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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal features and origins of carbonaceous aerosols at Syowa Station, Antarctica" *by* Keiichiro Hara et al.

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Reply to comments from Referee #1: We would like to thank your helpful and complaisant comments to improve our manuscript. All comments are responded and addressed in the revised manuscript. Details are listed as follows. Corrected parts on comments from Ref. #1 were marked by red characters in the revised manuscript.

Comment from Referee: In particular there is a tendency toward paragraphs that focus on multiple ideas and that lack clear topic sentences or conclusions. An examination of the order in which ideas are presented may also be helpful.

Reply from Authors: We rewrote abstract and conclusions to be clear. Because one of reasons for lack clear topic sentences may be length of paragraph, we divided long

paragraph into a few paragraphs. Additionally, some results and discussion (e.g., AAE) moved to last section.

Comment from Referee: I suggest that the Authors ensure that their stated conclusions follow directly from the data they have presented.

Reply from Authors: We rewrote conclusions on basis of results from field measurements and model simulation.

Comment from Referee: In particular it would be advantageous for the authors to clearly describe not only the observations made, or what was done, but also what these observations mean or what can be learned from them.

Reply from Authors: We added and modified text on basis of your specific comments.

Comment from Referee: The main point of the paper is not entirely clear from the abstract or conclusions.

Reply from Authors: We arranged and rewrote sentences in abstract and conclusions on basis of comments from you and Referee #2.

Comment from Referee: P1 L10: Indicate the end date of the analysis (2016?), even if measurements are continuing to present.

Reply from Authors: BC measurements have been made at Syowa station. In this study, we used BC data in 2005-2016. This was added in abstract.

Comment from Referee: P1 L17: "internally mixed"

Reply from Authors: Because of modification of abstract, this words was removed.

Comment from Referee: P1 L19: "chemical transport model simulations"

Reply from Authors: We changed to "chemical transport model simulations" here and in the other locations.

Comment from Referee: P1 L19-20: What is the implication of direct transport? That

higher concentrations arrive at Syowa? That there is less removal? The implications of this transport path may deserve mentioning either here or later in the text

Reply from Authors: To avoid confusion, we changed "direct transport" to just “transport” or more specific explanations.

Comment from Referee: P1 L22: “BC” is used here, but not defined in the abstract

Reply from Authors: BC was defined in abstract of the revised manuscript, although location of BC in the text was changed by modification of manuscript.

Comment from Referee: P1 L25: I suggest re-wording this introductory sentence, in particular, “aerosols” cannot be components of “aerosol”

Reply from Authors: We changed into "Carbonaceous aerosols are major aerosols in the troposphere".

Comment from Referee: P1 L26: There are many other references and review articles that deserve mentioning here.

Reply from Authors: We added some references such as Andreae and Gelencsér, 2006; Bond et al., 2013.

Comment from Referee: P1 L26-27: Without acknowledging that soot particles (or carbonaceous particles) are a complex mixture of elemental carbon and organic species this discussion of terminology is somewhat confusing. In addition, defining BC according to only Novakov misses a large amount of literature on this topic. Several recent reviews provide clear recommendations for consistent terminology in this area.

Reply from Authors: In addition to definition by Novakov (1984), we added some references (e.g., Andreae and Gelencsér, 2006; Bond et al., 2013). Short explanation of definition about carbonaceous aerosols and BC were added into the text, as follows.

Apart from secondary organics associated with biogenic cycles, most of carbonaceous aerosols (e.g., soot) can be released from combustion of biomass and fuels. Soot

particles consist of refractory and insoluble matter (aka EC) and organics (e.g., Andreae and Gelencsér, 2006). As defined by Novakov (1984), BC comprises particulate graphitic particles. Recently, BC is defined by the following physical properties; (1) strong light absorption, (2) refractory, (3) insoluble, (4) aggregates of small carbon spherules (e.g., Bond et al., 2013).

Comment from Referee: P2 L3-5: Are there references for this conclusion? Or do the Authors find this statement attributable to their own observations? This should be clarified.

Reply from Authors: This is consideration by researchers (including us) who observed BC in the Antarctica. We changed this sentence to " It has been considered that BC must be supplied from outside of Antarctica, i.e. long-range transport, to maintain the background BC level and that it has seasonal features in the Antarctic atmosphere because of the low BC source strength in the Antarctic region. "

Comment from Referee: P2 L15: This wording is somewhat confusing. Do the Authors mean that ice core records of BC are correlated with historical variations in grassland and biofuels BC emissions?

