

Response to Reviewer #1 (R. Wang):

We are grateful for the constructive comments. Our itemized responses are as follows:

“.... I have not found enough novelty in the present work. In the present manuscript, "using updated Asian anthropogenic BC emissions and global biomass burning emissions" seems to be the only improvement by this work, although it's still questionable that they have treated very well with the emissions. Aging of BC is from a published reference, but there is another study based on the same model but with a more complicated method to consider the aging process. Many important studies have not been discussed. I suggest the authors should explain clearly what have been done in previous studies, and what is being done for the first time in their work.”

We have added discussions to address these issues. Particularly, we added discussions in the introduction to clarify the novelty of the present work. We included additional discussions on BC emissions (Sect. 2.2.1 and Sect. 4) and BC aging (Sect. 2.2.3 and Sect. 5). We also added and discussed necessary references about BC emissions and aging. Additional model experiments using finer model resolution (Sect. 6) and upper and lower bounds of BC emissions (Sect. 3) have been conducted and discussed. More details are presented in the following response to each comment.

Major comments:

1. *“.... However, the surface concentrations and BC AAOD have been evaluated by many previous studies. For example, Fu et al. have evaluated the surface BC concentrations using the same model (Fu, 2012). The evaluation of BC AAOD has been done by Bond et al (2013). So, it's not very clear what is the novel contribution of this work. For aging of BC, there is another study, which is based on the same model but with a more complicated method (Huang, 2013). These important studies have not been discussed in the paper. The BC in snow is likely a new part in this study. However, it's not well documented, and some important relevant information are missed (e.g. where is the major source region for the snow BC over the Tibet Plateau?).”*

Good points. In this study we seek to understand the capability of a global 3-dimensional CTM (GEOS-Chem) in simulating BC in the Tibetan Plateau and the associated discrepancies between model results and observations. To our knowledge, this is the first attempt to systematically evaluate a global simulation of BC in the Tibetan Plateau using all three types of available *in situ* measurements: BC in surface air, BC in snow, and BC AAOD. We have added discussions to the last two paragraphs in the introduction to clarify the novelty and importance of the present work. We cited and discussed Fu et al. (2012) on surface BC simulations (Page 4, Line 12) and BC emissions (Page 8, Line 16). Additional discussions have been added to BC aging parameterization (Sect. 2.2.3 and Sect. 5), including the discussion of Huang et al. (2013) (Page 11, Line 27; Page 20, Line 21). We have added discussions

on BC in snow, particularly the uncertainties associated with its calculation, in the last paragraph of Sect. 2.2.2 (see also the response to Short Comments). The relevant information for BC in snow such as source regions, BC transport, BC deposition and precipitation has been discussed in Sect. 3.2 in our original manuscript. Now we've made modifications to the statements in this section to clarify it.

2. *“The authors are using a global 3-dimensional chemical transport model at a horizontal resolution of 2 degree by 2.5 degree (or close to 200 km). It should be noted such a coarse resolution is difficult to capture the high BC concentrations at local scale, especially for urban sites. There is a recent study which quantified the effects of model resolution on simulating the surface BC concentrations in East Asia and South Asia (Wang, 2014). This spatial scale effect is important when comparing the modeled concentration over a large model grid to the observed concentration at a local site. There is a nested version of GEOS-Chem, which has been used by Fu et al. (2012). The authors should test the effect of using a higher-resolution model in their study.”*

We agree with the reviewer that the spatial scale effect is important to model simulation of BC, particularly over the complex topography such as the Himalayas. We now include an additional model experiment conducted at $0.5^\circ \times 0.667^\circ$ resolution (nested) over Asia. The description for this experiment has been added to Sect. 2.2.4 (Page 13, Line 1) and Table 5. The model results have been discussed in Sect. 6 (Sensitivity to model resolution) and included in Tables 1, 2 and 6 and Figs. 2, 3 and 10. We also cited and discussed Fu et al. (2012) and Wang et al. (2014) (Sect. 6).

