Reply to referee 1

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 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 SO_2 is shown in Table 1. We have clarified this in the revised text.

Table 1. The values of effective Henry's law constant of SO_2.

	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

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 text.

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.

$$F = \frac{P_{\mathrm{r}}}{k \cdot \mathrm{ICCW}}\left(1 - e^{-k \cdot \Delta t}\right) = \frac{f_{\mathrm{c}} \cdot P_{\mathrm{r}}}{k\left(\mathrm{LCW{+}ICW} + P_{\mathrm{r}} \cdot \Delta t\right)}\left(1 - e^{-k \cdot \Delta t}\right)$$

This is clarified in equation 1 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 text.

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. SO_2 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 SO_2. 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. WETrev 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.

Reply to referee 2

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).

Interactive comment on "Further improvement of wet process treatments in GEOSChem 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.

Thanks for the positive comment about the importance of this work.

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.

Thanks for the constructive comments. The manuscript has been revised following the suggestions.

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.*

We added comparison of simulated wet deposition of sulfate+SO_2, nitrate+HNO_3, ammonium+NH_3 with available measurements. Associated discussions are in section 3.1.

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.

We clarified in the revised paper that wet process updates improve aerosol and aerosol precursor simulation over Asia remote regions.

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.

The criterion of observations used for model validation is that valid data are available for every month in 2011. For EANET observations, due to too much missing data, the criterion is loosen to monthly mean data available for each month during a 3-year period (2010-2012). Seto et al. (2007) pointed out that EANET observations at urban sites are much higher than those at remote sites. Since the number of the Asian sites is very limited, to make the validation more appropriate, only remote and rural sites are used for model validation. A brief description on these has been added at section 3.1.

Seto, S., Sato, M., Tatano, T., Kusakari, T. and Hara, H., Spatial distribution and source identification of wet deposition at remote EANET sites in Japan. Atmos. Environ.41,9386-9396, 2007.

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.

Normalized mean biases (NMB) NMB and correlation coefficient (r) have been used to quantitative evaluation for simulated vertical profiles of aerosol and aerosol precursors with AToM-1 and AToM-2 aircraft measurements.

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.

GEOS-Chem determined wet surfaces based on land use type from the Olson 2001 land map (Olson, 1992). They are not altered by precipitation. This has been clarified in the text.

Olson, J, World Ecosystems (WE1.4): Digital raster data on a 10 minute geographic 1080 x 2160 grid, in Global Ecosystems Database, version 1.0, Disc A, edited by NOAA Natl. Geophys. Data Center, Boulder, Colorado, 1992.

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.

Accepted.

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

It is caused by the overestimation of January SO4 at Primorskaya, Russia (43.63ºN, 132.24ºE) whose value is high up to 12 µg m^{-3}, 2.5 times higher than observation at this site. This overestimation is associated with aqueous phase chemistry over there.

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

Model values were sampled at sites where 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?

The present version of GEOS-Chem considers diurnal, seasonal, and interannual variability of ammonia emission.

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.

The unreasonable seasonal variation of ammonia in the Europe is caused by the updated emission treatment in GC12.6. GC12.6 replaced old EMEP emissions and seasonal scaling factors with CEDS global emissions. After switching back to EMEP emissions, seasonal variation of ammonia was captured by the model. We rerun the cases with EMEP emissions and updated the results in the revised manuscript.

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.

The two figures were provided to show the impacts of WETrev on a global scale. One can derive globally averaged absolute and relative differences from values given on top of each panel. Interested readers can use the enlarge function for pdf figures to zoom into specific regions. There will be too many figures if we provide zoomed subplots and plots for both 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.

Quantitative metrics have been added.

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.

The decrease in NMB of these species is mainly caused by the updated ICCW and empirical washout. Excluding the results in the US sounds strange here.

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.

As shown in the web site wiki, H^* of SO_2, H_2O_2, and NH_3 for dry deposition was originally in drydep_mod.F. We were not able to locate the peer reviewed literature on this.

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.

It is based on the work of Guo et al. (2018) cited in the text which suggested ammonium-sulfate aerosol molar ratio is 1.47±0.43 (≈1.5).

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

We will investigate this in our future work.

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.*

More detailed description on this has been added:

This iterative calculation is updated to use Newton's method in order to arrive at a consistent result (Moch et al., 2020). To implement Newton's method the equilibrium expressions for the concentrations of each soluble semi-volatile ion (SSVI) in terms of H+ and the derivatives for these equilibrium expressions are each solved explicitly so that the Newton's method equation is in the form of:

$$\mathrm{H}^{+}_{\mathrm{n+1}}=\mathrm{H}^{+}_{\mathrm{n}}+\frac{\left[\mathrm{SSVI}\left(\mathrm{H}^{+}_{\mathrm{n}}\right)\right]+\left[\mathrm{SNVI}\right]}{\frac{\mathrm{d}}{\mathrm{dH}^{+}}\left[\mathrm{SSVI}\left(\mathrm{H}^{+}_{\mathrm{n}}\right)\right]}, (7)$$

where SNVI is the concentrations of soluble nonvolatile ions. For equation (7) the concentrations of each ion are multiplied by the ion charge (e.g. the terms for SO_3^{2-} concentrations are multiplied by -2).

In tests with this new calculation the solution always converged to an answer in less than 20 iterations, but if a maximum of 50 iterations is reached we set it so that the last two solutions are averaged together. We here considered the solution to converge if the difference between $\mathrm{H}_{\mathrm{n}}^{+}$ and $\mathrm{H}_{\mathrm{n+1}}^{+}$ was less than 0.01. By default the initial guess for H^+ is set to 4.5, but we tested initial guesses ranging from a pH of 2 to 13 and found no change in the values at which the answer converged.

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

The assumption of 25 % is based on the work of Guo et al. (2018) cited in the text. More robust calculation of SNVC need to be investigated in future works.

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

Rainout is the removal due to formation of precipitation in cloud, while washout is the removal due to falling precipitation from upper layers. This has been clarified in the text.

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?*

Yes, it is right. Due to uncertainty of the thickness of aqueous layer of ice, we decide to only use equation 9 to calculate aqueous phase cloud fraction. The assumption of aqueous layer of ice does not increase the wet scavenging of nitric acid 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.

In GC12, rainout of SO_2 is limited by the aqueous phase oxidation of SO_2 by H_2O_2 rather than the absorption by cloud water (Chin et al., 1996). However, the conversion of SO_2 to sulfate in cloud has been accounted for in the aqueous phase chemistry. In this work, rainout of SO_2 is limited by the absorption by cloud water. More explanation of the processes has been added in the revised text.

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

Added.
Abdul-Razzak, H., and Ghan, S. J., A parameterization of aerosol activation: 2. Multiple aerosol types, J. Geophys. Res., 105(D5), 6837– 6844, doi:10.1029/1999JD901161, 2000.

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

Added.
Fassi-Fihri, A., Suhre, K., and Rosset, R.: Internal and external mixing in atmospheric aerosols by coagulation: impact on the optical and hygroscopic properties of the sulphate-soot system, Atmos. Environ., 10, 1393–1402, 1997.

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

Added.
Sahu, L. K., Y. Kondo, N. Moteki, N. Takegawa, Y. Zhao, M. J. Cubison, J. L. Jimenez, S. Vay, G. S. Diskin, A. Wisthaler, T. Mikoviny, L. G. Huey, A. J. Weinheimer, D. J. Knapp, Emission characteristics of black carbon in anthropogenic and biomass burning plumes over California during ARCTAS‐CARB 2008, J. Geophys. Res., 117, D16302, doi:10.1029/2011JD017401, 2012.

Zender, C. S., Bian, H., and Newman, D., Mineral Dust Entrainment and Deposition (DEAD) model: Description and 1990s dust climatology, J. Geophys. Res., 108, 4416, doi:10.1029/2002JD002775, D14, 2003.

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

GC12 assumed rainout efficiency of water-soluble aerosols by cold cloud is 100 %. This is clarified in the revised text.

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

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

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?

Uptake coefficients for heterogeneous chemistry on sulfate in the work of Holmes et al. (2019) are ~ 10 times smaller than those used in GC12.5 which lead to less nitric acid production in GC12.6 than that in GC12.5. Due to large uncertainties of uptake coefficients for heterogeneous chemistry, further investigations are needed. Yes, at this point, we are 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?

This is a good point and we agree it is because the increasing ammonia wet deposition is compensated by less reaction with decreased nitric acid in the air. We have modified the sentence to reflect this.

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

Added.

Mao, Y. H., Li, Q. B., Henze, D. K., Jiang, Z., Jones, D. B. A., Kopacz, M., He, C., Qi, L., Gao, M., Hao, W.-M., and Liou, K.-N.: Estimates of black carbon emissions in the western United States using the GEOS-Chem adjoint model, Atmos. Chem. Phys., 15, 7685–7702, https://doi.org/10.5194/acp-15-7685-2015, 2015.

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

Added.
Xu, J.-W., Martin, R. V., Morrow, A., Sharma, S., Huang, L., Leaitch, W. R., Burkart, J., Schulz, H., Zanatta, M., Willis, M. D., Henze, D. K., Lee, C. J., Herber, A. B., and Abbatt, J. P. D.: Source attribution of Arctic black carbon constrained by aircraft and surface measurements, Atmos. Chem. Phys., 17, 11971–11989, https://doi.org/10.5194/acp-17-11971-2017, 2017.

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?

The flight tracks of ATom-1 and ATom-2 are shown in Fig. 1, while vertical profiles over land are shown in Fig. 2. These figures have been provided in supplementary materials. 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.

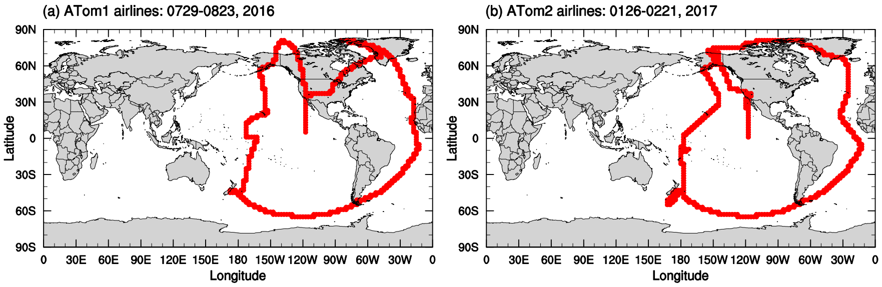

Figure 1. The flight tracks of (a) ATom-1 and (b) ATom-2.

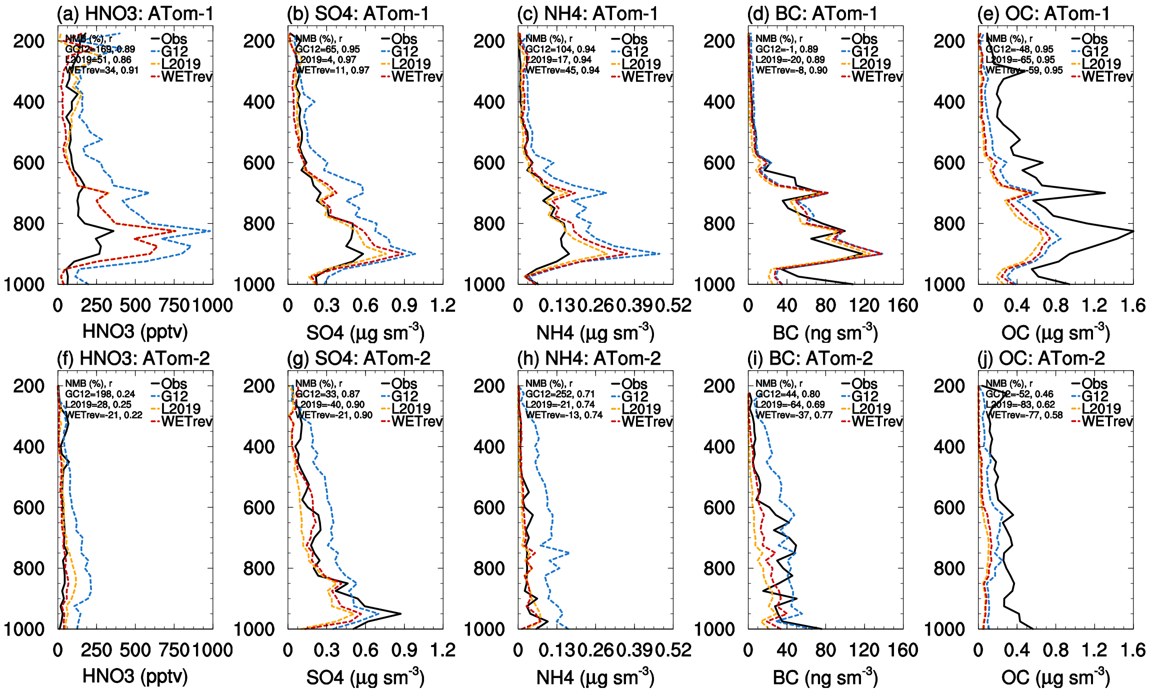

Figure 2. Vertical profiles of nitric acid, sulfate, ammonium, black carbon, and organic carbon from ATom aircraft observations (black, ATom-1: a-e; ATom-2: f-j) and GEOS-Chem simulations by GC12 (blue), L2019 (yellow) and WETrev (red) cases over the land.

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).

Added as suggested.

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.

We modified it as:
the updated wet process treatments in this work and L2019 can improve the agreements of simulated and observed vertical profiles of nitric acid and aerosols.

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.

Accepted. Quantitative metrics have been added. For 5th conclusion, we found NMB of SO_2 is increased from -23 % to 32 % at Nord and decreased from 27 % to 22% at Zeppelin. We modified the conclusion as:
(5) The updated wet surface uptake during dry deposition changes the performance of simulated SO_2 at Arctic sites. NMB of SO_2 is increased from -23 % to 32 % at Nord and decreased from 27 % to 22% at Zeppelin.

Reply to referee 3

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 H_{SO2}, H_{H2O2}, and H_{NH3} are the Henry's law constants (M atm^{-1}) for SO_2, H_2O_2, and NH_3, respectively. K_1 (M), K_2 (M), K_3 (M), K_4 (M^2), and K_5 (M) are rate coefficients for SO_2 reaction, HSO_3^- reaction, H_2O_2 reaction, H_2O reaction, and NH_3 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 SO_2 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.

Further improvement of wet process treatments in GEOS-Chem v12.6.0: Impact on global distributions of aerosols and aerosol precursors~~ and aerosols~~

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Abstract

Wet processes, including aqueous phase chemistry, wet scavenging, and wet surface ~~uptakes~~uptake during dry deposition, are important for global modeling of aerosols and aerosol precursors~~ and aerosols.~~. In this study, we ~~improved~~improve the treatments of these wet processes in the GEOS-Chem v12.6.0, including pH ~~calculation~~calculations for cloud, rain, and wet ~~surface,~~surfaces, the fraction of cloud available for aqueous phase chemistry, rainout efficiencies for various types of ~~cloud~~clouds, empirical washout by rain and snow, and wet surface ~~uptakes~~uptake during dry deposition. We ~~compared~~compare simulated surface mass concentrations of aerosols and aerosol precursors~~ and aerosols~~ with surface monitoring networks over the United States, Europe, Asia, and Arctic regions, and ~~showed~~show that ~~the~~ model results with~~ the~~ updated wet processes agree better with measurements for most species. With the implementation of these updates, normalized mean biases (NMB) of surface nitric acid, nitrate, and ammonium are reduced from 78%, 126%, and 45% to ~~13 %, 24~~ 0.9%, 15%, and ~~6.2~~ 4.1% over the US sites, from ~~56 %, 105~~ 107%, 127%, and ~~91~~ 90% to -~~20 %, -5.1~~ 0.7%, 4.2%, and ~~22~~ 16% over Europe sites, and from 121%, 269%, and 167% to -~~18 %, 40~~ 21%, 37%, and 86% over Asia remote region sites. Comparison with surface measured SO_2, sulfate and black carbon at four Arctic sites indicated that these species simulated with the updated wet processes match well with observations except for a large ~~underestimation~~underestimate of black carbon at one of the sites.~~ Furthermore, we compared~~ We also compare our model simulation with aircraft measurement of nitric acid and aerosols during the ATom-1 and ATom-2 periods and found ~~seasonal variation~~a significant improvement of modeling skill of nitric acid, sulfate, and ~~vertical~~

~~profile~~ammonium in the Northern Hemisphere during winter time. The NMBs of these species ~~have been successfully improved by considering the updated wet processes.~~are reduced from 163%, 78%, and 217% to -13%, -1%, and 10%, respectively. The investigation of impacts of updated wet process treatments on surface mass concentrations indicated that the updated wet processes have strong impacts on the global means of nitric acid, sulfate, nitrate, and ammonium and relative small impacts on the global means of sulfur dioxide, dust, sea salt, black carbon, and organic carbon.

