gmd-2021-404

We would like to thank the reviewers for the constructive comments. This revised manuscript has been further improved based on the reviewers' comments and suggestions. The point-by-point responses to each reviewer's comments are given below.

Referee 1

General Comments

The manuscript describes the development of WRF-GC-Hg model, based on the already developed WRF-GC model and also shows a case study of the application of WRF-GC-Hg model in understanding the high Hg wet deposition in Southeast US. I do have several major comments:

- Does the paper focus on the study of the high Hg wet deposition in Southeast US only or also the development of the WRF-GC-Hg model? I would suggest including the development of the WRF-GC-Hg model as an important component of the paper. If so, the title should be revised as something: Development of WRF-GC-Hg v1.0 and its application in studying Hg wet deposition in Southeast US. This will make the paper stronger and more applicable. With that, the paper will need to be reorganized to include one part to focus on the new development and its evaluation and another part to focus on the study of the high Hg wet deposition in the Southeast US. It will be also nice to extend the domain to the whole continental US for the WRF-GC-Hg evaluation part.
- We thank the reviewer for this helpful suggestion. Our motivation is to study high mercury wet deposition in southeastern US. The reason we chose WRF-GC is that this model has many advantages (like flexible resolution, better meteorology simulation, HEMCO inside the model for emission inventories, etc.)

  However, given that WRF-GC is a novel model without complimentary mercury libraries, some developments have been done for this research, so here we added some paragraphs to better describe the WRF-GC-Hg v1.0.

  Corresponding to our paper, the modifications are as follows:

  1. The advantage of WRF-GC.
     Line 64-70: Therefore, we chose WRF-GC (Feng et al., 2021; Lin et al., 2020) to develop a new Hg simulation capacity with a complimentary Hg library because WRF-GC has several advantages: 1) It has flexible resolution and widely accepted meteorology simulation provided by WRF model; 2) The Hg chemistry included by GEOS-Chem model is more up-to-date than many other models (Horowitz et al., 2017); 3) It is relatively easy to port Hg library from GEOS-Chem to WRF-GC-Hg.

  2. We added a figure to illustrate the structure of WRF-GC-Hg v1.0 and our modifications.
     Line 74: The model's framework is shown in Fig. 1.

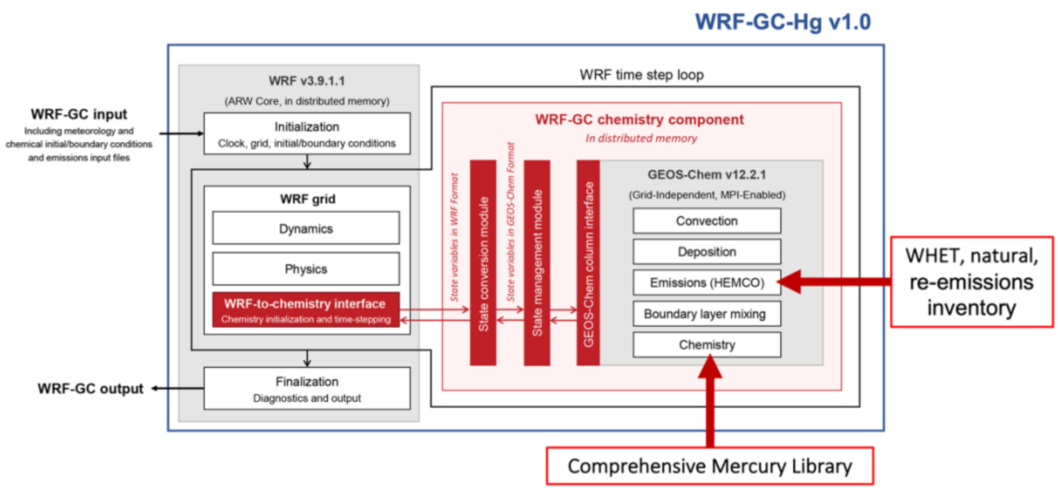

Fig. 1 WRF-GC-Hg v1.0 framework based on WRF-GC v1.0 (Lin et al., 2020)

3. Chemistry library
   Line 92: We implement a complimentary Hg chemistry library (Fig. 1) to the WRF-GC model by first introducing …

4. Emission module
   Line 99-100: We use the WHET emission inventory (1° × 1°) for the anthropogenic Hg emissions (Zhang et al., 2016) as well as natural emissions and re-emissions inventory (4° × 5°) from Horowitz et al. (2017) (see Fig. 1).

