

We thank the referee for the comments and suggestions to substantially improve our manuscript. The Point by Point Clarifications to referees comments and suggestions are as follows;

Response to comments of anonymous Referee #1

[C1] The authors present a study using the SPRINTARS global aerosol transport model, investigating three different treatments of the aging of black carbon from a water insoluble to a water soluble state. It is well known that current aerosol models vary greatly in their predictions for the global direct radiative forcing from BC, even when using harmonized emissions. Lifetime treatment is thought to be among the reasons, but its impact is not well quantified. The present paper is therefore a welcome addition to the literature, and should be published in ACP after minor revisions.

[A1] Thank you very much for reading our manuscript and giving us useful comments for improving the manuscript.

General comments:

[C2] I have two main comments which relate to making the results in the paper easier to compare to other models: Firstly, in section 2.2.1 the variable BC aging method is presented and a good set of constants for the methodology are given. However the authors do not present the actual lifetimes that the method yields, other than stating that there's a maximum value of 20 days. Without the SPRINTARS SO4 field it is hard to estimate these values. Arguably the results that follow on are more important, but if another model was to try the same method a comparison of the actual lifetimes would certainly be interesting – either as a pdf of as an annual mean map?

[A2] We feel that your suggestion is really important, so that we have made an independent section as 3.1 in the revised manuscript to show results of the conversion time (τ_{bc}) calculated by the AGV method. The global map of simulated τ_{bc} was illustrated in Figure 2 in the revised manuscript. Over the United State and Europe, the

τ_{BC} values range from 0.2 to 1 day, and the τ_{BC} values are less than 3 days over China and India because of the large emissions of SO_2 from anthropogenic activities. In biomass burning regions such as South America and Central Africa, the τ_{BC} value tends to be much higher, with a range of up to 20 days, because the BC concentration from biomass burning is much larger compared with the amount of condensed gases, i.e., sulfuric acid in the present study. If the present study considers other volatile gases, such as organic gases and nitric acids, as condensed matter on the BC particles, the τ_{BC} value is smaller (Oshima and Koike, 2013). In remote areas, the τ_{BC} value ranges from 3 to 5 days, especially in polar regions, where the τ_{BC} value is much higher, with a range of up to 20 days, because the solar incident flux and SO_2 concentration from anthropogenic activities and biogenic sources are generally low. However, we cannot know what is a realistic value of τ_{BC} over polar regions because of the lack of observations, but we can at least safely state that the conversion time, depending on actinic radiation, over polar regions tends to be large and that its value will be still high even if we explicitly consider another aging process of BC, such as coagulation, because the total aerosol concentration is low.

By the way, we have changed the word ‘conversion rate’ in the manuscript into ‘*e*-folding conversion time’ in the revised manuscript. Apparently, the inversion of the *e*-folding conversion time represents conversion rate.

[C3] Secondly, BC ARF normalized by either AOT or burden are very useful measures of the impact of BC in a given model. The authors present these in section 4, but especially for what they term β_s (BC ARF per change in column burden) I don't quite understand the numbers. The unit for this value should be W/g, and is commonly of the order 1500- 2000 W/g on global, annual average. The authors give no units here, and in figure 12a numbers between -0.1 and 0.7 are shown. Can the BC-ARF over Europe, relative to a no-aerosol run, really be negative for the AGF method as shown here? I probably misunderstand what the authors are actually presenting, but this is an indication that the presentations should be clarified and proper units given.