1. Introduction

Aqueous phase chemistry, wet scavenging, and wet surface uptake during dry deposition are the three major atmospheric wet processes for aerosols and aerosol precursors. Aqueous phase chemistry plays a role as reaction chamber which efficiently converts aerosol precursors to aerosols (Ervens et al.,2011; Walcek and Taylor,1986). Wet scavenging, a process by which chemicals accumulate in droplets and then are removed by precipitation, is the ~~predominantly~~predominant removal pathway of aerosols and aerosol precursors (Textor et al., 2006). Dry deposition, where chemicals settle out of the atmosphere in the absence of precipitation, is greatly enhanced due to the absorption of water soluble gases at wet surfaces associated with dew, fog, and rain (Garland and Branson, 1977; Wesely, 1989). These wet processes significantly impact global mass load and redistribute aerosols and aerosol precursors~~ and aerosols. Since aerosol~~. Aerosol mass load and its global distributions are important for ~~the ~~studies ~~on~~of aerosol optical properties (Kinne et al., 2006), aerosol direct radiative forcing (Myhre et al., 2013; Penner et al., 1994), and ~~particle matter's~~the health effects of particulate matter (Shiraiwa et al., 2017; Hopke et al., 2006~~), a~~). A better representation of wet processes in global modeling of aerosols and aerosol precursors ~~and aerosols is important~~can therefore enhance our ability to accurately simulate these different aerosol impacts.

GEOS-Chem is a widely used community model which is continuously being improved (Holmes et al., 2019; Keller et al., 2014; Martin et al., 2003; Bey et al., 2001). Luo et al. (2019), L2019 ~~thereafter~~hereafter, updated the GEOS-Chem wet scavenging scheme by using the Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) spatially and temporally varying cloud and rain water to replace the assumption of fixed in-cloud condensation water (ICCW) in the GEOS-Chem rainout parameterization and by using new empirical rates for nitric acid and water soluble aerosols in washout. These changes together reduced the normalized mean biases (NMB) of simulated nitric acid, nitrate, and ammonium mass concentrations at the United States' surface monitoring networks from 145%, 168%, and 81% to 24%, 25%, and 13%, respectively. However, the impacts of the updated wet scavenging scheme on ~~the~~ simulations over other regions (Europe, Asia, and remote areas) and free ~~atmosphere~~troposphere were not investigated. Moreover, L2019 only investigated the changes of nitric acid, nitrate, and ammonium. The impact of the updated wet scavenging scheme on other aerosols such as sulfate, sea salt, dust, and carbonaceous aerosols ~~were~~was not investigated in that work. Due to the large impact of updated wet scavenging on model simulations, a comprehensive validation of simulated aerosols and aerosol precursors~~ and aerosols~~ with ground based monitoring networks for surface mass concentrations and aircraft measurements for vertical profiles is needed.

In this study, we further update the treatments of wet processes (aqueous chemistry, wet scavenging, and wet surface ~~uptakes~~uptake during dry deposition) in GEOS-Chem and evaluate comprehensively simulated major inorganic aerosol precursors (sulfur dioxide, nitric acid, and ammonia) and aerosols (sulfate, nitrate, ammonium, black carbon, and organic carbon) by comparison with a large set of in-situ observations. The updates to the wet processes are detailed in Section 2. Comparisons of simulations with measurements from surface monitoring networks including the United States Environmental Protection Agency (USEPA), the Interagency Monitoring of Protected Visual Environments (IMPROVE), the Chemical Speciation Network (CSN), the Clean Air Status and Trends Network (CASTNET), the Ammonia Monitoring Network (AMoN), the National Trends Network (NTN), the European Monitoring and Evaluation Programme (EMEP), and the Acid Deposition Monitoring Network in East Asia (EANET) are given in section 3.1. Validations of aerosols and aerosol precursors ~~and aerosols at~~for the Arctic and the Atmospheric Tomography (ATom) mission are presented in sections 3.2 and 3.3. The impact of the updated wet processes on global surface ~~mass~~ concentrations or aerosols and aerosol precursors are discussed in section 3.4. A summary of our results is given in section 4.

2 Updates of wet process treatments in GEOS-Chem associated with aerosol precursor and aerosol modeling

In the ~~public~~publicly released GEOS-Chem version 12.6.0, GC12 thereafter, in-cloud aqueous phase chemistry ~~in cloud~~ was developed by Chin et al. (2000) for SO_2. ~~Wet~~The wet scavenging scheme, including rainout due to formation of precipitation from clouds and washout due to falling precipitation from upper layers, was developed by Jacob et al. (2000) and Liu et al. (2001) for aerosols and by Amos et al. (2012) for gases. Scavenging of aerosol by snow and cold–mixed precipitation was updated by Wang et al. (2011, 2014). Wet surface ~~uptakes~~uptake during dry deposition is represented with constant values of effective Henry's law coefficient for surface resistance ~~calculation~~calculations (http://wiki.seas.harvard.edu/geos-chem/index.php/Physical_properties_of_GEOS-Chem_species#Definition_of_Henry.27s_law_constants).

L2019 showed that the assumption of in-cloud condensation water with a fixed value (1 $g\cdot m^{-3}$) in the rainout parameterization in GC12 is one of the major reasons causing an overestimate in nitrate and ammonium mass concentrations compared to surface monitoring networks over the US. After replacing the fixed value of in-cloud condensation water with MERRA-2 cloud and rain water, we get an updated equation for rainout loss fraction (Luo et al., 2019):

$$F = \frac{P_\mathrm{r}}{k \cdot \mathrm{ICCW}}\left(1 - e^{-k \cdot \Delta t}\right) = \frac{f_\mathrm{c} \cdot P_\mathrm{r}}{k\left(\mathrm{LCW} + \mathrm{ICW} + P_\mathrm{r} \cdot \Delta t\right)}\left(1 - e^{-k \cdot \Delta t}\right), \quad (1)$$

where F is the fraction of a water-soluble tracer in the grid-box scavenged by rainout, Δt (s) is the model integration time step. k is the first-order rainout loss rate which represents the conversion of cloud water to precipitation water. ~~f_c,~~ ICCW ($g\cdot m^{-3}$) is in-cloud condensation water. P_r ($g\cdot m^{-3}\cdot s^{-1}$~~), and~~) is the rate of new precipitation formation. f_c, LCW ($g\cdot m^{-3}$), and ICW ($g\cdot m^{-3}$) are the grid-box mean cloud fraction, ~~the rate of new precipitation formation, and~~ , liquid phase cloud water content, and ice phase cloud water content, respectively.

L2019 also showed that the difference between observations and simulations can be further reduced, through (1) the update of empirical washout coefficients by rain for water-soluble aerosol with the value which was calculated by the parameterization of Laakso et al. (2003) for a 500 nm particle diameter, and (2) the new estimated washout coefficients for nitric acid by referring to field measurements for particles with a 10 nm diameter (Laakso et al., 2003) and the theoretical dependence of scavenging coefficients on particle sizes for particles < 10 nm (Henzing et al., 2006). L2019 only focused on warm cloud wet scavenging, and did not systematically consider the impact of wet process treatments on the simulated aerosols and aerosol precursors~~ and aerosols.~~. Here we show that a number of treatments in GC12 and L2019 can be further updated (as detailed below) to improve the performance of GEOS-Chem in simulating spatial and temporal variations of major aerosols and aerosol precursors ~~and aerosols in~~on a global scale.

2.1 pH for cloud, rain, and wet surface

Water pH is important for dissolution and subsequent aqueous phase reactions of water-soluble gases (Turnock et al., 2019; Ervens, 2015; Pandis and Seinfeld, 1989). Based on Henry's law, dissolution of water-soluble gases can be calculated as:

$$f_w = 1 - \frac{1}{1 + H^* \cdot R \cdot T \cdot \mathrm{LW}}, \quad (2)$$

where f_w is the dissolution fraction for water-soluble gases, H^* (mol·L^{-1}·atm^{-1}) is effective Henry's law constant, R (0.08205 L·atm·K^{-1}·mol^{-1}) is the gas constant, T (K) is the temperature, and LW (m^3·m^{-3}) is the liquid water content.

H^* represents the impact of temperature, water ~~acidy~~acidity, and aqueous phase equilibrium on solubility of water-soluble species (Seinfeld and Pandis, 2016). For SO_2, H_2O_2, and NH_3, which are important for aerosol precursor and aerosol simulation, H^* can be calculated as (Seinfeld and Pandis, 2016):

$$\left.\begin{aligned}
&\mathrm{H}^*_{\mathrm{SO2}} = \mathrm{H}_{\mathrm{SO2}}\left(1 + \frac{K_1}{[\mathrm{H}^+]} + \frac{K_1 \cdot K_2}{[\mathrm{H}^+]^2}\right),\\
&\mathrm{H}_{\mathrm{SO2}} = 1.22 e^{10.55\left(\frac{298.15}{T} - 1\right)},\\
&K_1 = 1.3 \times 10^{-2} e^{6.75\left(\frac{298.15}{T} - 1\right)},\\
&K_2 = 6.31 \times 10^{-8} e^{5.05\left(\frac{298.15}{T} - 1\right)}
\end{aligned}\right\} \quad (3)$$

$$\left.\begin{aligned}
&\mathrm{H}^*_{\mathrm{H2O2}} = \mathrm{H}_{\mathrm{HO2}}\left(1+\frac{K_3}{[\mathrm{H}^+]}\right),\\
&\mathrm{H}_{\mathrm{H2O2}} = 8.3\times10^{4}e^{24.82\left(\frac{298.15}{T}-1\right)},\\
&K_3 = 2.2\times10^{-12}e^{12.52\left(\frac{298.15}{T}-1\right)}
\end{aligned}\right\}\quad(4)$$

$$\left.\begin{aligned}
&\mathrm{H}^*_{\mathrm{NH3}} = \mathrm{H}_{\mathrm{NH3}}\left(1+\frac{K_5[\mathrm{H}^+]}{K_4}\right),\\
&\mathrm{H}_{\mathrm{NH3}} = 59.8e^{14.1\left(\frac{298.15}{T}-1\right)},\\
&K_4 = 1.\times10^{-14}e^{-22.5\left(\frac{298.15}{T}-1\right)},\\
&K_5 = 1.7\times10^{-5}e^{-14.5\left(\frac{298.15}{T}-1\right)}
\end{aligned}\right\}\quad(5)$$

where H_{SO2}, H_{H2O2}, and H_{NH3} are the Henry's law constants (M atm^{-1}) for SO_2, H_2O_2, and NH_3, respectively. K_1~~,~~ (M), K_2~~,~~ (M), K_3~~,~~ (M), K_4~~,~~ (M^2), and K_5 (M) are rate coefficients for SO_2 reaction, HSO_3^- reaction, H_2O_2 reaction, H_2O reaction, and NH_3 reaction, respectively. The values of the Henry's law constants and rate coefficients are the same as those used in GEOS-Chem aqueous phase chemistry. $[H^+]$ (M) is the hydrogen ion concentration in cloud/rain droplets and at wet surfaces, which is related to pH as:

$$[\mathrm{H}^+]=10^{-\mathrm{pH}},\quad(6)$$

GC12 calculates cloud water pH iteratively by using the concentrations of sulfate, total ammonium (ammonium + ammonia), total nitrate (nitrate + nitric acid), SO_2, and CO_2 based on their effective Henry's law coefficients and cloud liquid water content in corresponding grid box (Alexander et al., 2012). This iterative calculation is updated to use Newton's method in order to arrive at a consistent result (Moch et al., ~~2019).~~2020). To implement Newton's method the equilibrium expressions for the concentrations of each soluble semi-volatile ion (SSVI) in terms of H+ and the derivatives for these equilibrium expressions are each solved explicitly so that the Newton's method equation is in the form of:

$$\mathrm{H}^+_{n+1}=\mathrm{H}^+_{n}+\frac{\left[\mathrm{SSVI}\left(\mathrm{H}^+_{n}\right)\right]+[\mathrm{SNVI}]}{\frac{d}{d\mathrm{H}^+}\left[\mathrm{SSVI}\left(\mathrm{H}^+_{n}\right)\right]},\quad(7)$$

where SNVI is the concentrations of soluble nonvolatile ions. For equation (7) the concentrations of each ion are multiplied by the ion charge (e.g. the terms for SO_3^{2-} concentrations are multiplied by -2).

In tests with this new calculation the solution always converged to an answer in less than 20 iterations, but if a maximum of 50 iterations is reached we set it so that the last two solutions are averaged together. We here considered the solution to converge if the difference between H_n^+ and H_{n+1}^+ was less than 0.01. By default the initial guess for H^+ is set to 4.5, but we tested initial guesses ranging from a pH of 2 to 13 and found no change in the values at which the answer converged.

To represent the ~~removing~~removal of aerosols due to rainout, GC12 assumes 30~~-~~% of sulfate, nitrate, and ammonium are removed away from cloud water before cloud water pH calculation. To take into account the variations in the amount of these species rained out, we ~~propose to directly~~ use the real-time rainout fractions for corresponding species which are calculated during the treatment of wet scavenging to replace this constant value (i.e., 30%). Additionally, in GC12, sulfate is assumed to be the only ~~soluble nonvolatile ion (~~SNVI~~)~~ in cloud water, while ammonium and nitrate are treated as volatile species similar to ammonia and nitric acid:

$[\mathrm{SNVI}] = 2\left[\mathrm{SO}_4^{2-}\right]$, (~~7~~8)

Previous studies found that observed ammonium-sulfate aerosol molar ratio is lower than 2 over the US (Silvern et al., 2017; Hidy et al., 2014). Guo et al. (2018) found ammonium-sulfate aerosol molar ratio during the Wintertime Investigation of Transport, Emissions, and Reactivity (WINTER) study to be 1.47±0.43 and pointed out that this phenomena indicates an important role of soluble nonvolatile cations in aerosol thermodynamics. To reflect the impact of soluble nonvolatile cations on cloud water pH, we assume that total amount of soluble nonvolatile cations associated with aerosol thermodynamics (SNVC) is 25~~-~~% of sulfate. We also consider the contribution of calcium and magnesium based on simulated dust mass in GC12, assuming that 3% of dust mass is soluble calcium and 0.6% is soluble magnesium~~,~~ (Farlie et al., 2010; Moch et al., ~~2019)~~2020) , to soluble nonvolatile ions (SNVI):

$[\mathrm{SNVI}] = 2\left[\mathrm{SO}_4^{2-}\right] - 2[\mathrm{SNVC}] - 2\left[\mathrm{Ca}^{2+}\right] - 2\left[\mathrm{Mg}^{2+}\right]$, (~~8~~9)

Rainwater pH, which is used for the calculation of ~~water-soluble gases'~~ effective Henry's law constants ~~for~~of water-soluble gases in rain droplets (Eqs. 3-5), is assumed to be a constant value of 4.5 in GC12. Rainwater pH is determined by the cloud water pH where the rain is produced, ~~uptakes~~uptake of water and ions during rainfall processes, and evaporation of rain droplets. In addition, rainwater pH also depends on temperature (Smith and Martell, 1976). Although it is difficult to fully trace rainwater pH in the model based on current available information in GC12, we use cloud pH at where rainout occurs to represent rainwater pH for rainout process and rainwater-mass-weighted cloud pH above where washout occurs to represent rainfall water pH for washout process in this work. The calculated rainwater pH in this study varied from 4.3 to 6.9.

pH values also affect dry deposition of water-soluble gases ~~at wet surface~~ via its impact on the ~~uptakes~~uptake due to dissolution~~, therefore the~~ at wet surfaces. The origin of surface water where this uptake occurs is therefore important~~.~~ to account for the effect if varying pH. GC12 calculated effective Henry's constant for dry deposition by assuming temperature of 298.15 K and leaf water pH of 7. Surface water ~~of~~on land is dominated by leaf water whose pH is ~7. The pH of ocean surface water varies from 8 to 8.5 (Antonov, 2010; Jacobson, 2005). de Caritat et al. (2005) found the pH of the meltwaters of the Arctic snow varies from 4.6 to 6.1 with median value of 5.4. ~~So we~~We assume the pH values at wet surface are 7 for land, 8.2 for ocean, and 5.4 for snow in this work.

2.2 Fraction of cloud available for aqueous phase chemistry

In GC12, the fraction of cloud available for aqueous phase chemistry is assumed to be 100~~-~~% of grid box cloud fraction when temperatures are above 258 K and 0~~-~~% of grid box cloud fraction when temperatures are below 258 K. ~~It~~This means aqueous phase chemistry in mixed ~~cloud~~clouds where temperatures are often below 258 K is not considered in GC12. However, many studies have indicated that supercooled cloud water can exist when temperatures are above 237 K (Rosenfeld and Woodley, 2000; Sassen, 1985). Therefore, we~~ propose to~~ calculate aqueous phase cloud fraction based on MERRA-2 cloud liquid content and cloud ice content when temperatures are higher than 237 K~~ and lower than 263 K~~:

$$f_{\mathrm{aq}} = f_{\mathrm{c}} \frac{\mathrm{LCW}}{\mathrm{LCW+ICW}},\ (\mathrm{T} > 237\ \mathrm{K})$$, (10~~9~~)

where f_{aq} is aqueous phase cloud fraction, LWC (g m^{-3}) is grid box mean liquid phase cloud water content, and ICW (g m^{-3}) is grid box mean ice phase cloud water content.

