

Response to Referee #2:

We thank the reviewer for the acknowledgment and the helpful comments and suggestions. We have provided our responses to the referee's comments below (in blue).

1. Comment: Shah et al., (2021) had a major revision to the GEOS-Chem mechanism described in Horowitz et al., (2017), how do the authors account for those changes? It is relevant to include a discussion on the mercury chemistry implemented in this study with Shah et al., 2021, which is the most updated mercury chemistry in a global model.

Shah et al., (2021) integrated the recent laboratory data of mercury and implemented a new chemical mechanism for atmospheric Hg into GEOS-Chem. The new mechanism in Shah et al., (2021) has more tracers and more complicated chemical processes compared to Horowitz et al., (2017). But it should be noted that the main purpose of developing CAM6-Chem/Hg is to test whether the online model could simulate the Hg cycle and for future prediction and coupling. The online simulation of CAM6-Chem/Hg is able to reproduce the observed surface and seasonal variations of TGM concentrations and the wet deposition fluxes reasonably well. Future model would benefit from both the updated Hg chemistry and online meteorology and chemistry.

We added the following sentence in line 360-362:

“Shah et al., (2021) presented a new chemical mechanism for atmospheric Hg in GEOS-Chem, and the CAM6-Chem/Hg presented here provides a convenient platform to integrate the updated Hg chemistry with online meteorology and atmospheric chemistry.”

2. Comment: Why were not the emissions from Streets et al., (2015) used in the study? The Global Mercury Assessment 2018 used it in their report and in Shah et al (2021).

Since the natural/legacy emissions (and initial conditions) used in this study are from the result of GEOS-Chem (Horowitz et al., 2017), which is driven by the emissions from Zhang et al., (2016). Therefore, we used the same anthropogenic emission inventory instead of Streets et al., (2019) to keep consistency.

We clarified this by modifying the sentence in line 94:

“The anthropogenic emissions used in this study are consistent with Horowitz et al., (2017). They are based on an improved 2010...”

3. Comment: Which version of the GEOS-Chem model output was used for the natural emissions? Add the details in the manuscript.

Thanks for pointing it out. The natural and legacy emissions are derived from the GEOS-Chem v11 (Horowitz et al., 2017). We clarified this by adding the following sentences in line 99-101 in the method part:

“The natural emissions and re-emissions from previously deposited legacy emissions are derived from the average of a 5-year simulation in GEOS-Chem v11 (Horowitz et al., 2017), including geogenic, biomass burning, soil, ocean, snow, and vegetation emissions.”

4. Comment: Is there a specific reason for choosing the simulation period 2011–2013?

Globally, observations of Hg are scarce and unfortunately becoming more scarce (e.g. many monitoring stations in AMNet and CAMNet are becoming inactive). We summarize the available observations of Hg to date (see Table S1 in SI) and found the data around 2011–2015 are the most abundant. So we choose the simulation period 2011–2013 to better constrain our model.

We clarified this by modifying the sentence in line 141:

“...the following three years (i.e., 2011–2013, **with abundant observations during the past decades**) for analysis (unless explicitly stated)”

References

- Horowitz, H. M., Jacob, D. J., Zhang, Y., Dibble, T. S., Slemr, F., Amos, H. M., Schmidt, J. A., Corbitt, E. S., Marais, E. A., and Sunderland, E. M.: A new mechanism for atmospheric mercury redox chemistry: implications for the global mercury budget, *Atmos. Chem. Phys.*, 17, 6353-6371, 10.5194/acp-17-6353-2017, 2017.
- Shah, V., Jacob, D. J., Thackray, C. P., Wang, X., Sunderland, E. M., Dibble, T. S., Saiz-Lopez, A., Černušák, I., Kellö, V., Castro, P. J., Wu, R., and Wang, C.: Improved Mechanistic Model of the Atmospheric Redox Chemistry of Mercury, *Environ. Sci. Technol.*, 10.1021/acs.est.1c03160, 2021.
- Streets, D. G., Horowitz, H. M., Lu, Z., Levin, L., Thackray, C. P., Sunderland, E. M., and Argonne National Lab. ANL, A. I. U. S.: Five hundred years of anthropogenic mercury: spatial and temporal release profiles, *Environ. Res. Lett.*, 14, 84004, 10.1088/1748-9326/ab281f, 2019.
- Zhang, Y., Jacob, D. J., Horowitz, H. M., Chen, L., Amos, H. M., Krabbenhoft, D. P., Slemr, F., St. Louis, V. L., and Sunderland, E. M.: Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions, *Proceedings of the National Academy of Sciences*, 113, 526-531, 10.1073/pnas.1516312113, 2016.