3. *“The authors states correctly in the abstract that there are deficiencies in emissions, but they failed to give sufficient discussion and consideration for that. In fact, there are many update of the emission inventory of black carbon in Asia (Lu, 2011; Qin, 2012; Kurokawa, 2013; Wang, 2014). Notably, the spatial pattern of black carbon emission has been improved in these inventories. However, these progresses seem not to be noticed by the present study. Page 7324 (Sensitivity to BC emissions): In Lu's paper, the uncertainty of BC emission has been quantified by a Monte Carlo method. However, this important point also has not been discussed in the paper. Without considering the associated uncertainties, it's unreasonable to conclude whether an inventory underestimates the emissions or not. This uncertainty should be considered in the study and quantified by running the model with the lower and upper bounds of the inventory. This is especially important for BC AAOD, which is now underestimated by a factor of 2-4 in the model. In addition, there is a lack of uncertainty analysis for most discussions in the present paper.”*

Excellent points. We agree with the reviewer that more discussions should be added on the update of Asian BC emission inventories. Thus, we cited and discussed additional references (Qin and Xie, 2012; Kurokawa et al., 2013; Wang et al., 2014) and compared these updated inventories with those used in the present work (Sect.

2.2.1: Page 8, Line 33; Sect. 4: Page 20, Line 12). We've added discussions on the uncertainties of BC emissions estimated by the Monte Carlo method (Lu et al., 2011) in Sect. 2.2.1 (Page 8, Line 29). We have included additional model experiments using the lower and upper bounds of the BC emissions. The model results and the associated uncertainties have been discussed in Sects. 3.1, 3.2 and 3.3, and also included in Tables 1, 2 and 3 and Figs. 2 and 3.

Specific comments:

1. *“Abstract, Line 5: The authors state that they are using “updated Asian anthropogenic BC emissions”. It's not clear which inventory they are updating against?”*

Agreed. We altered *“updated Asian anthropogenic BC emissions”* to *“recent anthropogenic BC emission inventories for Asia”*.

2. *“Abstract, Line 7: The authors state that the model results are in good agreement with observations. However, this is not the case for surface concentrations at urban sites and BC AAOD.”*

Fixed. Now we have the following in the abstract: *“... model results of both BC in surface air and in snow are statistically in good agreement with observations (biases < 15%) away from urban centers.”*

3. *“Page 7312 (AERONET AAOD): The methodology for retrieving the BC AAOD from the AERONET observations is not very clear. First, which version of the AERONET data is used in the study (level 2.0 or 1.5)? Second, the AAOD are observed under conditions (clean-sky, daytime, ...) by AERONET, which are available for only part of the days throughout the year. However, it's not clear how the monthly and annual means are calculated in the model and observation? In fact, it makes senses when comparing the modeled and retrieved BC AAOD at the same days. Third, SSA is only available for days when the AOD is larger than a value. How do you get the SSA for low AOD days?”*

Good points. We've added discussions on retrieving BC AAOD in Sect. 2.1.3 (Page 7, Lines 1 to 9). Specifically, we use monthly averaged AOD data from AERONET (Version 2.0 Level 2.0 products). The monthly means are derived for months when there are five or more days with AOD observations. Our evaluation of modeled BC AAOD is based on monthly averages (Sect. 3.3). This likely results in a larger uncertainty than comparing the modeled and retrieved BC AAOD on a daily basis. In addition, the SSA data at low AOD values is removed from AERONET Version 2 products for data quality assurance. Thus, we did not include the SSA for low AOD days, which likely introduces a positive bias in the AAOD retrieval (Bond et al., 2013).

4. *“Page 7312 (Model description and simulations): Since you are evaluating the modeled BC AAOD, what optical parameters (e.g. mass absorption cross-sections) are used in the model? It should be explained with the associated uncertainty discussed.”*

Agreed. We added discussions on BC optical parameters in Sect. 2.2.4 (Page 13, Line 5).

