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Interactive comment

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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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). The manuscript has been revised accordingly.

The authors aim to improve wet deposition simulation of nitric acid, nitrate and ammonium over the United States using GEOS-Chem via updating both in cloud stratiform cloud scavenging and below cloud washout. For in cloud scavenging, they adopt the

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Discussion paper

dynamic varied condensed water content provided by MERRA2 meteorological fields needed in wet scavenging parameterization, instead of the current assumption of a global flat value. For below cloud washout, they derive a new set of empirical washout scavenging coefficient and exponential coefficient for nitric acid based on the size-resolved coefficients summarizing from field measurement and theoretical derivation. This is an interesting and valuable study. The study would have a potential impact on the broad atmospheric composition study via improving tracers' wet scavenging if the authors could validate their work for other aerosols and their precursors. A minor revision is required before the paper is published in GMD.

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

Major Comments The authors test the physical-based condensed cloud water for stratiform cloud rainout. Convective cloud removal is important and is necessary to be studied as well. Studying convective rainout is particularly important for using the current generation of NASA GEOS meteorological fields since its partitioning of large scale and convective clouds tilts more towards the latter. The convective cloud fraction and water content can be provided by the GEOS model.

This is a valid point. However, the convective cloud fraction and water content, while available in GEOS online simulation, is not available in GMAO reanalysis datasets (including MERRA2) used to drive GEOS-Chem. Therefore, as we have already pointed out in the last paragraph of Session 2.2, "the updated wet scavenging method discussed above for stratiform precipitation cannot be directly applied to convective precipitation rainout scavenging in GEOS-Chem". We agree with the referee that "Convective cloud removal is important and is necessary to be studied as well" and have pointed this out in the discussion session.

The authors are highly encouraged to evaluate and summarize the impact of their work on other aerosols and their precursors. Once the GEOS-Chem adopts the improvements in wet scavenging parameterization suggested by the authors, all aerosols and *their precursors undergoing wet scavenging will be impacted. To have confidence in using their work, they should at least provide a brief description of the model performance for all important aerosol fields in supplementary material. In addition, the authors' work focuses on the United States only. What is the anticipated influence of the improved wet scavenging on other regions?*

Yes, we are evaluating the impacts of updated scheme on other aerosols and other regions. Based on preliminary comparisons with relevant measurements we analyzed so far, the updated wet scavenging parameterization also improves the model performance over Europe and Asia. More in-depth analysis is being carried out and we are preparing another paper on the impacts of updated wet scavenging parameterization on global simulation of aerosols in GEOS-Chem. We have pointed this out in the discussion session.

To be more useful of the proposed work on wet deposition, more words are needed about the broader impact of the study on the whole atmospheric chemistry community. Can other global chemistry models adopt their improvement? Is there anything that other modelers should be cautioned of in adopting their work?

The updates of rainout can be adopted by any atmospheric chemistry models which assume constant cloud condensation water. The empirical washout can help to reduce the overestimation of nitric acid gas shown in the work of Bian et al. (2017) by most of atmospheric chemistry models. Corresponding discussions have been added in the Summary and Discussion section.

Specific comments 1. Page 3 line 26 equation 1: Should the k in exponential term differ with the k in the denominator of coefficient? For my understanding, the k in exponential term, which is the first-order rainout rate, is linked to specific tracer species. One the other hand, the k in denominator represents the generic conversion rate of cloud water to precipitation. Please double check this. Please also give units of these fields and parameters in equation 1.

We double-checked this. As shown in the work (session 1.1) of Jacob et al. (2000), the k in exponential term and the k in the denominator of coefficient are the same for soluble species. In GEOS-Chem, the model assumes the first-order rainout rate for water soluble aerosols and nitric acid gas equals the generic conversion rate of cloud water to precipitation. The units have been added.

2. Page 7 lines 4-19: How about aerosols? Should the washout scavenging coefficients of aerosols be adjusted accordingly?

We applied empirical washout rate from Laakso et al. (2003) for water soluble aerosols and washout rate from Feng (2007) for water insoluble aerosols. Corresponding information has been updated in the revised text.

3. Page 8 line 7: Please add one more case study. Similar to case study 4 but empirical washout rate of HNO3 is applied only to large scale precipitation. This case, combined with case 4, will give us further information about the relative washout contribution from large scale and convective scale precipitations.

As shown in Figure 1 below, the exclusion of wash out by convective precipitation in GEOS-Chem has negligible impacts on surface level HNO3, nitrate, and ammonium over the US. It is because convective precipitation is large over Tropics and small over middle and high latitude continent. Convective precipitation over the US is 10-100 times smaller than large-scale precipitation over there.

4. Page 8 line 7: Do the authors present the work of empirical washout rates for aerosols? Section 2.2 seems only give discuss for HNO3. What are the new empirical washout rates for aerosols?

In this study, empirical washout rate is from Laakso et al. (2003) for water soluble aerosols, while washout rate is from Feng (2007) for water insoluble aerosols. We modified corresponding sentences in the paper.

5. Page 9 lines 1-2: The change range shown here (from 150% to 24%) includes not *only using empirical washout rate, but also changing cloud condensation water.*

Corrected. We changed the value from 150 % to 125 %.

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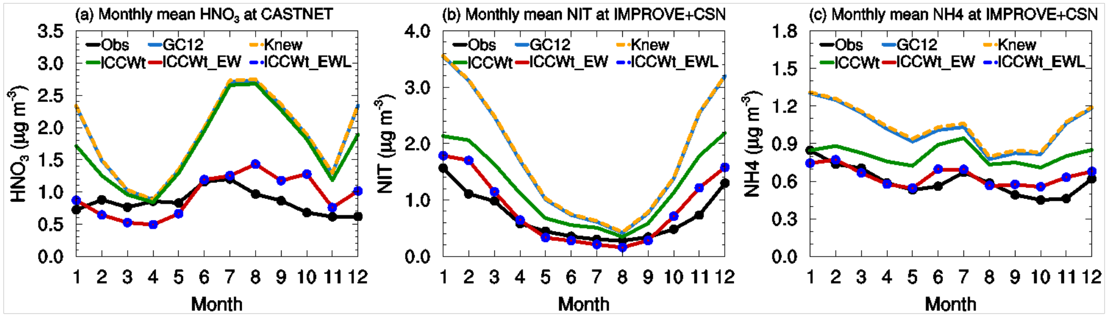

Fig. 1. Monthly variations of mean for year 2011 showing the comparison between nitric acid (a), nitrate (b), and ammonium (c) mass concentrations observed at ground-based sites (black) and simulated by GC12