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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source attribution of Arctic black carbon constrained by aircraft and surface measurements" *by* Junwei Xu et al.

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We would like to thank Referee #1 for his/her useful comments and suggestions that helped to improve the quality of this manuscript. Reponses to these comments are provided below.

Specific Comments:

P1L15: I found the abstract too long, written like a conclusion section. Please try to make it more concise and underline the key messages from your study.

Response: Thanks for the suggestions. The abstract has been condensed.

P1L27-28: I think you oversell the performance of the model in reproducing the vertical

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profile of BC. I don't think you can say that the simulations are "consistent" with the airborne measurements of BC. See for example Fig. 5 where the simulated profiles are flat, especially between 700 and 500 hPa, whereas the observations demonstrate a significant enhancement of BC concentrations. See more comments below (P15L14, P17L17 or P23L1).

Response: Thanks for pointing this out. We have clarified that our simulations underestimate BC concentrations in the middle troposphere in the abstract and the following paragraphs.

P6L4-7: The way it is phrased makes it difficult to understand what is new in this study. Do the authors develop the approach? Do they apply it to a different/longer period than that in Qi et al. (2017a)? Highlight the differences between the methodologies detailed in Kopacz et al. (2011) or Qi et al. (2017a).

Response: Qi et al. (2017a) has been revised to Qi et al. (2017b). We extended the time period from one month (April) in Qi et al. (2017b) to four months (each representing one season) and extended from the surface BC in Qi et al. (2017b) to the column BC that has implications for radiative forcing. We have highlighted these differences in the sentence as "We extend the application of this method to investigate the seasonal and annual responses of Arctic column BC to changes in regional emissions.". Kopacz et al. (2011) has been removed because this is a relatively old study and it not directly relevant to this study.

P6L8: Say that GEOS-Chem is a global CTM. It was only mentioned in the abstract.

Response: Done.

P7L5-6: Can you justify why no scattering corrections to the aethalometer measurements contrary to what is done for the PSAP data (P7L12)?

Response: Good question. The manufacturer's recommended MAC of 16.6 m2 g-1 is calibrated to account for multiple scattering (Sharma et al., 2017), thus no additional

scattering corrections are necessary. We have revised the description to the following in the manuscript: “This MAC value is recommended by the manufacturer for Model AE31 at 880 nm to account for absorption by BC and additional light scattering by both particles and filter fibers.”

P10L11-16: How are fire emissions injected in the model? Are the emissions confined to the surface or does the model takes into account the variability in injection heights (function of burnt vegetation, intensity of fires, buoyancy, ...)?

Response: Fire emissions are injected into the boundary layer in the model. We have clarified this as “Biomass burning emissions are injected into the boundary layer in our simulations” in the manuscript.

P10L22: The description of the simulation is not given in this study. The authors give only a reference to an “old” paper (Park et al., 2003), which cannot be taken as a good reference to fully decribe what has been really done in this work. Please give additional details: period of the simulation, horizontal and vertical resolutions, . . . A lot of modifications have been included in the GEOS-Chem model from the study of Park et al. (2003). Some of them are likely to produce substantial changes in the BC distributions (e.g. wet deposition). Try to underline the main improvements.

Response: Thanks for the suggestion. We have included a detailed description of wet and dry deposition with major improvements since Park et al. (2003) in the model as the following in the manuscript:

“Dry deposition of BC aerosols adopts a standard resistance-in-series scheme as described in Zhang et al. (2001) with improvements on BC dry deposition velocity over snow and ice following Fisher et al. (2010) and Wang et al. (2011). Wet deposition of BC aerosols is initially described in Liu et al. (2001) and developed by Wang et al. (2011) to distinguish between liquid cloud ($T > 268$ K) in which 100 % hydrophilic BC is removed and ice cloud ($T < 268$ K) in which only hydrophobic BC is removed.”