~~The surface pre-melt layer or quasi-liquid layer occurs when temperatures are above 263 K (Nenow, 1984; Ocampo and Klinger, 1983). Conklin et al. (1993) suggested that the ice surface can be modeled as an aqueous phase when temperatures are higher than 265 K. So for temperature higher than 263 K, we assume aqueous phase cloud fraction equals grid mean cloud fraction:~~

~~$f_{\mathrm{aq}} = f_{\mathrm{c}}$, (T > 263 K) , (10)~~

~~The 263 K cut-off used here is to reflect the cover of meltwater on ice when temperature is not too low.~~

2.3 Rainout efficiencies

2.3.1 Warm cloud

GEOS-Chem ~~used~~uses rainout efficiencies to represent the absorptions of water-soluble gasses and aerosols in the cloud condensate phase (Jacob et al., 2000; Mari et al., 2000; Liu et al., 2001). After applying ~~them~~these efficiencies with the updated parameterization for rainout loss fraction (Luo et al., 2019), we get the new equation as

$$F = \frac{f_{\mathrm{c}} \cdot P_{\mathrm{r}}}{k\left(\mathrm{LCW+ICW} + P_{\mathrm{r}} \cdot \Delta t\right)}\left(1 - e^{-E_{\mathrm{r}} \cdot k \cdot \Delta t}\right), (11)$$

where E_r is the rainout efficiency for corresponding species. Eq. (11) is the same as Eq. (1) except Eq. (11) contains E_r in the rainout calculation.

In GC12, rainout efficiencies for water-soluble aerosols are assumed to be 100~~-~~% while those for water-soluble gases, except nitric acid and SO_2, are calculated via Henry's law constants (Jacob et al., 2000). E_r of nitric acid is assumed to be the same as water-soluble aerosols due to its high solubility. ~~Due to the low solubility of SO_2 in water, rainout of SO_2 is limited by the aqueous phase oxidation of SO_2 by H_2O_2 rather than the absorption by cloud water (Chin et al., 1996). E_r of SO_2 in GC12~~E_r of SO_2 is assumed to be the same as water-soluble aerosols but limited by the availability of H_2O_2 in the precipitating grid box~~.~~ (Chin et al., 1996). It means rainout of SO_2 in GC12 is attribute to the aqueous phase oxidation of SO_2 by H_2O_2 rather than the absorption by cloud water. However, GEOS-Chem already accounted for in-cloud oxidation of SO_2 as part of the aqueous phase chemical calculation which converts in-cloud SO_2 to sulfate, so doing the same in the scavenging calculation would be double-counting the removal of SO_2. Considering the low solubility of SO_2 in water, it is more appropriate to calculate rainout efficiency for SO_2 based on Henry's law. In the present work, we assume E_r of SO_2 equals its dissolution fraction:

$$E_{r\_SO2} = f_{w\_SO2}, \quad (12)$$

with f_{w_SO2} calculated with Eq. (2).

In the present work, we also modified rainout efficiencies for hydrophilic black carbon (BC) and primary organic carbon (POC), from 100% in GC12 to 50%. The rationale for the modification is that, although the aging of BC and POC in the atmosphere converts these aerosols from hydrophobic to hydrophilic, they are not as ~~easy to be~~easily activated into cloud droplet as water-soluble aerosols (e.g. sulfate, nitrate, ammonium~~ and so on~~). The composition of the particles ~~decide~~decides the hygroscopic parameter kappa which is important for cloud activation calculation~~.~~ (Abdul-Razzak et al., 2000). If BC and POC are internally mixed with the sulfate, nitrate, ammonium (SNA) aerosols, then they all have similar compositions. However, in the actual atmosphere, many particles are externally mixed: some particles are pure SNA while others are primary particles (BC, POC, dust, etc.) coated with SNA~~.~~ (Fassi-Fihri et al., 1997). It takes time for primary particles to gain coating through condensation, coagulation, and aqueous chemistry. The amount of SNA coated on primary particles depends on the aging time and abundance of SNA in the air. Based on a detailed size and mixing state resolved advanced particle microphysics (APM) simulation which explicitly resolves the amount of SNA coating (Yu et al., 2012), the hygroscopic parameter kappa of coated BC and POC is roughly about half of that of SNA. More robust calculation of rainout efficiencies for BC and POC ~~shall~~should consider the amount of soluble species coated on these particles (Yu et al., 2012; Yu and Luo, 2009~~)~~), but this will be the subject of future work.

2.3.2 Mixed and cold clouds

In GC12, aerosols in mixed ~~cloud~~clouds ($237\ K \le T < 258\ K$) and cold

~~cloud~~clouds (T < 237 K) were assumed to be removed through heterogeneous and homogeneous freezing nucleation (Wang et al., 2014). GEOS-Chem assumed that heterogeneous nucleation dominates ice formation at 237 K ≤ T < 258 K (mixed cloud) and results in 100~~-~~% rainout efficiencies only for dust and hydrophobic black carbon which are considered as ice nuclei (IN). Homogeneous nucleation takes place at T < 237 K (cold cloud) and results in 100~~-~~% rainout efficiencies for both water-soluble aerosol and IN.

Ice nucleation processes and their impacts on aerosol wet scavenging by mixed and cold ~~cloud~~clouds are largely unclear. However, it is known that ice nucleation rates depend strongly on temperature (DeMott et al., 2015; Kanji and Abbatt, 2010). To take into account this, we ~~propose to~~ parameterize rainout efficiencies at warmer ~~temperature~~temperatures based on the fraction of dust in mixed ~~cloud~~clouds contributing to IN, which can be calculated as a function of T according to DeMott et al. (2015) as:

$$E_{\mathrm{r\_mixed\_dust}} = \frac{e^{0.46(273.16-\mathrm{T})-11.6}}{153.5}, \ (237\ \mathrm{K} \le \mathrm{T} < 258\ \mathrm{K}) \ ,(13)$$

In addition to T, ice nucleation efficiency of particles also ~~depend~~depends on their sizes and smaller particles (diameter < 500 nm) are less likely to act as IN (Niedermeier et al., 2015). While most of the mass of dust particles are dominated by those larger than 500 nm~~,~~ (Zender et al., 2003), a significant fraction of BC particles are smaller than 500 nm~~.~~ (Sahu et al., 2012). Based on sectional aerosol microphysics calculation in GEOS-Chem-APM (Yu and Luo, 2009), the mass fraction of BC particles with diameter > 500 nm is ~50~~-~~%. In this study, we assume E_{r} for hydrophobic BC in both mixed cloud (237 K ≤ T < 258 K) and cold cloud (T < 237 K) are 50~~-~~% of those values for dust.

Water-soluble aerosols are 100% removed via homogeneous freezing nucleation in cold cloud (Wang et al., 2014; Liu et al., 2001). Strom et al. (1997) observed that ~ 40~~-~~% of preexisting aerosol mass is incorporated in ice ~~crystal.~~crystals. In this work, we assume cold cloud rainout efficiencies are 40~~-~~% for water-soluble aerosol, 50~~-~~% for hydrophobic black carbon, and 100~~-~~% for dust, respectively. Additionally, rainout of cold clouds is limited to below the MERRA-2 troposphere since stratospheric water in MERRA-2 is known to have unphysical behavior.

In GC12, cold cloud wet scavenging of nitric acid is treated the same as water-soluble aerosol. However, in cold ~~cloud~~clouds (T < 237 K), nitric acid is removed by the ~~partition~~partitioning on ice ~~crystal~~crystals (Kärcher and Voigt, 2006; Voigt et al., 2006), while water-soluble aerosol is removed by homogeneous freezing nucleation. Kärcher et al. (2008) used a climatology of cirrus ice water content together with observed molar ratios of HNO_3/H_2O in cirrus ice particles to estimate the range of nitric acid content in cirrus ice (185-240 K). Their study showed that less efficient nitric acid uptake limits the nitric acid content in cirrus ice at higher temperatures and small ice water contents permit only little nitric acid in ice at low temperatures. ~~Fraction~~The fraction of nitric acid in ice generally increases with decreasing temperature. Kärcher and Voigt (2006) attributed this behavior to less efficient nitric acid trapping at higher temperatures despite faster ice growth rates, which is caused by increasingly rapid escape of adsorbed nitric acid into the gas phase. A parameterization of nitric acid partitioning in cold cloud developed by Kärcher et al. (2008) is employed here to calculate E_r of nitric acid in cold cloud when temperature is below 240 K:

$$E_\mathrm{r} = \frac{10^{-(-26.5\times1.00155^{T}+30.7)}\cdot\frac{63}{18}\cdot[\frac{\mathrm{LCW+ICW}}{f_\mathrm{c}}]_\mathrm{vmr}}{[\mathrm{HNO_3}]_\mathrm{vmr}}, (14)$$

where $[\frac{\mathrm{LCW+ICW}}{f_\mathrm{c}}]_\mathrm{vmr}$ is volume mixing ratio of in-cloud water and in-cloud ice, and $[HNO_3]_\mathrm{vmr}$ is volume mixing ration for nitric acid gas.

~~2.4 In-cloud condensation water for cold cloud~~

~~In GC12, ICCW for cold cloud (T < 237 K) is assumed to have a fixed value of 1 g·m^{-3} which is the same as that of warm cloud. This assumption significantly underestimates wet scavenging due to rainout in cold cloud (T < 237 K). L2019 replaced the fixed ICCW with cloud water and rain water as shown in Equ. 1. However, water-soluble aerosols in cold cloud (T < 237 K) can also exist in ice due to freezing of supercooled water, therefore, we calculate ICCW for cold cloud as:~~

~~$\mathrm{ICCW_c} = \frac{\mathrm{LCW+ICW}+P_\mathrm{r}\cdot\Delta t}{f_\mathrm{c}}$, (15)~~

~~2.5~~2.4 Empirical washout coefficients by rain and snow

Washout coefficients by rain and snow in GC12 were updated by Wang et al. (2011) by adopting the parameterization constructed by Feng (2007, 2009) for individual aerosol modes. Accumulation-mode washout coefficients were used for all aerosols except dust and sea salt, for which the coarse mode coefficients were used. Previous studies noticed that washout rates by rain derived from field measurements are 1 to 2 orders of magnitude larger than the values from theoretical calculation (Wang et al., 2010; Luo et al., 2019). Therefore, L2019 recommended using empirical washout coefficients for the simulation of washout by rain.

Wang et al. (2014) found that the large differences in washout rate between field measurements and theoretical calculation not only appear in washout by rain but also appear in washout by snow. In this work, we use the semi-empirical parameterization developed by Wang et al. (2014) for the calculation of nitric acid and aerosol washout by both rain and snow. Washout rate is calculated by an exponential equation:

$$k_{\mathrm{wash}} = \Lambda \left(\frac{P_{\mathrm{d}}}{f_{\mathrm{r}}} \right)^{b} \text{ , (~~16~~15)}$$

where k_{wash} (s^{-1}) is the washout rate, P_{d} (mm h^{-1}) is rain or snow falling from upper layers, f_{r} is rainfall area fraction, Λ is washout scavenging coefficient, and b is an exponential coefficient.

The values of Λ and b for nitric acid and aerosol washout by rain ($T > 268$ K) and snow ($248 \text{ K} < T < 268$ K) are shown in Table 1. We assume precipitation at temperatures lower than 248 K is dominated by ice. GC12 assumed washout of aerosol by ice is the same as that by snow. However, uptake of aerosol by ice and by snow ~~are~~is different. Schneider et al. (2019) found specific surface area (SSA) of ice crystal is ~1/5 of SSA of snow. Therefore, in this work, we roughly assume washout rate by ice ($T < 248$ K) is 1/5 of that by snow. Washout of nitric acid uses the same values as in ~~the work of Luo et al. (2019)~~L2019 but we extend the temperature limitation from 268 K to 248 K. Washout of nitric acid by ice is assumed to be 1/5 of that by snow. Empirical washout coefficients by rain and snow for coarse aerosol and hydrophobic fine aerosol in this work are based on the values in Wang et al. (2014). 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, we used the value of 1×10^{-5} to replace 5×10^{-7} for hydrophilic aerosol's washout by rain. The washout coefficient of hydrophilic aerosol by snow is replaced with the value of 2×10^{-4} which is 20 times higher than the value by rain. ~~Washout by ice is assumed to be 1/5 of that by snow.~~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 larger than that calculated by the semi-empirical parameterization. One of the possible reasons is that droplet–particle collection mechanisms for hydrophobic and hydrophilic aerosols are different. Washout by ice is assumed to be 1/5 of that by snow.

2.~~6~~5 Wet surface ~~uptakes~~uptake during dry deposition

~~Uptakes~~Uptake of water-soluble gases at wet ~~surface are~~surfaces is strongly influenced by dissolution processes. The solubility of SO_2, H_2O_2, and NH_3 at wet ~~surface~~surfaces needs to be calculated via effective Henry's law coefficient because it is associated with a series of aqueous phase reactions (Seinfeld and Pandis, 2016). In GC12, H^* of SO_2, H_2O_2, and NH_3 for dry deposition are assumed to be the constants with the values of 10^5 M atm^{-1}, 5×10^7 M atm^{-1}, and 2×10^4 M atm^{-1}, respectively (http://wiki.seas.harvard.edu/geos-chem/index.php/Physical_properties_of_GEOS-Chem_species#Definition_of_Henry.27s_law_constants). In this work, we consider the impacts of temperature and pH at wet surface on the values of H^* (Erisman et al., 1994; Wesely et al., 1990), and the values of H^* for SO_2, H_2O_2, and NH_3 are calculated with equations (3-5). Wet surface pHs discussed in section 2.1 are used to reflect the impact of wet surface ~~acidy~~acidity on dissolution during dry deposition. Wet surface pHs are only determined by land type and are not altered by precipitation. Ganzeveld et al. (1998) reported that observations and physical-chemical model simulations indicated SO_2 dry deposition velocity increases from a minimum value of 0.01 cm s^{-1} for a temperature of 253 K to a value of 0.15-0.25 cm s^{-1} for 273 K. Therefore, in this work, we assume SO_2 dry deposition velocity over snow and ice is 0.01 cm s^{-1} when temperatures are lower than 253 K.

3. **Results and discussions**

To investigate the impacts of updated wet processes on global simulation of aerosols and aerosol precursors~~ and aerosols~~, we run GEOS-Chem for 3 cases: (1) standard Geos-Chem version 12.6.0, called GC12; (2) the same as case GC12 except using wet scavenging described in the work of Luo et al. (2019), and this case is named L2019; (3) the same as the case L2019 except considering the updated wet processes described in section 2, and this case is called WETrev. All simulations are run with 2°×2.5° horizontal resolution and 47 layers from surface to 0.01 hPa. ~~Emissions~~Emission over Europe is based on the EMEP inventory. Other emissions are produced by the default setting of HEMCO (Keller et al., 2014~~).~~) for all simulations presented in this work.

3.1 Comparison with surface monitoring networks over the US, Europe, and ~~East~~ Asia

~~Figure 1 and Table 2~~To validate model results with surface monitoring networks, we use observational data taken at USEPA, CASTNET, AMoN, IMPROVE and CSN, EMEP and EANET sites. The criterion of observations used for model validation is that valid data are available for every month in 2011. For EANET observations, due to too much missing data, the criterion is loosened to monthly mean data available for each month during a 3-year period (2010-2012). Seto et al. (2007) pointed out that EANET observations at urban sites are much higher than those at remote sites. Since the number of the Asian sites is very limited, to make the validation more appropriate, only remote and rural sites are used for model validation. Table 2 shows number of sites with observations and number of sites satisfying these criteria. Figure 1 and Table 3 present the comparisons of observed secondary inorganic aerosol precursors and secondary inorganic aerosols at surface monitoring networks and the simulated mass concentrations by the GC12, L2019, and WETrev cases described above.~~ At the United States, SO_2, nitric acid, ammonia surface mass concentration measurements were collected from USEPA, CASTNET, and AMoN, respectively. Measurements of secondary inorganic aerosols were collected from IMPROVE and CSN. Surface mass concentrations of secondary inorganic aerosol precursors and secondary inorganic aerosols over Europe and Asia were observed by EMEP and EANET, respectively.~~ As shown in Fig. 1 (a-c), simulated SO_2 for the 3 cases is lower than observed values over the US but higher than the observations over Europe and Asia. Over the US, simulated SO_2 is ~ 20% lower than observations. One possible reason is that a large amount of USEPA observations are located at urban regions where SO_2 concentrations are much higher than rural and remote regions. There were 288 USEPA sites with valid data in each month of 2011. Only 69 of these sites were with the mark of 'Not in a city'. After considering the updates of wet scavenging by L2019, NMBs are increase from 20% to 23% over the US, reduced from ~~101~~ 74% to ~~84~~ 59% over Europe, and reduced from 63% to 43% over Asia, respectively. Considering of updated wet processes in this work further reduces NMBs to ~~73 % at~~51% in Europe and 23~~ % at~~% in Asia, respectively.

