

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 #2: We would like to thank your helpful and complimentary 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. #2 were marked by blue characters in the revised manuscript.

Comment from Referee: I would suggest authors provide a clear explanation of data screening procedure (from measured data to useful data for BC).

Reply from Authors: Specific procedures for data screening were added into Section of 2 (Measurements, modelling and analysis).

Comment from Referee: I found abstract and conclusion a little confusing, vaguely

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written, especially with the division of potential source areas (PSA), types of aerosols, and the CHASER model part is not mentioned explicitly.

Reply from Authors: Based on comment from you and referee #1, sentences in abstract were modified to simple and clear explanation.

Comment from Referee: While looking it at it, I got the first impression as high variability in daily EBC values between the years 2005 to 2009, which diminishes after that. It might be possible that more local influences during those years than in other years. I would suggest discussing if any change of location of instrument occurred. Is aethalometer calibrated? Or anything which authors wish to comment on that? Is the sampling line is heated or any changes?

Reply from Authors: At a glance, high EBC concentrations were often observed in winter–spring during 2005–2009. Measurement conditions (e.g., tube length and room temperature) and analytical procedures were the same from 2005–2016. Therefore, this change might result from variations of frequency or strength of EBC transport events rather than measurement and analytical reasons. These explanations were added to discussion in Fig. 3.

Comment from Referee: Air mass history and classification are explained very nicely, but it stands suddenly out of context. Authors have dedicated Figure 5, Figure 6 and Figure 7 especially for explaining this and they only connect it with EBC in Figure 8. I would suggest the authors make a good connection in meteorology and aerosol in these figures and sections. Also, classifications and their sub-classifications are not explained and connected for better readability.

Reply from Authors: Based on comment from you and referee #1, some statements were added in these explanations.

Comment from Referee: Based on the CHASER model, EBC origins were classified into three sections (biomass, fossil fuel, and other combustion). Later it is stated that

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"other combustion" is broadly biomass burning, which makes it as two classifications, which could be inferred from angstrom absorption exponents (AAE) of Figure 3. Is it possible to add information about the mixed state (Internal or external) or aging (fresh or aged) of BC, used in the CHASER model, which showed promising results in monthly values and seasonality of BC in Figure 9?

Reply from Authors: CHASER model indicated that internal mixing state of EBC was dominant. The short explanation was added in the text of the revised manuscript. Details of model results will be published in another paper (now on preparation for publication).

Comment from Referee: At many places, authors replace "Syowa" with "coastal Antarctica" or "Antarctica coast". It might be useful to replace "Antarctica" in the title with "coastal Antarctica". But I would leave this totally on author's choice.

Reply from Authors: At some places, "Syowa" were changed "coastal Antarctica". In title, we changed to "coastal Antarctica".

Comment from Referee: Page 1: Line 10: We measured equivalent black carbon (BC) [I think you measured BC and corrected it to make it EBC)

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

Comment from Referee: Line 10: Feb 2005 to Feb 2016 [adding the end month of measurement]

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

Comment from Referee: Line 25: First statement needs a reference

Reply from Authors: A few references (e.g., Gelencsér, 2004; Gilardoni, and Fuzzi, 2017) were added in the text.

Comment from Referee: Line 34: Antarctic regions; BC concentrations

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

Comment from Referee: Line 34: Page 2: The Antarctic is referred to here as one of the remote regions. I think calling it as “Antarctic region” throughout the manuscript is appropriate than “Antarctic regions”? I also think it is worth mentioning and dividing Antarctica as Eastern and Western Antarctica in the introduction as this section talks about tourism and transport from South America and the African region

Reply from Authors: “Antarctic regions” was changed to “Antarctic region” in the text. We added short statement using Eastern and Western Antarctica in the text.

Comment from Referee: Line 14: please specify where in Antarctica?

Reply from Authors: We added specific locations in the text.

Comment from Referee: Page 3: Section 2 heading could be “Measurements, Modelling, and Analysis”

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

Comment from Referee: Line 5: “Research” is missing in JARE expansion

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

Comment from Referee: Line 6: It would be worthy to mention the altitude of sampling station at Syowa with latitude and longitude

Reply from Authors: In addition to latitude and longitude of Syowa Station, altitude (elevation) was added in the text.

Comment from Referee: Line 7 To Syowa, the icebreaker ship Shirase approaches every summer (mainly between end-December to – early February) for the transportation of fuel and materials for wintering operations and scientific activity

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

Comment from Referee: Line 21: I think data screening procedure needs more clarity

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as it is not in Hara et al., 2010. Hara et al., 2010 cites Hara et al., 2008 and I would suggest citing the right paper here.

Reply from Authors: As mentioned above, we added specific procedures for data screening in the text.