Reply from Authors: To avoid confusion, this sentence changed to " In fact, BC records for the past 150 years in the Antarctic ice cores (WAIS core in Western Antarctica and Low dome core in Eastern Antarctica) showed influences by El Niño-Southern Oscillation (ENSO) and BC emissions from biomass burning and human activity in the source areas (Bisiaux et al., 2012)."

Comment from Referee: P2 L23-25: The logic here is a bit confusing, presumably anthropogenic BC emissions would have to originate not only from the surface but also from similar geographical regions as biomass burning emissions for this logic to hold. The claim in the previous sentence (i.e., that anthropogenic metals arrive in Antarctica), seems like a stronger suggestion that anthropogenic BC might be important.

Reply from Authors: We changed the explanation to " In contrast to the Arctic atmosphere, earlier works concluded that anthropogenic effects were only slight and negligible for aerosols in the Antarctic atmosphere (e.g., Weller et al., 2011, 2013), although some anthropogenic metals such as Pb have been found in snow and ice cores in the Antarctic region (Planchon et al., 2002; Vallelonga et al., 2002). Considering that biomass burning occurs on the ground in forests and grasslands, anthropogenic BC (derived mainly from fossil fuel combustion) can be transported to Antarctica."

Comment from Referee: P2 L26: Rather than simply stating that contributions of anthropogenic BC “must be assessed” it would be helpful for the Authors to delineate exactly why these sources need to be assessed. This is reasonably well done in the previous sentence.

Reply from Authors: We changed to "must be assessed", here.

Comment from Referee: P2 L38-41: We don’t expect BC to have the same sources so we can’t apply knowledge from mineral dust to BC. This could be stated much more clearly, potentially at the beginning of the paragraph, if this is the main idea being communicated here.

Reply from Authors: We changed the explanation in the paragraph as follows.

To elucidate BC transport from the low latitudes and mid-latitudes to the Antarctic region, we must ascertain the potential source area (PSA) and transport pathway. Actually, BC cannot be vaporized in ambient conditions. Therefore, BC must be transported from the origins (i.e. combustion processes) to the Antarctica. However, chemical analyses such as isotope ratio investigations are difficult to apply for identification of BC origins because the major BC component is graphite. Hara et al. (2010) described BC transport from South America and southern Africa to Syowa Station, Antarctica. Similarly to BC, mineral particles are transported from their origins to Antarctica, except for local emissions originating within the Antarctic Circle. For identification of the origins of mineral particles, earlier studies have been conducted to analyze and assess

PSA of mineral particles based on Nd/Sr isotope ratios (Smith et al., 2003; Delmonte et al., 2004, 2008; Bory et al., 2010; Valleloga et al., 2010; Aarons et al., 2016), Pb isotope ratios (De Deckker et al., 2010; Gilli et al., 2016), rare earth element patterns (Gabrielli et al., 2010; Valleloga et al., 2010; Wegner et al., 2012, Aarons et al., 2016), and trajectory/models (Perreira et al., 2004; Li et al., 2008; Albani et al., 2010; Gasso et al., 2010; Krinner et al., 2010; Neff and Bertler, 2015). From the aspect of mineral particles transported into Antarctica, South America (mostly Patagonia) has been identified as the most dominant PSA, whereas Australia and Africa respectively show minor and unimportant PSAs (e.g., Neff and Bertler, 2015). Although one must consider the following differences between BC and minerals, (1) geographical locations of PSA, (2) seasonality of source strength, and (3) size of aerosol particles containing BC and minerals, BC can be transported by outflow from the continents in the mid-latitudes to Antarctica. Here, we combine BC measurements with backward trajectory and chemical transport model simulation. This study was conducted to elucidate BC origins and PSA and to characterize BC concentrations and their seasonal features at Syowa Station, Antarctica located in the Indian Ocean sector.

Comment from Referee: P2 L41: What is meant by show difficulty?

Reply from Authors: Because of modification of text, this was removed from the text.

Comment from Referee: P2 L42: “Here, we combine...”

