Response to Editor

acp-2021-1091: "Contrasting source contributions of Arctic black carbon to atmospheric concentrations, deposition flux, and atmospheric and snow radiative effects" by H. Matsui et al.

We thank the Editor very much for reading the paper carefully and giving us valuable comments. We revised the paper by taking into account all the Editor's comments. Detailed responses to individual comments and suggestions are given below.

Editor's comment:

Having said that, I also agree with the assessment that the explanation of the enhanced mass absorption coefficient for Asian BC would still benefit from a bit more scrutiny and improved explanation.

Response:

As the reviewer pointed out, the large MAC values of anthropogenic BC from Asia in our model simulations may be because of fast aging processes near the source regions. Considering this point, we have revised the manuscript as follows: "*Compared with anthropogenic BC from Siberia, Europe, and North America (>50°N), anthropogenic BC particles from Asia have a higher fraction of thickly coated BC particles (which have higher* MAC_{BC}*) and a lower fraction of thinly coated BC particles (which have lower* MAC_{BC}*) (Fig. 12a). The higher fraction of thickly coated BC from Asia might be explained by fast aging processes near their sources, where the concentrations of condensable gases emitted with BC are high, and by the higher fraction of anthropogenic BC from Asia in the upper troposphere in the Arctic (Fig. S7) and its longer lifetime in the Arctic (24−30 days) (Fig. 12b).*" (Lines 365-371).

In addition, many sources may contribute to the observed thickly coated BC in the Arctic region, as in the reviewer's comment. Considering this point, we have added the following discussion to the revised manuscript: "*The fraction of thickly coated BC was observed to be high in the Arctic in recent aircraft measurements by Ohata et al. (2021b), consistent with our model simulations, although it is difficult to observe the dependence of BC mixing states on emission sources. Our simulation results indicate the importance of understanding the differences in BC mixing states among sources and the mechanisms that control them in evaluating the source contribution of BC to* RE_{BC_TOA} *in the Arctic.*" (Lines 374-378).

Editor's comment:

An understanding of this relies on the description of the aerosol microphysics. However, in the present form, insufficient detail is provided to understand key processes, in particular regarding the mixing state and the radiative properties:

- It is not entirely clear from the description, if all microphysical processes act on the full 2D bins or if they are estimated from simulating microphysical processes on the 17 size bins and subsequent allocation to mixing state bins.

Response:

Microphysical processes are calculated for the full 2D bins (47 bins). Changes in particle size and mixing state by microphysical processes are calculated for all the 47 bins, and bin shifting by these processes is calculated by a two moment (mass and number) advection scheme (Simmel and Wurzler, 2006) for particle size bins and the moving center approach (Jacobson, 1997) for mixing state bins (Matsui, 2017). We have added these sentences to the revised manuscript (Lines 98-105).

Editor's comment:

- The calculation of radiative properties as "calculated theoretically (Bohren and Huffman, 1998;" is not sufficient. What theories are used and what are the actual references (noting that Bohren and Huffman is a full textbook not a specific reference)? How is mixing represented (effective medium approximations, core/shell,...) and what are the related uncertainties?

Response:

For optical properties, we assumed the core/shell treatment for internally mixed BC in the fine particle size bins (40−1250 nm in diameter) and the well-mixed treatment for the other particles (for pure BC and BC-free particles in the fine particle size bins and for all particles larger than 1250 nm or smaller than 40 nm) (Matsui, 2017). CAM-ATRAS uses look-up tables of optical parameters (extinction coefficient, single scattering albedo, and asymmetry factor) calculated based on the codes for homogeneous and coated spheres (Appendices A and B in Bohren and Huffman (1998)). The core/shell treatment could underestimate the mass absorption cross section of BC (MAC_{BC}) for large particles (Forestieri et al., 2018), but as shown by Matsui et al. (2018b), the enhancement of BC absorption by the core/shell treatment is comparable to that by other mixing state assumptions such as the dynamic effective medium approximation (Chylek et al., 1984;

Jacobson, 2006) and the Bruggeman mixing rule (Jacobson, 2006). We have clarified these points in the revised manuscript (Lines 108-116).

Editor's comment:

- And finally, the representation of the mixing state in microphysical models tends to be heavily affected by assumptions made during emissions (which often dominate over the actual microphysical processes). While some BC sources emit fairly pure particles, other sources have already a high degree of internal mixing. How is the mixing state dealt with at point of emission (or of formation of secondary organics)?

Response:

Given the large uncertainty in the assumption of BC mixing states in emissions (Matsui, 2020), we assumed BC is emitted as pure BC and the other species as BC-free particles (Matsui et al., 2018b). In reality, the mixing state of emitted BC particles depends on the types of sources. Matsui et al. (2018b) made a simulation assuming that 50% of BC mass is emitted as pure BC and the other 50% of BC as internally mixed BC with the shell (organic aerosol) to core (BC) diameter ratio of 1.1 for fossil fuel sources and 1.4 for biofuel and biomass burning sources and showed that global mean RE_{BC_TOA} in this simulation is about 10% larger than that in the simulation assuming pure BC for all BC emissions. We have clarified these points in the revised manuscript (Lines 186-191).

Response to reviewer #1

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Reviewer’s comment:

The authors calculated a larger MAC for the BC from the Asian region, suggesting that it is due to its 30-day long transport and ageing process that causes the thickest coating of its BC.

Here I would argue that even if the coating of BC from Asian sources is thick, it does not necessarily mean that it is due to ageing and mixing in transport. It is also likely that the high concentration of condensable gas from Asian emissions would result in a thicker coating a few hours after BC is emitted.

Furthermore, the authors argue that the thicker BC coating observed in the Arctic shows the contribution of aerosol ageing in transport. However, this does not indicate that this observed thick-coated BC is of Asian origin. Other sources with shorter transport times can also contribute to thickly coated BC particles. After too long of a transport time, for example 30 days as estimated by the authors, coating thickness may decrease with photolysis. The authors cannot prove that coating is consistently increasing or not decreasing over such a long transport time. While this may be a common problem in the current models, I would propose to add a few sentences of discussion to clarify this possible uncertainty.

Response:

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of condensable gases emitted with BC are high, and by the higher fraction of anthropogenic BC from Asia in the upper troposphere in the Arctic (Fig. S7) and its longer lifetime in the Arctic (24−30 days) (Fig. 12b)." (Lines 365-371).

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