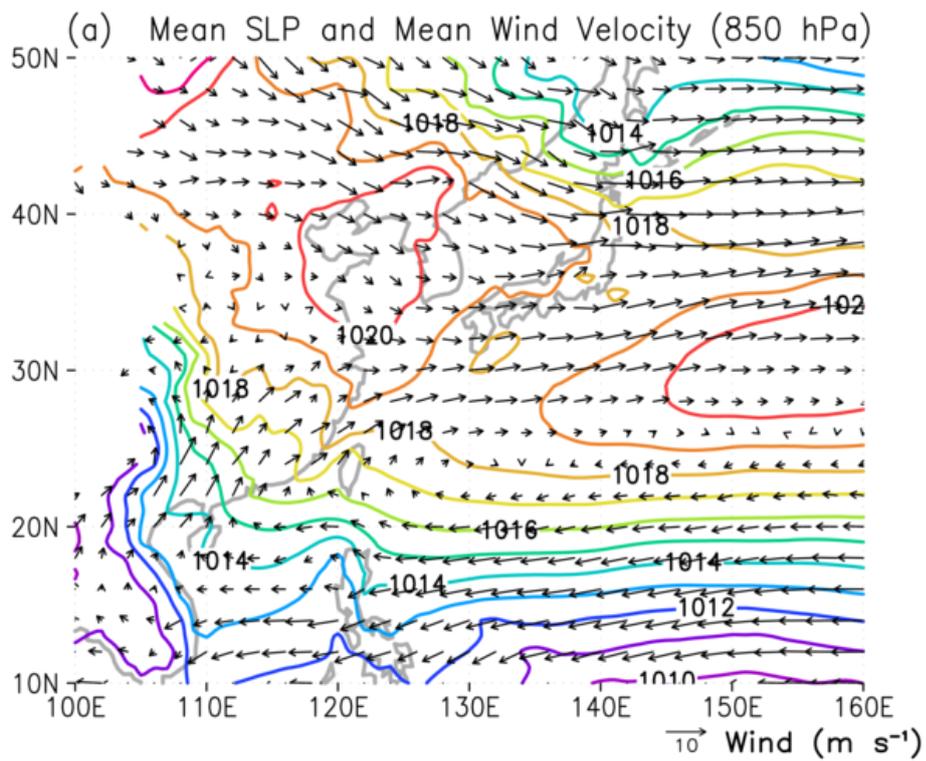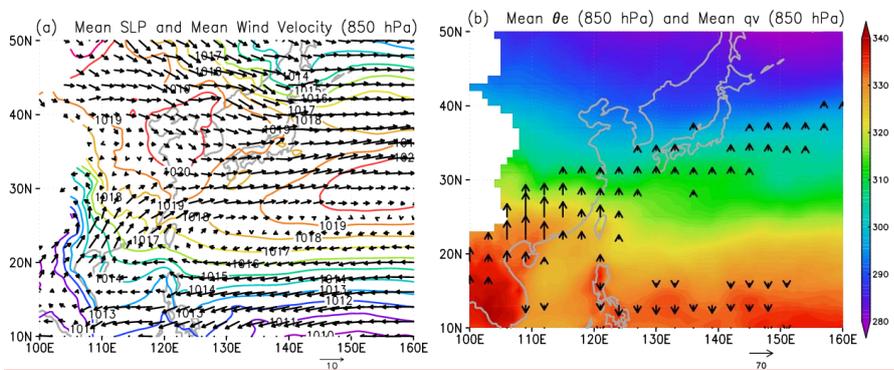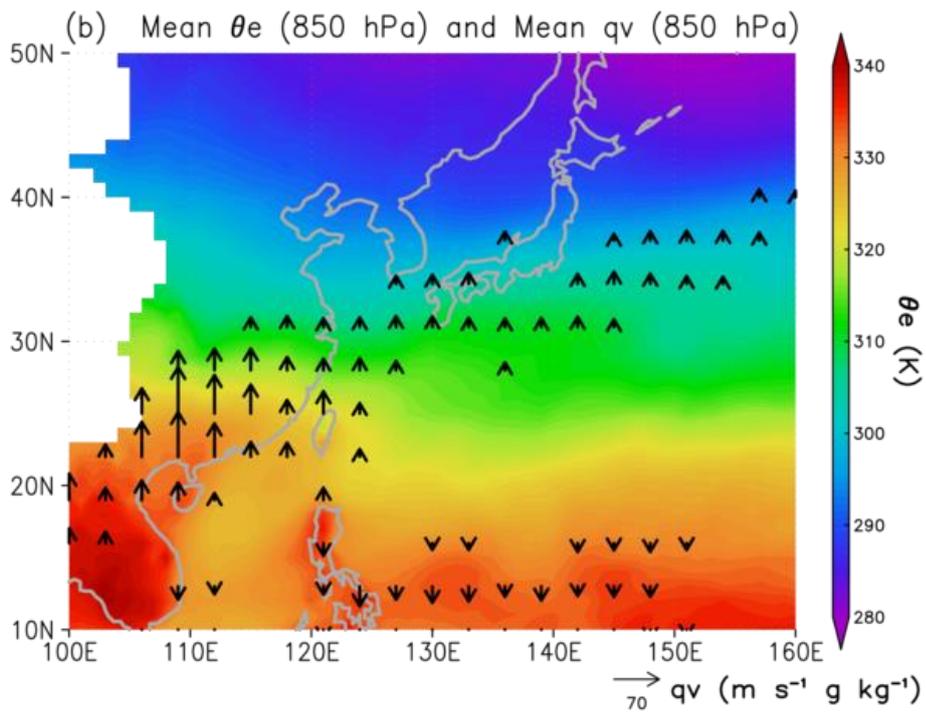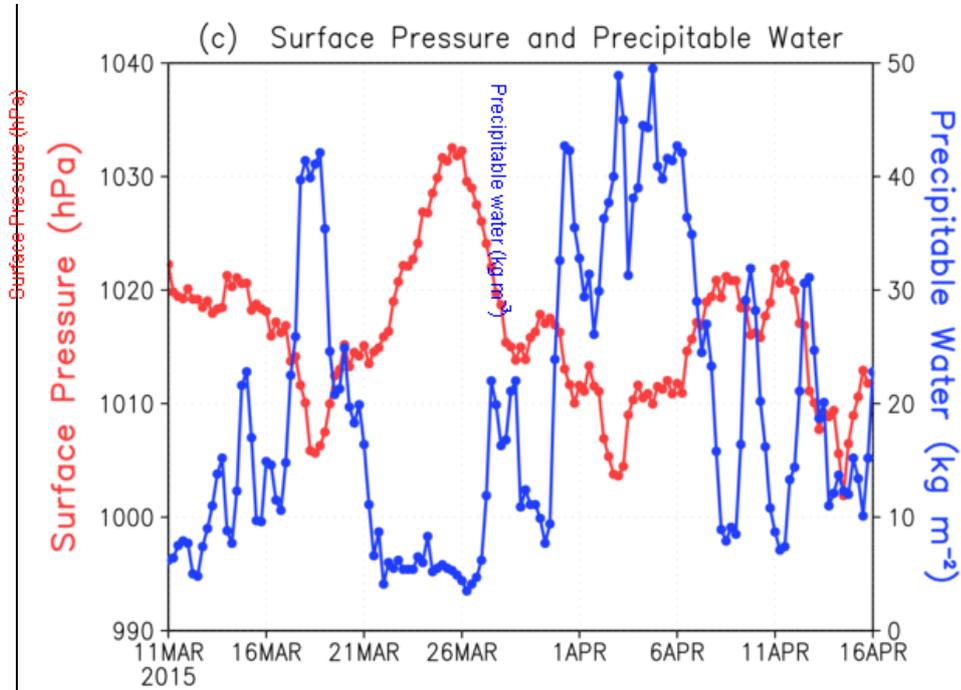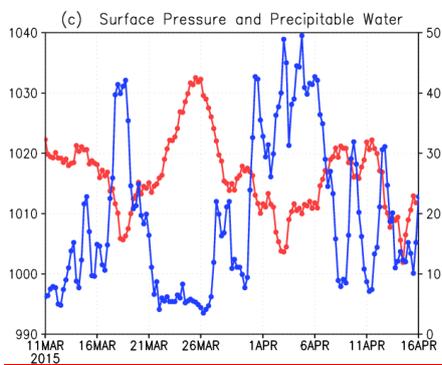
Figure 3. Meteorological fields in East Asia during the observation period (March 11–
–April 14, 2015) based on NCEP FNL data.   (a) Mean SLP (hPa, contours) and mean
horizontal wind velocity at the 850-hPa level (m s^{-1}).   Regions without data
correspond to those of high-altitude mountains.   (b) Mean θe (K) and total meridional

