

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

Response to comments of anonymous Referee #2

In this study, the authors examined how different treatments of BC aging process may affect BC spatial distribution, hygroscopicity and radiative forcing in a global aerosol model. This could be an interesting addition to the literature, as there are still large uncertainty in the treatment of BC aging process. However, I found that the experiments were not well designed, and the results presented also needs further clarification. I therefore can not recommend the publication in its current form.

Thank you very much for your suggestion. We have redesigned experiments to clarify our statements that we estimated BC radiative forcings caused by differences in the treatment of BC aging process. Especially, we have changed the experimental conditions used in the ORIG method to minimize the differences in the settings among the experiments in the present study. Also we have added some explanation to help the readers to understand the results in our study. Please see the following answer to your comments.

General comments:

1. Experiment design: The goal of this study is to examine how different aging treatment affects BC and its radiative forcing. However, in three numerical experiments, there are many differences other than differences on aging treatment. For example, size distributions are different (the ones used in AGV and AGF are different from the ones used in ORIG). Refractive index of BC is also different in different experiments. Another difference is the mixing state. In AGV, BC and sulfate can be internally mixed, while in ORIG, BC and OC can be internally mixed, while in AGF, BC does not mix with either OC or sulfate. What is more, the initial mixing states are also different. In the AGV and AGF, the WIBC and WSBC compounds constituted 80 and 20% of the total BC emission in all sources,

while in ORIG, the assumption on this are different (see lines 1-5, page 29811). Though these additional differences can be interesting by themselves, these make it difficult to interpret the results of the three experiments in the study. It is not clear to me whether the large differences in forcing comes from differences in aging treatment or differences in size distribution, refractive index, or different mixing state with sulfate or OC. I would suggest the authors to eliminate these additional differences and rerun their experiments.

Thank you for your comments. First, differences between the AGV and AGF methods are two points; (1) BC conversion time and (2) mixing state of water-soluble BC (WSBC) particles with/without attached compounds. Because only the BC aging process in the AGV method can simultaneously determine both the conversion time and the amount of attached compounds onto WSBC particles, in the present study we use the term 'BC aging process', which can directly affect (1) BC conversion time and (2) mixing state of water-soluble BC particles. Therefore, we can say that the differences in the BC distribution and radiative forcings between the AGV and AGF methods are attributed to the differences in the 'BC aging process'. Second, in the manuscript, differences between the AGV and ORIG methods are (1) BC conversion time and (2) compounds attached to WSBC particles, (3) emission ratio of WIBC and WSBC, (4) size distribution of WSBC and WIBC, (5) refractive index of BC, and (6) mass density of BC. As the reviewer suggested, the differences make it complex to analyze their contributions to the differences in the ARF among the experiments. Therefore, in the revised manuscript we have modified the ORIG method, which is still based on the original SPRINTARS but executed under the same condition of (4), (5), and (6) as those used in the AGV and AGF methods. In the original SPRINTARS, the WIBC particles cannot be aged in the atmosphere. Instead of them, the original SPRINTARS assume that most BC particles are internally mixed with OC within one GCM time step and one grid as mentioned in section 2.2.2 in the manuscript. Therefore, the terms of (1), (2), and (3) are one connected condition, which can represent the original SPRINTARS. These new experiments are summarized at the revised Table 1, which will be much easier for readers to understand the whole figures of the present study than before. In addition, we have revised all the figures related to results by the ORIG method.

The estimation of the ARF value has also been modified in the revised manuscript. In addition to the change in the experimental design, 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 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.

