Geosci. Model Dev. Discuss.,
https://doi.org/10.5194/gmd-2020-11-AC3, 2020

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.

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Received and published: 24 April 2020

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).

This paper presented updated treatments of wet processes in GEOS-Chem, including rainout efficiencies for warm, mixed-phase and cold clouds, empirical washout by rain/snow, aqueous phase chemistry and wet removal for SO2 and sulfate, and wet surface uptakes during dry deposition. Model simulated concentrations of aerosols and aerosol precursors were evaluated with various surface observational data sets

over the U.S., Europe, Asia, and Arctic as well as aircraft measurements of nitric acid and aerosols during two ATom campaigns. Results showed significant improvement over previous version of the model and better agree with the observations. Although mentioned in various places in the paper, the roles of individual wet processes in the improvements were not systematically quantified. This paper is well organized and overall well written, but needs careful proofreading. I recommend publication after the following comments are addressed.

We appreciate the positive comment about the paper. The revised manuscript has been carefully proofread.

P4, Line 3, eqn 1: Pr is the grid-box large-scale precipitation (rain+snow) formation rate. LCW is liquid phase cloud water content. But the total condensed water content should also include ice cloud water content, which is missing from this equation.

Yes, it is right. The equation and associated discussions have been modified.

P8, Line 7, eqn 11: same issue as for eqn 1. For T>=258K (warm clouds), this equation assumes zero ice cloud water (ICW), which is probably not true in MERRA-2. Since the model uses temperature ranges to separate scavenging due to warm/mixedphase/cold clouds, the cloud condensed water (for all T) needs to include ICW. This is expected to have a significant impact on the model results of this paper.

Thanks for pointing this out. We have modified the equation and code. We rerun the WETrev case with these updates.

P10-11, Section 2.4: For rainout in cold cloud (T<237K), do you limit it to below the MERRA-2 tropopause?

We did not. After discussed with the GEOS-Chem Steering Committee, we decided to limit rainout to below the MERRA-2 troposphere since stratospheric water in MERRA-2 is known to have unphysical behavior. We rerun the WETrev case with these updates. This has been clarified in the revised text.

P16, L17-19, and Fig.5: Please double check. It looks like the L2019 and WETrev lines for HNO3 are switched. What aspect of the "old treatments in GC12" do you mean here?

The lines of L2019 and WETrev cases in Fig.5 are right. GC12 and L2019 only considered washout of nitric acid by rain. WETrev also considered washouts of nitric acid by snow and ice which were absent in L2019 and GC12, therefore nitric acid concentrations of WETrev between 500 hPa and 300 hPa are lower than those of L2019. Rainout efficiency of nitric acid by cold cloud in WETrev is lower than that of L2019, therefore, nitric acid concentrations of WETrev above 300 hPa are higher than those of L2019. Old treatment referred to cold cloud wet scavenging of nitric acid in GC12 is treated the same as water-soluble aerosol with 100 % rainout efficiency. Cold cloud rainout efficiency in WETrev is based on the parameterization of nitric acid partitioning in cold cloud developed by Kärcher et al. (2008). We modified the sentence as: As we mentioned earlier, L2019 may overestimate cold cloud wet scavenging of nitric acid due to treat cold cloud rainout of nitric acid the same as water-soluble aerosol with 100 % rainout efficiency.

P19, Code and data availability: the revised GEOS-Chem v12.6.0 code and model output need to be made available at a public data depository. Also it's not clear where the various observational data sets used in this work were downloaded from.

We have updated the Code and data availability. Links of observational data sets have been provided.

Minor comments: P4, Line 28: is LW different than LCW in eqn 1.

LW in equation 1 is liquid water content for Henry's law. It equals liquid cloud water content (LCW) in the atmosphere.

P5, L1 (and other places): Do you mean "acidity"?

Yes, you are right. Modified.

P5, L3: H can be calculated . . .*

Modified.

P5, L8-9: what are the units for these constants and coefficients?

We modified the sentence as: where HSO2, HH2O2, and HNH3 are the Henry's law constants (M atm-1) for SO2, H2O2, and NH3, respectively. K1 (M), K2 (M), K3 (M), K4 (M2), and K5 (M) are rate coefficients for SO2 reaction, HSO-3 reaction, H2O2 reaction, H2O reaction, and NH3 reaction, respectively.

P6, L19: the comma is misplaced.

Modified.

P7, L20: LCW not LWC

Modified.

P13, L3: Emissions are produced by the default setting of HEMCO. Does this mean that emissions are specific to the periods of ATom-1 and ATom-2 campaigns?

We used the default setting of HEMCO to produce emissions for all simulations presented in this work. We modified the sentence as: Emission over Europe is produced by EMEP inventory. Other emissions are produced by the default setting of HEMCO (Keller et al., 2014) for all simulations presented in this work. EMEP emission over Europe is used in our rerun cases of GC12, L2019, and WETrev. It is because we found the replacement of EMEP emission with CEDS global emission in GC12.6.0 leads unreasonable performance of ammonia seasonal variation over Europe.

P13, L16: Is there a reference for "a large amount of USEPA observations are located at urban regions"?

We didn't find related reference. There were 288 EPA's Air Quality System sites with valid data in each month of 2011. Only 69 of these sites were with the mark of 'Not in

a city'. More information can be found at https://www.epa.gov/outdoor-air-quality-data.

P14, L20: remove "– ".

Modified.

P15, L7: low dissolution

Modified.

P15, L27 L29: "at Alert during spring" – during winter / early spring?

For BC at Alert, it is winter and spring. For sulfate at Alert, it is spring. We have modified the sentence.

P16, L1: converted

Modified.

P16, L10-11: Why are the flight tracks over the land filtered out for comparison?

ATom observations over the land, whose values vary greatly, only account for 28 % of total measurements. To make the comparison more appropriate, we filtered out the flight tracks over the land.

P18, L18-24: this sentence needs a break.

We rewrote the sentence as: In this study, we updated aqueous phase chemistry and wet scavenging for SO2 and sulfate, rainout efficiencies for warm, mixed, and cold cloud, empirical washout by rain and snow, and wet surface uptakes during dry deposition in GEOS-Chem version 12.6.0. Systematic validations of simulated aerosol precursors and aerosols with ground based monitoring networks over the US, Europe, and Asia, in-site observations at Arctic for surface mass concentrations and aircraft measurements during ATom-1 and ATom2 for their vertical profiles were presented.

P19, L3, L12: remove"an"; exist.

Modified.

Table 1: refer the reader to eqn 16.

Accepted.

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