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711   moisture transport (qv values) at the 850-hPa level (m s^{-1} g kg^{-1}).   Only qv vectors

712   with magnitudes greater than 10 m s^{-1} g kg^{-1} were plotted.   (c) Temporal variations in

713   the surface pressure (hPa, red line and markers with left axis) and precipitable water

714   (kg m^{-2}, blue line and markers with right axis) at the Fukue observation site (32.75°N,

715   128.68°E).

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(a)

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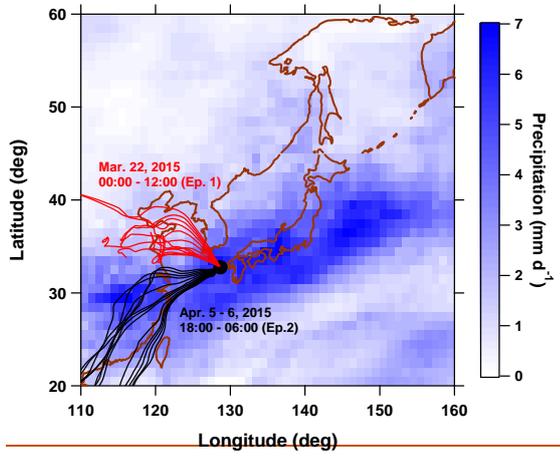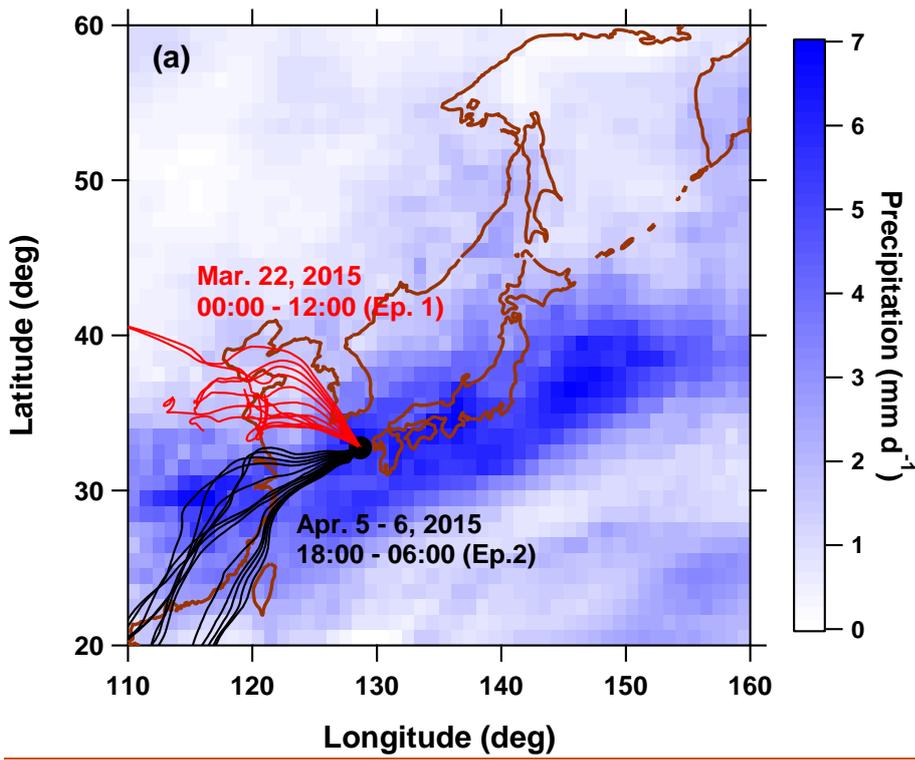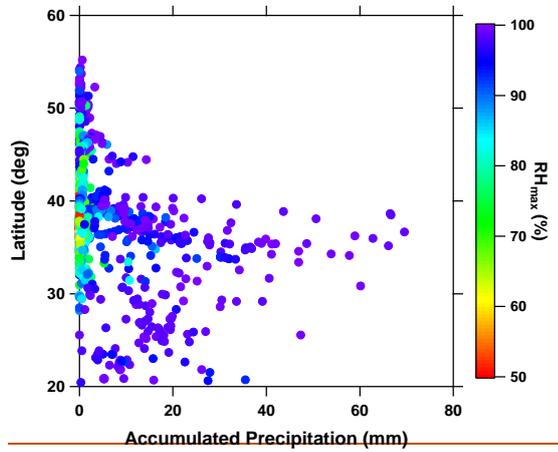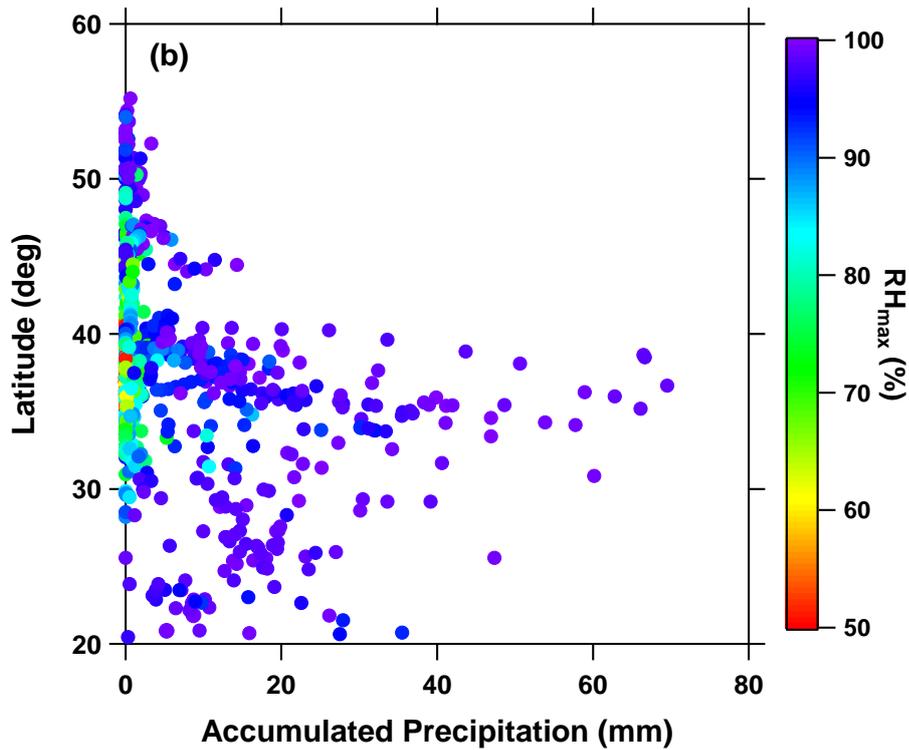
Figure 4. (a) Mean precipitation derived from GPCP during the observation period (March 11—April 14, 2015). 3Three-day backward trajectories for selected periods