Comment from Referee: Line 23 to 34: what is the value of multiple scattering and loading parameter used in making BC to EBC? Would be a good idea to mention it explicitly here.

Reply from Authors: Specific parameters for BC correction by Weingartner's method were given in statements in Supplementary. To avoid confusion, we added statement of "correction factor for multiple scattering of light and shadowing effects in Weingartner's correction" in the Supplementary.

Comment from Referee: Line 28: Attenuation at 880 nm is used widely for BC retrievals, I would suggest making changes in the statement accordingly

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

Comment from Referee: Line 35: the statement "We use a multi-wavelength. . ." could be rephrased like "using the spectral (or multiwavelength) aerosol absorption values retrieved from aethalometer, we estimated AAE

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

Comment from Referee: Page 4: Line 10-14: CHASER could be expanded. There are other acronyms also need to be expanded. I think it would be helpful for readers who are not modelers.

Reply from Authors: Acronyms in explanation on CHASER model were defined in the text and added into the Acronym list in Appendix.

Comment from Referee: Line 39: "cooking" not "cocking"

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

Comment from Referee: Page 5: Section heading “Discussions”

Reply from Authors: Because “Discussion” is used usually in paper, we use “Discussion”.

Comment from Referee: Line13-14: Any seasonal long-term trend at Syowa? It would be worth seeing whether there is an increase or decrease in summer (like Neumayer) or spring

Reply from Authors: Thank you very much for nice comment. We checked long-term trend in each month, as shown in Fig. S4. Trends were not clear except July. The following explanation and discussion were added into the revised manuscript.

Although a decreasing trend of EBC concentrations in summer (November and December) was found at Neumayer (Weller et al., 2013), no seasonal long-term trend was clear at Syowa except for July (Fig. S4 in Supplementary Information). At a glance, EBC concentrations in July showed an increasing trend for 2011–2016 (0.325 ng m⁻³ yr⁻¹ in monthly median and 0.363 ng m⁻³ yr⁻¹ in monthly mean). However, we must consider the likelihood that EBC concentrations in winter (June–August) declined in 2010–2012 rather than following the increasing trend by EBC emissions at middle and low latitudes. Indeed, this variation in July might be related to changes of air mass origins (details are discussed in section 3.2).

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: Line 25-42: It is not much clear. I would suggest defining seasons and maintain uniformity in the discussion of seasonality and comparison with

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other stations. The possible sources in each season could be also be highlighted.

Reply from Authors: Specific months were added in the text, instead of definition of seasons.

Comment from Referee: Page 6: Line 21: BrC Brown Carbon (BrC)

Reply from Authors: BrC was defined in Introduction. Thus, we did not change it, here.

Comment from Referee: Line 29: larger negative values (<-0.4)

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

Comment from Referee: Line 24: What is a high correlation means here? R2 values are lesser for June-Aug, in comparison to other months

Reply from Authors: We added explanation of “Particularly, high correlation ($R^2 > 0.7$) was obtained in March, September-December.”

Comment from Referee: Page 7: Line 6: Slopes >1 but AAE was lesser in spring (Figure 3d), so how you suggest it is biomass burning aerosol of organic origin? Please clarify

Reply from Authors: The following explanation was added to the revised manuscript.

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 CH_3SO_3^- derived from oceanic bioactivity increased during summer.

Comment from Referee: Line 15: “First, we compare EBC data to the air mass history at Syowa” this line does not seem appropriate here

Reply from Authors: The statement of “First, we compare EBC data to the air mass history at Syowa” was removed from the text.

Comment from Referee: Line 17: for the 3rd classification, do you mean outflow from

the high-latitude Antarctic continent to coastal Antarctica? Please clarify

Reply from Authors: This explanation was changed to “(3) outflow from the high-latitude Antarctic continent to the coasts.”

Comment from Referee: Line30: It appears that the probability density of air mass arriving at Syowa shows an East-West spread from one month to another month, as compared to North-South spread. In that case, transport from inland Antarctica is more important than long-range transport from populated continents. Is it the case?

Reply from Authors: This explanation was changed to “This difference implies that the transport strength of the outflow from the Antarctic continent had remarkable seasonal change in addition to important contribution of the poleward flow patterns from the ocean.”

Comment from Referee: Line 41: could specify an approximate tropopause height.

Reply from Authors: We checked approximate tropopause height from previous work by Tomikawa et al. (2009). The sentence was modified in the revised manuscript, as follows.

Considering tropopause height (8–10 km) identified by O3 profiles in the Antarctica during the winter (Tomikawa et al., 2009), the air mass history implies that air masses near tropopause over the continent can flow to the boundary layer (BL) at the Antarctic coasts during winter.

