

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

Anonymous Referee #1

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This work describes 11 years of equivalent black carbon (eBC) measurements at Syowa Station in Antarctica and contributes to our understanding of eBC source regions, optical properties and its seasonal cycle in Antarctica. This work represents a set of measurements that are crucial to understanding the composition of the remote atmosphere and is therefore deserving of publication in Atmospheric Chemistry and Physics. Following are suggestions to improve the paper.

General Comments:

(1) This paper requires a thorough edit for clarity. 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.

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(2) This paper make some inferences based on indirect observations, which are identified specifically below. I suggest that the authors ensure that their stated conclusions follow directly from the data they have presented.

(3) The text would benefit from more clear and concise explanations in a number of locations (detailed in specific comments), 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. One example is the discussion of transport pathways in the abstract (boundary layer and lower FT transport versus upper FT). From the abstract alone it is difficult to understand why these findings are relevant. The authors could adapt these sentences to focus more on how the transport pathways identified in their work helps us better understand the seasonal cycle of Antarctic eBC, for example.

(4) The main point of the paper is not entirely clear from the abstract or conclusions. Several possibilities seem clear, such as the lack of significant trend in eBC concentrations between 2005-2016, or the dominance of biomass burning sources, or the seasonal change in eBC optical properties based on changes in AAE. The paper may benefit from more clear statement of which finding(s) is/are the most novel or interesting outcome of this work. That these findings are presented based on a decade of observations also deserves highlighting. If the authors find it appropriate, they may also consider adapting their title to highlight what was found or learned rather than what was done.

Specific Comments:

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

P1 L17: “internally mixed”

P1 L19: “chemical transport model simulations”

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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

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

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

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

P1 L26-27: I appreciate the authors efforts to confront this difficult terminology straight away; however, 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 1984 misses a large amount of literature on this topic. Finally, using “carbonaceous particles” to describe soot of BC-containing aerosol is confusing because it suggests that non-combustion derived organic species are not types of carbonaceous particles. Several recent reviews provide clear recommendations for consistent terminology in this area.

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.

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?

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

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suggestion that anthropogenic BC might be important.

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.

P2 L38-41: I believe what the authors are trying to say here is that a lot of work has been done on finding the sources of long-range transported mineral particles in Antarctica, but 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.

P2 L41: What is meant by show difficulty?

P2 L42: “Here, we combine..”

P3 L1: “chemical transport model simulations”

P3 L11: 2005 - 2016

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.

P3 L30: “BC mass”

P3 L34: How frequently was the DL determined? What is the detection limit at a 15 minute time resolution?

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?

P5 L7: What is meant by “frequent”? The mode value?

P5 L10: Showing a plot of the frequency distribution of eBC (e.g., in the supplement) is

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more illustrative than reporting the log-normal distribution and fitting parameters in the text.

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?

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?

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)

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

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

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

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.

P5 L36-37: This sentence contains problematic wording; it sounds like biomass burning comes from the Antarctic troposphere

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.

P6 L6-7: Mixing month range (here and elsewhere) and season name is a bit confus-

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ing, especially since we are discussing the southern hemisphere

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.

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

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

P6 L31: “spring eBC maximum”

P6 L31-32: Since a lot of this discussion depends of knowledge 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

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?

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.

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.

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

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completely discounted here.

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.

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

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

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?

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

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).

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.

P8 L29-30: The authors' meaning is not clear in this sentence

P8 L35: "southern Atlantic MBL"

P8 L40: Has MFT been defined?

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

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?

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

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

P9 L12: the Antarctic plateau?

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

P9 L31: “Therefore, we discuss. . .”

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.

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?

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

P11 L9-10: “The following possibilities are contributing factors:. . .”

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?

P11 L25: 2005-2016

P11 L30: Are these species internally mixed with eBC?

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

P11 L32: “eBC was emitted”

P11 L34: “chemical transport model simulations”

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

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)

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

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.

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?

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