are overlaid (rRed lines, 00:00—12:00LT March 22, 2015 (Ep. 1); Black black lines, 08:00LT, April 5—06:00LT, April 6, 2015 (Ep. 2)).   (b) The relationship between APT and Lat_{ORIG} (see text for details) colored by the maximum RH along the backward trajectories.
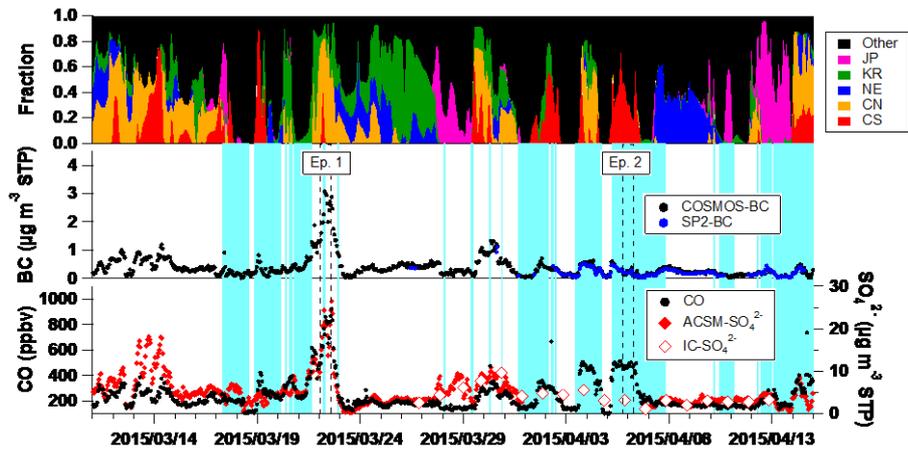
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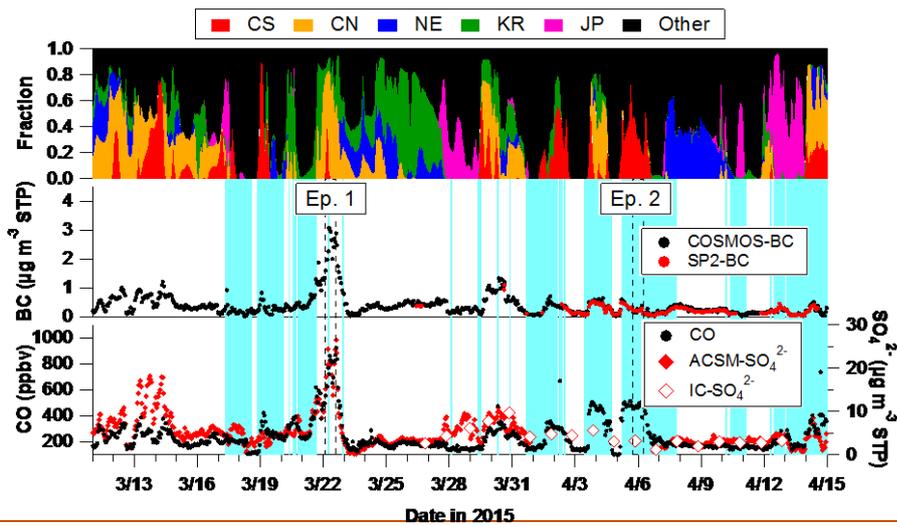
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731 **Figure 5.** Temporal variations in air mass origin and concentration of trace species.
732 (Top panel) Fractional residence time of air masses passed over selected area (Red,
733 Central South China; Orange, Central North China; Blue, Northeast China; Green,