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P3 L1: “chemical transport model simulations”

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P3 L11: 2005 – 2016

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P3 L25-26: While a full description of the correction is well

placed in the supplement, some estimation of the uncertainty introduced into the measurement from this correction would be useful, along with an estimate of the total uncertainty.

Reply from Authors: The following explanation was added into the end of this paragraph.

Uncertainty of the measured EBC concentrations relates to (1) stability of the optical signal, (2) flow rate control, (3) spot area, and (4) scattering and shadowing effects. The detection limit value corresponds to uncertainty resulting from processes of (1)–(3). Uncertainty by the process (4) depends on the aerosol number concentrations and optical properties (single scattering albedo). The EBC concentrations corrected using Weingartner's method were mostly lower by 0.5–2% compared to the uncorrected EBC concentrations in this study (Fig. S1). Less difference between the corrected and uncorrected EBC concentrations might derive from higher single scattering albedo and replacement of the filter spot before optical attenuation reaching to 10% in most cases in our measurement conditions at Syowa.

Comment from Referee: P3 L30: "BC mass"

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P3 L34: How frequently was the DL determined? What is the detection limit at a minute time resolution?

Reply from Authors: DL was determined several times during our measurements. In this study, we did not DL at one-minute resolution, because EBC concentrations were too low to be measured with time resolution of one minute unlike urban areas.

Comment from Referee: P4 L24: Since biomass burning is such a large source of Antarctic BC, was any comparison between biomass burning emission inventories done? Do the Authors expect significant differences in their results with different inventories?

Reply from Authors: Regional biomass burning emission and its seasonal trend in MACC are similar to those in other inventory (e.g., GFED), although there are slight difference in regional distribution of biomass burning in each inventory. We added this into the text in the revised manuscript.

Comment from Referee: P5 L7: What is meant by "frequent"? The mode value?

Reply from Authors: Modal value is correct. We change from "frequent" to "Modal".

Comment from Referee: P5 L10: Showing a plot of the frequency distribution of eBC (e.g., in the supplement) is more illustrative than reporting the log-normal distribution and fitting parameters in the text.

Reply from Authors: The histogram of EBC concentrations with regression line was added in Figure of Supplementary (Fig. S2).

Comment from Referee: P5 L13: By "not clear" do the Authors mean that there was not statistically significant trend? Can this be shown, for example in the supplement?

Reply from Authors: We added results and figure of trend analysis in supplementary. On basis of the results, we added the following explanation in the revised manuscript.

From trend analysis (Supplementary and Fig. S3), a very slight decreasing trend (-0.036 ng m-3 yr-1, $p = 0.0145$) was observed in our measurements for 2005–2016. However, an increasing trend (0.105 ng m-3 yr-1, $P < 0.001$) was obtained in 2010–2016. These trend values included temporal trends, as explained below. Therefore, we concluded only slightly whether these trends were long-term EBC trends or not. More continuous EBC measurements must be taken at Syowa Station to analyze long-term trends.

Comment from Referee: P5 L17-19: Is Ferraz at higher altitude or is it just closer to South America? Why is long range transport more likely at Ferraz than Syowa?

Reply from Authors: Ferraz is located at northern part of the Antarctic Peninsula closer

to the South America. In addition, air masses at Ferraz were transported frequently from South America (Pereira et al., 2004), so that the long-range transport from South America might engender higher EBC concentrations at Ferraz than those in the other Antarctic coasts. This explanation was added in the text.

Comment from Referee: P5 L19-20: Are all of these stations using the exact same measurement and definition of BC (or EBC)? Are the same corrections used? This should be discussed, as the same issues have created challenges for comparison of BC measurement in the Arctic (e.g., see Sharma et al., 2017).

Reply from Authors: We corrected EBC concentrations in this study. However, EBC (or BC) concentrations measured at other Antarctic stations were not corrected. Also, difference between the corrected and uncorrected EBC concentrations at Syowa Station was discussed in the revise manuscript, as follows.

In earlier works, EBC concentrations were uncorrected, unlike this study. The EBC concentrations corrected using Weingartner's method decreased mostly by 0.5–2% compared to the uncorrected EBC concentrations in this study (Fig. S2). The lesser difference between the corrected and uncorrected EBC concentrations might result from (1) higher single scattering albedo and (2) replacement of filter spot before optical attenuation reaching to 10% in most cases in our measurement conditions at Syowa. Therefore, we can compare EBC concentrations in this study to the uncorrected EBC concentrations measured at other Antarctic stations in previous works.

