

## ***Interactive comment on “Revised treatment of wet scavenging processes dramatically improves GEOS-Chem 12.0.0 simulations of nitric acid, nitrate, and ammonium over the United States” by Gan Luo et al.***

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Received and published: 17 June 2019

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 a revised wet scavenging parameterization that considers the spatiotemporal variability of cloud liquid water content and an empirical washout (below-cloud scavenging) rate in the GEOS-Chem global chemical transport model. The au-*

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*thors showed that the updated parameterization significantly improves simulated annual mean (and seasonal) mass concentrations of nitric acid, nitrate, and ammonium as compared with surface observations over the U.S. This is an important contribution to the improvement of GEOS-Chem. Minor revision is recommended before publication on GMD.*

We appreciate the referee's positive comments about the importance of this study.

*Major comments: The impact of updated wet scavenging on model simulations was only assessed at the surface level and for nitric acid, nitrate, and ammonium over the U.S. It's not shown how the updated treatment of scavenging affects the global aerosol simulations, especially the vertical profiles and other aerosol species (e.g., sulfate). Consider discussing this in the Summary and Discussions section. Lead-210 aerosol tracer has been used to test wet deposition in GEOS-Chem (e.g., Liu et al., 2001), and this updated scavenging parameterization will need to be tested with (at least) lead-210 before it is incorporated into the standard version of the model.*

Agree. We added additional discussions on these issues in the summary and discussions section. More in-depth analysis is being carried out and we are preparing another paper on the impacts of updated wet scavenging parameterization on all major aerosol species over the whole globe.

*Page 5, equation 4: 1). "CW is grid-box mean cloud water content". What's the corresponding variable name in MERRA-2? Does it include both cloud liquid (QL) and ice (QI), or QL only? 2). It's not clear why the rain water term "Pr\*DeltaT" is needed. There is no prognostic precipitation (no raining condensate) in MERRA-2 or GEOS-5. Prognostic cloud liquid and ice are autoconverted to estimate precipitation. Are "CW" values for pre-conversion or post-conversion? More explanation as well as references are needed.*

CW is "QL" in MERRA2. It only includes cloud liquid. As shown in Equation 6 in MERRA2's file specification (Bosilovich et al., 2016), QL is the residual condensation

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water after precipitation. Due to large fraction of cloud water converted to rain water, cloud water in MERRA2 is low when precipitation is occurring. Because the fraction of soluble species rained out should equal to the fraction of total condensed water (or ICCW in our case) converted to rain water, we think that ICCW in Eq (3) should include rain water (i.e., Eq 4). The following reference is added to the reference list. Bosilovich, M. G., R. Lucchesi, and M. Suarez, 2016: MERRA-2: File Specification. GMAO Office Note No. 9 (Version 1.1), 73 pp, available from [http://gmao.gsfc.nasa.gov/pubs/office\\_notes](http://gmao.gsfc.nasa.gov/pubs/office_notes).

*Page 7, lines 20-23: The first-order rainout parameterization is not used for convective precipitation scavenging in GEOS-Chem driven by MERRA-2. Instead, scavenging in convective updrafts are coupled with convective transport (e.g., see section 2.3.1 of Liu et al., 2001).*

We have modified the text to reflect this.

*Minor comments: Title: Suggest adding “surface” to the title since this study examined the impact of revised scavenging on surface aerosol concentrations only.*

Accepted.

*Page 1, line 21: typo “mentoring” (“monitoring”)*

Revised.

*Page 3, lines 6-7: are there references for this statement?*

We did the simulations with  $4^\circ \times 5^\circ$  and  $2^\circ \times 2.5^\circ$  horizontal resolutions in GEOS-Chem and found the switching of model resolution has small impact on simulated nitrate over the US.

*Page 3, line 14: change “in-site observations” to “surface observations”*

Accepted.

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*Page 3: A brief description of the GEOS-Chem model is needed here before discussing the wet scavenging scheme.*

Accepted.

*Page 3, section 2: See this webpage [http://acmg.seas.harvard.edu/geos/geos\\_chem\\_narrative.html](http://acmg.seas.harvard.edu/geos/geos_chem_narrative.html) for “Narrative description (and how to cite GEOS-Chem)”, which provides guidance on citing relevant model components. “The wet deposition scheme in GEOS-Chem is described by Liu et al. [2001] for water-soluble aerosols and by Amos et al. [2012] for gases. Scavenging of aerosol by snow and cold/mixed precipitation is described by Wang et al. [2011, 2014].” Suggest citing Jacob et al. (2000) along with one of these publications, where appropriate, since it is an unpublished document. The first-order rainout parameterization (equations 1 and 2) is based on Giorgi and Chameides (1986), which also needs to be referenced.*

Added description and citation as suggested.

*Specify the units for variables in all equations in the text.*

Accepted.

*Page 4, line 3: condensed water content includes liquid and ice phases. Do you revise warm cloud scavenging only here? Does “Pr” (rate of new precipitation formation) include snow? How about ice cloud scavenging?*

We only revised rainout for warm cloud in this study. Pr, DQRLSAN in MERRA2, only includes rainwater. Ice cloud scavenging is applied to aerosols via washout by snow following the approach suggested by Wang et al. (2011).

*Page 4, lines 23-25: Croft et al. (2016) previously used GEOS-5 cloud liquid and ice water content to replace the fixed value in their GEOS-Chem-TOMAS simulations. Consider citing that work here. (Croft, B., Martin, R. V., Leaitch, W. R., Tunved, P., Breider, T. J., D’Andrea, S. D., and Pierce, J. R.: Processes controlling the annual cycle of Arctic aerosol number and size distributions, Atmos. Chem. Phys., 16, 3665-*

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3682, <https://doi.org/10.5194/acp-16-3665-2016>, 2016.)

Thanks for pointing us to this work. The major difference of rainout treatment between Croft et al. (2016) and our work is the assumption of ICCW. Croft et al. (2016) used cloud liquid and ice water content to replace the fixed ICCW, while we used the sum of cloud liquid water and rain water to replace the fixed ICCW which is critical for rainout calculation (Eqs. 2-3). Corresponding discussions have been added in the revised paper.

*Page 8, line 4: these references are not for rainout and washout parameterizations, but for the standard GEOS-Chem model (or other model components).*

Accepted. These references are cited at the brief description of the GEOS-Chem model in revised paper.

*Page 9, line 13: CCW or ICCW?*

It is ICCW. Revised.

*Page 9, line 17: concentrations OF; line 20: showS*

Revised.

*Fig. 2 caption: indicate the year and number of sites over the U.S., and note the small differences between blue and green dashed lines.*

Modified as suggested.

*Fig.3 caption: annual mean surface*

Modified as suggested.

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Interactive comment on Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2019-58>, 2019.