734 Korea; Pink, Japan; Black, other regions such as Ocean). (Middle panel) mass
735 concentrations of BC measured using the COSMOS (black markers) and SP2 (blue red
736 markers). (Bottom panel) concentrations of CO (black markers) and SO_4^{2-} (red
737 circles and open diamond for ACSM and IC, respectively). The periods with the APT
738 > 3 mm are highlighted in light blue in the middle and bottom panels. The periods
739 denoted as Ep.1 and Ep.2 (see the text for details) were enclosed by dashed lines.
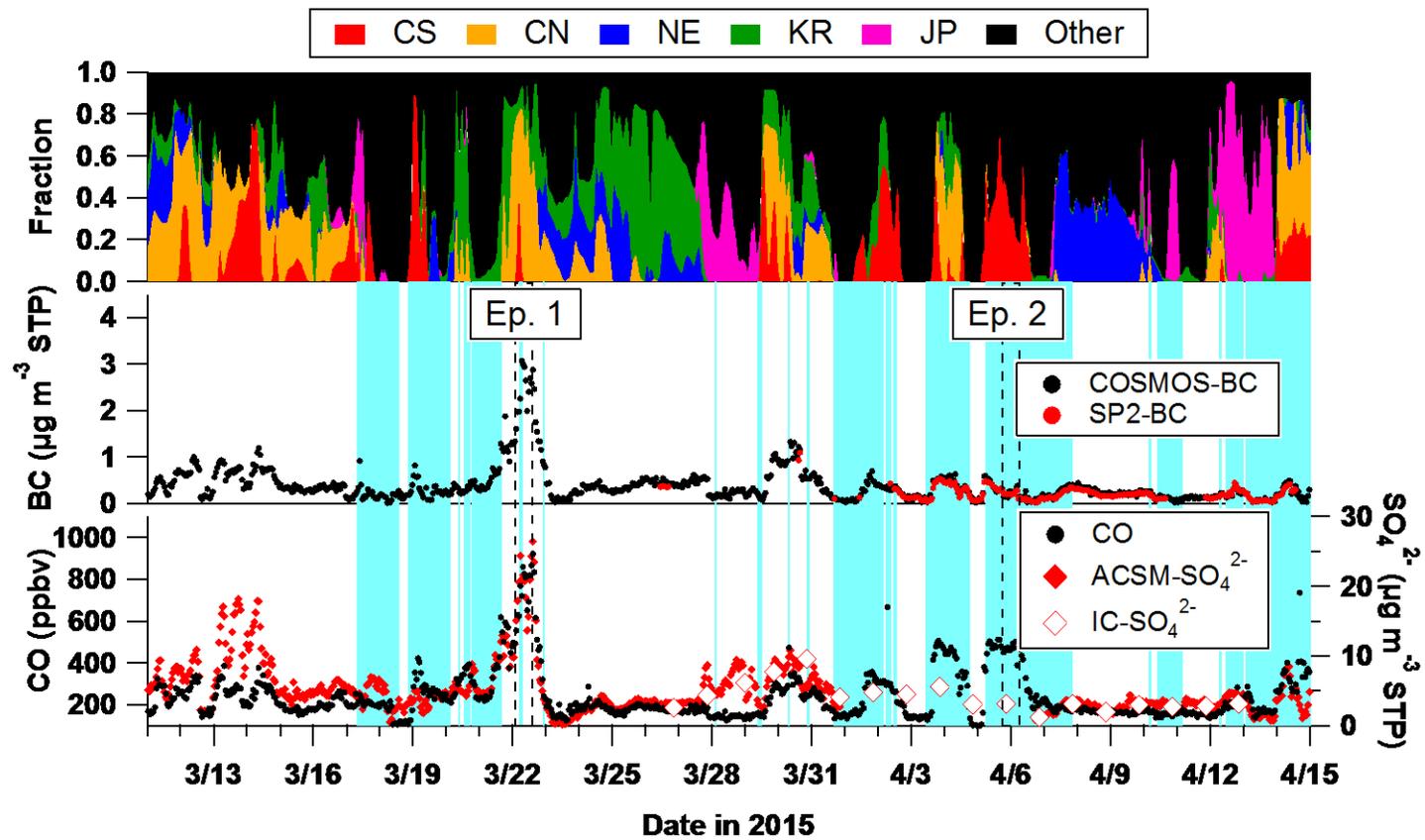
Figure 5. Temporal variations in air mass origin and concentration of trace species. (Top panel) Fractional residence time of air masses

