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Interactive comment on "Tagged tracer simulations of black carbon in the Arctic: Transport, source contributions, and budget" *by* Kohei Ikeda et al.

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Reply to Referee #1

Thank you very much for carefully reading our manuscript and providing valuable suggestions. We have thoroughly revised the manuscript following the comments by the reviewers.

Major Comments: In abstract authors seem to claim that the new scheme has improved comparison with observations, however, this does not seem to be true when looking at figure 2 for Barrow and Zeppelin where the so called standard scheme suggests a

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better comparison with observation. Also why there is no blue line in Fig. 3 similar to Fig. 2?, how does blue line compare here? Why Fig. 4 does not show comparison with standard and new scheme?

Answer: We agree that the new scheme did not improve the reproducibility at Barrow and Zeppelin. We have modified the expression claiming that the model reproducibility has been entirely improved by the new scheme in abstract. The statement about the model performance in abstract was modified to "Firstly, we evaluated the simulated BC by comparing it with observations at the Arctic sites and examined the sensitivity of an aging parameterization and wet scavenging rate by ice clouds" (Page1, Lines 11-12). We have added the results of the standard scheme to Fig. 3 and Fig. 4 and discussions on comparisons with standard and new schemes as described below.

Page 2, lines 30-33: I agree that Eckhardt et al. (2015) found that BC is still underestimated in several models. The potential reasons for this were investigated by Mahmood et al. (2016) who used data from same model used by Eckhardt et al., 2015 and found that one major reason is convective wet deposition process outside the Arctic which influences transport of BC into Arctic. This is a major study for Arctic BC processes and should be included in the introduction. Mahmood, R., K. von Salzen, M. Flanner, M. Sand, J. Langner, H. Wang, and L. Huang (2016), Seasonality of global and Arctic black carbon processes in the Arctic Monitoring and Assessment Programme models, J. Geophys. Res. Atmos., 121, doi:10.1002/2016JD024849

Answer: We have added the following description about the study of Mahmood et al. (2016) to the introduction section (Page 2, Line 34-Page 3, Line 2).

"Mahmood et al. (2016) pointed out that convective wet deposition outside the Arctic influenced vertical distribution and seasonal variations of BC in the Arctic by analyzing the same models used by Eckhardt et al. (2015)."

Page 4: Emission inventory: I wondering why the authors are using an older version of GFED fire emission data when a new version (GFED4 and GFED4s) are available?

Answer: We compared BC emissions of GFEDv3.1 with GFEDv4.1s for 2007-2011 (simulation period) and confirmed that the difference of global BC emissions between these inventories is 9%. For boreal forests, BC emission from Siberia (defined as SIB-BB in this study) of GFEDv4.1 is only 8% larger than that of GFEDv3.1. The emission from Alaska and Canada (ALC-BB) of GFEv4.1s is 32% smaller compared with GFEDv3.1. Therefore, we think that the main conclusion of this study will not be influenced by the difference in the version of GFED.

Page 4, lines 10-25: It is not quite clear which anthropogenic emission inventory the authors are using. At beginning they say that "In this study, we adopted the BC emissions of HTAPv2.2", however later on they mention that they used an inventory by Huang et al., 2015. In addition, the authors also claim, without any proof, that the inventory of Huang et al., 2015 improved comparison with observation. I do not see any such results of their so called "preliminary simulations".

Answer: Because the emission of Huang et al. (2015) is the regional inventory for Russia, we adopted this inventory only for Russia and HTAPv2.2 was used for all regions except Russia. We have added the results of the preliminary simulation as Fig. S1. This sentence has been modified to "Our preliminary simulations found that the model result replacing HTAPv2.2 emission in Russia by the inventory of Huang et al. (2015) improved the reproducibility of the observed BC concentrations at the Arctic sites (see, Supplemental Fig. S1), and thus we used this emission dataset as the anthropogenic BC emissions for Russia" (Page 4, Lines 27-30).