[A3] First of all, we have redesigned experiments to clarify our statements that we estimated BC radiative forcings caused by differences in the treatment of BC aging process, as suggested by the reviewer #2. We have changed the experimental conditions in the ORIG method to minimize differences in the settings among the experiments in the present study. In the revised manuscript, we executed the ORIG method under the same condition of the particle size distribution of WIBC and WSBC, refractive index of BC, and mass density of BC, as those used in the AGV and AGF methods. In addition, we found that the ARF estimation in the manuscript was included some small but unexpected cloud feedbacks even though we used a nudging technique of the meteorological fields. Therefore, we have re-calculated the BC-ARF as a difference in net radiative fluxes with and without BC compounds within one simulation, that means that twice radiative transfer calculation with/without BC compounds in the one simulation have been executed, as in the method of Takemura et al. (2009). As a result, the results obtained by the ORIG method in the revised manuscript are very different from those obtained by the ORIG method in the manuscript, whereas the results obtained by the AGV and AGF methods in the revised manuscript are generally comparable to those obtained by the AGV and AGF methods in the manuscript. Therefore, we have modified most of the figures in the revised manuscript. The ARF values at the tropopause have been changed from +0.300 to +0.311 Wm^{-2} in the AGV method, from +0.047 to +0.111 Wm^{-2} in the AGF method, and from +0.356 to 0.526 Wm^{-2} in the ORIG method. Therefore, possible differences in the treatment of the BC aging process between aerosol modeling studies produce a difference of approximately 0.4 Wm^{-2} in the magnitude of BC-ARF, which is larger than the uncertainty suggested by results from a global aerosol modeling intercomparison project, AeroCom, as mentioned in the revised manuscript. At the same time, the all of BC-ARF values at various regions in the revised manuscript are positive, so we have modified Figure 11 and 12 in the manuscript. Although the results showed in the figures have been largely modified, our conclusion from this figure is not so changed. With regards to Figure 12 in the manuscript (or Figure 13 of the revised manuscript), we have showed the results only at the tropopause, because the main conclusion from the results at the surface is similar to that obtained at the tropopause.

Second, in Figure 13 in the revised manuscript, the relation between β_n and β_s obtained by the AGF and ORIG methods are approximately linear. A slope of the linear

line represents $\Delta\text{AOT}/\Delta\text{BC}$, defined as a ratio of a change in the AOT to a change in the BC column burden, which in the AGF method is lower than that obtained by the ORIG method. This slope is used as an averaged mass extinction coefficient (MEC) of total BC (e.g., Myhre et al., 2013). Therefore, we have modified/added some comments about the results of the AGV method and have led to conclusions in the revised manuscript; The results indicate that the presence of the attached compounds onto the WSBC causes the differences in the averaged MEC of the total BC. At the same time, these results show that the BC aging process used in the AGV method can only produce the large variability in the MEC of the total BC over the BC source regions. Please read the revised manuscript in detail.

Third, although we find the differences in the MEC of the total BC among the experiments and the regions, the values of β_s and β_n obtained in the present study tend to be smaller than those estimated by previous studies. With respect to β_s in the present study, for example, the values obtained by the AGV and AGF methods, within the range of less than 700 Wg^{-1} , tend to be smaller than those by the previous studies of the AeroCom results (Schulz et al., 2006; Myhre et al., 2013), with a range of 500-2000 Wg^{-1} . This tendency of our model was also shown in the previous study of Goto et al. (2011a), which focused on sulfate aerosols and their radiative forcings. However, the values, especially for absorbing aerosols, in global climate models are highly influenced by cloud fraction, which is very uncertain among the models, as suggested by Stier et al. (2013). Therefore, even though both the values of the averaged MEC of the total BC and the values of β_n and β_s obtained by the present study could be different from those obtained by other studies, we can lead to the same conclusion that the way in which mixtures of BC and other compounds are accounted for has a significant impact on the normalized ARF, depending on whether regions are controlled by biomass burning and/or by industrial areas. The above comments have been inserted to the revised manuscript.

Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevag, A., Lamarque, J.-F., Lin, G., Liu, X., Lund, M. T., Luo, G., Ma, X., van Noije, T., Penner, J. E., Rasch, P. J., Ruiz, A., Seland, O., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu,

H., Yu, F., Yoon, J.-H., Zhang, K., Zhang, H., and Zhou, C.: Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, *Atmos. Chem. Phys.*, 13, 1853-1877, 2013.

Minor/technical comments:

[C4] P29802, line 3: particle -> particles

[C5] P29802, line 7: one of main -> one of the main

[C6] P29802, line 8: an impact -> the impact

[C7] P29803, line 4: thus have -> thus has

[A4-A7] Thank you very much. We have corrected as you suggested.

[C8] P29807, line12-13: You claim that 11.4Tg/y emissions are higher than the aerocom phase 1 mean of 11.9 Tg/y. This doesn't add up – please reformulate to make clear what you mean.

