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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 #2

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 this paper, the authors claim that using the new scheme "the model reproducibility of the seasonal variations is increased" or " the simulated seasonal variations were improved". However, based on figure 2 this claim is only correct for Alert and Tiksi sites. I believe this needs further clarifications. For example, for Zeppelin site, the above claims are not true at all and the standard scheme shows significantly better performance in capturing both values and seasonality of BC. For Barrow, the standard scheme captures the summer, fall, and winter-time BC concentration better than the new scheme and we only see the improvement in the simulations for spring. Also for Tiksi, although the new scheme values are closer to observations, they are still under predicting BC very significantly. I would recommend adding some statistical analysis and more discussion for backing up this claim.

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 and conclusions.

In section 3.1, we have modified the discussion on the model performance of seasonal variations based on the 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).”

I would highly recommend comparing the results of the new scheme vs. the standard scheme for the vertical distributions along the ARCTAS flight path. Also, did you make any comparisons for each flight? Have you checked the performance of you model for the ARCTAS flights below 66N?

Answer: We have added the results of the standard scheme to Fig. 3 and discussions about differences between the standard and the new schemes. 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 description has been added (Page 7, Line 32-Page 8, Line 2). This comparison showed averaged vertical distributions of five flights in April and did not include the observations below 66N.

“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 probably caused by the longer lifetime of BC in the high latitudes as discussed above.”

Finally, I would recommend adding more description on the transport mechanisms from each sector and the reasons behind the seasonality. The paper shows interesting results, but it needs more discussion on how the transport pathways change in different seasons.

Answer: We added the description on the transport patterns from each source and their seasonal variations to section 3.2 based on meteorological fields. A new figure of horizontal winds in the lower and middle troposphere and precipitation was added as Figure 7. For the low-level transport from Europe and Russia, northeastward winds prevailing over northern Europe and western Russia probably played an important role

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Discussion paper

on the poleward transport in winter and spring. Low precipitation (< 1 mm day-1) over Russia also contributed to the effective transport to the Arctic from northern Eurasia in the cold season. In contrast, during summer the circulation pattern changed to southeastward winds and was not preferable for the poleward transport. In addition, precipitation increased over high-latitude Eurasia in summer leading to effective wet removal. The weak transport to the Arctic from Europe and Russia in summer was attributed to these meteorological conditions. The poleward transport from East Asia in the middle troposphere was attributed to northward winds blowing over the Okhotsk Sea, East Siberia and the Bering Sea in winter. Although seasonal mean northward winds in spring over these regions were weaker than those in winter, the contribution of East Asia BC in spring was larger than that in winter. This enhancement of EAS-AN BC during spring was not sufficiently explained by only the seasonal mean winds, suggesting that synoptic-scale disturbances on shorter time scales had an important role on the poleward transport from East Asia to the Arctic. Based on these results, we have added the discussions on seasonal variations of transport patterns from individual sources to the first three paragraphs in section 3.2 (Page 8 Line 31-Page10 Line 22).

Minor Comments: Page 6, Lines 17-20: Please add a reference or citations for the observation data used for this section.

Answer: We have added a reference for the observation data (Page 6, Lines 23-24). “The measurement data at the Arctic sites were obtained from EMEP and WDCA database (http://ebas.nilu.no).”

Page 6, Lines 30-31: I would recommend removing the “expect in summer” phrase from the following sentence and add further clarifications to it. “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.” Based on figure2 the new scheme shows higher values for summer as well, but the increase is smaller than other seasons.

Answer: We have removed “except in summer” from this sentence. The following statement was added to discuss the seasonal difference of the sensitivity (Page 7, Lines 9-11). “The sensitivities by changing these parameterizations were the 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.”

Page 7, Line 13: What would be the possible reasons for “a too effective transport to Zeppelin”?! I would recommend adding more clarifications on why the model overestimated BC in Zeppelin.

Answer: We have removed this sentence. Our simulations suggested that it is difficult to reproduce the seasonal variations at the all Arctic sites in the current model. Although the cause of the discrepancies remains unclear, it is useful to show the sensitivities of aging and wet removal by ice clouds processes at the Arctic sites. One reason is that the sensitivities of these processes at Zeppelin were larger than those at Barrow and Alert, leading to the overestimation of the new scheme in winter and spring. (Page 7, Lines 19-20)

Page 7, Lines 15-23: What would be the possible reasons for underestimation below 3k and overestimation in mid-troposphere? Adding more discussion and statistical analysis in this section will help. Also please add the standard scheme results to this analysis and figure3.

Answer: We have added the result of the standard scheme and discussion as described above. The possible reason was added (Page 8, Lines 7-8). “The simulated vertical gradient from the surface to the middle troposphere was slightly weaker than that of the observations. One possible reason is that upward transport of BC was underestimated by the model.”

Page 7, Line 17: Please add the dates of flights used for this analysis.

Answer: We have added the dates of flights used for this analysis (Page 7, Line 31). "The dates of flights used for the comparison were April 8, 9, 12, 16, and 17."

Page 7, Lines 28-30: I would recommend adding references here or in page 6-lines17-20. Please see the above comment. Also please add a map with the locations of the sites that are selected for this study.