Figure 1 (d-f) are the results for nitric acid. NMBs of simulated nitric acid by GC12 for the US, Europe, and Asia are ~~89 %, 44~~ 78%, 107%, and ~~136~~ 121%, respectively. G12~~ simulation~~ significantly overestimates surface mass concentration of nitric acid at these regions. Simulations by L2019 and WETrev indicate that wet scavenging is the dominant process causing the overestimation of nitric acid in GEOS-Chem. NMBs of simulated nitric acid in WETrev for the US, Europe, and Asia are reduced to ~~13 %, -20 %, -18~~ 0.9%, -0.7%, and -21%, respectively. We also notice that WETrev underestimates nitric acid at low temperatures ~~fort~~for the US and Europe sites. These underestimates may be associated with the updated uptake coefficients by Holmes et al. (2019) for heterogeneous chemistry. If we switch back to the old heterogeneous chemistry in GEOS-Chem version 12.5, the underestimation of nitric acid at low temperatures is reduced (not shown). Figure 1 (g-i) show that the biases of model simulated ammonia by the 3 cases over the 3 regions are small.~~ The aqueous concentration of~~ Since the increasing ammonia wet deposition is ~~much lower than~~compensated by less equilibrium partitioning with decreased nitric acid~~, and therefore~~ in the air, wet processes show relatively small impact on the simulation of ammonia.

Figure 1 (j-l) are observed and simulated sulfate ~~at~~over the US, Europe, and Asia. NMBs of the GC12 case over the 3 regions are -1.1~~ %, 27 ~~%, 6.9%, and 5.5%, respectively. The application of updates ~~of~~to wet scavenging in L2019 leads to ~~significantly~~a significant underestimation of sulfate during winter time, reaching up to 50% over the 3 regions. Based on our investigation, we found that the missing of aqueous phase chemistry in mixed cloud appears to be the main reason of underestimated sulfate at low temperatures. As we discussed in section 2, aqueous phase chemistry in GC12 is only simulated when temperatures are higher than 258 K. Conversely, in WETrev case, the temperature limitation of aqueous phase chemistry is extended from 258 K to 237 K. This change allows aqueous phase chemistry to be simulated when temperatures are low. After employing the new approaches of cloud water pH and aqueous phase cloud fraction calculation, NMBs of the WETrev case at the 3 regions are -~~11 %, 15~~ 10%, 4.3%, and ~~-7.0~~ 6.3%, respectively. It significantly reduces the bias shown in the L2019 case. The absence of aqueous phase hydroxymethanesulfonate chemistry may also be a potential reason for the remaining model biases with sulfate, but this is not explored here (Moch et al., 2018). ~~As shown in Figure 1 (m-r), simulated nitrate and ammonium by~~ NMB of sulfate simulated by WETrev in the US is higher than that of GC12. However, the good agreement between GC12 sulfate and the observation can be attributed to the coincidental offsets of the higher sulfate mass due to the underestimation of sulfate wet scavenging and the lower sulfate mass due to the absence of aqueous phase chemistry in mixed cloud and hydroxymethanesulfonate chemistry. As shown in Figure 1 (m-r), simulated nitrate and ammonium in the GC12 case over the 3 regions are much higher than observations. As discussed in Luo et al. (2019), the overestimation is associated with the underestimation of rainout and washout of nitric acid and nitrate. Updated wet scavenging in L2019 successfully reduces NMBs of nitrate over the 3 regions from 126% to 10~~%, 105~~%, 127% to ~~-14~~ 7.5%, and 269% to 47%, respectively. NMBs of ammonium over the 3 regions are reduced from 45% to -13~~%, 91~~%, 90% to -7.3~~.3~~%, and 167% to 42%, respectively. Updated wet processes in this work show relatively small impact on simulated nitrate and ammonium surface mass concentrations over the 3 regions.

For simplicity, the WETrev case includes all updates to wet processes as described in Section 2. To understand the contribution of various updates to the overall changes in the predicted concentrations of aerosols and aerosol precursors, we carry out five numerical sensitivity study cases (RO, WO, RP, DD, and AC). 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. Relative contributions to the changes are calculated as:

$$RC_i = \frac{\sum_{j=1}^{nsite}\left|C_{i,j}\text{-}C_{WETrev,j}\right|}{\sum_{j=1}^{nsite}\left|C_{RO,j}\text{-}C_{WETrev,j}\right| + \sum_{j=1}^{nsite}\left|C_{WO,j}\text{-}C_{WETrev,j}\right| + \sum_{j=1}^{nsite}\left|C_{PR,j}\text{-}C_{WETrev,j}\right| + \sum_{j=1}^{nsite}\left|C_{DD,j}\text{-}C_{WETrev,j}\right| + \sum_{j=1}^{nsite}\left|C_{AC,j}\text{-}C_{WETrev,j}\right|}, \quad (16)$$

where RC is the relative contribution (%), C is simulated surface mass concentration (μg m^{-3}), i is the numerical sensitive study case index (e.g. when i=1, $C_{i,j}$ refers to $C_{RO,j}$), j is the site index.

Relative contributions of RO, WO, RP, DD, and AC to the changes of January and July surface concentrations over the USA, Europe, and Asia sites are summarized in Table 4. In the US, the changes of SO_2 are mainly caused by DD and AC whose contributions are up to 54.2% and 25.0% in January and 50.5% and 22.3% in July. Rainout and washout both show a relatively small impact on the changes of SO_2. In contrast, rainout and washout are important to the changes of nitric acid, sulfate, nitrate, and ammonium. The contribution of wet scavenging to the changes of these species exceeds 50% in both January and July. For nitric acid, nitrate, and ammonium, the contribution of wet scavenging can be as high as 70-90%. For sulfate, AC also plays an important role with relative contributions in January and July of 29.5% and 17.5%, which is comparable to the contributions of RO and WO. For ammonia, most of the changes are caused by DD and AC, with the sum of the 2 processes contributing > 50% of the changes. The contribution of RP to SO_2, sulfate, ammonia, and ammonium is small in January and large in July. In July the contribution of RP to SO_2, sulfate, ammonia, and ammonium is 8.5%, 4.4%, 13.4%, and 4.1%, respectively. The relative contribution from RO, WO, RP, DD, and AC at the sites over Europe and Asia are similar to those over the US (Table 4).

Figure 2 is ~~the~~a comparison of observed BC and OC over the US and Europe. Simulated BC over the US is close to ~~the~~ observations except for a 10-20% underestimate during summer and fall. The underestimate is likely associated with the underestimated wildfire emissions in the western US~~.~~ (Mao et al., 2015). Simulated OC over the US is close to ~~the~~ observations during summer but 50-60% lower than observations during spring and fall. GEOS-Chem (all three cases) significantly ~~underestimated~~underestimates BC and OC over Europe and the possible reasons behind the bias remain to be investigated. NMBs of the ~~2 species~~BC and OC in Europe are ~~high~~ up to -37~~ ~~% and -61~~ ~~%, respectively. The differences of simulated BC and OC in the 3 cases are small for the US and Europe which indicates wet processes have a small impact on the simulation of BC and OC in these regions. The small impact of wet processes on BC in the US and Europe is because 80~~ ~~% of emitted BC is assumed to be hydrophobic aerosol which needs 1.15 days to be converted to hydrophilic BC. Updated wet processes ~~has~~have little impact on hydrophobic aerosol in ~~low~~the lower troposphere where wet scavenging is dominated by warm ~~cloud.~~clouds. OC consists of primary organic aerosol (POA) and SOA which is formed through the oxidation of organic gaseous precursors. Due to low ~~solution~~dissolution of POA and organic gaseous precursors in water, wet processes ~~can~~also have little impact on these species.

Wet deposition of simulated SO_2+SO4, HNO_3+NIT, and NH_3+NH4 are compared with NTN observations over the US (Fig. 3), EMEP observations over Europe (Fig. 4), and EANET observations over remote region in Asia (Fig. 5). The criteria of observations used for model validation are (1) valid data are available for each month in 2011and (2) the difference between observed and simulated monthly precipitation is within a factor of 4 (Paulot et al., 2014). Number of sites with observations and number of sites satisfying these criteria are shown in Table 5. For the comparison shown in Table 6, model simulated wet depositions are corrected following Paulot et al. (2014) to remove bias due to precipitation. As shown in Figure 3 and Table 6, GC12 underestimates SO_2+SO4 wet deposition over the US and Europe. NMBs of SO_2+SO4 wet deposition simulated by GC12 over the two region are -21% and -46%, respectively. After considering the updated wet processes in WETrev, NMBs of SO_2+SO4 wet deposition are reduced to -9.0% over the US and -6.2% over Europe, respectively. However, all the three cases significantly underestimate SO_2+SO4 wet deposition over Asia. One possible reason is that GEOS-Chem may underestimate eruptive volcanic emission nearby the four Japanese sites. For HNO_3+NIT wet deposition over the US, the values simulated by GC12 are close to observations, while the values simulated by WETrev are ~ 2 times higher than observations. However, wet deposition data are collected weekly at NTN sites. It is hard to estimate the uncertainty due to the evaporation of HNO3 from the collected precipitation water. Over Europe and Asia, wet deposition fluxes are observed daily at most of EMEP and EANET sites. The values of HNO_3+NIT wet deposition simulated by GC12 are lower than observations, while the values simulated by WETrev are higher than observations. For NH_3+NH4, GC12 underestimates wet deposition over the US, Europe, and Asia. NMBs over the 3 regions are -10%, -33%, and -10%, respectively. NMBs of NH_3+NH4 wet deposition simulated by WETrev are reduced to -7.7% over Europe and -2.5% over Asia, respectively.

3.2 Comparison of SO_2, sulfate and BC mass concentrations at Arctic sites

We also ~~studied~~study the impact of updated wet processes on SO_2, sulfate and BC surface mass concentrations at several Arctic sites where measurements are available. Figure ~~3~~6 shows the comparison of SO_2 at Nord (81.6ºN, 16.7ºW) and Zeppelin (78.9ºN, 11.9ºE). GC12 ~~case~~ matches well with the observed SO_2 at Nord but ~~3 times overestimated~~overestimates SO_2 at Zeppelin in January and December by a factor of 3. The updated wet scavenging (yellow line) shows a small impact on simulated SO_2 ~~simulation at~~ in the Arctic~~. Simulated~~., with simulated SO_2 ~~is~~ reduced slightly ~~reduced~~ during winter and spring. In WETrev ~~case~~, we assumed SO_2 dry deposition velocity is 0.01 cm s^{-1} when temperatures are lower than 253 K. ~~It~~WETrev slightly enhances SO_2 at the higher latitude site Nord during winter. At Zeppelin, temperature in December is higher than that in January and February, and SO_2 concentration 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 SO_2. By switching from GC12 to WETrev, NMB of SO_2 is increased from -23% to 32% at Nord and decreased from 27% to 22% at Zeppelin. Figure ~~4 is~~7 compares the observed and simulated sulfate and BC at Alert (82.5ºN, 62.5ºW), Barrow (71.3ºN, 156.6ºW), and Zeppelin. Observations at the ~~3~~three sites show that both sulfate and BC are high in spring and low in summer. ~~Model simulation~~ The model simulations generally ~~captures~~capture seasonal variation at these Arctic sites. However, GC12 overestimates sulfate mass concentration at the 3 sites by a factor of 2-3. Simulated BC by GC12 is 50~~–~~% lower than observation at Alert during winter and spring and a factor of 2 higher than observations at Barrow and Zeppelin during winter. Updated wet scavenging significantly impacts simulated sulfate and BC in Arctic regions. Simulated sulfate by L2019 is much closer to observations except for a 50~~-~~% underestimation at Alert during winter and spring, while simulated BCs at the 3 Arctic sites by L2019 is much lower than observations. The comparison with model results from WETrev shows the underestimation of sulfate at Alert during spring is compensated by considering aqueous phase chemistry in mixed clouds. Most of BC at Arctic regions is transported from middle-low latitude source regions with open fire and anthropogenic emissions~~,~~ (Xu et al., 2017), and during the long-range transport hydrophobic BC is aged and ~~covered~~converted to hydrophilic BC. The assumption of reduced hydrophilic BC rainout efficiency in the WETrev case increases simulated BC mass concentration and enhances agreement with observations at these Arctic sites. NMBs of BC are reduced from -67% to -40% at Barrow and from -75% to -46% at Zeppelin due to the switch from L2019 to WETrev.

3.3 Vertical profiles of nitric acid and aerosols: Comparison with ATom-1 and ATom-2 aircraft measurements

To evaluate the impact of updated wet processes on simulated vertical profiles of aerosols and aerosol precursors~~ and aerosols~~, we compare simulated nitric acid and aerosols for the 3 cases with the aircraft measurements ~~during~~of ATom-1 in July-August 2016 and ATom-2 in January-February 2017 over the ~~North~~Northern Hemisphere (Fig. ~~5~~8) and the ~~South~~Southern Hemisphere (Fig. ~~6~~9). Flight tracks over the land or in the stratosphere are filtered out for the comparison~~.~~ (see Figure S1 in supporting materials for flight tracks of ATom-1 amd ATom-2). We filter out the flight tracks over the land is because ATom observations over the land, whose values vary greatly, only account for 28% of total measurements. The exclusion of these data makes the comparison more appropriate. Vertical profiles of nitric acid and aerosols over the land, which are similar to Fig. 8 and 9, are shown in Figure S2.

As shown in Figure ~~5~~8, GC12 overestimates nitric acid and underestimates black carbon and organic carbon over the ~~North~~Northern Hemisphere during both ATom-1 and ATom-2. NMBs of the 3 species are 66%, -77%, and -55% during ATom-1 and 163%, -10%, and -27% during ATom-2. GC12 simulated sulfate and ammonium match well with observations during ATom-1 but are much higher than observations during ATom-2~~.~~ whose values are high up to 78% for sulfate and 217% for ammonium. After considering the updated wet scavenging in ~~Luo et al. (2019),~~L2019, the ~~overestimations~~overestimates of nitric acid, sulfate, and ammonium during ATom-2 and nitric acid during ATom-1 are reduced~~.~~ to 5%, -11%, -30%, and -36%, respectively. However, L2019 significantly underestimates nitric acid at the upper troposphere where pressure is lower than 300 hPa. As we mentioned earlier, L2019 may overestimate cold cloud wet scavenging of nitric acid due to ~~the old treatments in GC12.~~ treatment of cold cloud rainout of nitric acid as same as water-soluble aerosol with 100% rainout efficiency. With updated cold cloud scavenging in WETrev, the bias of nitric acid simulated by L2019 at the upper troposphere is reduced during ATom-2 and is enhanced during ATom-1. This indicates further understanding regarding ice uptake and removal of nitric acid are needed. Nitric acid concentrations simulated by WETrev between 500 hPa and 300 hPa are much lower than those simulated by L2019 and GC12. This is because WETrev considers washouts of nitric acid by snow and ice which were absent in L2019 and GC12. Figure ~~5~~8 (g) shows the impact of updated aqueous phase chemistry in mixed ~~cloud~~clouds on the sulfate vertical profile. Considering aqueous phase chemistry in mixed ~~cloud~~clouds significantly enhances sulfate mass concentration within the range of 700-500 hPa during ATom-2 which makes the simulated sulfate much closer to observed values. Figures ~~5~~8 (d) and (i) indicate that the impact of updated wet scavenging on the black carbon vertical profile during ATom-2 is more obvious than that during ATom-1. ~~It~~This is because there is much less black carbon emitted from open ~~fire~~fires in January ~~is much less~~ than ~~that~~there is in July. Black carbon observed during ATom-2 is dominated by hydrophilic black carbon which is more affected by wet scavenging processes, while black carbon observed during ATom-1 is dominated by hydrophobic black carbon. Updated wet scavenging shows a small impact on organic carbon vertical profiles during both ATom-1 and ATom-2.

Figure ~~6~~9 shows ~~the~~ comparisons over the ~~South~~Southern Hemisphere. Updated wet scavenging reduces overestimated nitric acid especially during ATom-1 period. NMB is reduced from 80% to -25%. For sulfate, ammonium, black carbon, and organic carbon, the differences among the 3 cases are relative small. NMBs of WETrev for these species are larger than those of GC12. All cases significantly underestimate black carbon from open fire and organic carbon in the upper troposphere~~ organic carbon.~~. Based on the comparisons with ATom-1 and ATom-2 measurements, it is clear that the updated wet process treatments in this work and L2019 can improve the agreements of simulated and observed vertical profiles of nitric acid ~~and aerosols~~(Fig. 8a, Fig. 8f, Fig. 9a, and Fig. 9f). The simulated of winter time sulfate and ammonium in the Northern Hemisphere are also improved by WETrev.

3.4 Impact on global distributions of surface mass concentrations

The impacts of updated wet process treatments on global simulation of surface mass concentrations are shown in Figures ~~7-11.~~10-14. Figures ~~7-9 are~~10-12 show simulated surface mass concentrations of secondary inorganic aerosol precursors (SO_2, nitric acid, and ammonia), secondary inorganic aerosols (sulfate, nitrate, and ammonium), primary inorganic aerosols (sea-salt, dust, and black carbon), and organic carbon (primary organic aerosol and secondary organic aerosol) simulated by GC12 case and WETrev case, while figures ~~10-11~~13-14 are the percentage differences.