Furthermore, some discussion about comparison of BC measurement in the Arctic (e.g., see Sharma et al., 2017) was added in the text, as follows.

In addition to filter-based EBC measurements, a single particle soot photometer (SP2) has been used for the measurement of refractory BC (rBC) (e.g., Bond et al., 2013; Sharma et al., 2017). According to Sharma et al. (2017), high correlation with R2 = 0.8–0.9 and slopes = 1.2–1.6 was observed between rBC and EBC in aerosols in the Arctic, where aerosol concentrations and anthropogenic effects were greater and

stronger than those in Antarctica. Considering different conditions of aerosol chemistry and optical properties between those in Antarctica and the Arctic, correlation in the Antarctica is expected to be different from that in the Arctic. No report of the relevant literature has described SP2 used to measure rBC year-round in the Antarctic region. Because of higher single-scattering albedo and lower aerosol concentrations in the Antarctica, differences between rBC and EBC might not be greater than in the Arctic.

Comment from Referee: P5 21-23: Why might there be more human activity at these two stations? More explanation is useful here.

Reply from Authors: High BC concentrations at Maitri and Larsemann Hills did not result from not more human activity but insufficient screening of local contaminated data. This was added in the text.

Comment from Referee: P5 23-24: What is meant here by "EBC concentrations at the Antarctic coasts in the Indian Ocean sector."? do the Authors assert that they are measuring background EBC concentrations not impacted by local human activity? If so, this could be explicitly stated

Reply from Authors: We mean "EBC concentrations observed at Syowa corresponded to background EBC concentrations at the Antarctic coasts in the Indian Ocean sector." The sentence was modified.

Comment from Referee: P5 L29-30: This sentence is somewhat repetitive compared to previous lines.

Reply from Authors: This repetition was removed from the text.

Comment from Referee: P5 L30: Use of "this," rather than an explicit statement or a paraphrase of previously presented ideas, makes it difficult to understand what the authors are referring to. Reply from Authors: Instead of "this", we used specific statement in the revised manuscript.

Comment from Referee: P5 L36-37: This sentence contains problematic wording; it

sounds like biomass burning comes from the Antarctic troposphere

Reply from Authors: This sentence changed to “biomass burning in the mid- and low latitudes has been regarded as having dominant origins of EBC measured in the Antarctic troposphere”.

Comment from Referee: P5 L40-42: I believe that the Authors are saying that the relative importance of biomass burning will depend on where a particular station is located in Antarctica. This is reasonable but could be made more clear.

Reply from Authors: This sentence changed to “the contribution of biomass burning from each PSA likely depends on where the respective coastal stations are located (e.g., sectors of Atlantic, Indian, and Pacific Oceans).”

Comment from Referee: P6 L6-7: Mixing month range (here and elsewhere) and season name is a bit confusing, especially since we are discussing the southern hemisphere

Reply from Authors: We showed month and season in the revised manuscript.

Comment from Referee: P6 L10-20: The discussion of AAE is quite confusing. If an AAE less than one indicates coated eBC, then the Authors need to be more clear about what they mean by assess the effects of organic aerosol and mineral particles. Do the Authors expect these species to be internally mixed or partially mixed with BC? Or do these species need to be externally mixed to produce an AAE greater than one? This can be explained clearly.

Reply from Authors: On basis of comments from you and referee #2, we corrected the discussion and statement as follows.

The CHASER model also indicates that internal mixing states of BC were dominated through the year (not shown, details published elsewhere). Therefore, lower AAE in April–October might result from the dominant presence of coated EBC particles (internal mixtures) at Syowa. The slight AAE increase corresponded to the spring maximum

of EBC concentrations. The following possibilities were considered for the slight AAE increase: (1) change of mixing states of BC and (2) contribution of other light-absorbing materials such as organic aerosols and minerals. The organic aerosols and minerals have high AAE, for instance, 3.5–7 for organics and typically 2–3 for minerals (e.g., Bond et al., 2013, and references therein). Although the internal mixing states of BC were dominant in CHASER model simulation, external mixtures of BC increased in spring EBC maximum (not shown, details published elsewhere). Considering AAE of external mixing of BC, increase of external mixing of BC can engender an AAE increase. Additionally, spring EBC maximum at the Antarctic coasts was associated closely with biomass burning. Organic aerosols with high AAE derived from biomass burning were expected to be transported simultaneously into Antarctica. Consequently, transport of organic aerosols might contribute to the slight AAE increase in September – October.