Additional details including the period of the simulation, horizontal and vertical resolutions are described on page 10 line 11-19.

P10L26: Does the model include wet deposition also in subgrid convective updrafts?

Response: Yes. This is described in Liu et al. (2001).

P13L8: The authors explain that EBC concentrations are biased high relative to rBC measurements because of absorbing components and errors in the coefficient used to derive BC concentrations from absorption coefficients. But the fact that EC concentrations are always larger than rBC is never explained. Is it similar to what was observed in other studies? Can it be caused by a difference in sampling the plumes?

Response: Thanks for point this out. Sharma et al. (2017) showed that some of the difference between EC and rBC could be explained by the presence of pyrolysis OC and carbonate carbon that might remain in aerosols after heating to 870 C in the thermal method but were removed in aerosols at 3600C in the refractory method. We have included this reason in the manuscript as "EC concentrations are lower than EBC concentrations from the Aethalometer, yet still high relative to rBC partly due to the presence of pyrolysis OC and carbonate carbon (Sharma et al., 2017)."

P13L14-16: May this discrepancy between aethalometer and PSAP measurements be also ascribed to differences in the calibration method: scattering corrections applied to PSAP but not to aethalometer data? And/or spectral dependency of the absorption coefficient of other substances than BC?

Response: The questions on scattering corrections were explained in the response to P7L5-6.

To understand the effect of spectral dependency of the absorption coefficient of other substances (i.e. brown carbon) on our results, we examined the Aethalometer EBC measurements at 370 nm at Barrow to see if the summer peak still exists at 370 nm. The result showed a distinct peak in July and August with a concentration increased by

10 ng m-3 compared to June and September, indicating the existence biomass burning. Thus, the summer peak in Aethalometer measurements at Barrow was not affected by the wavelength of measurements, but was influenced by biomass burning that was missing in PSAP measurements. We have revised the manuscript to the following to clarify this point.

"The summer peak is also observed in Aethalometer EBC measurements at 370 nm that is sensitive to brown carbon, indicating the influence of biomass burning. Unintentional exclusion of biomass burning plumes in the local pollution data screening performed for PSAP measurements at Barrow could contribute to the bias between the PSAP and the Aethalometer there (Stohl et al., 2006)."

P14L11-14: The peak observed in model simulations at Barrow is explained by a wrong timing of biomass burning emissions. Shouldn't it be the case for all models compared in the Eckhardt et al. (2015) paper if they all use the same fire emissions (with the same timing)? What is the role of injection heights in summer? And what is the contribution of wet deposition processes in the different models?

Response: Eckhardt et al. (2015) used identical biomass burning emissions (GFED 3.1) for all the models in their study. They attributed the difference of simulated surface BC concentrations from different models not only in summer but also the whole year to the treatment of aerosol wet scavenging in the models.

It is not clear what the effect of injection heights is on Arctic surface BC concentrations because some models (i.e. FLEXPART and HadGEM3) do not show the summer peak with biomass burning emissions distributed evenly within the boundary layer, while some other models (i.e. ECHAM6-HAM2) do show the summer peak with the same biomass burning emissions algorithm.

We have revised the sentence from "At Barrow all simulations show a distinct peak in July, which is due to the timing of biomass burning." to "At Barrow all simulations show a distinct peak in July, which is partly due to the timing of biomass burning."

P15L14: I think this sentence oversells the model prediction. The main feature noticed in the observed BC profile is the significant enhancement between 700 and 500 hPa, which is not captured at all by the simulations. So the model does not "generally represent "the relative vertical distribution of BC. The next sentence in the manuscript is a much better way to say this.

Response: We have revised the sentence to "All simulations generally represent the near constant vertical distribution of BC measurements from the surface to 700 hPa, and the decrease above 500 hPa, yet none represent the enhancement between 700-500 hPa."