As shown in Figure ~~7~~10, high values of secondary inorganic aerosol precursors are mainly located ~~at~~over continental regions with high anthropogenic and natural emissions. After considering the updated wet process treatments in this study, global mean surface mass concentrations (GMSMC) of SO_2, nitric acid, and ammonia are changed from 0.~~76~~73 μg m^{-3}, 0.~~55~~56 μg m^{-3}, 0.32 μg m^{-3} to 0.~~78~~75 μg m^{-3}, 0.~~28~~26 μg m^{-3}, 0.~~43~~42 μg m^{-3}, respectively. The updated wet process treatments slightly impact GMSMC of SO_2 but strongly impact GMSMC of nitric acid. The impact on ammonia is small over land but ~~strong~~large over ocean. The weak impact of the updated wet process treatments on SO_2 is because its wet removal is dominated by aqueous phase chemistry. The strong impact of the updated wet process treatments on ammonia over ocean is due to the changes of rainwater pHs over remote regions whose values are higher than the assumed 4.5 rainwater pH in GC12. Some large changes of surface mass concentration at Arctic and Antarctic regions, as shown in Figure ~~10~~13 (a-c), are associated with the updated treatments of wet surface ~~uptakes~~uptake during dry deposition at snow and ice. However, due to low mass concentrations ~~at~~for Arctic and Antarctic regions, their impacts on GMSMC are small. The updated wet process treatments ~~exhibit significant~~significantly impact~~ on~~ GMSMC of secondary inorganic aerosols whose water solubility is high. After considering the updated wet process treatments, GMSMC of sulfate, nitrate, and ammonium are changed from 0.~~87~~84 µg m^{-3}, 0.~~41~~42 µg m^{-3}, 0.~~34~~33 µg m^{-3} to 0.~~76~~74 µg m^{-3}, 0.~~22~~21 µg m^{-3}, 0.~~27~~26 µg m^{-3}, respectively. Their global mean relative changes are high up to -25~~%, -51~~%, -53%, and -22%, respectively. Most of the reductions of these species happen at middle-high latitude regions with high mass concentrations.

Figures ~~9~~12 and ~~11~~14 show the ~~impacts~~impact of updated wet process treatments on primary inorganic aerosols and organic carbon. It is clear that the updated wet process treatments have little impact on GMSMCs of these species. For sea salt, its high concentrations are mainly located at middle latitude regions in both the ~~North~~Northern Hemisphere and ~~South~~Southern Hemisphere where in cloud condensation water values are close to the assumed constant value in GC12. Therefore, the differences of wet scavenging in GC12 and WETrev cases at these regions are small. For dust, due to its low water solubility, the updated wet processes show a small impact in the lower troposphere where wet scavenging is dominated by warm ~~cloud.~~clouds. Most of black carbon and organic carbon are emitted as hydrophobic aerosols and then converted to be hydrophilic aerosols due to aging. Therefore, the updated wet process treatments show only a small impact at source regions but show a strong impact ~~at~~for remote regions.

4. Summary

In this study, we updated aqueous phase chemistry and wet scavenging for SO_2 and sulfate, rainout efficiencies for warm, mixed, and cold ~~cloud~~clouds, empirical washout by rain and snow, and wet surface ~~uptakes~~uptake during dry deposition in GEOS-Chem version 12.6.0~~ and presented the~~. Systematic validations of simulated aerosols and 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. Based on these validations, we found:

(1) The model results with the updated treatment of wet processes agree better with measurements for most species in different regions, especially for nitric acid, nitrate, and ammonium whose NMBs were improved, respectively, from

78%, 126%, and 45% to 0.9%, 15%, and 4.1% over US sites, from 107%, 127%, and 90% to -0.7%, 4.2%, and 16% over Europe sites, and from 121%, 269%, and 167% to -21%, 37%, and 86% over Asia remote region sites;

(2) ~~The~~Comparing to Luo et al. (2019), the updated aqueous phase chemistry and wet scavenging of SO_2 and sulfate significantly improve the agreement of simulated SO_2 and sulfate ~~at~~over the US, Europe~~,~~ and Asia remote region, especially during the winter time~~;~~. NMBs of sulfate in the 3 regions are reduced from -30%, -33%, and -36% to -10%, 4.3%, and 6.3%;

(3) The updated wet process treatments significantly improve the performance of sulfate wet deposition simulation over the US and Europe. NMBs are reduced from -35% to -9% over the US and from -46% to -6.2% over Europe, respectively;

~~(3)~~(4) The updated rainout efficiencies enhance BC mass concentration ~~at~~for remote regions and successfully reduce the bias between simulation and observation at Arctic sites. NMBs of BC are reduced from -67% to -40% at Barrow and from -75% to -46% at Zeppelin due to the switch from L2019 to WETrev;

~~(4)~~(5) Cold cloud scavenging plays ~~an~~ important roles in the simulation at the upper troposphere, especially for nitric acid~~;~~.

~~(5)~~(6) The updated wet surface uptake during dry deposition ~~improves~~changes the ~~agreement~~performance of simulated SO_2 at Arctic sites. NMB of SO_2 is increased from -23% to 32% at Nord and decreased from 27% to 22% at Zeppelin.

Wet processes are important for atmospheric chemistry modeling. Our study indicates that the updated wet process treatments introduced in this study have strong impacts on global means of water soluble aerosols and aerosol precursors~~ and aerosols~~ such as nitric acid, sulfate, nitrate, and ammonium. The updated wet process treatments exhibit relatively small impacts on the simulated global means of SO_2, dust, sea salt, black carbon, and organic carbon. Although ~~we tried to make~~there are clear improvements derived from the updated treatment of wet ~~process treatments to be the state-of-the-art~~processes, there still ~~exit~~exist limitations of the work presented in this study. For example, washout efficiencies of water soluble species such as SO_2 and ammonia are sensitive to rain water pH values. In this study, we simply assumed rainwater pHs for rainout and washout are cloud pH at where rainout occurs and rainwater-mass-weighted cloud pH above where washout occurs, respectively. However, rain water pH needs to be calculated by tracing the cloud process and precipitation process of rain water lifecycle. The impact of traced rain water pH on wet scavenging needs to be further investigated.

Code and data availability. The code of GEOS-Chem 12.6.0 is available through the GEOS-Chem distribution web-page http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_12. ~~All measurement data are publicly available~~The updated wet process code can be obtained by contacting the author directly. All measurement data are publicly available. USEPA data are download from https://www.epa.gov/outdoor-air-quality-data; CASTNET, AMon, IMPROVE, CSN data are download from http://views.cira.colostate.edu/fed/; NTN data are download from http://nadp.slh.wisc.edu/data/ntn/ntnAllsites.aspx; EMEP date are download from http://ebas.nilu.no/default.aspx and https://projects.nilu.no//ccc/emepdata.html; EANET data are download from https://monitoring.eanet.asia/document/signin; ATom data are download from https://espoarchive.nasa.gov/archive/browse/atom.

Author contributions. GL and FY proposed and implemented the improved wet processes schemes and validated model simulations with surface observations and ATom aircraft measurements. ~~MJ~~JM provided the new cloud pH approach in GEOS-Chem. All authors contributed to the writing and editing of the paper.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgments. This work is supported by NYSERDA under contract 137487, NASA under grant NNX17AG35G, and NSF under grant 1550816. The authors thank Daniel J. Jacob, Harvard University, whose comments and suggestions greatly helped improve and clarify this paper. We would like to acknowledge the United States Environmental

Protection Agency (USEPA), the Interagency Monitoring of Protected Visual Environments (IMPROVE), the Chemical Speciation Network (CSN), the Clean Air Status and Trends Network (CASTNET), the Ammonia Monitoring Network (AMoN), National Trends Network (NTN), the European Monitoring and Evaluation Programme (EMEP), and the Acid Deposition Monitoring Network in East Asia (EANET) for the in-site measurement data. We would like to acknowledge the Atmospheric Tomography Mission (ATom) for the aircraft measurement data. GEOS-Chem is a community model maintained by the GEOS-Chem Support Team at Harvard University.

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Table 1. List of Λ and b values in equation 15 for rain and snow washout parameterizations.

	Rain		Snow	
	T>268 K		248 K<T<268 K	
	Λ	*b*	Λ	*b*
GC12				
HNO_3	2.8×10^{-5}	1.0	0	0
Coarse aerosol	2.6×10^{-4}	0.79	4.2×10^{-4}	0.96
Fine aerosol	4.3×10^{-6}	0.61	8.8×10^{-6}	0.96
This work				
HNO_3	3×10^{-3} [‡]	0.62 [‡]	3×10^{-3} [‡]	0.62 [‡]
Coarse aerosol	2×10^{-4} [†]	0.85 [†]	2×10^{-3} [†]	0.7 [†]
Hydrophobic fine aerosol	5×10^{-7} [†]	0.7 [†]	1×10^{-5} [†]	0.66 [†]
Hydrophilic fine aerosol	1×10^{-5} [⁑]	0.7 [†]	2×10^{-4} [⁑]	0.66 [†]

[†] from Wang et al. (2014) assuming fine aerosol with diameter of 100 nm and coarse aerosol with diameter of 6 μm; [‡] from Luo et al. (2019); [⁑] this work.

Table 2. Number of sites with surface concentration observation (NVO) and number of sites satisfying criterion (NSC) at surface monitoring networks in the US, Europe, and Asia.

	USA		Europe		Asia	
	NVO	NSC	NVO	NSC	NVO	NSC
SO2	USEPA		EMEP		EANET	
	464	288	42	20	14	3
HNO3	CASTNET		EMEP		EANET	
	84	77	25	8	25	5
NH3	AMoN		EMEP		EANET	
	53	17	40	15	25	10
SO4	IMPROVE+CSN		EMEP		EANET	
	371	214	52	21	25	9
NIT	IMPROVE+CSN		EMEP		EANET	
	371	213	66	22	25	8
NH4	IMPROVE+CSN		EMEP		EANET	
	371	178	66	24	25	9
BC	IMPROVE		EMEP			
	168	122	11	5		
OC	IMPROVE		EMEP			
	168	118	11	5		

Table 3. Observed and simulated annual mean surface concentrations of aerosols and aerosol precursors in the US, Europe, and Asia. Comparisons include annual mean surface concentrations (M, µg m^{-3}), normalized mean bias (NMB, %), and correlation coefficient (*r*, when # of samples > 10) between observed and simulated annual mean values for the 8 species by G12, L2019, and WETrev cases.

		USA				Europe				Asia		
		G12	L2019	WETrev		G12	L2019	WETrev		G12	L2019	WETrev
SO2	M: 5.61	4.48	4.29	4.32	M: 1.36	2.36	2.16	2.05	M: 2.51	4.08	3.58	3.08
	NMB	-20	-23	-23	NMB	74	59	51	NMB	63	43	23
	r	0.49	0.49	0.48	*r*	0.53	0.50	0.50	*r*			
HNO3	M: 0.83	1.47	0.83	0.83	M: 0.67	1.40	0.66	0.67	M: 0.86	1.90	0.64	0.68
	NMB	78	0.9	0.9	NMB	107	-2.4	-0.7	NMB	121	-26	-21
	r	0.57	0.59	0.60	*r*				*r*			
NH3	M: 1.00	1.02	1.04	1.21	M: 0.83	0.84	0.91	1.07	M: 0.96	0.95	0.88	1.06
	NMB	2.6	4.4	21	NMB	0.9	8.7	28	NMB	-1.7	-8.6	10
	r	0.26	0.28	0.29	*r*	0.90	0.89	0.91	*r*			
SO4	M: 1.30	1.29	0.91	1.17	M: 1.29	1.38	0.87	1.24	M: 2.63	2.77	1.69	2.46
	NMB	-1.1	-30	-10	NMB	6.9	-33	-4.3	NMB	5.5	-36	-6.3
	r	0.92	0.92	0.92	*r*	0.92	0.90	0.92	*r*			
NIT	M: 0.71	1.60	0.78	0.81	M: 1.66	3.77	1.54	1.73	M: 0.60	2.23	0.89	0.83
	NMB	126	10	15	NMB	127	-7.5	4.2	NMB	269	47	37
	r	0.53	0.58	0.61	*r*	0.85	0.86	0.86	*r*			
NH4	M: 0.61	0.89	0.54	0.64	M: 0.88	1.67	0.82	1.02	M: 0.58	1.55	0.82	1.08
	NMB	45	-13	4.1	NMB	90	-7.3	16	NMB	167	42	86
	r	0.76	0.79	0.79	*r*	0.79	0.81	0.81	*r*			
BC	M: 0.20	0.18	0.16	0.17	M: 0.51	0.38	0.32	0.34				
	NMB	-7.0	-20	-14	NMB	-25	-37	-32				
	r	0.54	0.54	0.54	*r*							
OC	M: 1.01	0.80	0.68	0.72	M: 1.97	1.00	0.77	0.85				
	NMB	-20	-33	-29	NMB	-49	-61	-57				
	r	0.63	0.65	0.65	*r*							

Table 4. Relative contribution (%) of modified rainout (RO), washout (WO), rain pH (RP), dry deposition (DD), and aqueous chemistry (AC) to the changes of January and July surface concentrations at the US, Europe, and Asia sites.

	USA					Europe					Asia				
	RO	WO	RP	DD	AC	RO	WO	RP	DD	AC	RO	WO	RP	DD	AC
	January														
SO2	5.0	15.3	0.5	54.2	25.0	11.7	24.1	12.0	19.0	33.1	3.6	15.3	0.2	27.7	53.2
HNO3	15.5	73.4	0.5	5.3	5.2	25.2	60.1	1.3	2.4	11.0	8.7	63.1	0.1	8.4	19.6
NH3	7.9	23.7	1.6	30.5	36.3	9.0	20.4	31.3	14.9	24.4	3.9	7.0	5.8	26.2	57.1
SO4	46.6	17.3	0.4	6.2	29.5	74.3	8.5	0.9	2.1	14.3	29.4	17.5	0.1	5.8	47.3
NIT	37.7	46.7	0.7	5.3	9.6	56.5	34.1	1.4	1.5	6.5	17.4	43.9	0.3	10.7	27.6
NH4	48.7	34.3	0.7	6.0	10.3	78.3	13.2	1.0	2.2	5.2	40.6	22.9	0.3	3.1	33.0
	July														
SO2	5.6	13.1	8.5	50.5	22.3	3.0	31.3	1.3	31.0	33.4	13.3	15.9	15.2	23.5	32.1
HNO3	5.8	91.3	0.5	2.0	0.4	5.2	93.8	0.2	0.7	0.1	11.4	86.8	0.5	1.2	0.1
NH3	6.7	21.7	13.4	49.9	8.2	5.7	53.2	11.5	26.6	3.0	4.8	17.9	28.7	45.0	3.6
SO4	48.7	16.5	4.4	12.9	17.5	66.0	11.7	0.7	3.0	18.7	63.9	16.2	2.6	8.4	8.8
NIT	16.1	68.7	2.7	10.9	1.6	12.3	82.6	1.5	3.2	0.4	24.4	64.7	3.3	6.9	0.7
NH4	35.7	36.4	4.1	13.0	10.8	27.2	63.7	1.0	2.6	5.5	52.6	29.1	3.3	8.9	6.2

Table 5. Number of sites with wet deposition observation (NVO) and number of sites satisfying criterion (NSC) at surface monitoring networks in the US, Europe, and Asia.

	USA		Europe		Asia	
	NTN		EMEP		EANET	
	NVO	NSC	NVO	NSC	NVO	NSC
SO2+SO4	250	86	62	25	53	4
HNO3+NIT	250	86	67	30	53	4
NH3+NH4	250	85	64	29	53	4

Table 6. Observed and simulated annual mean wet depositions of aerosols and aerosol precursors in the US, Europe, and Asia. Comparisons include annual mean wet depositions (M, kg ha^{-1} year^{-1}), normalized mean bias (NMB, %), and correlation coefficient (*r*, when # of samples > 10) between observed and simulated annual mean values by G12, L2019, and WETrev cases. Simulated values at sites were corrected following Paulot et al. (2014) to remove bias due to precipitation.

