Geoscientific
Model Development
Discussions

Interactive comment on "Further improvement of wet process treatments in GEOS-Chem v12.6.0: Impact on global distributions of aerosol precursors and aerosols" by Gan Luo et al.

Anonymous Referee #2

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Interactive comment on "Further improvement of wet process treatments in GEOS-Chem v12.6.0: Impact on global distributions of aerosol precursors and aerosols" The authors aim to improve the wet processes simulation of aerosols and aerosol precursors in GEOS-Chem v12.6.0 by further revising their previous work Luo et al. (2019), including updates to aqueous-phase chemistry and wet scavenging of aerosols and aerosol precursors in and below different types of cloud and during different types of precipitation, as well as dry deposition to different wet surface. The authors evaluate their updated wet process simulations using surface and aircraft measurements of aerosols and aerosol precursors concentrations from the US, Europe, Asia and Arctic, as well as AToM-1 and AToM-2 campaigns. This work is interesting and this topic is

important for regional and global modeling of aerosol and aerosol precursors.

However, major revision is recommended before being suitable for publication. While this work represents an admirable set of updates that are ostensibly improvements, the main drawback is that there is no systematic exploration of the impacts of any of the updates included in this paper. All changes are updated simultaneously, and then the model is evaluated in a rather generic fashion, without for example seeking out spatiotemporal subsets of data that would be most useful for isolating the impacts of any of the processes studied here. Notably, neither wet deposition measurements nor precipitation measurements are considered in the model evaluation. While the overall simulation updates are indeed an improvement, the paper leaves a bit to be desired in terms of explanations and scientific analysis. I believe addressing these requires more targeted use of the observations, additional simulation that examine the impact of subsets of the model updates tested individually, and evaluation of these to potentially refine some of the assumptions made during the model development. The work needs extensive proofreading throughout (every paragraph contains several grammatical errors; it goes beyond what I'm willing to edit myself), and several of the references are inappropriate. Further comments are described below.

Major comments: 1. As this work focus on improving the wet processes (mainly wet scavenging) simulation of aerosols and aerosol precursors, validation by measurements of wet deposition of aerosols and aerosol precursors is quite necessary given the availability of a bunch of wet deposition measurements over the US (http://nadp.slh.wisc.edu/ntn/), Europe (https://projects.nilu.no//ccc/emepdata.html) and China (https://www.nature.com/articles/s41597-019-0061-2) and also from EANET network (https://www.eanet.asia/about/site-information/). Please at least validate your simulated wet deposition of sulfate + SO2, nitrate + HNO3, ammonium + ammonia using available measurements. Precipitation itself should also be evaluated.

2. Although the authors claim that they have surface measurement-based validation for Asia, the number of the Asian sites is very limited and none of these sites is in China or

India, where there are high emissions of aerosols and aerosol precursors and a large amount of precipitation. So robust validations in China and India are necessary if the authors claim that their updates also improve aerosol and aerosol precursor simulation for Asia.

3. The number of AMoN/EANET sites in Figure 7 seems to be much fewer than the total number of AMoN/EANET sites. Please give a brief description of all the measurements (IMPROVE, CSN, CASTNET, AMoM, EMEP, EANET, AToM-1 and AToM-2), number of valid sites, and data filtering you are using.

4. Quantitative evaluation for simulated vertical profiles of aerosol and aerosol precursors using AToM-1 and AToM-2 aircraft measurements is necessary to support your conclusions about improvements using your updated wet processes.

5. Section 2.6: How are wet surfaces defined in GEOS-Chem? Are these based on land-type or some other classification? Are they altered by precipitation? Overall this strikes me as a level of details beyond what this model can actually resolve.

6. Fig 1: a. It seems evident that the updates in both L2019 and WETrev degraded the model performance for SO2 and SO4 in the US, especially for SO2 This needs to be mentioned, explicitly, and discussed.

b. Why is the modeled SO4 seasonality incorrect in comparison to EANET?

c. Fig 1: Are model values for entire region or only sites at which observations are available?

d. Other factors mentioned in previous studies that may possibly impact overestimated HNO3 and nitrate concentrations are the constant hourly emissions of NH3. Has that been addressed here?

e. The authors seem to gloss over the impacts on NH3. First, the initial model performance compared to the observed NH3 concentrations is surprisingly good, given uncertainties in NH3 emissions. Second, there does seem to be significant difference

between WETrev and the other simulations, in comparison to the observations. In many months it would appear the bias compared to the measurements has increased by up to a factor of 2. None of the simulations correctly replicate the spring time NH3 maximum, most notably in Europe.

7. Figs 7 - 8: It is hard to get much out of the comparison to observations in these figures. Those are better represented by the previous figures, or require zoomed subplot of the US, Europe and E Asia. What would be more useful in Figs 7 - 8 would be to see the base case model (GC12) and the differences between this model and WETrev, as absolute and relative differences.