To more clarify the difference in the ARF among the experiments, especially because the radiative impact of the compounds attached to WSBC particles on the ARF value is expected to be large, we have added two new experiments (named as AGV_EXT and ORIG_EXT shown in Table 4 in the revised manuscript due to the assumption of external mixture of BC and sulfate or OC in the estimation of the radiative forcing), which are conducted under the same condition as the AGV (or ORIG) method but without taking account for sulfate (or OC) compounds attached to the WSBC particles in the radiative transfer model. The results of the estimated ARF values were summarized in Table 4 in the revised manuscript. At the tropopause, the annual and global mean BC-ARF values under all-sky conditions is estimated to be +0.311 Wm^{-2} (AGV), +0.111 Wm^{-2} (AGF), +0.140 Wm^{-2} (AGV_EXT), +0.526 Wm^{-2} (ORIG), +0.152 Wm^{-2} (ORIG_EXT), and +0.356 Wm^{-2} (ORIG_REF), respectively. As a reference, we calculated the results obtained by the 'ORIG_REF', which represents the ORIG method using the physical and optical properties used in the original SPRINTARS. Compared with the original SPRINTARS, the BC-ARF value in the ORIG method increases by +0.17 Wm^{-2} due to the differences in the size distributions and refractive index of BC. The difference in the BC aging process between the AGV and AGF methods causes both a difference in the ARF values between the AGV_EXT

and AGV methods, with a value of 0.17 Wm^{-2} , and a difference in the ARF values between the AGV_EXT and AGF methods, with a value of 0.03 Wm^{-2} . The former values are derived from the difference in the conversion time of WIBC particles between the AGV and AGF methods, whereas the latter values are derived from the difference in the sulfate compounds attached to WSBC particles within the radiative transfer calculation. With regard to the ORIG method, the difference between the ORIG and ORIG_EXT methods causes both a difference in the ARF values, with a value of 0.38 Wm^{-2} , because of the difference in the OC compounds attached to WSBC particles within the radiative transfer calculation.

These comments have been added to section 4 in the revised manuscript. Therefore, in section 4 in the manuscript, we often mentioned that the way in which mixtures of BC and other compounds are accounted for has a significant impact on the ARF value.

2. The forcing efficiency calculation is quite interesting. However, more clarification is needed, especially in terms of changes in AOT (Figure 11) (see specific comments #22, 23, 24).

Thank you very much for your interest. We have answered to your comments #21-24 in detail. In addition, we have modified the comments about the results of the forcing efficiencies due to the revision of the ORIG method and correction of the ARF calculation in the experiments, as mentioned in our answer to your general comment #1. Although the results showed in Figure 12 in the manuscript (or Figure 13 of the revised manuscript) have been largely modified, our conclusion from this figure is not so changed. The details are described in the revised manuscript.

Specific comments:

1. p. 29804, lines 12-14: I would argue that, to date, not “some” but many global models take into account the internal mixing and related processes. Listed references are incomplete.

As you suggested, we have added several references (Adams et al., 2002; Easter et al., 2004; Seland et al., 2008; Aquila et al., 2011; Liu et al., 2005; 2012) to the revised manuscript. If we look at chemical transport models, GLOMAP (Spracklen et al., 2005) and GEOS-Chem-APM (Yu and Guo, 2009) can be also included. However, we feel most of global models with predicting aerosols are still simplified in terms of aerosol dynamic processes. According to international intercomparison of aerosol transport models such as the AeroCom, ACCMIP, and CMIP5 projects, more than half of the models in the AeroCom and most models in ACCMIP and CMIP5 do not treat such complex processes. Therefore, we used not “many” but “some” in the manuscript.