		USA				Europe				Asia		
		G12	L2019	WETrev		G12	L2019	WETrev		G12	L2019	WETrev
SO2+SO4	M: 10.3	6.8	8.0	9.4	M: 6.3	3.4	5.0	5.9	M: 28.6	10.3	11.1	13.2
	NMB	-35	-23	-9.0	NMB	-46	-21	-6.2	NMB	-64	-61	-54
	r	0.81	0.79	0.81	*r*	0.56	0.55	0.49	*r*			
HNO3+NIT	M: 9.5	9.6	18.1	19.1	M: 9.9	6.8	14.3	14.0	M: 14.6	13.3	15.8	15.5
	NMB	0.6	89	100	NMB	-31	45	42	NMB	-9.2	8.1	6.2
	r	0.9	0.85	0.88	*r*	0.84	0.59	0.64	*r*			
NH3+NH4	M: 3.6	3.2	4.0	4.2	M: 3.9	2.6	3.9	3.6	M: 3.9	3.5	3.3	3.8
	NMB	-10	12	16	NMB	-33	-1.6	-7.7	NMB	-10	-14	-2.5
	r	0.85	0.87	0.85	*r*	0.75	0.55	0.67	*r*			

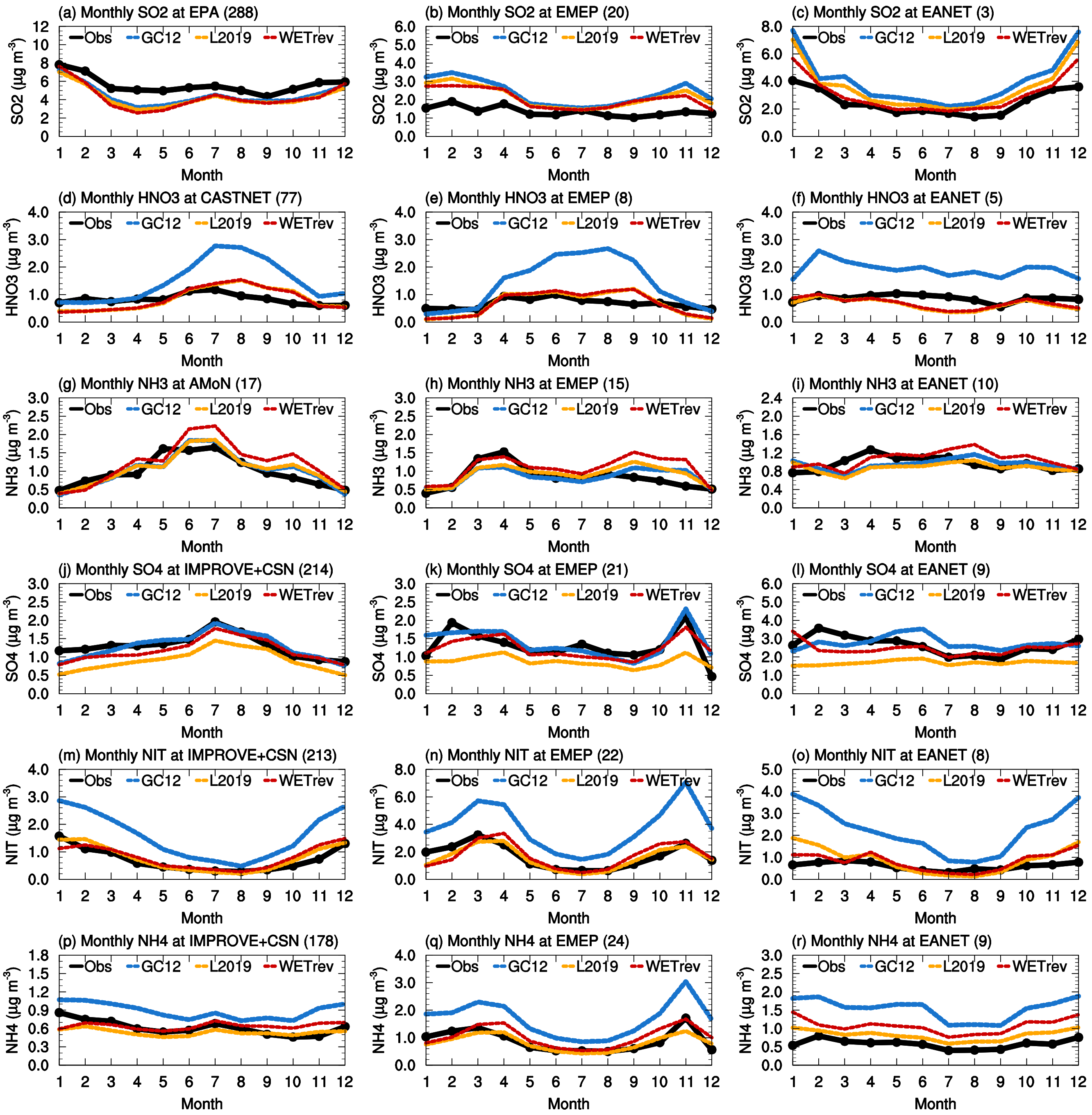

Figure 1. Variations of monthly means for year 2011 showing the comparisons of SO_2, nitric acid, ammonia, sulfate, nitrate, and ammonium surface mass concentrations which are observed over (left column) the US, (center column) Europe, and (right column) Asia sites (black) and simulated by GC12 (blue), L2019 (yellow), and WETrev (red) cases.

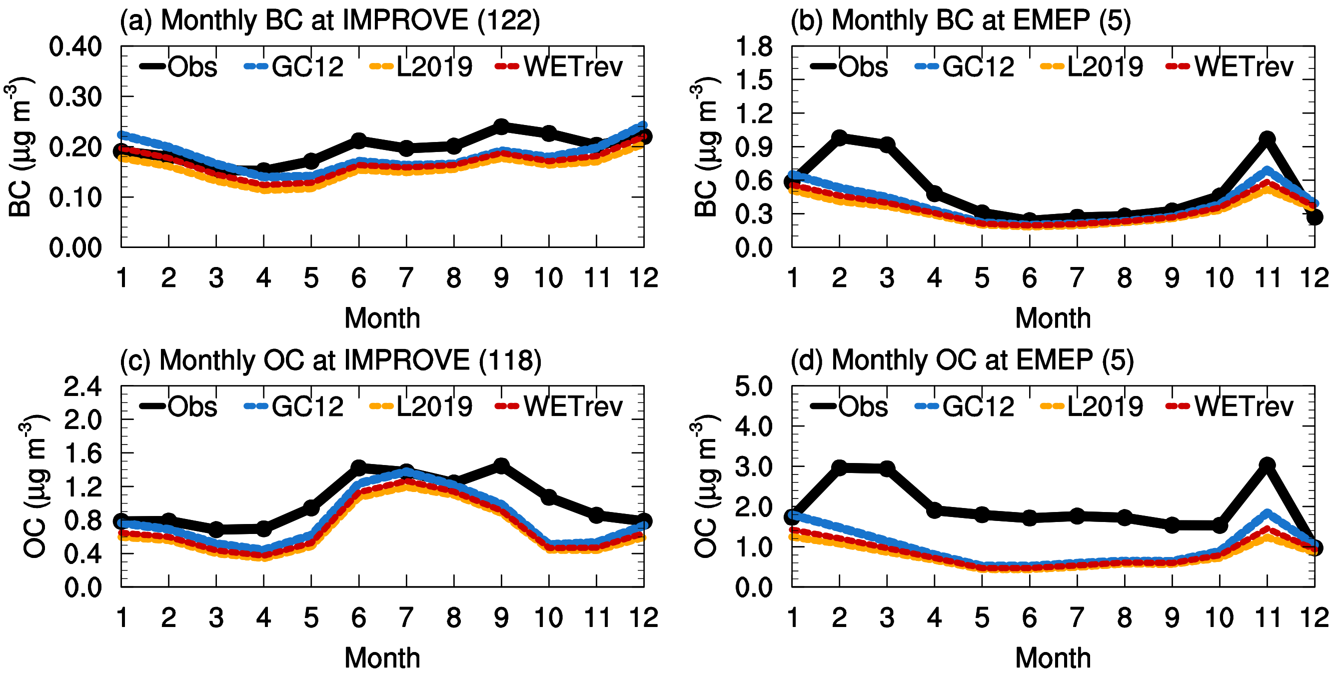

Figure 2. Variations of monthly means for year 2011 showing the comparisons of black carbon and organic carbon surface mass concentrations which are observed over (left column) the US and (right column) Europe sites (black) and simulated by GC12 (blue), L2019 (yellow), and WETrev (red) cases.

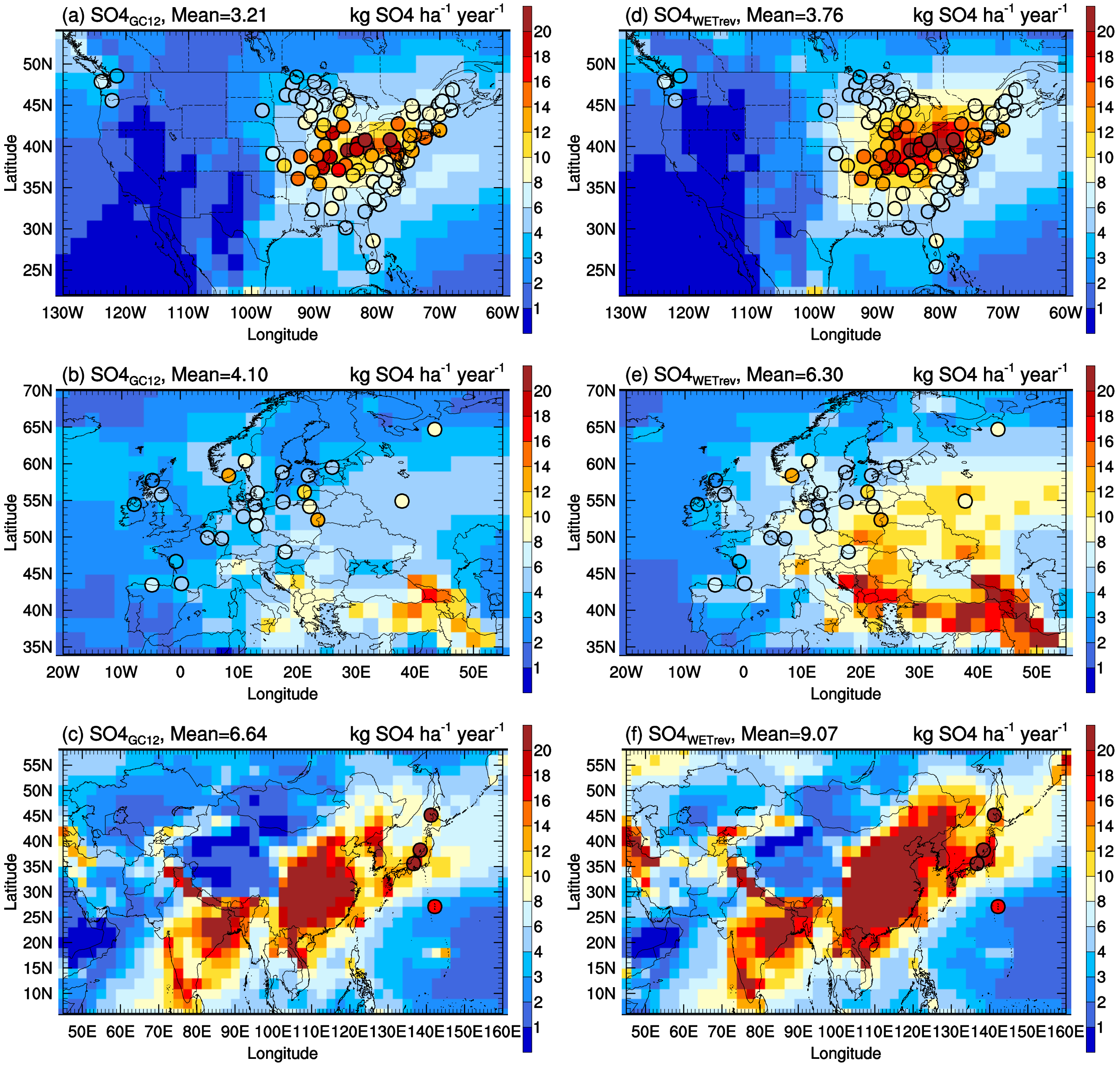

Figure 3. Horizontal distributions of SO_2+SO4 deposition over the US (top), Europe (middle), and Asia (bottom). Filled circles are annual mean wet depositions at NTN, EMEP, and EANET corrected following Paulot et al. (2014) to remove bias due to precipitation.

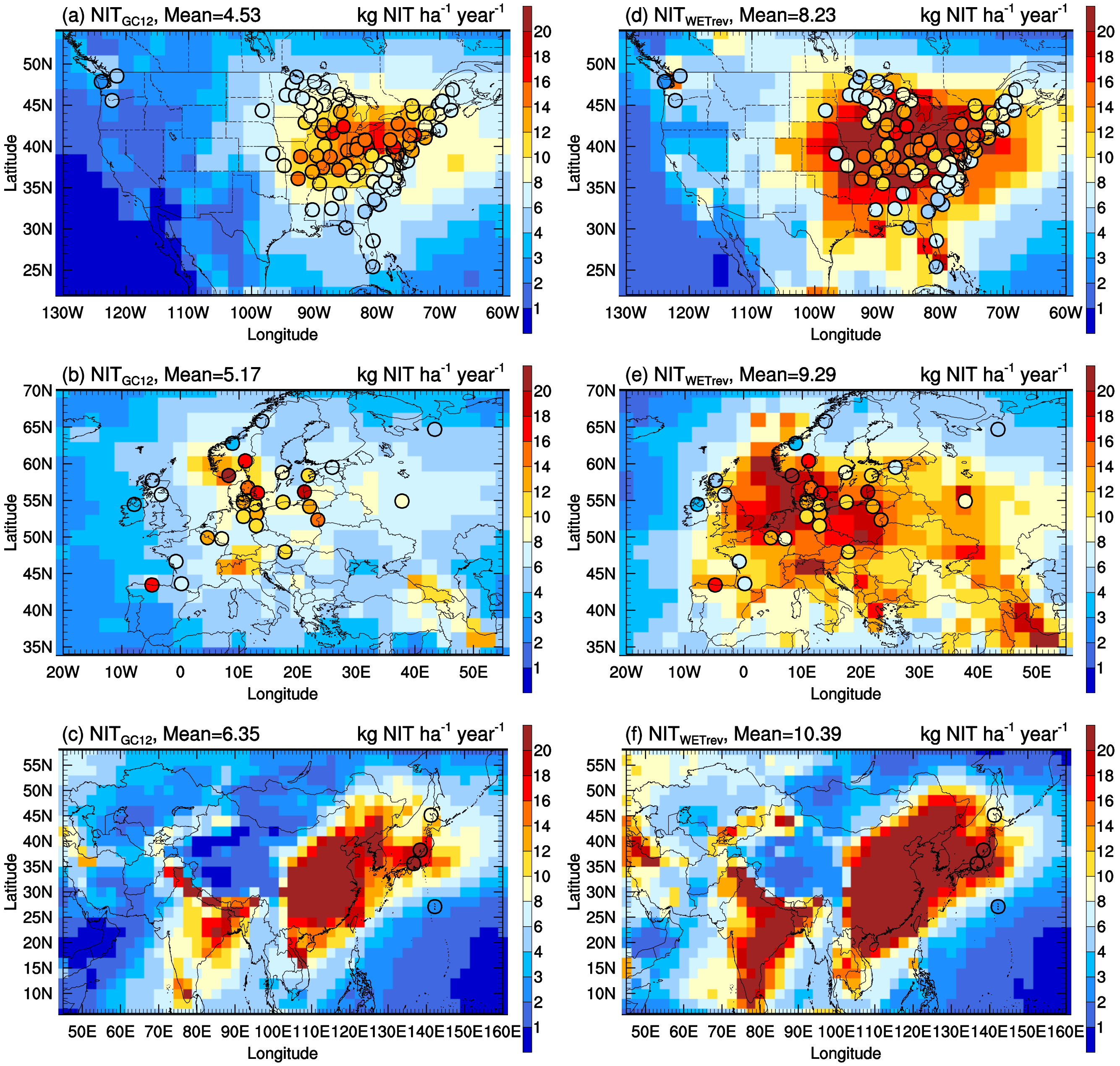

Figure 4. The same as Fig. 3 but for HNO_3+NIT.

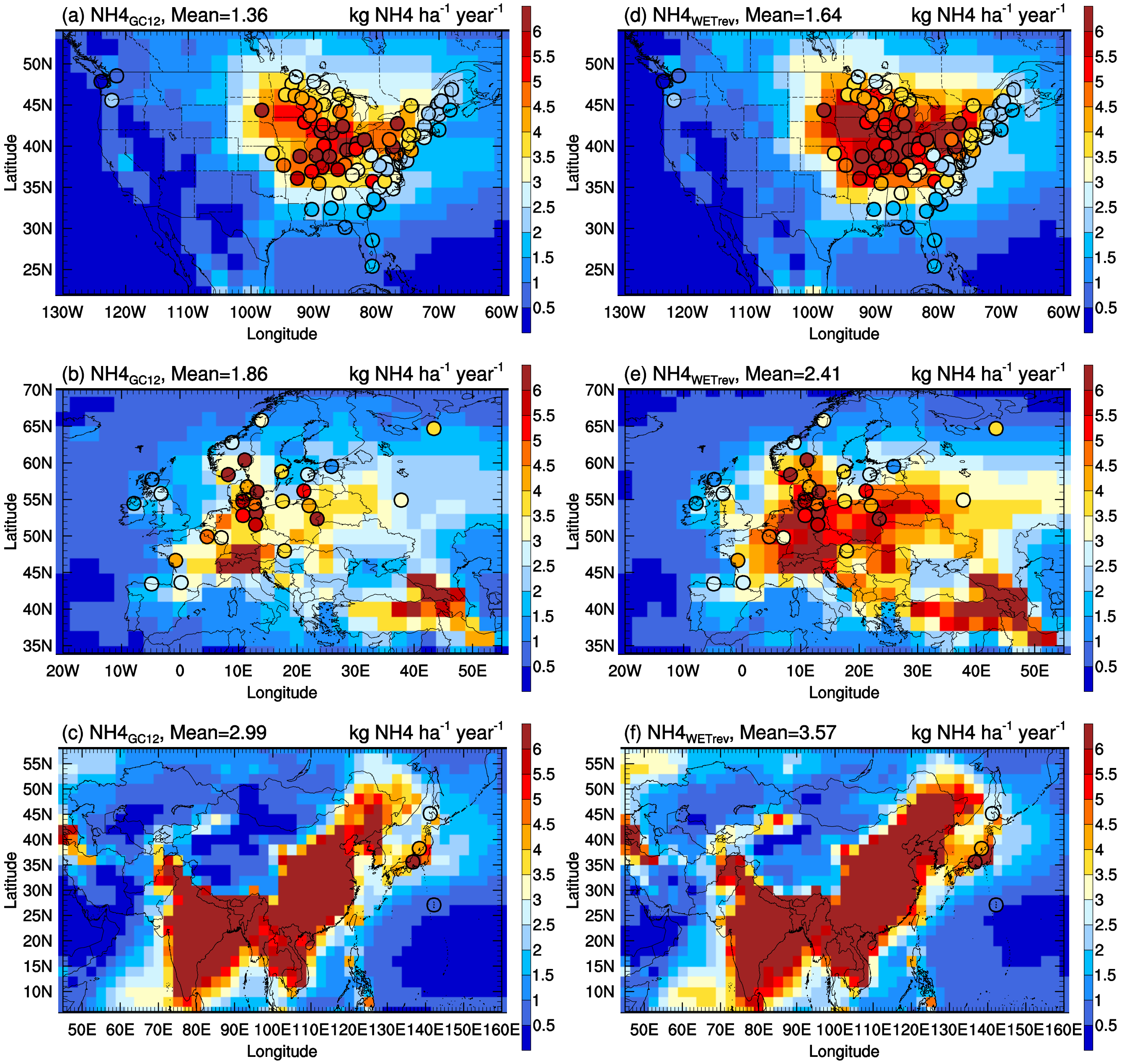

Figure 5. The same as Fig. 3 but for NH$_3$+NH4.