P15L17-18: The way the sentence has been written gives the impression that most models also strongly underestimate BC in the mid or upper-troposphere. You cannot say this without also referring to more recent studies, highlighting either an overestimation aloft (e.g. Sharma et al. 2013 ; Wang et al., 2014), an improvement in the mid-troposphere (e.g. Breider al., 2014 ; Raut et al., 2017) or a very distinct behaviour between models (as in Eckhardt et al., 2015). In particular both Breider al. (2014) and Wang et al. (2014) used the GEOS-Chem model to represent BC in the Arctic and the results are not similar. This latter study suggested an incorrect production of convective precipitation in the summer in the Arctic. Can you give also some possible reasons to explain the underestimation of BC in your paper (e.g. emissions, plume injection, numerical diffusion, aerosol scavenging, ...) and some ways of improvements?

Response: Thanks for the suggestion. We have referred to Koch et al. (2009), Eckhardt et al. (2015), Breider et al. (2014) and Wang et al. (2011) in the discussion and have proposed some possible reasons for the underestimation as the following:

"The remaining underestimation of 14 ng m-3 RMSE in 500-700 hPa in the HTAP+flaring simulation is possibly due to insufficient magnitude or altitude comparisons of model with ARCTAS and ARCPAC measurements (Koch et al., 2009; Wang et al., 2011; Breider et al., 2014; Eckhardt et al., 2015) as proposed based on preferential

sampling by the aircraft of plumes discussed further below."

P16L5-8: I cannot believe that missing plumes during one specific day (8 April 2015) could explain such a substantial discrepancy. Furthermore, writing "is perhaps due" is "perhaps" a bit too colloquial for a paper (but not a review!). I don't see what this sentence brings to the analysis.

Response: We have rephrased this sentence to indicate the potential sampling bias of plumes and have given another possible reasons as described in the response to the previous comment.

P16L24: What do you mean exactly by the "misrepresentation of these plumes"? Give rather the physical processes responsible for the strong BC underestimation: emissions? numerical diffusion?

Response: By misrepresentation, we meant low in magnitude. Emissions or numerical diffusion might be responsible for the underestimation. We have revised the sentence to "The underestimated magnitudes of these plume, likely related to emissions or numerical diffusion, may contribute to the underestimation of BC concentrations between 500-700 hPa in Fig. 5".

P17L12-13: It does not seem right in summer. Why are the BC columns so low in July near northern Russia? Are the flares off in summer? Is it realistic?

Response: We have included emissions for Jan, Apr and Jul at the bottom panel of Fig. 6. As shown by the figure, flaring emissions in July are very similar to those in the other months. The minor flaring contribution in summer is shown not only for the column but also at the surface where the flaring contribution is usually the largest, as shown in the red shadings of Fig. 3, which is consistent with Stohl et al. (2013). Thus, the flares are not off in summer but the effective wet scavenging in summer likely removes most of BC from anthropogenic sources including flaring. We have included this explanation in the manuscript as the following:

"In July, the enhanced concentrations in western Siberia due to flaring are less obvious, due to more effective wet scavenging in summer."

P17L17: I would remove the word "vertical" and keep only the term "spatial" distributions.

Response: Done.

P18L2-4: This is not only caused by different transport patterns along which air masses reach the Arctic region, but also by different transport efficiencies due to scavenging.

Response: We have revised the sentence to "partly reflecting different transport pathways and scavenging efficiencies".

P19L8: I disagree with the use of the word "generally" here. A lot of studies have been focused on the same objective and they are not necessarily in agreement. How do the results of this study compare for example to Table A1 of Wang et al. (2014) who summarized twelve studies focusing on the source attribution of Arctic BC?