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that passed over the 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 by using COSMOS (black markers) and SP2 (red markers).   (Bottom panel) Concentrations of CO (black markers) and SO_4^{2-} (red circles and open diamonds for ACSM and IC, respectively).   The periods with the APT >3 mm are highlighted in light blue in the middle and bottom panels.   The periods denoted as Ep. 1 and Ep. 2 (see the text for details) were enclosed by dashed lines.

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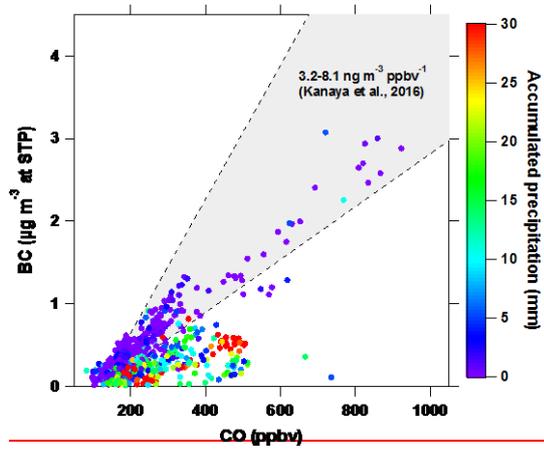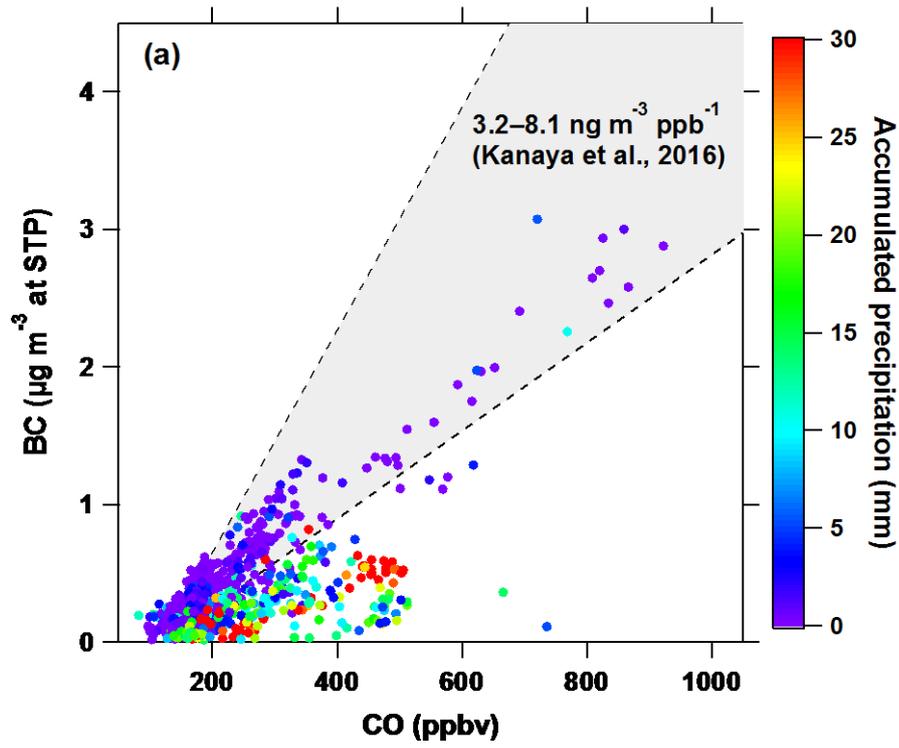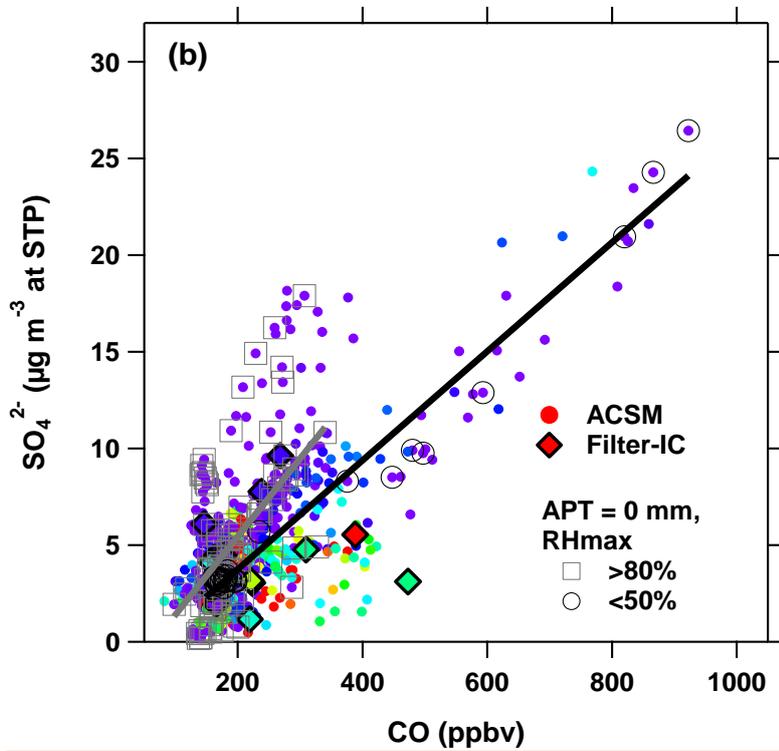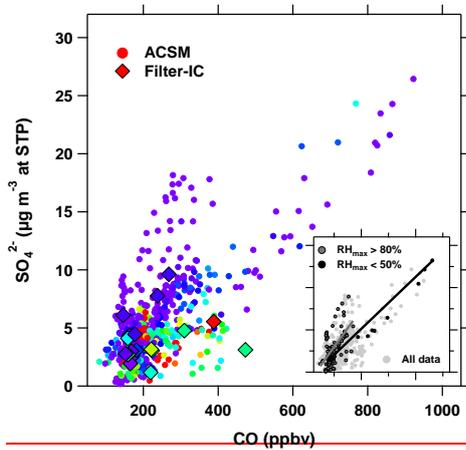
Figure 6. Correlation between aerosol mass concentrations and the CO mixing ratio colored according to the APT.   (a) BC measured by COSMOS and (b) SO_4^{2-} measured by ACSM and IC (circles and diamonds markers, respectively).   ACSM--SO_4^{2-}/CO