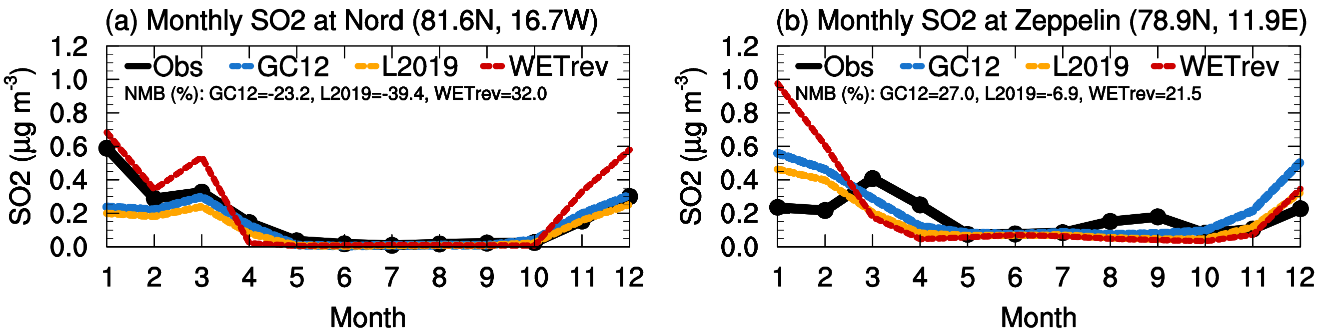

Figure 6. Variations of multiyear monthly means showing the comparisons of SO_2 surface mass concentrations which were observed at (a) Nord (2008-2013) and (b) Zeppelin (2008-2013) sites (black) and simulated (2011) by GC12 (blue), L2019 (yellow), and WETrev (red) cases.

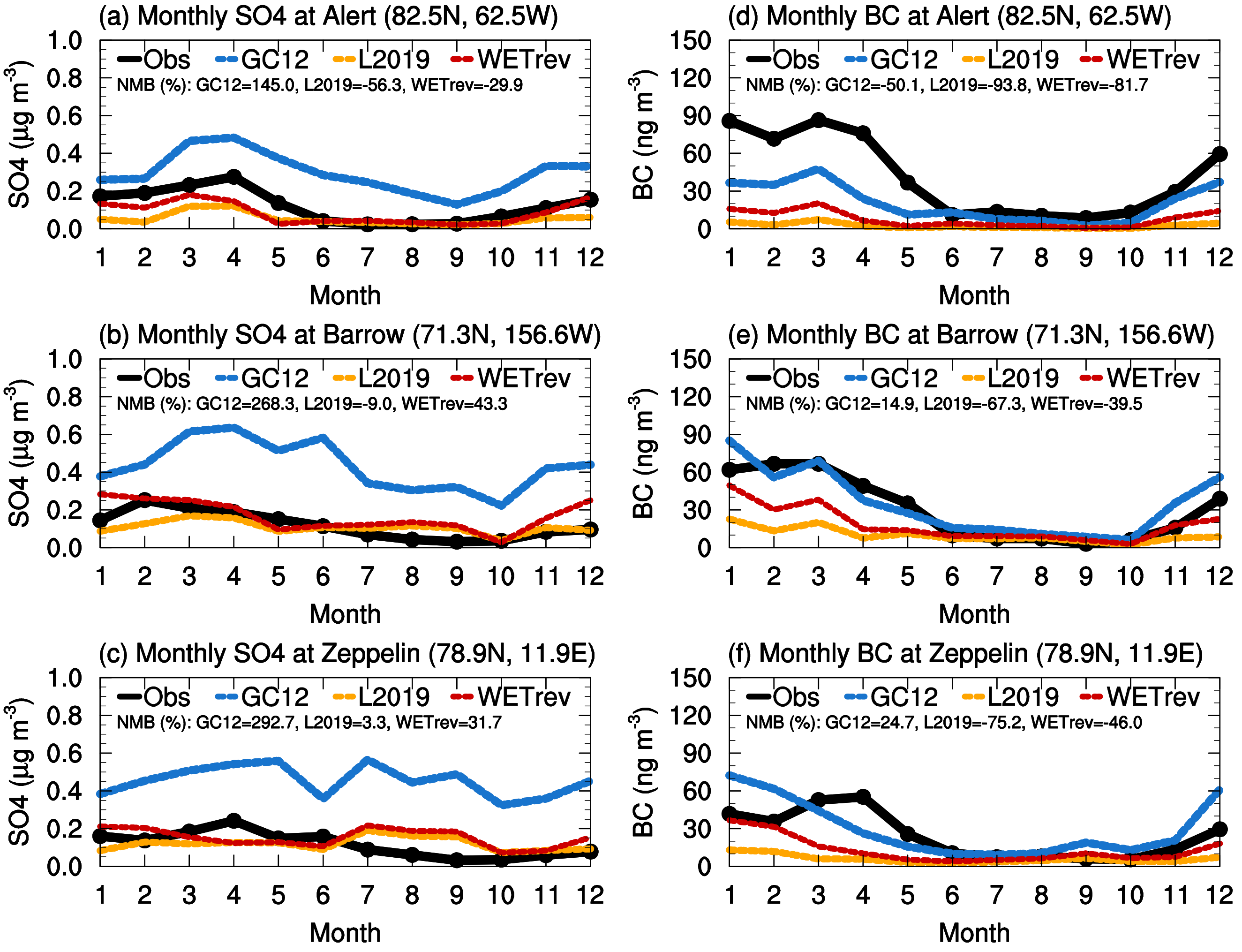

Figure 7. Variations of multiyear monthly means showing the comparisons of (a-c) sulfate and (d-f) black carbon surface mass concentrations which were observed at (top) Alert (2008-2012), (middle) Barrow (2008-2013), and (bottom) Zeppelin (2008-2013) sites (black) and simulated (2011) by GC12 (blue), L2019 (yellow), and WETrev (red) cases.

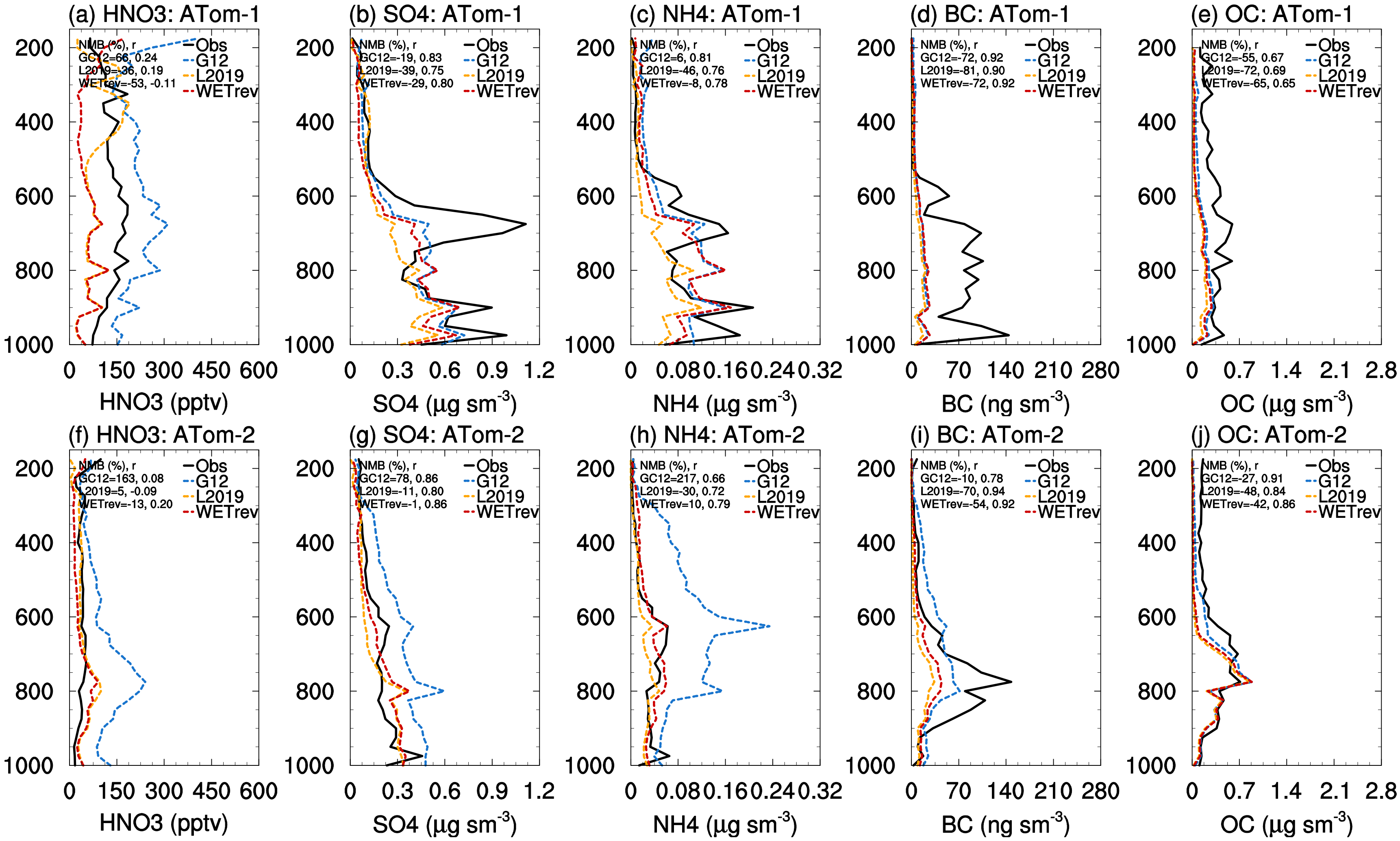

Figure 8. Vertical profiles of nitric acid, sulfate, ammonium, black carbon, and organic carbon from ATom aircraft observations (black, ATom-1: a-e; ATom-2: f-j) and GEOS-Chem simulations by GC12 (blue), L2019 (yellow) and WETrev (red) cases over the Northern hemisphere.

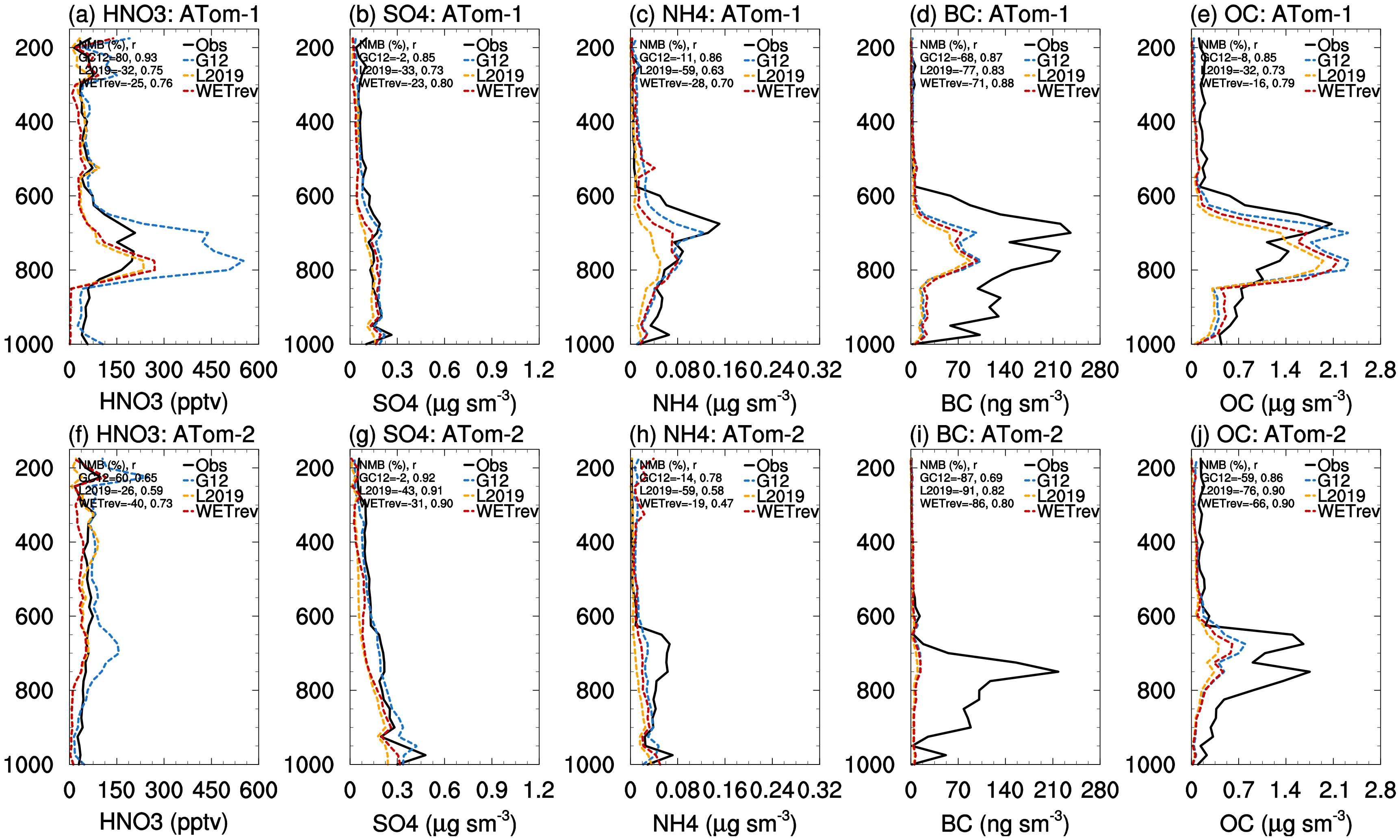

Figure 9. The same as Fig. 8 but over the Southern Hemisphere.

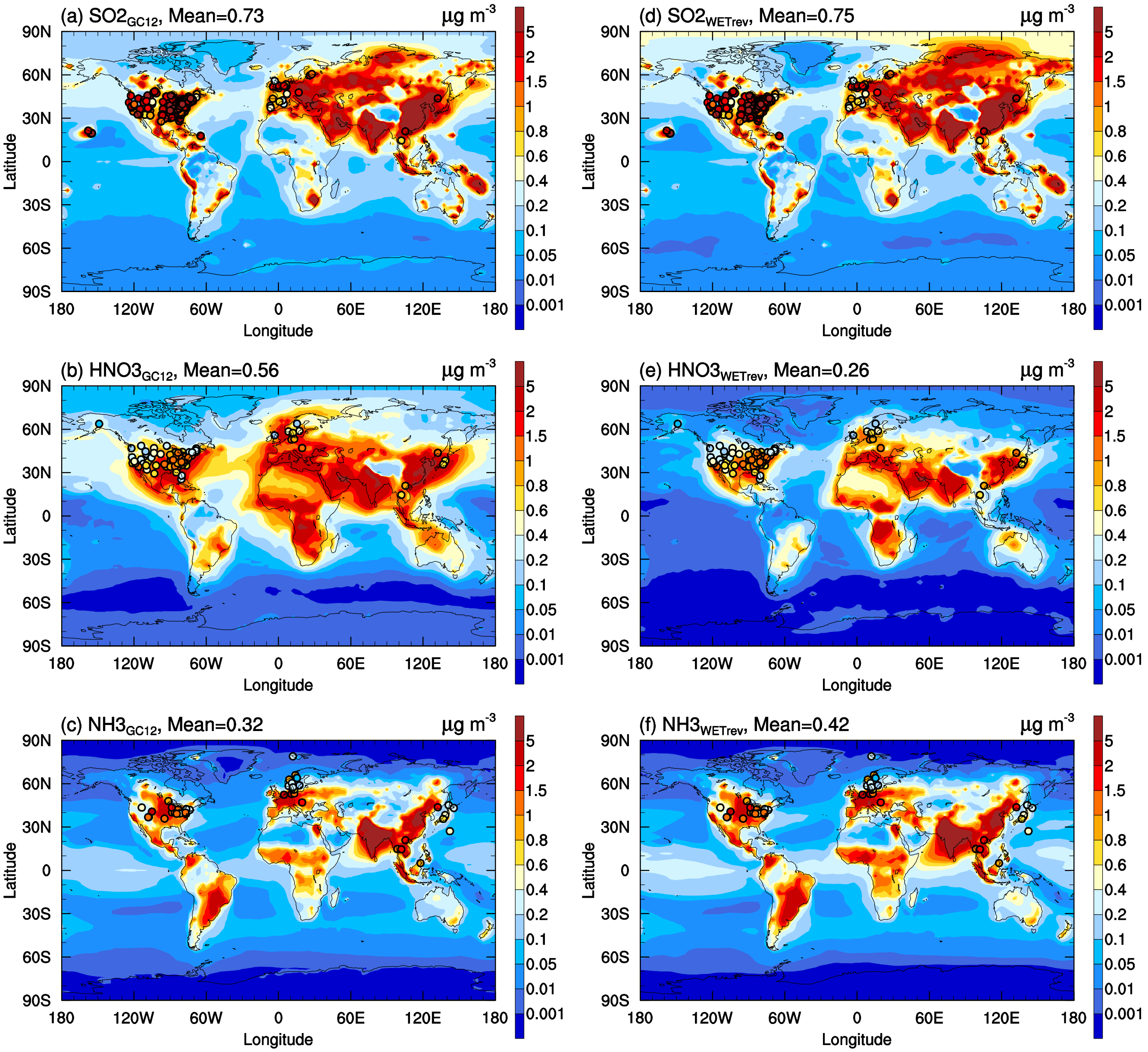

Figure 10. Horizontal distributions of SO_2, nitric acid, and ammonia surface mass concentrations simulated by (a-c) GC12 case and (d-f) WETrev case. Filled circles are annual mean surface mass concentrations observed at IMPROVE, CSN, CASTNET, AMoN, EMEP, and EANET for corresponding species.