- (1) Adams, P. J., and Seinfeld, J. H.: Predicting Global Aerosol Size Distributions in General Circulation Models, *J. Geophys. Res.*, 107(D19), 4370, doi:10.1029/2001JD001010 (2002)
- (2) Aquila, V., Hendricks, J., Lauer, A., Riemer, N., Vogel, H., Baumgardner, D., Minikin, A., Petzold, A., Schwarz, J. P., Spackman, J. R., Weinzierl, B., Righi, M., and Dall’Amico, M.: MADE-in: a new aerosol microphysics submodel for global simulation of insoluble particles and their mixing state, *Geosci. Model Dev.*, 4, 325–355, doi:10.5194/gmd-4-325-2011 (2011)
- (3) Easter R. C., Ghan, S. J., Zhang, Y., Saylor, R. D., Chapman, E. G., Laulainen, N. S., Abdul-Razzak, H., Lenug, L. R., Bian, X., and Zaveri, R. A.: MIRAGE: Model Description and Evaluation of Aerosols and Trace Gases, *J. Geophys. Res.*, 109, D20210, doi:10.1029/2004JD004571 (2004)
- (4) Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J.-F., Gettelman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M. J., Bretherton, C. S., Flanner, M. G., and Mitchell, D.: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, *Geosci. Model Dev.*, 5, 709–739, doi:10.5194/gmd-5-709-2012 (2012)
- (5) Liu, X. H., Penner, J. E., and Herzog, M.: Global Modeling of Aerosol Dynamics: Model Description, Evaluation, and Interactions between Sulfate and Nonsulfate Aerosols, *J. Geophys. Res.*, 110, D18026, doi:10.1029/2004JD005674 (2005)
- (6) Seland, Ø., Iversen, T., Kiekevåg, A. and Storelvmo, T.: Aerocol-Climate Interactions in the CAM-Oslo Atmospheric GCM and Investigation of Associated

- Basic Shortcomings, *Tellus*, 60A, 459-491 (2008)
- (7) Spracklen D. V., K J. Pringle, K. S. Carslaw, M. P. Chipperfield, and Mann, G. W.: A Global Off-line Model of Size-Resolved Aerosol Microphysics: I. Model development and prediction of aerosol properties, *Atmos. Chem. Phys.*, 5, 2227-2252 (2005)
- (8) Yu, F. and Luo, G.: Simulation of Particle Size Distribution with a Global Aerosol Model: Contribution of Nucleation to Aerosol and CCN Number Concentrations, *Atmos. Chem. Phys.*, 9, 7691-7710 (2009)

2. p. 29805, Croft et al. (2005): can you highlight the main conclusion of Croft et al. (2005) here? Also, it will be good to refer back to these two literatures when discussing BC burden changes from different aging treatment Section 3.

Thank you for your suggestion. We have added the important message of Croft et al. (2005) to section 1 in the revised manuscript. Croft et al. (2005) examined four separated parameterizations of the BC aging process and concluded that (1) the traditional parameterization using a fixed τ_{BC} was shown to reasonably represent BC column burdens in a global scale and (2) that the global and annual mean τ_{BC} of insoluble BC to soluble BC is less than the 24 hours. In addition, as you suggested, we have inserted new small discussion of BC mass distribution over remote areas using these literatures (Croft et al., 2005, Vignati et al., 2010 and Liu et al., 2011) to section 3 in the revised manuscript as follows; “In previous works, Vignati et al. (2010) and Liu et al. (2011), which mainly improved removal processes of BC, successfully simulated BC mass concentrations over the Arctic, although Croft et al. (2005) did not simulate them well. Therefore, the BC removal process could be an important key to reasonably simulate BC mass concentrations especially over the Arctic.”

3. P. 29808, line 16: what is the fixed tau_bc for “AGF”?

We set the value to be 1.2 days in the present study and add this statement to section 2 in the revised manuscript.

4. P. 29810, lines 3-17: It is not clear to me why size distribution and density are updated in AGV and AGF case. I would think this introduced additional complexity and make it difficult to interpret the differences among different experiments. Also see general comments.

As you suggested, we have redesigned the experiments. Please see our answer of your general comment.

5. P. 29810, line 12: the hygroscopicity of WSBC was the same as that of pure sulfate. Is this for all three experiments? This seems not a good assumption. How about sulfate predicted from Eq. (6)? Is there any difference between AGF and AGV on this?