[A8] The total BC emission used in the present study is estimated to be 14.0 Tg/yr including 11.4 Tg/yr of non-biomass burning and 2.6 Tg/yr of biomass burning. So we have modified this sentence as follows; “As a result, the global mean BC emission amount from anthropogenic sources *including* biomass burning is estimated to be 14.0 Tgyr⁻¹, which is higher than the mean of 11.9±1.4 Tgyr⁻¹ used in the AeroCom Phase I model (Textor et al., 2006), 7.9 Tgyr⁻¹ used in the AeroCom Phase II model (Diehl et al., 2012), and the mean of 4.4 Tgyr⁻¹ used in Bond et al. (2007).”

Diehl, T., Heil, A., Chin, M., Pan, X., Streets, D., Schulz, M., and Kinne, S.: Anthropogenic, biomass burning, and volcano emissions of black carbon, organic carbon, and SO₂ from 1980 to 2010 for hindcast model experiments, *Atmos. Chem. Phys. Discuss.*, 12, 24895-24954, 2012.

[C9] P29807, line 20: “The global mean BC emission amount is estimated to be 2.6Tg/y. . .” You just stated it was 11.4Tg/y. Do you mean biomass emissions here,

as discussed above? Also, I wasn't able to locate this number in Bond et al. (2007), but this may be because I don't quite see what you're describing. Please clarify.

[A9] This is our mistake. The referred literature is not "Bond et al. (2007)" but "Diehl et al. (2012)", which presents emission inventories including BC during 1980-2010 as a standard emission used in AeroCom Phase II project.

[C10] P29814, line 11: "These models can also calculate the radiative forcing under clear- sky and all-sky conditions at any vertical levels". A linguistic point: RF is defined as a change in net top-of-atmosphere fluxes, or similarly at the surface. The RF at a given vertical level isn't really meaningful. Maybe change to "exerted at any vertical level"?

[A10] Yes, you are right. We have modified it as follows; "In the model, the radiative forcing under clear- sky and all-sky conditions are exerted at any vertical level"

[C11] P29815, line 14: "the higher BC emissions" Are they higher? See comments above.

[A11] Yes, the BC emissions used in the present study are higher than those used in the AeroCom project. Please see our answer to comment C8.

[C12] P29818, line 25: This description of sulfate treatment, sizes, internal/external mixing etc. seems like it belongs in Methods.

[A12] Thanks for your suggestion. We have moved this statement to section 2 in the revised manuscript.

[C13] P29819, line 27: Second ref. is not in References.

[A13] We have modified it to Samset and Myhre (2011) and added it to the references as you suggested.

Samset, B. H., and Myhre G.: Vertical dependence of black carbon, sulphate and biomass burning aerosol radiative forcing, *Geophys. Res. Lett.*, 38, L24802, doi:10.1029/2011GL049697, 2011.

[C14] P29822, line 27: 80-90% (AGV) stated twice, missing AGF?

[A14] Our mistake. We have corrected it.

[C15] P29823, line 15: “could be underestimated”. Why under and not over? Please give the reasoning.

[A15] We showed the ARF values obtained by the AGF method were smaller than those obtained by the AGV method. As we mentioned in the revised manuscript, the differences in the ARF between the AGV and AGF methods are mainly attributed to the sulfate compounds attached to WSBC particles, because the AGF method ignores the condensed compounds in WSBC particles. We think that the results obtained by the AGV method are more reliable in the present study than those obtained by the AGF method. In addition, the BC column burdens obtained by both the AGV and AGF methods are not so different; therefore, the normalized ARF values obtained by the AGF method are smaller than those by the AGV method, as shown in Figure 12 in the manuscript (Figure 13 in the revised manuscript). Therefore, we mentioned that, the magnitude of the normalized forcing efficiencies obtained by the AGF method, which is widely used in most global climate models, could be underestimated.

[C16] P29836: “where Si represents. . .” Variables Si and Oi are not used in the table.

[C17] P29845: “. . .with a random var.” Please either explain or remove this statement.

[C18] P29848: Figure is missing (a), (b) etc. which are referenced in the caption.

[C19] P29849: As commented above, please add units to this plot.

[A16-A19] Thank you very much for correcting our mistake. We have changed them as you suggested.