Minor comments:

1. Page 1, line 25-28: "we compared model simulation . . . successfully improved by considering the updated wet processes." Please give quantitative metrics to support this conclusion.

2. Pag1, line 20: So results for the decrease in NMB of these species in the US are from L2019? If so, probably should't be presented in the abstract as results from the current study.

3. Page 3, line 26 and page 12, line16: I don't think the web site wiki is a suitable citation. Please refer the peer reviewed literature upon which such material is based.

4. Page 6, line 16-17: "we assume that total amount . . .aerosol thermodynamics (SNVC) is 25 % of sulfate". Why do you use 25% here? You should have some rational for this 25% although it is assumed.

5. Page, line 22-30: You are encouraged to validate your rainwater pH using precipitation pH measurements over the US, Europe and Asia.

6. Page 6, line 2: I'm curious how this problem is formulated and how the updated solution method using Newton's method is applied. These are the sorts of details that should be described explicitly here, at the level at which they are reproducible

from reading the text. Citation of unpublished preliminary work from a conference presentation (Moch 2019) is not an adequate reference nor explanation.

7. Page 6, line 17: What is the basis for picking 25% here? No explanation or reference has been provided.

8. Section 2: It's not clear how this particular model distinguishes / defines the processes of rainout vs washout âĂŤ could this be clarified?

9. Page 7, line 22-27: I can understand that you take the ice surface as an aqueous layer when temperature higher than 263K in the mixed cloud, and this means aqueous chemistry can happen at the surface layer of the ice cloud. But I don't understand why it is reasonable to assume aqueous phase cloud fraction equals grid mean cloud fraction: faq=fc. This formula means you assume the aqueous chemistry happen in the ice cloud the way as it does in an aqueous cloud, not just at the pre-melt layer of the ice cloud, most of which is ice-phase. The ice-phase cloud water in one grid is 3 dimensional and I can accept the assumption that the aqueous chemistry can happen at the surface pre-melt layer, but it doesn't make sense that the aqueous chemistry can happen in an ice-phase cloud the way in aqueous-phase cloud. Also, does this assumption of aqueous layer of ice also increase the wet scavenging of HNO3 in the same grid cell?

10. Page 8, line 14 − 18: It's not clear to me this is double counting. It seems one process describes the absorption and the other the oxidation, with both steps being necessary. Am I missing something here? If so, could the authors explain the model treatment of these processes in more detail? Schematics could be helpful.

11. Page 8, line 25-30: "the rationale . . . as water soluble aerosols . . .. The composition . . . for cloud activation calculation." Please include some appropriate references here.

12. Page 9, line 1-3: "However, in the actual atmosphere, . . . coated with SNA". Please also include some appropriate references here.

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13. Page 9, line 29-30: "While most of . . .smaller than 500 nm." Please include some appropriate references here.

14. Page 10, line 4 − 8: How was this treated in the original model?

15. Page 13, line 15-17: "One possible reason is . . . at urban sites . . .remote regions." Please include some appropriate references here.

16. Page 13, line 29: Is there a scientific explanation why one would revert this change, in terms of understanding of heterogeneous chemistry? Or are the authors alternatively suggesting that these parameters be adjusted so that the model estimates better fit the data?

17. Page 14: line 1-2: "The aqueous concentration of ammonia is much lower than nitric acid, . . . small impact on the simulation of ammonia." How does this conclusion come? Isn't it because the increasing ammonia wet deposition is compensated by less reaction with decreased HNO3 in the air?

18. Page 14, line 27-28: "The underestimate is likely . . . wildfire . . . US." Please include some appropriate references here.

19. Page 15, line 30-31: "Most of BC at Arctic . . . anthropogenic emissions," Please include some appropriate references here.

20. Page 16, line 7-11: Please provide figures for the tracks of ATom-1 and ATom-2. And why do you filter out tracks over land? Could you provide profile comparison over land?

21. For the whole section 3.3, please give quantitative metrics (e. g. NMB and R) to show how your updated wet scavenge schemes work in reproducing the observed profiles of these chemical species. Figure 5 (a) shows WETrev underestimate HNO3 throughout the whole layers except for high bias at upper layer (nearby 200 hpa). Figure 6 shows that L2019 and WETrev largely underestimate HNO3 at lower troposphere (> 800 hpa).

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22. Page 17, line 6: With regards to "in this work," it seems that a bulk of the improvements are from the updates in L2019, more so than the present work.

23. Page 18 line 25 - page 19 line 6: please try to quantitively and appropriately show how your updates improve the simulation. Like your 5th conclusion on page 19 line 5-6: "The updated wet surface ... SO2 at Artic sites", I am not so sure whether your conclusion is fully supported by figure 3 or not.

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