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correlations for the zero-APT air masses (no precipitation during transport) with symbols indicating if the RH$_{max}$ ~~greater than~~above 80% (~~dark~~ shaded ~~crosse~~open ~~squress~~markers) or ~~less than~~below 50% (black open circles~~crosses~~) ~~are in the subset of 6b~~overlaid.

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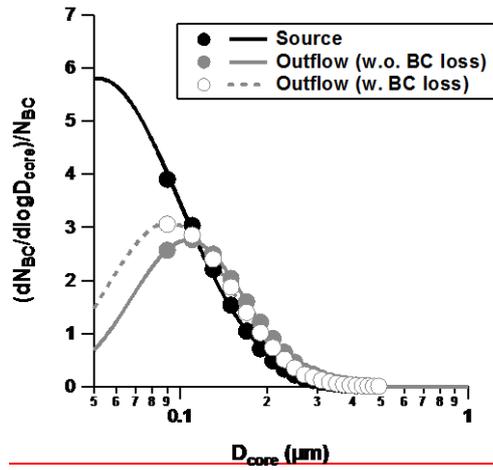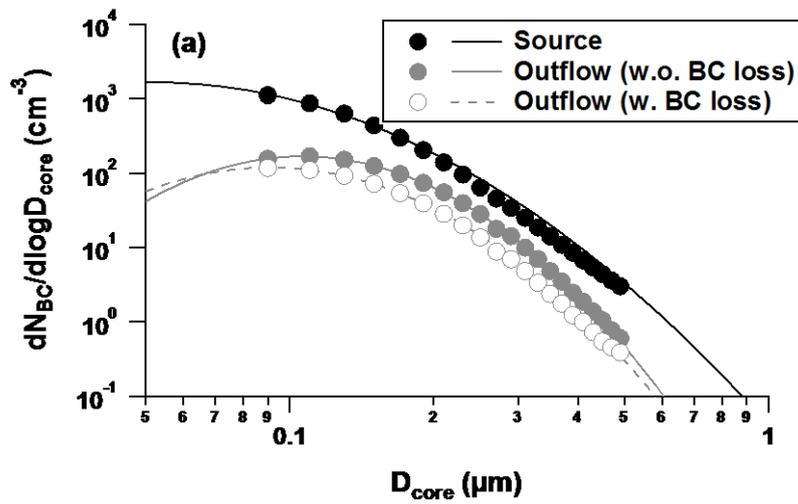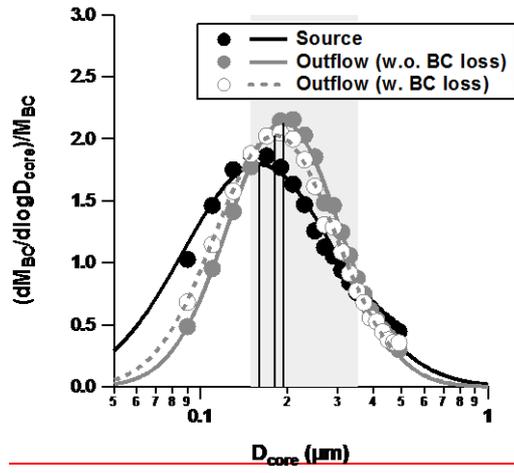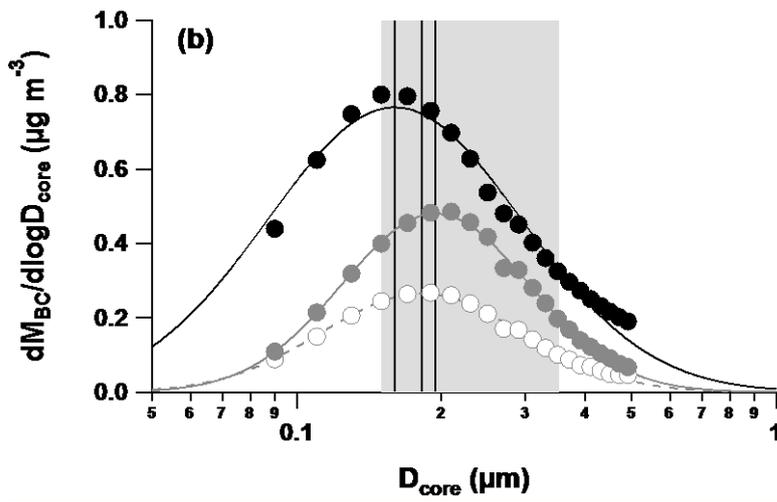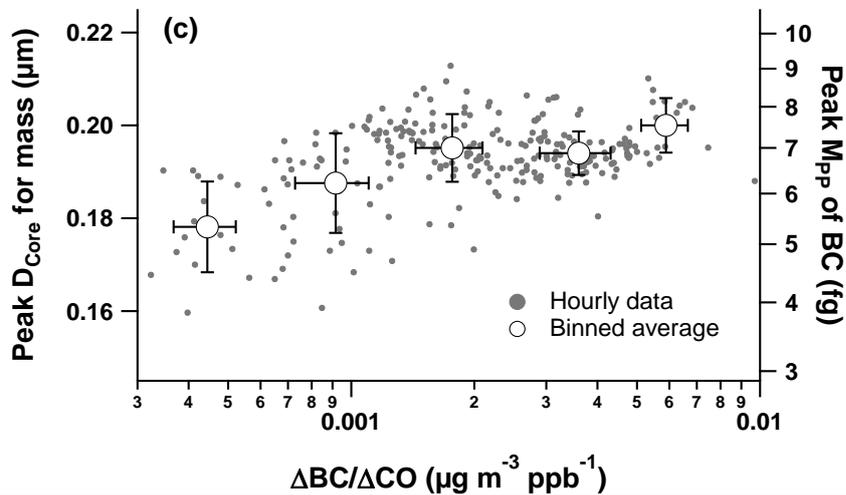
Figure 7. The (a) number and (b) mass size distributions of BC measured at Yokosuka (black markers) and at Fukue Island (gray markers). (c) The evolution of the peak D_{core} as a function of the degree of removal of BC (i.e., ΔBC/ΔCO ratios). ~~All the size distributions are normalized by the number or mass concentrations of BC integrated for the diameter range of 0.08-0.5 μm.~~ The size distributions at Fukue Island include the data for the outflow air masses with (open markers) and without (closed markers) BC loss. Lines are the lognormal fitting