Page 4, lines 15-20: The doubling of BC emissions in Asia and Russia, How realistic that would be? The authors argue that it is necessary to match modeled BC with observations in Arctic, but could this not be due to other modelling errors/discrepancy? How certain the authors are about this? A recent study showed that the differences in modeled aerosol processes in different models can contribute to overall concentrations in the Arctic (Mahmood, R., K. von Salzen, M. Flanner, M. Sand, J. Langner, H. Wang, and L. Huang (2016), Seasonality of global and Arctic black carbon processes in

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theArctic Monitoring and Assessment Programme models, J. Geophys. Res. Atmos., 121, doi:10.1002/2016JD024849.)

Answer: This description is about the previous study of Wang et al. (2011) who used the inventory of Bond et al. (2007), and not for this study. We do not make any changes from the original emission data (i.e., HTAPv2.2 and the inventory of Huang et al., 2015 for Russia).

Page 4 lines 20-22: "which was about 20%", Is 20% correct? It seems to be ∼22.2%from the numbers given in that line? Even after rounding it would be 22%? Please also check the subsequent numbers.

Answer: We corrected it and subsequent numbers. This part has been modified to "The target year of HTAPv2.2 was 2010 and global annual emissions were estimated to be 5.5 Tg yr−1, which was about 22 % larger than that of Bond et al. (2007) (4.5 Tg yr−1). On a regional basis, the emissions from China were 40 % larger than those of Bond et al. (2007), and the emissions from Europe and North America were 34 % and 11 % smaller than those in Bond et al. (2007), respectively" (Page 4, Lines 21-24).

Page 5 paragraph 25: the authors say "We separated the major source regions of anthropogenic BC such as Europe, Russia, Asia and North America into different tracers", which different tracers?

Answer: We have modified this sentence to "We separated Europe, Russia, Asia and North America to examine transport patterns and contributions to the Arctic from the major source regions." (Page 5, Lines 28-29)

Page 5, lines 25-30: "Asia was separated into three regions (i.e., East Asia, Southeast Asia and India)" , According to Fig. 1, the region named "India" contains several other countries, e.g. Sri Lanka, Pakistan, Nepal, Bangladesh, Myanmar, so this region should be named "South Asia : SA".

Answer: We have changed the name of this region to "South Asia" in the text, the figures, and the tables.

Page 6, lines 25-30: The authors claim that the correlations between observed and model BC has improved with new scheme. However, at least from Figure 2, it does not seem to appear that the new curve changed in its shape compared to standard scheme, only the magnitude seems to have changed then how the correlation is improved?

Answer: We have modified the discussion on the model performance of the seasonal variations based on correlation coefficients (R) and root mean square error (RMSE) at each Arctic site. R values were improved by the new scheme from 0.89 to 0.92 at Alert and from 0.935 to 0.944 at Tiksi, respectively. At Barrow, R was increased from 0.69 to 0.81, but RMSE was not improved by the new scheme. At Zeppelin, the standard scheme (R=0.89) showed a good agreement compared with the new simulation (R=0.83). Based on these results, the discussion about the model reproducibility has been modified to the following statement (Page 7, Lines 11-16). These statistics (R and RMSE) have also been added to Fig. 2.

“The standard scheme underestimated observed BC in winter and spring at Alert and Tiksi. The model negative biases were reduced by the new scheme in these seasons, and R values were improved from 0.89 to 0.92 at Alert and from 0.935 to 0.944 at Tiksi (Fig. 2). At Barrow, while the new simulation improved the negative biases in spring, the observed concentrations were overestimated during winter. As a result, the correlation coefficient was increased from 0.69 to 0.81, but root mean square error (RMSE) was not improved by the new scheme at Barrow. Whilst there was an improvement at Alert and Tiksi, the observations at Zeppelin showed a reasonably good agreement with the standard simulation (R=0.89) rather than the new simulation (R=0.83).”

Page 6, lines 30-35: the authors say: “This is mainly because the new scheme yielded an increase in BC concentrations except in summer with maximum effects in winter at the all four Arctic sites.”. Figure 2 clearly shows that BC is also increased in summer, though relatively small. Thus I think this sentence is not correct. Similarly from Figure 2, I do not think that the new scheme improved BC values at Barrow as the authors seem to claim. There is clearly way more over-estimations for 9 months in new scheme than the standard scheme. How can the authors claim it an improvement when it is overestimating more than the standard scheme for most months of the year including, November, December, January, February?