- Regarding the domain of simulation, previous research (Holmes et al., 2016; Fulkerson and Nnadi, 2006) have shown the underestimation of Hg wet deposition in the southeastern US. Therefore, we only chose the southeastern US region for this research considering that WRF-GC high-resolution simulation is highly computationally intensive.

- The paper conducts two WRF-GC-Hg sensitivity simulations with a horizontal resolution of 50 km and 25 km respectively and compare the results to the GEOS-Chem simulation with a spatial resolution of 4º x 5º. As expected, the WRF-GC-Hg simulations with a finer spatial resolution will resolve more spatial signals. The comparison will be more meaningful to include the GEOS-Chem nested Hg simulation results, which are comparable to WRF-GC-Hg simulations.

Thanks for your suggestion. We had considered your suggestion before, but we find the currently existing GEOS-Chem Hg nested-grid simulation over North America is out of date (Zhang et al, 2012). The simulation is based on chemistry from Holmes et al. (2010), but we use an updated Hg chemistry from Horowitz et al. (2017). Therefore, the GEOS-Chem simulation nested Hg simulation is not off-the-shelf but would still require significant development and validation works. In addition, our main focus is not to compare GEOS-Chem and WRF-GC, but to find the simulation performance improvement with increasing resolution. We also expect the changes caused by increasing resolution to be similar between different models. We thus think it should be enough to compare a coarse resolution of GEOS-Chem with different resolutions of WRF-GC-Hg.

- Some of the sentences are confusing. I would suggest improving the English language throughout the manuscript.

Thanks for your suggestion. A grammar check has been done during this revision. For major changes, we have highlighted contents with yellow color.

Specific comments

1. Line 23-26, can you be more explicit here about the heights, different types of precipitation etc.?

Line 23-26: We divided simulation results by heights (2km, 4km, 6km, 8km), different types of precipitation (large-scale and convective), and combinations of these two variations together and find most of mercury wet deposition concentrates on higher level and caused by convective precipitation.

2. Line 28: It is atmospheric Hg that can undergo long-range transport, it is not accurate to say that Hg goes through long-range transport here.

Line 29-30: Mercury (Hg) is one of the most toxic heavy metals in our environment. Atmospheric Hg can undergo long-range transport (Ariya et al., 2015) in three major forms: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particle-bound mercury (PBM).

3. Line 46-48: I am not sure what it means here. What is the 80% of rainfall?

Line 48: approximately 80% of rainfall amount

4. Line 108: The model is run as one domain with 50 km and 25 km, right? “The model horizontal resolution is set as ranged from 50 km to 25 km, with 50 vertical layers.” It sounds like the model is run as a nested domain. Please clarify here.

Line 120: We ran simulations with different horizontal resolutions (50 km × 50 km and 25 km × 25 km for WRF-GC and 4° × 5° for GEOS-Chem) rather than using nested domains.

5. Line 120: what are the CMAP data? Are they merging observation and model data? Please provide more information here.

Line 133-134: The CMAP is 2.5° × 2.5° monthly analyses of global precipitation, generated from merging rain gauges and several satellite-based algorithms (Xie and Arkin, 1997).

6. Line 131-132: I do not quite understand the sentence here.

This is a problem we found during analyzing the data, but it is not related to this research, so we deleted it.

7. In Table 1, please spell out the lw and sw.

Table 1: Longwave and shortwave

8.	Line 140: the average total precipitation increases to 4.63 mm/day and 4.33 mm/day, so is it 4.63 or 4.33?
Apologies for not clarifying this. What we are trying to express is when we narrow down the domain to the southeast-most region, the changes in total precipitation, convective precipitation and large-scale precipitation.

Line 149-151: the average total precipitation increases to 4.63mm/day and convective precipitation increases to 4.33 mm/day, while the large-scale precipitation decreases to 0.29 mm/day.

9.	Line 152: Are the eight AMNet sites shown in Figure 1? If so, can you use different symbols to differentiate them from the MDN sites?
We added AMNet sites with different symbols to Fig. 2.