Comment from Referee: P6 L22-23: It is worth stating here which wavelengths correspond to the UV-IR and Vis-IR channels.

Reply from Authors: Wavelengths were added into the text.

Comment from Referee: P6 L26-28: This result could use more explanation and support from the literature.

Reply from Authors: We changed the explanation as follows.

High optical absorption by organic aerosols in the UV ranges engenders an increase of AAEUV-IR and the higher ratios (e.g., Bond et al., 2013). Therefore, the difference suggests that organic aerosols, rather than effects of mineral particles, contributed to optical absorption and AAE.

Comment from Referee: P6 L31: “spring eBC maximum”

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P6 L31-32: Since a lot of this discussion depends of knowl-

edge or source regions, the Authors may consider moving the discussion of source regions to before the discussion of optical properties and inferences about particle composition

Reply from Authors: Paragraph on AAE and optical properties were moved to last section.

Comment from Referee: P6 L33: While I agree entirely that the measurements of oxalate are useful to show the seasonal cycle that likely takes place for organic aerosol, it is unclear whether the inference can be made that this indicates more biomass burning organics or BrC. Is there any literature to show that oxalate correlated with brown carbon?

Reply from Authors: Yes. Previous work (Zhang et al. 2012) showed that high concentrations of oxalate and brown carbons were associated with secondary organic aerosol formation in a condensed phase. This work was referred in the revised manuscript. Also, short explanation was added to discussion.

Comment from Referee: P6 L34: It is somewhat difficult to understand how this conclusion follows from the discussion. The Authors need to better explain what it means physically for the AAE_Vis- IR to be negative when the AAE_UV-IR is zero.

Reply from Authors: We changed the explanation as follows.

Additionally, high concentrations of oxalate and brown carbons are associated with secondary organic aerosol formation in a condensed phase (e.g., Zhang et al., 2012). When optical absorption in UV regions was increased by organic aerosols (i.e. BrC), correlation between AAEUV-IR and AAEVis-IR can be shifted to larger AAEUV-IR region. This change might engender larger negative intercept values. Therefore, the larger negative intercepts in October–November might result from effects of organic aerosols derived from biomass burning.

Comment from Referee: P6 L37-38: Was MSA found to be a major aerosol constituent

in these filter measurements? If so, these data need to be shown along with the oxalate data.

Reply from Authors: MSA feature was added to Fig. S6 which showed the oxalate data.

Comment from Referee: P7 L1-2: This discussion seems somewhat speculative given that the MSA data is not shown, it is not clear that BrC or any other organic species that could absorb can be completely discounted here.

Reply from Authors: We modified the sentence into “Therefore, AAE in the summer (December–February) might be associated with EBC aging processes and with the presence and mixing of organic aerosols (e.g. CH3SO3H) derived from oceanic bioactivity.”

Comment from Referee: P7 L6-8: This statement is based on very little direct evidence (i.e., no direct observations of MSA or BrC organic aerosol). More evidence needs to be provided, or this statement needs to be adjusted to better reflect the data presented in this paper.

Reply from Authors: We added direct evidence of MSA feature in Supplementary. The explanation was changed into “The concentrations of EBC and organic aerosols derived from biomass burning increased in the spring maximum as described above, whereas the EBC concentrations decreased and the concentrations of organic aerosols such as CH3SO3- derived from oceanic bioactivity increased during summer.”

Comment from Referee: P7 L15-16: Are 5 day backward trajectories adequate to describe the origins from long range transport?

Reply from Authors: From aspect of identification of PSA, 5 days are not enough. Because uncertainty of trajectory analysis particularly in troposphere tends to be larger in longer analysis time. In this study, we calculated 5 day backward trajectory. However, the trajectory can provide important information on transport pathway, for example via

MBL and continental FT.

Comment from Referee: P7 L29-30: Doesn't this say that Syowa would be more sensitive to station sources in winter?

Reply from Authors: We did not think so. Although density of air masses from the Antarctic continent increased in winter, transport from ocean (mid-latitude) was major transport pathway.