Answer: We have added references for IMPROVE and EUSAAR sites as follows (Page 8, Lines 14 and 16). A map of the sites used in this study was also added to Figure 4. "For North America, the data from the IMPROVE network for 2007–2011 was used (http://views.cira.colostate.edu/fed)." "The measurement data at EUSAAR sites were obtained from EMEP and WDCA database (http://ebas.nilu.no)."

Page 7, Lines 25-30: Adding discussion on possible reasons on why the model underestimates the observations over Europe and East Asia. Also, please add the results of new scheme vs. standard scheme. How was the performance of the standard scheme for these selected sites?

Answer: We have added the following discussion on possible reasons over Europe and East Asia (Page 8, Lines 22-24). "The possible reasons for the model underestimation over Europe and East Asia are that BC emissions from these regions are underestimated and removals around the source regions are overestimated by the model." We have added the result of the standard scheme to Fig. 4 and discussions about differences between the standard and the new schemes. The sensitivities were small in the major anthropogenic source regions, 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 statement (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 8, Lines 15-30: Please add some description on how you calculated meridional fluxes for these plots.

Answer: We have added the following description (Page 9, Lines 3-4). "The horizontal fluxes were calculated by multiplying 6-hourly BC mass concentrations by horizontal wind speeds and were averaged for three months. "

Page 9, Lines 15-32: I would recommend adding more discussions here and summarize some previous studies on Transport pathways and why there is a strong aloft meridional flux. (For example adding more discussions on location of polar dome and relative vertical mixing in different seasons).

Answer: We have added discussion on uplifting of East Asia and North America BC during long-range transport including the influence of the polar dome. The following statements were added.

"The Arctic lower troposphere is isolated by the closed polar dome which is formed by isentropic surfaces of lower potential temperatures and pollutants cannot easily be penetrated into the Arctic from outside of the polar front (Barrie, 1986). East Asia is located at south of the polar dome and EAS-AN BC is emitted from at higher potential temperatures. As a result, the low-level transport of East Asia BC into the Arctic was weak and it was transported at higher altitudes (Klonecki et al., 2003; Stohl, 2006)." (Page 11, Lines 7-11) "This is because North America BC is also emitted from higher potential temperatures and was transported to the Arctic above the polar dome." (Page 11, Lines 14-15)

Page 12, Lines 17-20: I have found the following sentence very confusing. Please modify this sentence. "Although the efficiency of the EAS-AN BC transport to the Arctic was lower than that of the other anthropogenic sources (EUR-AN, RUS-AN and NAMAN) due to the effective wet removal (Fig. 9), the inflow flux was the largest among the four major sources. “

Answer: We have modified this sentence to “Although the fraction of BC from East Asia transported to the Arctic was lower than those of the other anthropogenic sources (EUR-AN, RUS-AN and NAM-AN) due to the effective wet removal (Fig. 9), the inflow flux of EAS-AN was the largest among the four major sources.” (Page 14, Lines 5-7)

Page 13, Line 24: The second largest what? Maybe “The second largest was the contribution” -> “The second largest contributor to the Arctic BC was”?

Answer: This sentence was modified to “The second largest contributor to the BC burden over the Arctic was Russia (21.0 %)”. (Page 15, Lines 17-18)

Page 15, Lines11:15: Please add the % contributions of BB emission from Siberia and Alaska and Canada during summer.

Answer: We have added the relative contributions of BB from Siberia (32 %) and Alaska and Canada (31 %) to BC deposition on the Arctic during summer. (Page 17, Lines 19-21) “However, for BC deposition on the Arctic, the contributions of biomass burning emissions from Siberia and Alaska and Canada that became substantial during summer were important, accounting for 15 % (32 %) and 12 % (31 %) in annual mean (during summer), respectively.”

Figure 1-a: The plot would be easier to read if you mark the whole East Asia as well, maybe adding a zoomed map for that section to show the East Asian regions (i.e. Korean Peninsula, South China, etc.) It was difficult to locate the region of East Asia and its sub-regions in the emission plot.

Answer: We have added a zoomed map for the East Asian region to Fig. 1(a) for clarity.

Figure 3: Please add the standard scheme vs new scheme comparison with observation in Figure 3. Also, it would be nice, if you can add the error bars and NMB (or

RMSE).

Answer: We have added the result of the standard scheme as described above. The error bars and NMB were added to Figure 3.

Figure 4: It would be great to add the locations of the observations site on a map. For example, it is not obvious which IMPROVE sites were chosen for plotting and this comparison.

Answer: We have added a map of the observation sites used in this study to Figure 4.

Figure 5 and Figure 6: I would recommend removing wet scavenging lines from these plots or export the plots at a higher resolution. The font of these plots was very small and very hard to follow. What do the numbers in the white squares represent? The numbers are very hard to read.

Answer: We modified Figure 5 and Figure 6 to high-resolution figures.

Figure 8 and Figure 9: Please add a description if this is the area average concentration for the Arctic or the concentration at a specified location in the Arctic?

Answer: We have added the averaged area (66-90N) to the top of these figures.

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