results. The shaded band in 6(b) corresponds to the size range analyzed to estimate D_s/D_{core} ratios. Vertical lines in 6(b) represent the peak diameter of the lognormal fit for each of three mass size distributions. Note that the peak diameter of the ~~Log~~log-normal fit for the BC number size distributions at Yokosuka was estimated from the peak diameter of its mass size distribution (**Table 1**).
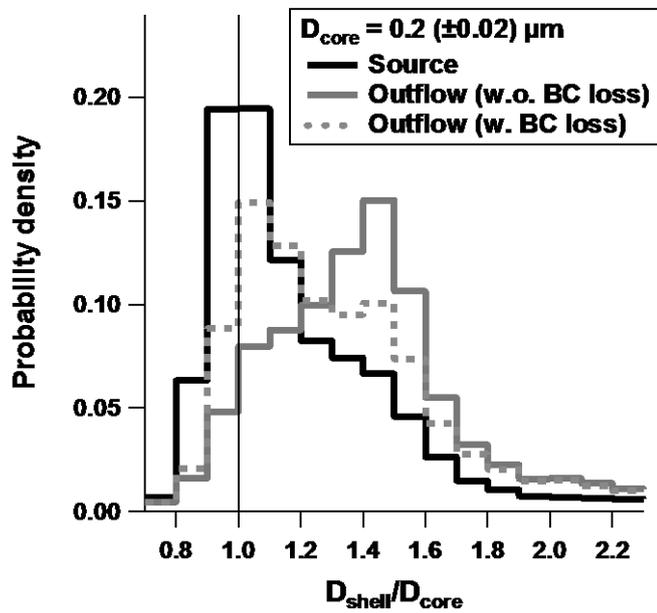
Figure 8. Probability density function of the estimated D_s/D_{core} ratios for BC-containing particles with the size 0.2 (±0.02) μm at Yokosuka (~~Black~~ black line) and in the air masses of continental outflow with (gray dashed line) and without (gray solid line) BC loss.

Tables

Table 1. Summaries of BC microphysical parameters measured at Yokosuka and Fukue Island

Site	Air mass type	Averaging time* (hrs)	$\Delta BC/\Delta CO$ (ng m^{-3} ppb^{-1})	APT (mm)	Log Normal Fit Parameters Avg. (1σ) MMD (μm)	σ_g	1-hr Median D_S/D_{core} for selected D_{core} Avg. (1σ) 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 the averaged statistics of the microphysical properties of BC-containing particles.