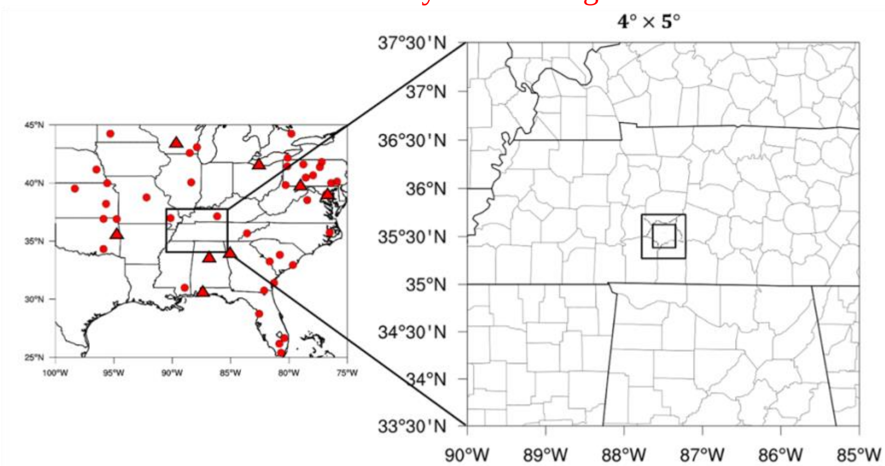

Fig. 2 Model simulation domain (Left panel: black box represents a single grid of GEOS-Chem 4° × 5° simulation, red circle dots represent MDN sites and triangle dots represents AMNet sites within this domain; Right panel: boxes from outside to inside respectively represents one grid of the resolution of 4° × 5°, 50km × 50km, 25km × 25km)

10.	Line 205: However, we find that this increase of resolution is finite. What do you mean here?
Line 225-226: However, we find that this increase of resolution is finite because the improvement of the increase of wet deposition flux is not that obvious as WRF-GC resolution increases.

11.	Line 254: whilst?
Here we want to compare CONV between different models at the same time. To better understand this sentence, we replaced “whilst” to “but”.

Line 275: CONV of GEOS-Chem is all lower than 0.15 μg m^{-2} but WRF-GC can reach 0.8 μg m^{-2} at ~4 km

12.	Line 156-158, 273-275: WRF-GC-Hg simulated Hg^0 concentration is 1.61±0.20 ng m^{-3}, which does not quite agree with the GEOS-Chem and observation concentration. Do you know why WRF-GC-Hg simulated higher Hg^0 concentration than both GEOS-

Chem and the ground observation, even though WRF-GC-Hg simulated better Hg wet deposition? It is due to the atmospheric redox chemistry or something else?
The development of WRF-GC-Hg (Hg chemistry library) coupling GEOS-Chem full-chemistry library with offline Br simulation. Even though all parameters were set the same as running GEOS-Chem, aqueous reductions and aerosol concentration may not be the same as GEOS-Chem's results.

Line 168-172: The average Hg^0 concentrations are 1.25±0.22 ng m^{-3} for the eight sites in the southeast US, which agree well with GEOS-Chem results of 1.27±0.06 ng m^{-3}. WRF-GC (1.61±0.20 ng m^{-3}) model does not agree with the observations or GEOS-Chem relatively well but is close. This might be due to the development of WRF-GC (Hg chemistry library) coupling GEOS-Chem full-chemistry library with offline Br simulation. Even though all parameters were set the same as running GEOS-Chem, aqueous reductions and aerosol concentration may not be the same as GEOS-Chem's results.

13. Line 279: in this area, which area do you mean here?
Line 300-301: Southeast-most area (states of Mississippi, Alabama, Georgia, South Carolina, and Florida)

14. Line 450-454: (Zhang et al., 2016a) and (Zhang et al., 2016b) are the same.
Sorry for the mistake, we have corrected the reference by deleting the repeated one.

15. Figure 7, 9, 10 are hard to read.
What we tried to express here is the comparison between different models at different heights (~2,4,6,8 km). From left to right is a comparison between different models. From top to bottom is a comparison between different heights. Here, we want to use this comparison to better see the distribution of Hg wet deposition on different levels. We moved the labels for heights to the left of the figure and added compasses to different panels.