Yes, we set the hygroscopicity of WSBC particles to be in pure sulfate. In terms of this point, we have three main assumptions. First, BC component in the WSBC particle is fully coated by other compounds, i.e., sulfate in the AGV method and unspecific component in the AGF method. Second, the BC component cannot be dissolved into sulfate. We think the second assumption is generally allowed, so that we ignore any change of the hygroscopicity through internally mixing. Third, we assume the hygroscopicity of WSBC in the AGV and the AGF methods are equal to each other, because we cannot define the component of WSBC in the AGF method. Therefore, we assume the hygroscopicity of WSBC is the same as that of pure sulfate in the present study.

6. P. 29812, lines 8-29: It is not clear to me how extinction coefficients and absorption coefficients are calculated in different cases. Here are several questions: i) what is the unit of extinction coefficient and absorption coefficients, /gram BC or /gram aerosols? ii) how are these calculated for WSBC? Will these depend on how much sulfate predicted by the model? Eq. (6) suggested that sulfate

can be different for different situations. In AGV, BC is internally mixed with sulfate, while in AGF, BC does not internally mix with sulfate. I would expect this will affect extinction/absorption coefficients in these two cases?

The answer to your first question is that the unit of the coefficients is ‘per gram aerosols’. We have added them to caption in Figure 1 in the revised manuscript. With regard to your second question, we assume the extinction and absorption coefficients in WSBC particles in the AGV and AGF methods are equal to each other. As we mentioned in section 2.2.1, in the present study we assume that the radius and the hygroscopicity of WSBC particles are common in both AGV and AGF methods with ignoring any change of the particle size through the BC aging process. Surely the equation (6) suggests a change in the sulfate components attached to WSBC particles, in the present study the particle size of shell in WSBC particles is constant. Furthermore, although in the AGF method WSBC particles internally mix with unspecific compounds, which cannot be explicitly defined in the model, we assume that the physical property of the WSBC in AGF method is the same as that of the WSBC in the AGV method. We believe this matter is generally used in GCM using the AGF method. Therefore, the extinction and absorption coefficients in WSBC particles in both the AGV and AGF methods are calculated using the same particle size distribution, particle density, refractive index, and hygroscopicity.

7. P. 29812, line 23: again, difference in refractive index introduces additional complexity.

As you suggested, we have redesigned the experiments. Please see our answer of your general comment.

8. P. 29813, lines 8-10: It is not clear to me how the model treats the internal mixing of BC and sulfate in AGV and the internal mixing of BC and OC in ORIG. Section 2.2.1 seems to suggest that WSBC has the same hygroscopicity as pure sulfate.

As you suggested, we have redesigned the experiments and modified Table 1, which will be easier for readers to understand the experimental conditions before.

9. Section 3.1: I agreed with referee #1 that it will be helpful to show the actual tau_bc generated from AGV method.

We have added a global map of tau_bc calculated by the AGV method as Figure 1 in the revised manuscript. From this figure, we have found that 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 SO₂ 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 SO₂ 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.

10. Section 3.1: a table to compare BC global burden, lifetime, wet deposition rate, and dry deposition rate can be very helpful for readers to understand how three

different schemes perform.

We have added a new table to show the global budget fluxes of WIBC, WSBC and BC in the experiments as Table 3 in the revised manuscript. In the AGV and AGF methods, most WSBCs are formed from the aging process of WIBC in the atmosphere, with amounts of 8.7 (AGV) and 9.5 Tgyr⁻¹ (AGF). This result means that 78% (AGV) and 85% (AGF) of WIBC are converted into WSBC before they are removed by wet and dry deposition. The lifetime of WIBC, as defined by the ratio of the column burden of WIBC to the emission flux of WIBC, is estimated to be 1.7 (AGV) and 1.0 days (AGF), respectively, whereas it is estimated to be 9.6 days (ORIG) because of the lack of conversion of WIBC to WSBC in the atmosphere. As for WSBC, the lifetime is estimated to be 6.2 (AGV), 6.1 (AGF), and 5.7 days (ORIG). To include these results as part of the total BC budget, we also estimate the lifetime of total BC to be 5.4 (AGV), 5.6 (AGF), and 4.6 days (ORIG), which are smaller than the averaged values reported in the AeroCom study by Textor et al. (2006), who indicated average values of 7.12±2.35 days. With regard to the budget of the deposition, the ratio of the budget of the wet deposition to those of the total deposition in the AGV method is estimated to be 81%, which is comparable to the values of 80-81% obtained by the AGF and ORIG methods and the averaged values of 78% reported in the AeroCom study by Textor et al. (2006).