Answer: We have removed the phrase "except in summer" from this sentence. The following statement about the seasonal variation of sensitivities (winter maximum) was added (Page 7, Lines 9-11).

"The sensitivities by changing these parameterizations were largest in winter because wet removal by ice clouds was most important in this season and aging time scale which depends on OH number concentrations also became longer than other seasons." We have modified the statement of the model performance at Barrow as described above. Please also see our reply to the above comment.

Page 7 lines 10-15: Is there any evidence for overestimation of BC emission from Russia?

Answer: We have deleted this sentence.

Page 7, lines 15-30: Why there is no discussion of standard scheme in Fig. 3? If the authors want to claim that the new scheme is better than the standard scheme then all model and observations comparisons should include results from both schemes. Same comments for next paragraphs about Figure 4.

Answer: We have added the results of the standard scheme to Fig. 3 and Fig. 4 and discussions about the comparison between the standard and the new schemes. For vertical profiles (Fig. 3), the standard scheme underestimated the observations especially in the middle troposphere. The new scheme improved the model performance by increases BC concentrations from the surface to the upper troposphere. The following

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description has been added (Page 7, Line 34-Page, 8 Line 4).

“Although the standard scheme reproduced the increase from near the surface to the middle troposphere and the decrease from 5 km to the upper troposphere, the observed concentrations were underestimated 24–42 % in the middle troposphere. The negative biases were improved by the new scheme by increasing BC concentrations 18–23 ng m-3 in the middle troposphere. These increases by the new scheme were caused by the longer lifetime of BC in the high latitudes as discussed above.”

The sensitivities were small in the major anthropogenic source regions (Europe, East Asia, and North America), because BC aging time of the new scheme is similar to that of the standard scheme ($\sim$1 day) in the mid-latitudes and wet scavenging by ice clouds is not so important in these regions. We have added the following description (Page 8, Lines 24-27).

“The differences between the standard and new schemes were small in the all three regions (Fig. 4). This is because BC aging time by the new scheme is similar to that of the standard scheme ($\sim$1 day) around the source regions in the mid-latitudes and wet scavenging by ice clouds is not so important in these regions. Because the BC concentrations tended to slightly increase in the new simulation, NMB were improved by the new scheme from -14–-43 % to -6–-42 % (Fig. 4).”

Page 9, lines 10-15: The authors write “The stable condition by cold temperatures near the surface suppresses the upward transport of BC over Russia especially in winter”, I agree that stable conditions would suppress vertical transport of BC, but it would also depend on source. For example, if source is gas flaring or forest fires then emissions could reach middle troposphere?

Answer: Because fires in boreal forests occur from late spring to autumn, BC emitted from forest fires is not included in the discussion on the transport process in the cold season. Injection heights of anthropogenic sources including gas flaring are not provided by emission inventories. Thus, it is difficult to investigate the source dependence of vertical distribution at the present stage. We would like to make this issue to address in the future research.

Page 11, lines 17-18: “The relative importance to the BC concentrations on an annual basis will be discussed later (Table 2)” This sentence does not make much sense and therefore need be rewritten.

Answer: We have removed this sentence.

Page 12, lines 25-27, How is the BC lifetime defined here? More importantly how this discussion is related to the current study which is primarily about regional BC processes. It would more relevant if the lifetime of BC in the Arctic is given here since this study is focused on Arctic (for a multi-model comparison of lifetimes see Mahmood et al., 2016)

Answer: The BC lifetime is defined as the BC burden divided by the annual total (wet and dry) deposition. The definition of BC lifetime was added to the text (Page 14, Line 13). We have added the BC lifetimes in the Arctic to Table 1 and Table S1 and a comparison with the lifetimes reported by Mahmood et al. (2016). The following statement was added (Page 14, Lines 16-21).