Comment from Referee: P8 L3-5: Are flow patterns that demonstrate interactions with areas of open ocean either along the coast or in the MBL associated with times when the Authors suggest that MSA mixing with eBC was important?

Reply from Authors: In summer, transport from open water area (mid-latitude) showed high density as shown in Fig. 4. This air mass history might be associated with MSA concentrations at Syowa and mixing states of aerosol particles containing EBC. However, this explanation was not added in the text because discussion on AAE was move to last section, and lengthy explanation should be avoided.

Comment from Referee: P8 L7-13: The Authors may consider connecting these observations with those in the previous section more explicitly.

Reply from Authors: To explain more explicitly, we changed these sentence as follows.

As described above, EBC is expected to be supplied mostly from outside of Antarctica. Plausible transport pathways are transport via MBL and FT. We must know the EBC concentrations of each air mass origin (MBL, coastal BL, continental BL, continental FT, coastal FT, and MFT) to elucidate EBC transport pathway to the Antarctica. Figure 6 depicts seasonal features of air mass origins in each month and monthly mean and median EBC concentrations at Syowa during our measurements (2005–2016). The dominant air mass origins were MBL, coastal BL, coastal FT, and continental FT. The most dominant air mass origins were MBL and coastal BL in November–February. In addition to MBL and coastal BL, the contributions of transport from coastal FT and

continental FT increased in February/March – October at Syowa, although year-to-year differences were found in the seasonal variations of air mass origins. Particularly, the contribution of transport from continental FT in March–October was higher than that in other years. This change corresponded to lower EBC concentrations in July of 2010–2012, as described above. Therefore, the increasing trend of EBC concentrations in July of 2010–2016 might not be a long-term trend but a temporal trend resulting from year-to-year variations of air mass history.

Comment from Referee: P8 L15: The Authors need to clarify what is meant by "compared" here. How is the eBC apportioned to these trajectories? Is the dominant trajectory origin assumed to supply all the eBC in each hour? Or is the eBC apportioned based on the percentage of each trajectory category for each hour? This should be explained a bit more clearly in the methods section (and referenced here).

Reply from Authors: The sentences were modified as follows.

For comparison between EBC concentrations and air mass origins, hourly mean EBC concentrations were estimated. Then, hourly EBC data were classified into each air mass origin.

Comment from Referee: P8 L21: That these stations all use different measurement techniques needs to be discussed much earlier in the text, when the Authors make general comparisons between BC concentrations measured at different stations.

Reply from Authors: This explanation was moved to discussion on Fig. 3 (seasonal features of EBC conc.). Some explanation about EBC data at other stations were also added in the text.

Comment from Referee: P8 L29-30: The Author's meaning is not clear in this sentence

Reply from Authors: This sentence was changed as follows.

Seasonal features of EBC concentrations in MBL (Fig. 7a) might correspond to those in the MBL in the Southern Ocean in Atlantic and Indian sectors, considering that air

masses were transported dominantly via MBL from the mid-latitudes by the cyclone approach.

Comment from Referee: P8 L35: “southern Atlantic MBL”

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P8 L40: Has MFT been defined?

Reply from Authors: Not yet. I defined MFT in the revised manuscript.

Comment from Referee: P9 L1: The wording “difference was similar to differences” is very confusing, and obscures the meaning of this sentence.

Reply from Authors: To avoid confusion, this sentence was changed as follows.

Seasonal variations of CO concentrations and fire counts (Gros et al., 1999; Edwards et al., 2006a, 2006b) were similar to the seasonal features of EBC concentrations at Syowa, as described above.

Comment from Referee: P9 L5: Since these trajectories ended in the Southern Ocean, does that suggest the trajectory duration is not sufficient to reach source regions or that marine sources (e.g., ships) could also be important?

Reply from Authors: Ship operation in MBL can release EBC to the atmosphere. Density of marine traffic (i.e. ship operation) in the Southern Ocean and near the Antarctic coasts was too low to engender increase of EBC concentrations in air mass from MBL, although ship emission can have an influence locally on EBC concentrations, for examples ship-borne tourism in the Antarctic Peninsula during summer. These explanations were added into the revised manuscript.

Comment from Referee: P9 L8: What is meant by “seasonal features of eBC concentrations were maximum”?