11. Figure 2: which aging method is used in model results in Figure 2?

Sorry for missing the name of the method. This result corresponds to that in the AGV method. As we mentioned in the manuscript, a difference in surface BC mass concentration among the experiments was so small that we only showed one result as Figure 2 (Figure 3 of the revised manuscript).

12. P. 29814, lines 11-28: Here the paper presented the differences in sulfate BC mass concentrations among different treatments. But what is missing is the explanation what might cause the differences.

As the reviewer suggested, we have inserted an explanation of the differences in the surface BC mass concentrations in the revised manuscript. At first, although over the industrial regions the differences in the BC distribution among the experiments are within 10%, those are up to 50% over remote areas. As shown in Figure 2 in the revised manuscript (related to our answer A9), the τ_{bc} value in the AGV methods tends to be higher over the remote areas compared with that in the AGF method. Therefore, the water-insoluble BC (WIBC) in the AGV method is less fully converted to water-soluble BC (WSBC) and is therefore less scavenged by precipitation compared with that in the AGF method. Ultimately, the surface BC mass concentrations in the AGV method are larger than those in the AGF method. With respect to the difference between the AGV and ORIG methods, however, over the remote areas the surface BC mass concentrations in the AGV method are lower than those in the ORIG method by more than 50%, mainly because in the ORIG method, almost all BC-containing particles over these areas are suspended as WIBC particles, which are relatively difficult to remove from the atmosphere (shown Figure 8 in the revised manuscript). In contrast, over the West African coast, where biomass burning affects the air quality, the BC mass concentration simulated by the AGV method is larger than the concentrations simulated by the ORIG method by up to 50%. The reason is that almost all BC in the ORIG method is emitted from biomass burning as WSBC particles, which are easily removed by wet deposition and are not increased through the BC aging process, whereas in the AGV method BC particles are both in WIBC and WSBC. These explanations have been added in the revised manuscript.

13. P. 29815, line 7: project

Thanks for your correction.

14. Figures 5 and 6: again, differences among different experiments warrant further explanations.

We have added some explanations in the revised manuscript, as mentioned in the

answer of C12. The differences in BC vertical distribution and BC column burden are caused by the difference in the conversion time of WIBC and in the scavenging efficiency of WIBC and WSBC. The conversion time of WIBC in the AGV method depends on regions, so that the results were different. More details were described in the revised manuscript.

With regard to Figure 5 of the annual and zonal mean BC mass concentrations in the manuscript, we did not show them in an annual mean due to our mistakes. So this figure (Figure 6 in the revised manuscript) has been revised and the results are consistent to those shown in the figure of annually averaged surface BC mass concentration (Figure 4 in the manuscript or Figure 5 in the revised manuscript). With regard to Figure 6b and 6c of the BC column burden in the manuscript, we have revised them to show absolute difference among the experiments near the aerosol source where the difference could be more important than that over the remote areas.

15. P. 29816, line 14: become smaller → become larger?

Thanks for your correction. Yes. You're right.

16. Figure 7a: the AGV method produces increasing WIBC ratio over the Arctic (up to 70%). Is this feature realistic? I would expect WIBC ratio over the Arctic is similar to those over the remote oceans.