“The BC lifetimes of each tracer in the Arctic (66°–90°N) were estimated to be 8.6–92.7 days. The lifetime of EAS-AN BC in the Arctic (57.5 days) was longer than those of EUR-AN (14.2 days) and RUS-AN (12.9 days), because East Asia BC was distributed mainly in the middle troposphere (Fig. 9) and its deposition to the Arctic was smaller than those of EUR-AN and RUS-AN (Table 1). The average lifetime of 21.3 days in the Arctic was close to 20.0 days of the multi-model mean in the AMAP (Arctic Monitoring and Assessment Programme) models (Mahmood et al., 2016).”

Page 13 lines 32-33: the authors write “We also quantitatively estimated the relative contributions to the total deposition of BC to the Arctic region (Table 2)”. relative contributions of what?

Answer: We modified this sentence to "We also quantitatively estimated the relative contributions of each source to the total deposition of BC to the Arctic region (Table 2)" (Page 15, Line 26).

Page 14: Conclusions: This section has conclusions which I would find hard to agree with. For example, the authors seem to claim that they have identified important pathways for BC transport to the Arctic. Stohl (2006) had discussed the transport pathways to the Arctic. I am not satisfied that the authors provide adequate discussions of transport pathways and how they would differ with those discussed by Stohl 2006. Again authors seem to claim that new scheme improved bc simulation in Arctic which is not obvious for at least two of the four sites for which observation data they used. Also there is no discussion of uncertainty about BC simulation results. Using just one model simulation can have problematic results. It is advisable that the author either use nudging technology or ensemble members or both to minimize the influence natural variability or at least provide some uncertainty range.

Answer: We have added discussions on transport pathways from individual sources and their seasonal variations in section 3.2. A new figure of horizontal winds in the lower and middle troposphere and precipitation was also added as Figure 7. In conclusions, we have modified the expression on transport pathways as follows. (Page 16, Lines 31-32) "We examined detailed transport pathways from the individual source regions to the Arctic and identified important regions where inflow from the individual source regions to the Arctic occurred."

We agree that the new scheme did not improve the reproducibility at Barrow and Zeppelin as replied above. We modified the expression claiming that the model reproducibility has been entirely improved by the new scheme in conclusions. The statement on the model performance in conclusions was modified as follows. (Page 16, Lines 20-24) "We introduced a parameterization of BC aging into GEOS-Chem and changed the wet scavenging ratio by ice cloud (T<258 K) to examine the sensitivities of these processes to the Arctic BC. By using these new schemes, the BC concentrations were increased at the Arctic especially in winter and spring. Although the new scheme over-estimated the observations at Zeppelin and Barrow especially during winter, model the negative biases in the cold season were improved at Alert and Tiksi."

Because the model used in this study (GEOS-Chem) is a chemical transport model (not a chemical climate model), meteorological fields are not calculated in the model and assimilated meteorological fields GEOS-5 are used to drive it. Thus, nudging and ensemble simulations are not required in this study. We agree that it is important to provide uncertainty range. We have added the interannual variations of the source contributions to annual mean BC concentrations at the surface and 5 km altitude, annual deposition and burden in the Arctic (Table S3). Because the anthropogenic emissions used in this study had no interannual trends, interannual variations in source contributions were caused by those in meteorological conditions and biomass burning emissions. We found that results of each year were similar to that of the 5-year averaged contributions. The following description of the interannual variations of the relative contributions from each source to the Arctic BC was added in section 3.4. (Page 16, Lines 5-9)

"We estimated interannual variations of relative contributions from individual sources to the Arctic BC and found that results of each year were similar to that of the 5-year averaged contributions (see, supplemental Table S3). The differences of the relative contributions from each source to the BC concentrations between maxima and minima were lower than 12 %. For BC total deposition, the relative contribution from biomass burning in Siberia (SIB-BB) showed the variation from 8.2 % to 24.0 % (Table S3)."

Minor comments: Several figures can be improved. For example for Figure 4, 5 and 6 only color bar may be used instead of repeating same color bar for individual plots. The font size of numbers of color bars is too small.

Answer: We have modified these figures, according to the referee comment.

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