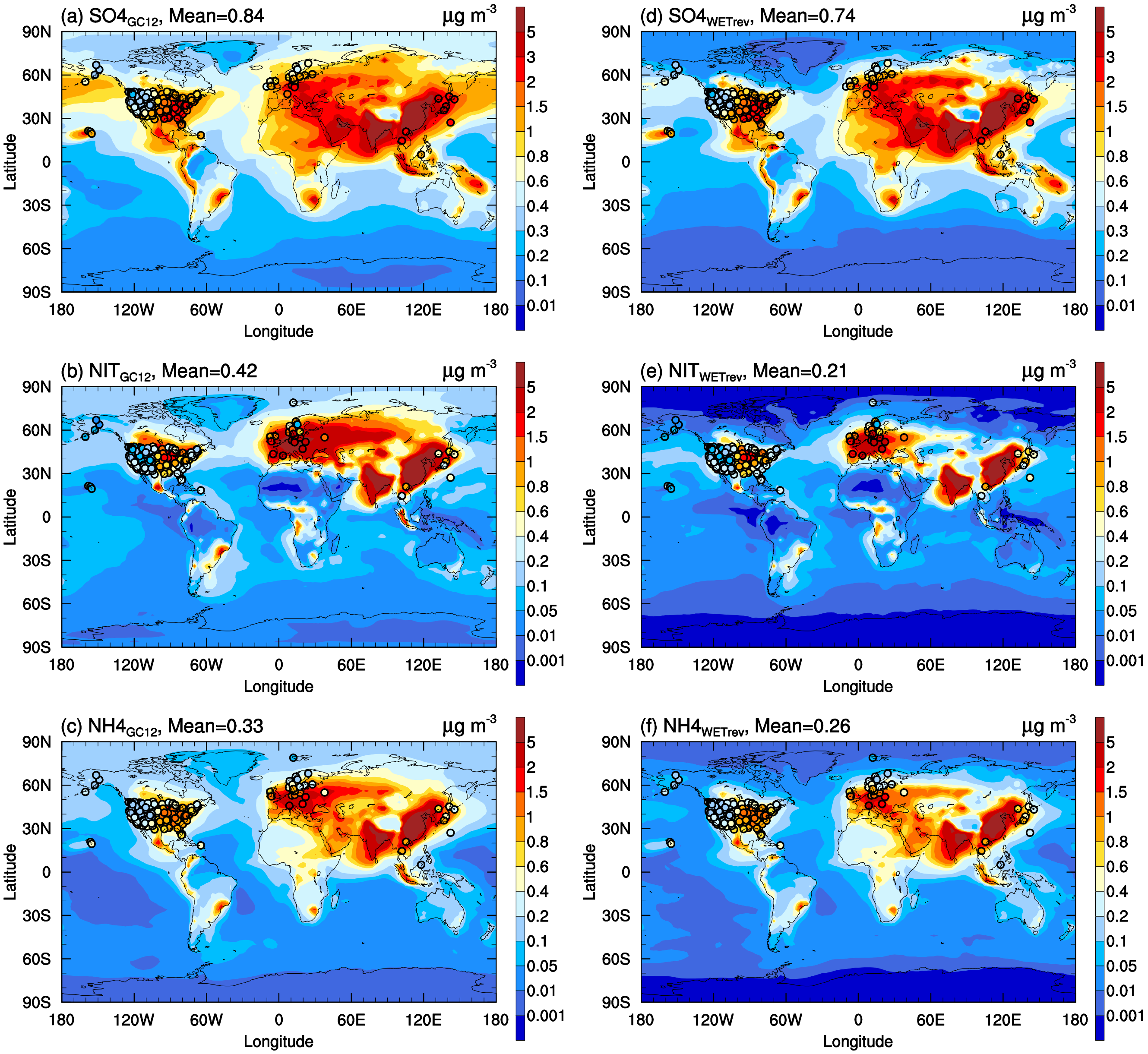

Figure 11. The same as Fig. 10 but for sulfate, nitrate, and ammonium surface mass concentrations.

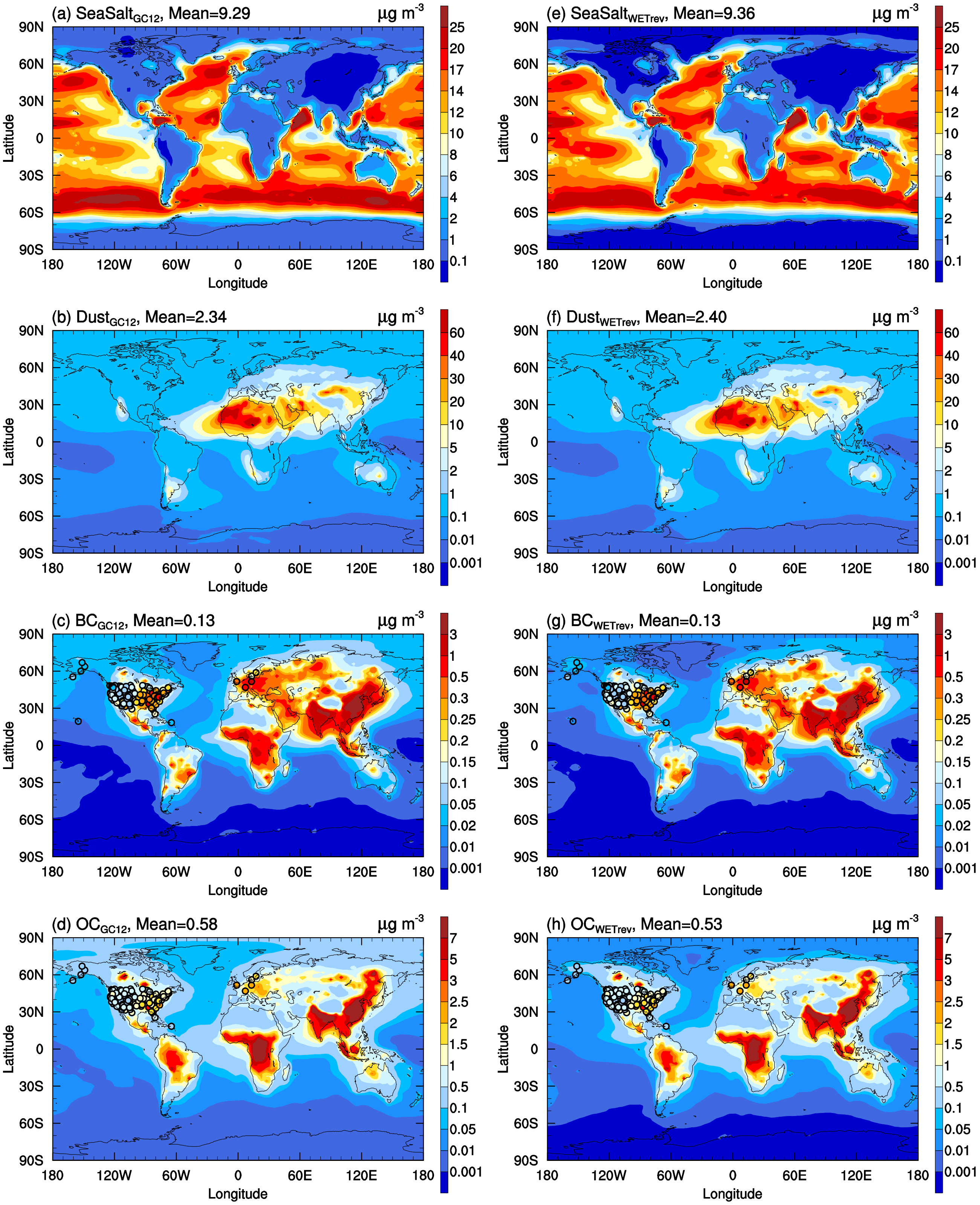

Figure 12. The same as Fig. 10 but for black carbon, organic carbon, sea salt, and dust surface mass concentrations.

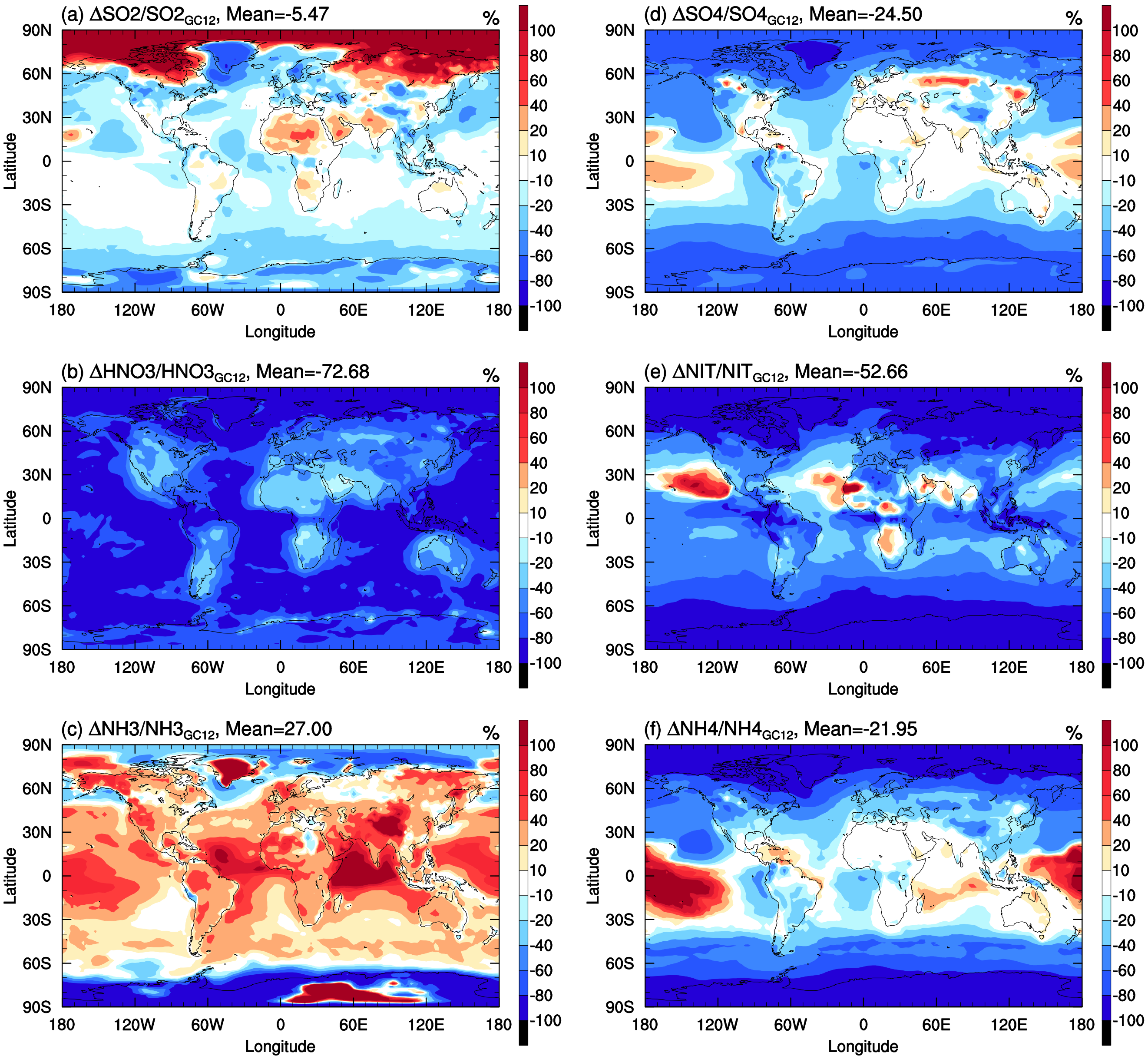

Figure 13. Horizontal distributions of percentage changes in annual mean (a) SO_2, (b) nitric acid, (c) ammonia, (d) sulfate, (e) nitrate, and (f) ammonium surface mass concentrations due to the switching of GC12 case to WETrev case.

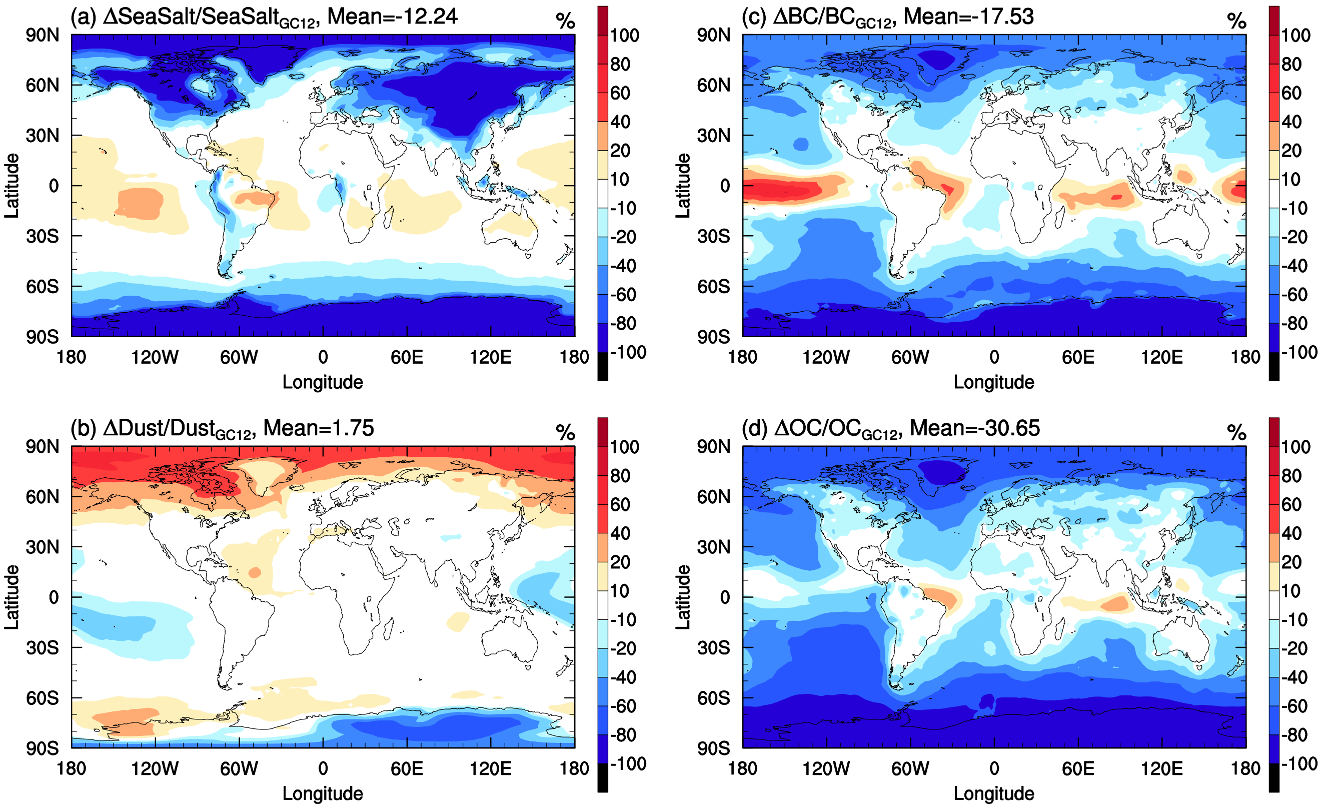

Figure 14. The same as Fig. 13 but for (a) sea salt, (b) dust, (c) black carbon, and (d) organic carbon surface mass concentrations.