Comment from Referee: Line 35-43: From Figure 6, it appears that Syowa is influenced by high-latitude inland Antarctica air mass during all months (which is relatively less in January). What is the final take from Figure 6?

Reply from Authors: Fig. 6 (Fig. 5 in the revised manuscript) indicates vertical motion of air mass in each latitude. Vertical mixing was varied largely depending on latitudes. This was already stated in the text. Some explanation was added in this section.

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Comment from Referee: Page 8: Line 3-5: Is this subclassification of the classification on page 7(Line 16-18)? I think it is 2 sub-classifications of the previous classification, but it is not clear in the text.

Reply from Authors: To make clear explanation, the statement was changed as follows.

With suggestion of vertical motion and geographical classification of air mass origins as described above, the following transport patterns and air mass origins at Syowa are finally classifiable in this study: (1) poleward flow from MBL, (2) poleward flow from LFT, (3) westward flow along the coastal line via BL, (4) westward flow along the coastal line from LFT, (5) outflow from the FT over the Antarctic continent, and (6) outflow from BL over the Antarctic continent.

Comment from Referee: Line 6: classification of air mass origin > 75S could be re-named as remote continental or Antarctic continental, as naming it continental confuses with polluted and populated continents.

Reply from Authors: This was changed based on your suggestion (we used “Antarctic-continental”). Because of suggestion by referee #1, this classification was moved to Section 2.2.

Comment from Referee: Line 8: statement is not clear

Reply from Authors: From comments from you and referee #1, we add explanation to classify air mass origins. This explanation was also added in Section 2.2.

Comment from Referee: Line 29-30: Is an increase in MBL air mass origin EBC, could be due to Ship emissions in the Antarctic Circle (for fishing or tourism)?

Reply from Authors: Ship operation can emit BC (or EBC) to the atmosphere. However, the contribution may be negligible due to lower density of marine traffic in the Southern Ocean and near the Antarctic coasts. We add the following discussion and explanation in the revised manuscript.

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Although high EBC concentrations were obtained in air masses from MBL, we must consider EBC origins in air masses from MBL. Additionally, 120-hr backward trajectory analysis was too short to reach to contributable PSA because it took longer than one week for transport from the coasts of South America and southern Africa to Syowa (Hara et al., 2010). Density of marine traffic (i.e. ship operation) in the Southern Ocean and near the Antarctic coasts was too low to engender an increase of EBC concentrations in air mass from MBL, although ship emissions can have an influence locally on EBC concentrations, for example ship-borne tourism in the Antarctic Peninsula during summer.

Comment from Referee: Page 9: Line 25-26: Filter biased problems and related uncertainty were not discussed while detailing EBC. I am glad that the authors bring it up here.

Reply from Authors: Discussion on filter biased problems and related uncertainty were added to sections of 2-1 (Page 3) and 3.1 (Page 6-7).

Comment from Referee: Line 28-29: This is already discussed in section 3.1

Reply from Authors: This sentence was removed in the revised manuscript.

Comment from Referee: Line 34-36: So basically, this classification is biomass and fossil fuel? As authors said other combustion is also biomass in the broad sense, so how this is different than AAE of section 3.1, besides it is from CHASER model?

Reply from Authors: We showed seasonal features of aerosol optical properties such as AAE. As shown in the manuscript, features of AAE was influenced by organic aerosols derived from combustion (dominantly biomass burning) and oceanic bioactivity, in addition to mixing states of EBC. Also, CHASER model provided us important knowledge on EBC origins and PSA. However, relation between AAE and each EBC origin contribution was not clear. This might result from the dominant contribution of biomass burning on EBC. Therefore, we did not add description about this in the re-

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vised manuscript.

Comment from Referee: Line 37-39: As the text says biomass burning is dominant in spring, and Figure 8 says it is Marine BL and Marine FT contributing to the EBC at Syowa, so what would be the conclusion? It is not clear

Reply from Authors: MBL and MFT were just transport pathway from EBC-PSA to Syowa. To avoid confusion and miss-understanding, discussion on origins of EBC in MBL was added in Section 3.3 in the revised manuscript.

Comment from Referee: Page 10: Line 1: It is difficult to identify the August-October peak in Figure 10b

Reply from Authors: To identify easily months, minor ticks every two months were added in Fig. (Fig. 9 in the revised manuscript).

Comment from Referee: Line 6: “contribution of BB” or “contribution from BB”. Also, the difference is significant if you compare magnitudes of BC, which showed a 25 to 50 % decrease from 2011-2012 to 2015-2016

Reply from Authors: Magnitude of their contributions was added in the text, as follows.

The contributions of BB in South America and southern Africa in August–November were, respectively, 18.1–62.3% (mean 42.1%) and 15.9–71.7% (mean 43.3%). Relative importance of BB in South America and southern Africa showed a slight year-to-year difference.

