Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmd-2020-11-AC1, 2020 © Author(s) 2020. This work is distributed under the Creative Commons Attribution 4.0 License.



## 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.

## Gan Luo et al.

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We thank the referee for the detailed reviews and constructive comments that help to improve the manuscript. Below we respond to the comments in detail. (Referee's comments are in Italic).

General comments The study attempts to improve the simulation of aerosol precursors and aerosols in GEOSChem via multiple updates in wet processes in the model. The updates in the treatment of wet processes have been described in details and the results are also evaluated with a large set of in-situ measurements from both surface monitoring networks and aircraft campaign. While the evaluation shows significant

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improvement in model results, it is not immediately clear how significant the update in each wet process actually is. First, the updates in wet processes in this study including pH calculation for cloud, rain and wet surface, fraction of cloud available for aqueous phase chemistry, rainout efficiencies, washout efficiencies and wet surface uptakes during dry deposition. Evaluation of each update is necessary to understand the factors contributing to the uncertainties in the simulation of aerosol precursors and aerosols so that similar improvement could be applied to other models.

Because of the lack of detailed process diagnostics in GEOS-Chem, it is difficult to trace the contributions from each modifications. To address the referee's comment, we carried out 5 numerical sensitivity study cases (RO, WO, RP, DD, and AC) to understand the factors contributing to the uncertainties in the simulation of aerosol precursors and aerosols. RO case is the same as case WETrev except using rainout rate in GC12; WO case is the same as case WETrev except using washout rate in GC12; RP case is the same as case WETrev except assuming pH of rainwater for wet scavenging is 4.5; DD case is the same as case WETrev except using dry deposition treatment in GC12; and AC case is the same as case WETrev except using aqueous phase chemistry treatment in GC12. The results of the sensitivity study and associated discussions have been added to section 3.1.

Second, the update in aqueous phase chemistry seems important for aerosol precursors and the corresponding aerosol species. But to what extent is the cloud/rain pH and subsequent dissolution in WETrev different from those in GC12?

In GC12, pHs of cloud and rain for wet deposition are assumed to be 4.5. The calculated rainwater pH in this study varied from 4.3 to 6.9. The impact of pH on effective Henry's law constant of SO2 is shown in Table 1. We have clarified this in the revised text

Specific comments p. 6, line 27-30: what is the range of the calculated rainwater pH in this study?

The calculated rainwater pH in this study varied from 4.3 to 6.9. This is clarified in the

p.11, line 3-4: In which way is ICCW related to wet scavenging? In other words, which equation is ICCW applied for?

ICCW is applied for equation 1. This is clarified in equation and the text.

p.12, line 2: why are the washout coefficients different between hydrophobic and hydrophilic aerosols? The mechanism associated with washout includes processes such as diffusion, interception, and impaction. Not sure how and to what extent it is affected by the water solubility of the aeosols.

The assumption of different washout coefficients for hydrophobic and hydrophilic aerosols is because the rain washout rate for water-soluble aerosols measured by Laakso et al. (2003) is still 20 times larger than that calculated by the semi-empirical parameterization. One of the possible reasons is droplet-particle collection mechanisms for hydrophobic and hydrophilic aerosols are different. This is clarified in the

p.15, line 19: it also enhances so2 at Zeppling in January and February, but not December, why?

At Zeppelin, temperature in December is higher than that in January and February. SO2 is enhanced due to the modification of dry deposition in this work. However, there is more aqueous phase chemistry in December which consumes the enhanced SO2. This is clarified in the text.

p.16, line 17-21: I see the opposite way, where WETrev significantly underestimates nitric acid at the upper troposphere from Fig. 5

For nitric acid above 300 hPa, the values simulated by WETrev are higher than those by L2019. For nitric acid between 500 hPa and 300 hPa, the values simulated by WETrev are lower than those by L2019. It is because L2019 only considered washout of nitric

acid by rain. WETrey also considered washouts of nitric acid by snow and ice which were absent in L2019 and GC12. This is clarified in the text.

p. 34: reduce the xrange of the figure so that the difference among these lines can been seen more clearly

X-ranges of Figure 5 and Figure 6 were determined by maximum values of each species during ATom-1 and ATom-2. It lets readers easily find the spatial and temporal variations of these species at the North Hemisphere and South Hemisphere during boreal summer (ATom-1) and boreal winter (ATom-1). We keep the original x-range as we think it is more suitable for what we want to present.

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	273K	283K	293K	303K
pH=4.5	2479.7	1271.7	682.7	382.0
pH=5	7888.3	4039.5	2165.9	1210.4
pH=5.5	25474.2	12995.7	6947.4	3873.3
pH=6	85913.1	43355.2	22980.1	12724.6
pH=6.5	325309.2	159736.0	82801.8	45021.4

Fig. 1. Table 1. The values of effective Henry's law constant of SO2.