5. *“Page 7315 (BC aging): The authors state that “in the absence of nucleation and coagulation, the BC aging rate can be parameterized as a linear function of OH concentration.” However, there is no evidence that the nucleation and coagulation are not important in the studied region, especially close to the source regions.”*

Good point. We now provide additional discussions on nucleation and coagulation in the BC aging parameterization in Sect. 2.2.3 (Page 11, Lines 22 to 27).

6. *“Page 7315 (BC aging): In fact, in addition to Liu et al.(2011), there is another recent study of the aging of BC (Huang, 2013). Huang et al. have improved the parameterization for the BC aging in the GEOS-Chem model. According to Huang et al., there should be at least two parts: oxidation effects; condensation-coagulation effects, and the aging rate should be the sum of the two effects.”*

We now cite and discuss Huang et al. (2013) in Sect. 2.2.3 (Page 11, Line 27) and Sect. 5 (Page 20, Line 21).

7. *“Page 7319, line 15: The residual error doesn't make sense for low concentrations (also for Fig. 4).”*

Fixed. Now we have the following in Sect. 3.1 (Page 15, Line 6): *“We note that the residual errors at very low BC concentrations may not be particularly meaningful.”*

8. *“Page 7324 (Sensitivity to BC aging parameterization): In Liu's paper, the new parameterization of aging has a significant impact on the seasonality. Does it also influence the seasonality of surface concentrations and BC AAOD in your study? It should be discussed.”*

Agreed. We've added discussions on the impact of Liu et al. (2011) parameterization on the seasonality of surface BC concentration and AAOD in Sect. 5 (Page 20, Lines 23 to 27).

9. *“Table 6: Units are missed for mean error, mean absolute error, and RMSE.”*

Fixed. Now we have the following in the footnote of Table 6: *“Units for mean error, mean absolute error, RMSE and bias-corrected RMSE are $\mu\text{g m}^{-3}$ for BC in surface air*

and $\mu\text{g kg}^{-1}$ for BC in snow.”

Response to Short Comments (Hans-Werner Jacobi):

Short comments:

“... Since their model does not explicitly simulate the snow cover, the BC in snow concentrations are derived from the ratio between simulated total BC deposition and total precipitation. I believe that this assumption introduces additional uncertainties for the BC in snow concentrations that need to be discussed in more detail as is the case in the current manuscript.

Coarse resolution models like the one used for this study show biases in the simulation of the precipitation. ... Nevertheless, the bias in the total precipitation, which translates directly into a bias for the derived BC in snow concentrations, will probably remain high.

Moreover, the authors use the total precipitation without distinguishing between solid and liquid precipitation. However, rain has a two-fold impact on the BC in snow concentrations: first, rain should be subtracted from the total precipitation before calculating the snow concentrations, and, second, rain can lead to a significant melting of the snowpack further increasing the BC in snow concentrations. ... Finally, the model uses different parameterizations for the BC deposition in the case of rain or snow. Therefore, the model may generate cases when the BC is removed according to wet deposition by rain, while for the calculation of the BC in snow concentration the accumulated rain is then considered as snow.

I believe that these uncertainties in the derived BC in snow concentrations should be discussed in more detail.”

Thank you for the valuable comments. Now we've added discussions on the uncertainties in the calculation of snow BC concentrations in the last paragraph of Sect. 2.2.2, including the bias of precipitation from a coarse model resolution, the bias from the use of total precipitation without distinguishing between snow and rain, and the uncertainty from the effects of rain on snow melting.

Response to Reviewer #2 (Hongyu Liu):

We are grateful for your constructive comments. Our itemized responses are as follows:

Major comments:

1. “Abstract: Improvement is needed. Overall the statements are too general. There are more interesting points and results (in the text) that are worth being included here. Mention GEOS-Chem and the meteorological data set (GEOS-5 DAS) used to drive the model; both are important to the results presented. Avoid citing references in the

abstract. "model results of both surface BC and BC in snow are statistically in good agreement with observations (biases < 15%)" – is this conclusion for the whole region studied? If so, say so. "Model results are in general agreement with observations..." —this is vague."