Response: Thanks for the suggestion. We have compared to Huang et al. (2010), Sharma et al. (2013), Shindell et al. (2008), Stohl (2006) and Ma et al. (2013) in Table A1 of Wang et al. (2014) and have revised the paragraph to the following

"The largest contribution from eastern and southern Asia to Arctic BC burden in this study is consistent with Ma et al. (2013) and Wang et al. (2014). However, some prior studies suggested that Europe had the largest contribution to Arctic BC burden (Stohl, 2006; Shindell et al., 2008; Huang et al., 2010a; Sharma et al., 2013) The difference likely arises from trends in anthropogenic emissions with reductions from Europe and increases in eastern and southern Asia as discussed further below."

P20L6-7: Another possible explanation could be that biomass burning plumes have been transported at a too low altitude. This would explain the strong overestimation at the surface and the marked underestimation aloft. Is it a possible scenario?

Response: In the response to P13L14-16, we have described that biomass burning plumes are likely missed by the PSAP measurements. Thus, it is not clear to us whether the discrepancy should be attributed to the simulation or the measurements.

P23L1: Here again, the simulated vertical BC profile is not consistent with that measured. The following sentence is a good way to say this.

Response: Revised.

P24L5-8: This is an odd conclusive remark for a study focused on model simulations. It may be better to insist on the model uncertainties in the result mentioned above mentioned and how they can be addressed in future work.

Response: Thanks for the suggestion. We have added "The considerable impact of emissions from China and Indo-Gangetic Plain on the Arctic deserves further investigation." to the conclusion.

Fig. 3: What is the uncertainty on simulated monthly BC concentrations? It could be estimated by the standard deviations of the model results used to derive the monthly averages. Please add them on Fig. 3 at least for the HTAP+flaring emission inventory.

Response: Done.

Fig. 4: Could you add an errorbar on the ground mean concentrations?

Response: Done. But only one year (2009) of ground measurement is available for Ny Alesund, so no error bar presents in the figure.

Fig. 5: This caption is good, but some of this text should also be reported in the manuscript, in particular L6-7 to understand how the analysis is performed.

Response: Done.

Fig. 6: This is not any flaring plume in July. Is it normal?

Response: There is flaring plume in July but the magnitude is smaller than the other

seasons because of more effective wet scavenging in summer. This has been described in more details in the response to P17L12-13.

Fig. 8: Why aren't there error bars at Barrow and Ny Alesund stations as at Alert?

Response: Thanks for pointing out this error. Corrected.

Fig. 9: Why don't you take into account 12 months to build the annual map? I think this is confusing. It is fine to present maps for particular months (Jan, Apr, Sept) but I don't undertand the purpose of computing an annual average based on only 3 months, when 12 have actually been simulated.

Response: We do not have a full year simulation with the adjoint model because it is time consuming. We assume the four months (Jan, Apr, Jul and Sept) are representative of four seasons. We have revised the caption to "The annual map is the average of contributions in January, April, July and September calculated with the adjoint model."

Table 1: Why don't you also include rRMSE in this stable? The discussion relative to this section would be clearer.

Response: We have included rRMSE in Table 2.

Technical comments: In many places, add a space between value and unity, e.g. P2L6, P7L22, P8L4, P11L4, P15L8, P15L16, P15L17, P16L1, P16L18, P16L26, P18L9, P18L22, P23L3, P23L11, P23L15.

Response: Done.

P3L8: Remove the dot before the parenthesis.

Response: Done.

P7L21: Neodymium dopped YAG is generally abbreviated as Nd:YAG, not Ni-YAG.

Response: Done.

P9L25: ACP journal recommands to write Figure instead of Fig. at the beginning of a

sentence.

Response: Done.

P10L6: Replace semi-colon by comma.

Response: Done.

P17: section should be abbreviated as Sect. in ACP.

Response: Done.

P13L10-15: There is a typo in aethalometer L10, L11 and L15.

Response: Done.

P22L4: “to a half of the impact”: this is not very clear. Please reformulate.

Response: Done.

P27L10-14: Evangeliou et al. (2016): the paper has been published in ACP in June 2016. It is not a discussion paper any longer.

Response: Done.

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