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 the reviewer's comments are given below.

Referee 2

This study presents a representative demonstration of GEOS-CHEM multi-phase, multi-species mercury atmospheric chemical transport algorithms in the WRF-GEOS-Chem model and addresses one extant scientific question through multi-scale regional simulations. This novel tool is a major incremental advance in regional mercury modeling capabilities in scope for GMD and for scientific questions within the scope of EGU. This is also a first demonstration of the ease of transferring GEOS-CHEM algorithms to WRF-GC without the additional model development, porting, and QA/QC efforts usually required, another large incremental advance in regional atmospheric chemical transport modeling beyond the mercury application shown here. Thus, the title reflects the contents of the article as demonstration of model development. The overall presentation well-structured and clearly written, and ready for copy editing for fluency and precision. The original contributions are highlighted, and the number and quality of references are appropriate.

The manuscript nearly meets criteria for initial model demonstration, with a few important omissions. I recommend acceptance following minor revisions to evidence, context, methods and assumptions, and supplemental materials to support reproducibility:

1. Comparison of capabilities and performance to prior regional Hg modeling in the domain and elsewhere is a basic expectation, and notably absent. First, the authors must identify the differences and advantages of this model's atmospheric chemical transport mechanisms to other regional models that resolve Hg (e.g., CMAQ, CAMx, WRF-CHEM, STEM-Hg), cite them, and clearly communicate the value and novelty. A table comparing mechanism features would help. Performance comparison to regional scientific and regulatory modeling over the study domain is essential to communicating the applied value of this tool and would best be achieved by comparing to contemporary community model performance benchmarks (e.g., Emery et al., 2017) for the same season. Qualitative and quantitative summaries of the performance advantages of this regional model should appear in the abstract.

Thanks for your suggestion. We added content to better describe the difference between models and why we chose WRF-GC as our working model:

First, WRF-GC takes advantage of flexible resolution, better meteorology simulation from WRF, and updated Hg chemistry library from GEOS-Chem, while other models have not updated Hg chemistry mechanisms for a long time.

Second, this model will potentially benefit a large number of users that are familiar with WRF-Chem and GEOS-Chem.

Third, the WRF-GC-Hg model has relatively good portability. WRF-GC is off-the-shelf ready to work, and all we need to do is port GEOS-Chem Hg chemistry to WRF-GC-Hg.

Fourth, we wanted to focus on a scientific problem of high Hg wet deposition in the southeastern US, so we did not spend much effort on the intercomparison of simulation performance between different models.

However, considering that WRF-GC-Hg is a novel model for Hg simulation, we decided to add more description of its development in the revised paper:

Line 60-70: Except for GEOS-Chem (Zhang et al., 2012), Hg simulation was implemented into many models like WRF-Chem (Gencarelli et al., 2014), CMAQ (Bullock and Brehme, 2002), and STEM-Hg (Pan et al., 2010), etc. Models like WRF-Chem and CMAQ also use WRF for meteorology simulation with different Hg chemistry libraries that have not been updated in recent years. 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.



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

Line 76: The model's framework is shown in Fig. 1.

Line 94: We implement a complimentary Hg chemistry library (see Fig. 1) to the WRF-GC model by first introducing ...

Line 101-102: We use the WHET emission inventory $(1^{\circ} \times 1^{\circ})$ for the anthropogenic Hg emissions (Zhang et al., 2016) as well as natural emissions and re-emissions inventory $(4^{\circ} \times 5^{\circ})$ from Horowitz et al. (2017) (see Fig. 1).

2. The multi-scale comparison presented requires additional evidence to support the conclusions and more information on data and assumptions. Fig. 3 must include results from all three scales. The article must describe the spatial and temporal surrogates and processes used to allocate emissions from each sector. The authors are strongly

encouraged to present maps of total and sectoral emissions maps at each scale in the supplemental materials. In Fig. 4, panels 2 and 3 appear identical—this should be addressed to provide evidence of different resolutions, spatial patterns, and the differences in magnitude described in the manuscript. The discussion (206-213) should reflect the roles of the emissions inventory and downscaling as limiting factors in resolution for this study rather than an inherent process resolution issue below 50 km.



• Multiscale of Hg surface concentration are shown in Fig. 3 now.

Fig. 2 Comparison of monthly average Hg surface concentration of Hg⁰ (top), Hg² (middle), HgP (bottom) from July to September 2013. The left panel is GEOS-Chem $4^{\circ} \times 5^{\circ}$ simulation. The other two columns from left to right correspond to different WFF-GC resolutions: 50 km × 50 km, 25 km × 25 km. Dots on the top row of the panel represents Hg⁰ observation data from AMNet of NADP (<u>http://nadp.slh.wisc.edu/AMNet/</u>).

• For emission inventories, WRF-GC uses HEMCO (Lin et al, 2021). HEMCO is a software component designed for computing emissions. Users are allowed to select an ensemble of emission inventories, as well as state-dependent emission algorithms with the ability to re-grid, combine, over-write, scale, etc. emissions from various inventories by a configuration file, even adjusting model species with proper units. No further modification is needed to the model source code. Therefore, we do not need to do like using WRF-Chem, etc. to prepare emission files. HEMCO can do work, like linear

interpolation for simulations, automatically, so this is an advantage for GEOS-Chem and WRF-GC.

• Thanks for pointing this out. We also recognize this problem. Simply increasing resolution is only better for meteorology simulation because it can help the model to resolve small-scale weather conditions, but it is not that useful for emission. Since the resolution of emission inventories is fixed, the higher Hg wet deposition at higher resolution is mainly caused by meteorological precipitation but not emission.

Line 231-234: Here the increase of resolution is only better for meteorology simulation because the finer resolution can help the model resolve small-scale weather conditions. Since the resolution of emission inventories is fixed ($1^{\circ} \times 1^{\circ}$ and $4^{\circ} \times 5^{\circ}$), with higher resolution, more Hg wet deposition will be shown in our result because more convective precipitation is captured by the model.

3. The arbitrary filter of 70% data availability for site inclusion should be justified, and other observational data QA/QC steps described.

We chose this threshold following Holmes et al. (2010). The MDN and AMNet data have been quality checked before releasing. For example, all MDN data have a quality index like "A", "B", "C". ("D" is omitted).

Line 162: We only take sites that contain at least 75% availability of data for three months (Holmes et al., 2010).

4. The article and the GitHub and Zenodo repositories include all files necessary and sufficient for reasonable reproducibility. Addition of a WPS pre-processor file for the domains and scripts for the analysis and figures presented would bring the study close to full reproducibility.

Thanks for your advice. The whole WRF-GC model is too large to upload the whole folder. For parent model WRF-GC, Lin et al, 2020 and GitHub repository (<u>https://doi.org/10.5281/zenodo.3550330</u>) provided a detailed guide of porting WRF-GC model. Our research is based on the WRF-GC model. Anyone interested can first install WRF-GC model (https://fugroup.org/index.php/Installing_WRF-GC) and replace the necessary file from our GitHub repository. You may set your desired domain area and simulation period, while the other settings are the same as a normal WRF-GC simulation setup.

Line 325-326: The latest WRF-GC-Hg v1.0 is permanently archived at https://doi.org/10.5281/zenodo.6366777 (last access: 18th Mar 2022).

Reference:

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