Agreed. We rewrote the abstract. We added more details to the results presented in the abstract. We specifically mentioned that we use GEOS-Chem model driven by GEOS-5 assimilated meteorological fields. The references in the abstract have been removed. We've made our statements more specific and accurate in the abstract.

2. *"Introduction, p7309: In this section, the readers would be interested in knowing which paper in the literature used the same model (GEOS-Chem) and studied what aspects or properties of black carbon over the Tibetan Plateau or other regions of the world. This is currently lacking in the text. Also lacking is a specific list of what's new in this study, in terms of science questions to answer, approach taken, observational data sets used, and/or application of GEOS-Chem."*

Excellent points. We've added discussions in the last two paragraphs of the introduction. Specifically, we included the references which used GEOS-Chem to study different aspects of BC in different regions. We included additional statements to clarify what is new in this study.

3. *"Summary and conclusions: This section is too brief and appears even shorter than the abstract of this manuscript. The first paragraph of this section needs to state the scientific objectives of the study, what model with what meteorological data set used, observational data sets used, etc. The rest of the section can be organized in terms of BC in surface air (section 3.1), BC in snow (section 3.2), BC AAOD (section 3.3), and sensitivities (sections 4 & 5). Some of the questions raised above for Abstract also apply here. "The retrieved AAOD has a positive bias" — this was not mentioned in previous sections. "... This implies that the modeled BC AAOD probably should be scaled to AERONET observations ..." — I don't understand why "this implies...probably...". In a word, this section needs rewriting and should summarize what's presented in the results sections with some discussions on uncertainties, implications, and recommendations for further research. For the latter, most has actually been discussed in the results sections, but they just need to be briefly summarized here."*

Agreed. We rewrote the summary and conclusions (Sect. 7). We added more details to the first paragraph of this section, such as scientific objectives and information about the model, meteorological fields and observational data used. We re-organized the paragraphs presenting model results and included more details, following the reviewer's comments. We revised the confusing statements in this section and made them more specific and clear.

4. "p.7349 (Fig.5a): It looks like Fig.5a presents the total BC deposition in unit of kg / month / gridbox. If so, correct the unit in the caption (note that "kg/month" indicates gridsizes dependency). Actually, it's more appropriate to plot the total BC deposition in unit of kg/month/area (e.g., kg/month/m²), which would be more useful to those readers who may want to make comparisons."

Good point. We revised Fig. 5a by using a unit of kg month⁻¹ m⁻².

Minor comments:

1. "p.7312, L24: better use "GEOS-5 DAS" here (DAS: data assimilation system). Also see p.7320, L26."

Agreed. We altered "GEOS-5" to "GEOS-5 DAS" in the text.

2. "p.7313, L7 (and elsewhere): cite older references first."

Fixed.

3. "p.7316, L27: mention that 5.5 days is at the lower end of the 5-11 days range. On the other hand, "wet scavenging in the model is too weak" (p.7321). Any comments on the impact of such uncertainty on the results of this study?"

Fixed. Now we have the following in Page 12, Line 17: "The tropospheric lifetime of BC against deposition is 5.5 days, at the lower end of the range (5-11 days) reported by Koch et al. (2009)." We've included additional comments on the weak wet scavenging in the model in Sect. 3.2 (Page 17, Lines 10-16).

4. "p.7318, L1: If these are "urban" sites and a global model is used, do you need to include Fig.2 in the first place?"

Good point. We rearranged Figs 2 and 3 and the paragraphs in Sect. 3.1.

5. "p.7319, L1-2: "Fig. 3e" should be "Fig.3g" and vice versa."

Fixed (Page 14, Line 8).

6. "p.7321, L6: check wordings."

Fixed (Page 16, Line 26).

7. "p.7323, L12-14: It appears worth showing a figure for the case of "50% increase in BC absorption", where the model-observation discrepancy would be largely reduced."

Agreed. We added a panel in Fig. 9 for the case (50% increase of BC absorption).

8. *“p.7339, Table 1 (and elsewhere): footnote - “See text for details”.*”

Fixed. We altered *“See text for more details”* to *“See text for details”*.