Here are the changes for the three figures:

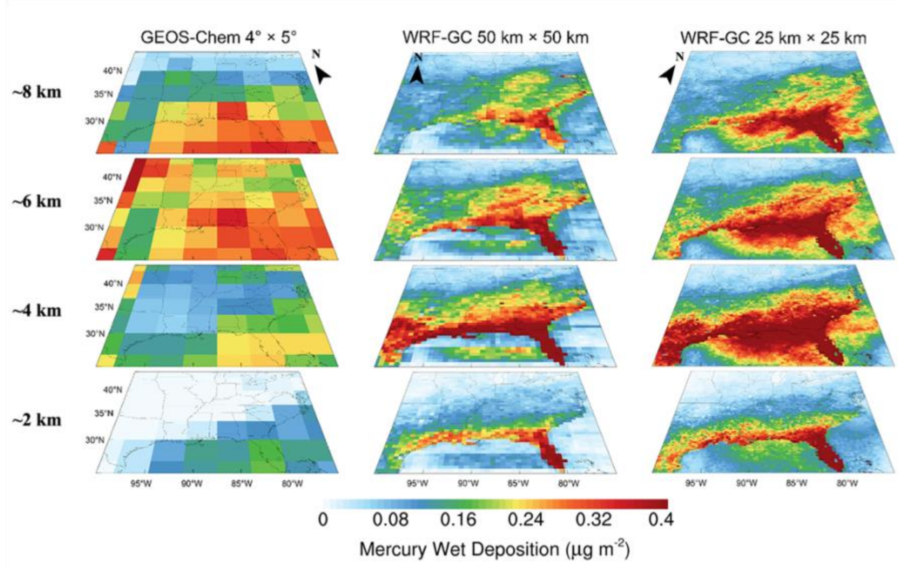

Fig. 8 Comparation of total Hg wet deposition of GEOS-Chem and WRF-GC at different levels and resolution. From top to bottom is the simulation results for ~2 km, ~4 km, ~6 km, ~8 km level, respectively. The first column is GEOS-Chem 4° × 5° simulation results. The other two from left to right correspond to different WFF-GC resolutions: 50 km × 50 km, 25 km × 25 km.

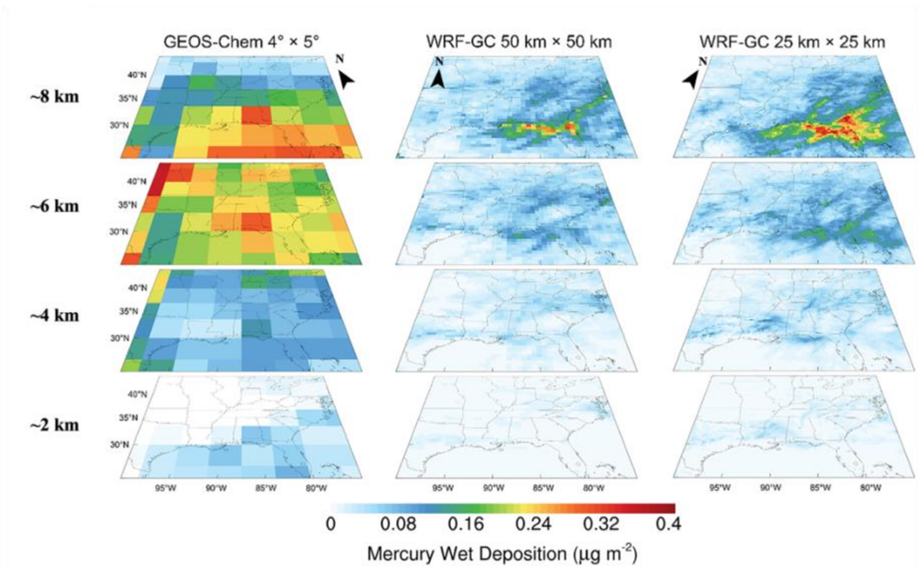

Fig. 10 Comparison of LS of GEOS-Chem and WRF-GC at different levels and resolutions. From top to bottom stands for Hg wet deposition at ~2 km, ~4 km, ~6 km, ~8 km, respectively. The first column is GEOS-Chem 4° × 5° simulation results. Other columns from left to right correspond to different WFF-GC resolutions: 50 km × 50 km, 25 km × 25 km.