In the AGV method, the conversion time in the BC aging process depends on both BC amount and sulfuric acid formed from the oxidation of SO₂ by hydroxyl radical, which is produced by actinic solar flux. Therefore, the BC aging process does not occur in nighttime and the midnight sun in wintertime polar regions. In addition, in polar regions, the solar incident flux is lower than that in usual remote areas over the tropical ocean. In conclusion, because of slow aging time caused by sulfuric acid in the AGV method, over the Arctic the WIBC ratios obtained by the AGV method tend to be larger compared with those obtained by the AGF method. However, we cannot know what is a realistic value of the conversion time and WIBC ratios over polar regions because of the

lack of observations.

17. Figure 9: And why AOD in ORIG is smaller by more than 5% than that in AGV over many regions?

First, the results of the ORIG method in the revised manuscript are different from those of the ORIG method shown in the manuscript. Second, we have modified the figure to show absolute (NOT relative) differences in AOT as well as AAOT in the revised figures in order to outstand the difference among the experiments near the aerosol sources. In addition, the differences in the AOT among the experiments in the revised manuscript are larger than those shown in the manuscript, because of an increase in the AOT values in the ORIG method. Third, we have inserted an explanation of the difference in the AOT among the experiments, because in the manuscript we did not explain them at all. As a result, the corresponding figure has been presented as Figure 10 in the revised manuscript.

Originally, these differences in the AOT among the experiments are explained by the difference in the BC column burden in Figure 7 in the revised manuscript, the difference in the attached compound onto the WSBC particles, and to a small extent, the difference in the WIBC ratio in Figure 8 in the revised manuscript. The negative values of the difference in the AOT value between the AGF and AGV methods in Figure 10b in the revised manuscript are mainly due to the difference in the simulated BC column burden between the AGV and AGF methods shown in Figure 7b of the revised manuscript and due to the sulfate compound attached to the WSBC particles in the AGV method.

In the case of the difference in the AOT between the AGV and ORIG methods, the AOT of the attached compounds in the WSBC particles become important in each region. In the ORIG method, OC compounds attached to WSBC particles largely contribute to the AOT values, especially over biomass burning areas, whereas in the AGV method, sulfate compounds attached to WSBC particles contribute to the AOT values all over the world. In Figure 10c, the difference in the simulated AOT value between the AGV and ORIG methods is calculated to be positive, with values of up to 0.2 over Asia and biomass burning regions. In Asia, both the BC column burden and the

AOT value obtained by the AGV method are smaller than those obtained by the ORIG method. In a large part of the Northern Hemisphere, however, although the BC column burden obtained by the AGV method is smaller than that obtained by the ORIG method, the AOT value in the AGV method is comparable to that in the ORIG method, most likely because in the AGV method, the sulfate compounds on the WSBC particles are distributed largely with higher AOT values compared with those of OC on the WSBC particles in the ORIG method. In contrast, in South America, the BC column burden obtained by the AGV method is larger than that obtained by the ORIG method (Figure 7c), whereas the AOT value obtained by the AGV method is lower than that obtained by the ORIG method (Figure 10c). This observation likely occurs because in the ORIG method over the biomass burning areas the OC compounds attached to WSBC particles have a great contribution to the total AOT value.

For AAOT, differences among the experiments can be explained by the same factors as those described for the difference in the AOT value. The AAOT values simulated by the AGV method are all larger than those simulated by the AGF method, most likely because of the larger BC column burden obtained by the AGV method and because of the sulfate compounds attached to the WSBC particles (Figure 11b). In contrast, in Figure 11c, the AAOT values simulated by the AGV method are generally smaller than those obtained by the ORIG method, with values of more than 50%, as shown in Figure 10c of the AOT comparison.

Above comments and explanations have been inserted to the revised manuscript.

18. Figure 10a: the legend of color map is incorrect.

Thanks for your correction. Figure 10a in the manuscript showed absorption AOT values by multiplying one thousand. We have modified it in the revised manuscript.