Reply from Authors: “seasonal features of EBC concentrations had maximum in

October–” is correct. This was modified in the revised manuscript.

Comment from Referee: P9 L10: “in” or “from” the continental FT?

Reply from Authors: “from” is correct. This was modified in the revised manuscript.

Comment from Referee: P9 L12: the Antarctic plateau?

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P9 L14-16: Transport patterns inferred from CO2 gradients may not be reflective of BC transport pathways, comparing to a shorter lived species may be important to draw these conclusions

Reply from Authors: This explanation was removed from the manuscript.

Comment from Referee: P9 L31: “Therefore, we discuss. . .”

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P10 L23-24: I suggest the Authors consider re-wording this sentence so that it is not in the form of a question, but rather outlines what was done and why, or the hypothesis.

Reply from Authors: This sentence was changed as follows.

We must ascertain the transport pathway from Australia to the Syowa to understand the high BB-mBC concentrations in Australia.

Comment from Referee: P10 L33-35: What is learned from these correlations? Would that not suggest that ’other combustion’ is more related to anthropogenic activities than biomass burning?

Reply from Authors: The following sentence was added here.

The good correlation found between FFC-mBC and OC-mBC implies strongly that seasonal features of FFC-mBC and OC-mBC might reflect variations of transport strength

from each PSA to Syowa.

Comment from Referee: P11 L 5-6: What is being suggested here? Further measurements beyond eBC? This could be explicitly stated.

Reply from Authors: We modified the sentence as follows.

Therefore, continual EBC measurements must be conducted at the Antarctic coasts to monitor the atmospheric substances (e.g. EBC) originating from combustion in the Southern Hemisphere.

Comment from Referee: P11 L9-10: "The following possibilities are contributing factors:. . ."

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P11 L12-13: How does this large precipitation amount impact outflow of BC from source regions? Is part of the seasonal cycle in eBC driven by precipitation removal near the source region or along the transport path? what does the model suggest?

Reply from Authors: We added the following explanation.

In the CHASER model, fresh BC immediately after release from BB is assumed as hydrophobic, so that few fresh BC might be removed by precipitation near source areas. Aging processes during transport can engender gradual change into internal mixtures of BC with a hydrophilic surface. Then, BC can be scavenged through wet deposition during transport. Considering clear seasonal features of CO with longer residence time than BC (Edwards et al., 2006a, 2006b; van den Werf et al., 2006), seasonal variation of BC emissions might have a greater contribution to seasonal features of mBC and EBC at Syowa than wet deposition of BC during transport.

Comment from Referee: P11 L25: 2005-2016

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P11 L30: Are these species internally mixed with eBC?

Reply from Authors: Aethalometer cannot provide ambient mixing states of aerosols. Thus, we need to compare to earlier work (e.g., Ueda et al., 2018). However, this explanation was removed from the text in conclusions because of modification of the sentences in conclusions.

Comment from Referee: P11 L31-32: In spring? Or at any time?

Reply from Authors: We mean winter – spring. This was added in the text of conclusion.

Comment from Referee: P11 L32: "eBC was emitted"

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P11 L34: "chemical transport model simulations"

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P11 L34-35: These conclusions are somewhat vague. What were the magnitude of these contributions? Is what seasons?

Reply from Authors: To avoid length explanation, we changed as follows.

The CHASER model simulation showed that the most important origins and PSA of EBC at Syowa Station were biomass burning in South America and southern Africa.

Comment from Referee: P11 L37-38: This statement should be the other way around: the eBC minimum might be attributable to general transport patterns (higher contributions of the free troposphere and coastal boundary layer)

Reply from Authors: This was changed based on your suggestion.

Comment from Referee: P12 L6-7: Given wavelengths that these channels correspond to

Reply from Authors: Wavelengths were added in the revised manuscript.

Comment from Referee: P18 Figure 1: Labels directly on this map might be more useful than numbers, given that there is ample space and the number of stations is relatively large.

Reply from Authors: Labels of each station were added in Fig. 1.

Comment from Referee: P22 Figure 7: Can a time series of eBC, or AAE be added to the right-hand axis of this plot? Would this shed any light on the influence of marine sources of aerosol absorption characteristics?

Reply from Authors: We added seasonal features of EBC concentrations in this figure (Fig. 6 in the revised manuscript).

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-1190/acp-2018-1190-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1190, 2019.