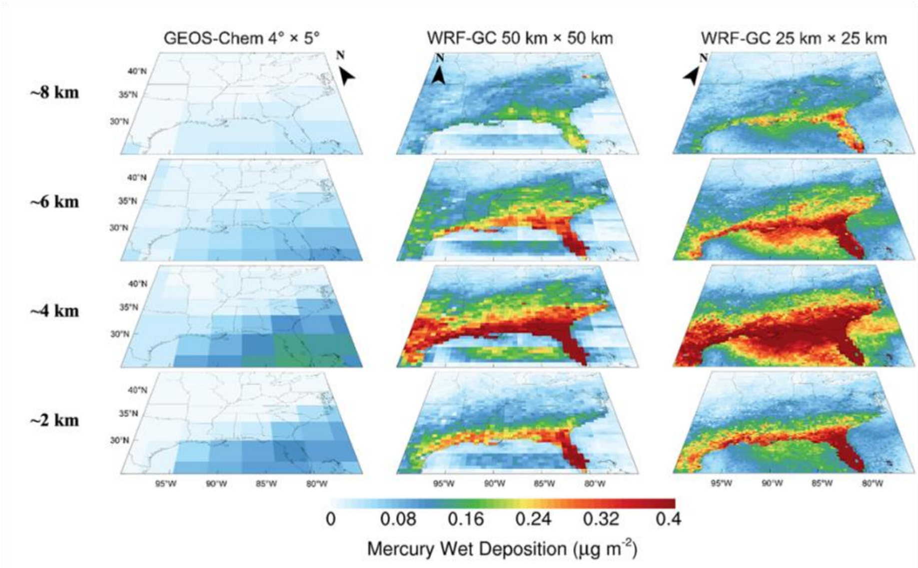

Fig. 11 Comparison of CONV of GEOS-Chem and WRF-GC at different levels and resolutions. From top to bottom stands for Hg wet deposition at ~2 km, ~4 km, ~6 km, ~8 km, respectively. The first column is GEOS-Chem 4° × 5° simulation results. Other columns from left to right correspond to different WFF-GC resolutions: 50 km × 50 km, 25 km × 25 km.

16. In section 3.2-3.3: did you compare the model simulated precipitation vs precipitation measured at MDN sites?

Thanks for your suggestion. Here, we did not compare model simulated precipitation to MDN sites' measured precipitation because MDN sites only have total precipitation data. What we focus on in this paper is the Hg wet deposition caused by different types of precipitation. Thus, we thought it should not be necessary to compare precipitation site by site.

To prove our assumption, Fig. 3 shows the comparison of precipitation between CMAP, GEOS_FP meteorological dataset, and WRF-GC-Hg simulation results, with different types of precipitation (total, large-scale, and convective precipitation).

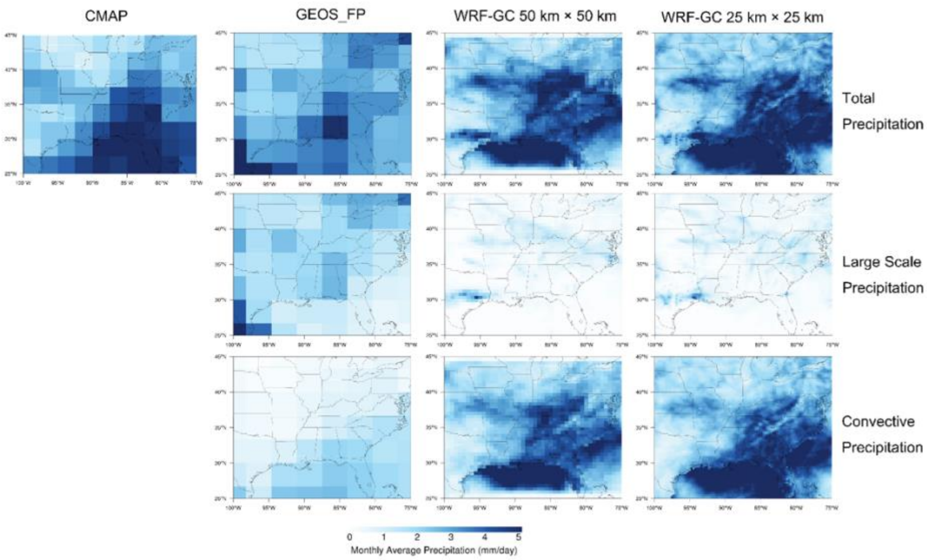

Fig. 3 Monthly average precipitation from July to September 2013 (Left top corner: CPC Merged Analysis of Precipitation; From the second to fourth column: GEOS_FP offline meteorological dataset, WRF-GC precipitation in 50km × 50km, 25km × 25km resolution; From top to bottom: three-month average precipitation, non-convective precipitation, convective precipitation)

17. This work only focuses on only one year (2013) study, what do you think of the interannual variability of the precipitation and Hg wet deposition?

Although WRF-GC has many advantages, as we mentioned before, for this research, when we increase the resolution to 25 km × 25 km, a three-month simulation was already computationally intensive. With this, we only applied the year 2013 as a case study, but it was able to prove our assumption. In our future work, we definitely could simulate more years.

Reference:

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