19. P. 29818, line 25: Fig. 10b → Fig. 9b?

Thank you for your suggestion, but we have largely modified the manuscript around

here after we have redesigned the experiments. So in the revised manuscript, this sentence has been deleted.

20. P. 29819, lines 25-29: the difference in sulfate explained here again introduces additional complexity

As the reviewer #1 also suggested, we have moved this description of sulfate to section 2 in the revised manuscript.

21. P. 29820, lines 4-10: I am not convinced that you can draw this conclusion here. Again, differences other than aging process may play a role here. Also, I do not think the underestimation is because the compounds mixed with BC is ignored. If you mean the difference between AGF and ORIG, it is clear from Figure 6, that BC burden in ORIG is significantly higher than AGF.

22. Fig. 11: several questions here. i) why BC column burden are quite similar among different aging treatment for these 9 regions, while Figure 6 shows large difference? ii) delta AAOT are quite similar (this is consistent with BC burden change), while delta AOT are quite different among three aging treatment. What might explain this discrepancy? iii) the smaller positive forcing in AGF than in ORIG seems due to larger delta AOT in AGF.

23. P. 29820, lines 22-24: again, I am not convinced by this this conclusion. Fig. 11 suggested to me that the difference in TOA forcing seems due to difference in delta AOT, but not from delta AAOT.

24. P. 29821, lines 11-14: I am not sure I agree with this statement. As I mentioned before, I would think this difference is caused by difference in delta AOT in three experiments, as shown in Figure 11. It is also not clear to me why attached compounds would increase beta n or beta s (noted that delta AAOT are quite similar for three different experiments, as shown in Figure 11).

We think the point in these comments is very close to each other, so we like to answer them all at once.

First, as for the question 22 i) of BC column burden, the results in Figure 9 and those in Figure 11 in the manuscript are actually consistent with each other, even though Figure 9 in the manuscript illustrated the relative difference and Figure 11 in the manuscript illustrated the absolute difference. In Figure 7 (BC column burden) of the revised manuscript, for example, a maximum of the absolute difference in BC column burden among the experiments is estimated to be about 0.2 mg/m^2 , which are also shown over East Asia in Figure 12a of the revised manuscript.

Second, as we mentioned in the answer to your general comment C1, we have revised these figures after we rerun the ORIG method and fixed the estimation of the ARF values. Ultimately, negative values of the ARF in some regions shown in Figure 11 in the manuscript are disappeared. In the revised figure (Figure 12 in the revised manuscript), the differences of both AOT and AAOT among the experiments are large. The reason has been added to the revised manuscript as represented in our answer of your comment C17. This is our answer for your second question 22 ii) of AAOT and AOT.

Third, we like to answer your comment of 21, 22 iii), 23 and 24. As you suggested, the relationship of the different parameters (BC column burden, AOT, AAOT, and ARF) and different experiments (AGV, AGF, and ORIG) was unclear and required some explanation in the manuscript. But after the revised results of them, the relationship shown in Figure 12 of the revised manuscript has been much clear. Although the absolute magnitude of the differences in the ARF has been changed, the ARF values obtained by the AGV method still lie between those obtained by the AGF and ORIG methods. In addition, the global mean ARF values obtained by the experiments in the present study have been shown both in the answer to the general comment C1 and in Table 4 in the revised manuscript. The difference in the ARF value among the experiments can be explained by the same factors as those described for the difference in the AOT and AAOT, as shown in our answer to your comment 17 in this document (Figures 10 and 11 in the revised manuscript), and for the difference in the global mean ARF value among the experiments, as shown in our answer to your general comment 1 in this document (Table 4 in the revised manuscript). The latter explanation has represented the importance of the compounds attached to WSBC particles on the ARF value. Therefore, in section 4 we often mentioned that the way in which mixtures of BC and other compounds are accounted for has a significant impact on the ARF

value.

25. P. 29822, line 27: the second AGV should be AGF?

Yes, thanks for your correction.