

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

Anonymous Referee #1

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General comments : This work investigates the source attribution of Arctic black carbon using a global CTM and surface (at 3 stations) and airborne (NETCARE, PAMARCMiP) observations. Using different simulations switching off some of the emission sources (e.g. flares, seasonal heating) as well as a global CTM adjoint, the authors determine the spatially resolved source contributions to receptor locations. They highlight the influence of different pollution sources and the resulting vertical and spatial distribution of BC. This study is interesting, scientifically important and sound for ACP. My main comment is that the authors oversell a bit the performance of the model to reproduce the vertical gradient observed in the BC concentrations. The model is actually doing a reasonable job of capturing the general features of the spatial BC distribution, though not doing a perfect job of simulating the concentrations. Most comments listed below are minor clarifications. Once these points are addressed satisfactory, the paper should

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in my opinion be suitable for publication in ACP.

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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.

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

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).

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

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

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

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 describe what has been really done in this work. Please give additional details : period of the simulation, horizontal and vertical resolutions, ... A

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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.

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

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 ?

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 ?

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 ?

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.

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 overestima-

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tion aloft (e.g. Sharma et al. 2013 ; Wang et al., 2014), a 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 ?

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.

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 ?

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 ?

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

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.

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 ?

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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 ?

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

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.

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.

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

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.

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

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

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 understand the purpose of computing an annual average based on only 3 months, when 12 have actually been simulated.

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

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Sharma, S., Ishizawa, M., Chan, D., Lavoué, D., Andrews, E., Eleftheriadis, K., and

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Maksyutov, S.: 16-year simulation of Arctic black carbon: Transport, source contribution, and sensitivity analysis on deposition, *Journal of Geophysical Research: Atmospheres*, 118, 943–964, 2013.

Wang, H., Rasch, P. J., Easter, R. C., Singh, B., Zhang, R., Ma, P.-L., Qian, Y., Ghan, S. J., and Beagley, N.: Using an explicit emission tagging method in global modeling of source-receptor relationships for black carbon in the Arctic: Variations, sources, and transport pathways, *Journal of Geophysical Research: Atmospheres*, 119, 12,888–12,909, doi:10.1002/2014JD022297, <http://dx.doi.org/10.1002/2014JD022297>, 2014JD022297, 2014.

Raut, J.-C., Marelle, L., Fast, J., Thomas, J. L., Weinzierl, B., Law, K. S., Berg, L., Roiger, L., Easter, R., Heimerl, K., Onishi, T., Delanoe, J. and Schlager, H. : Cross-polar transport and scavenging of Siberian aerosols containing black carbon during the 2012 ACCESS summer campaign, *Atmos. Chem. Phys. Disc.*, doi:10.5194/acp-2016-1023, 2017.

Breider, T. J., Mickley, L. J., Jacob, D. J., Wang, Q., Fisher, J. A., Chang, R., Alexander, B., et al.: Annual distributions and sources of Arctic aerosol components, aerosol optical depth, and aerosol absorption, *Journal of Geophysical Research: Atmospheres*, 119, 4107–4124, 2014.

Eckhardt, S., Quennehen, B., Olivie, D. J. L., Berntsen, T. K., Cherian, R., Christensen, J. H., Collins, W., Crepinsek, S., Daskalakis, N., Flanner, M., et al.: Current model capabilities for simulating black carbon and sulfate concentrations in the Arctic atmosphere: a multi-model evaluation using a comprehensive measurement data set, *Atmospheric Chemistry and Physics*, 15, 9413–9433, 2015.

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Technical comments :

In many places, add a space between value and unity, e.g. P2L6, P7L22, P8L4, P11L4,

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P15L8, P15L16, P15L17, P16L1, P16L18, P16L26, P18L9, P18L22, P23L3, P23L11, P23L15.

P3L8 : Remove the dot before the parenthesis.

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

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

P10L6 : Replace semi-colon by comma.

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

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

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

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

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-236, 2017.