Comment from Referee: Line 24-25: I am not clear about the statement which ends with a question mark. The explanation is given in the next lines and I consider that statement as a misfit. Is it possible to use a symbol for “BB-model-BC concentrations”, something like the BB (BC) model? Similarly, for FFC and OC

Reply from Authors: Here, we changed the description to “We need to know transport pathway from Australia to the Syowa to understand the high BB-mBC concentrations

in Australia.”

Comment from Referee: Line 30-35: It is quite difficult to follow the month to month explanation from Figure 10. Authors should either include minor labels or ticks or any other way to the identification. The authors might consider a stacked column chart (by normalizing it with total concentrations) for all panel 10b, c, and d. So, the stacked column length would be total, partitions in the column would represent the contributions of South America, Southern Africa, and Australia. I think in that way, all the description in the text would be clearer. But I leave this to authors.

Reply from Authors: We need information about mBC concentrations to explain seasonal features. Seasonal features of contributions of BC origins and PSA were shown in Fig. S5. Therefore, we did not change these figures.

Comment from Referee: Page 11: Line 1-6; I think as the southern America coastline extends much to the southern latitudes (near to western Antarctic peninsula), and the westward transport along coastal Antarctica, might be also a reason for the higher influence on the Antarctic BC, in addition to the GDP. This could be also clarified and detailed in the manuscript.

Reply from Authors: Based on your comment, the following description was added into discussion.

The relevant likelihoods must be discussed to elucidate this difference: (1) difference of transport pathway of anthropogenic EBC from South America and southern Africa to the Antarctica and (2) differences of EBC emission from anthropogenic combustion (i.e. fossil fuel use) in South America and southern Africa. Because of eastward cyclone movement in the Southern Ocean, air masses outflowed eastwardly from the continents of South America and southern Africa. Unlike the Africa continent, the South American continent extends to ca. 55°S. This geographical difference can engender higher contributions of anthropogenic EBC emitted from South America. Indeed, direct evidence of EBC transport from South America was reported in earlier works (Pereira

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et al., 2006; Fiebig et al., 2009; Hara et al., 2010). In addition, higher contributions of South America were observed in transport of mineral dusts to the Antarctica (e.g., Delmonte et al., 2004, 2008; Gassó et al., 2010; Li et al., 2010).

Comment from Referee: Conclusion section: It should be rephrased to highlight important data set period, seasonality of BC, transport patterns at Syowa, model comparison and regional contribution from South America, South Africa, and Australia.

Reply from Authors: From suggestion from you and referee#1, statements in conclusion were modified to understand easily highlight results.

Comment from Referee: Figure 1: I think it would be better to place Syowa station as a different symbol or by placing the name next to the current symbol. Identifying regions like South America and Africa could be also a good idea as it comes quite often in the manuscript

Reply from Authors: Names of each station and others were labeled in Fig. 1.

Comment from Referee: Figure 2: I would suggest adding first and last labels in the Y axis too. Authors may consider writing Syowa near the red circle.

Reply from Authors: We added label in y-axis and name of Syowa in Fig. 2.

Comment from Referee: Figure 3: Y-axis scale for panel c is missing. It doesn't seem matching with panel a. Blue line mentioned in the caption is not visible in the panel a

Reply from Authors: Scales of y-axis were incorrect. These figures were modified. Also, blue line was added in the figure.

Comment from Referee: Figure 5: Syowa location could be shown in a different color/symbol for better visibility. I would suggest using a latitude scale too for this figure

Reply from Authors: Symbols for location of Syowa station were changed. Latitude scale was added in one of figure (not all fig.).

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Comment from Referee: Figure 9: In panel b, the regression coefficient could be shown

Reply from Authors: Regression coefficient and relation (equation) were given in the text. Thus, we did not add them in Fig. 8 (in the revised manuscript).

Comment from Referee: Figure 10: Caption for the panels are not clear. Do authors mean BB aerosols from South America as a whole or BB from South America, contributing to EBC at Syowa.

Reply from Authors: The caption of Fig was modified in the revised manuscript as follows.

Figure 9: Seasonal features of (a) contribution of potential origins of mBC at Syowa Station, (b) the concentrations of mBC released from biomass burning in major PSA, (c) the concentrations of mBC released from combustion of fossil fuels in major PSA, and (d) the concentrations of mBC released from the others in major PSA.

Comment from Referee: List of Acronyms: Some Acronyms from the manuscript are missing in the list, like HYSPLIT, CHASER.

Reply from Authors: We added them and others into List of Acronyms.

Please also note the supplement to this comment:

<https://www.atmos-chem-phys-discuss.net/acp-2018-1190/acp-2018-1190-AC2-supplement.pdf>

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

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