#egusphere-2025-4018

Response to the reviewers

Reviewer 2

Reviewer Comment 2.1 — This manuscript presents a modeling study that quantifies the influence of uncertainties in anthropogenic mercury (Hg) emissions, chemistry, and meteorology on simulated atmospheric Hg concentrations and wet deposition. The authors use a signal-to-noise ratio (SNR) approach to identify regions where model-observation comparisons are most suitable for evaluating anthropogenic emissions uncertainties. The topic is timely and relevant, the experimental setup is interesting, and the results are valuable for improving future Hg modeling and monitoring efforts. However, several issues need to be clarified and discussed in more depth before the paper can be considered for publication. My main concerns relate to (1) the comparison between modeled and observed Hg species, (2) the regional aggregation choices that affect the interpretation of SNR, and (3) the discussion of results and their practical implications for monitoring. Detailed comments are provided below.

Reply: We thank the reviewer for the positive assessment of the manuscript and for recognizing the current relevance of the study. We appreciate the constructive feedback and the identification of key areas where further clarification and discussion would strengthen the paper.

Reviewer Comment 2.2 — Consistency between GEM and TGM

In the Methods, the authors refer to "GEM" observations but later discuss "TGM" in the Results and Discussion. It is unclear what is actually being compared. Are the authors comparing observed GEM to modeled GEM + GOM + PBM?

Since atmospheric observations primarily represent GEM (as GOM and PBM are usually negligible and highly uncertain), the model–observation comparison should be limited to modeled GEM, not total gaseous mercury. This distinction must be clarified throughout the manuscript and in the figures.

Reply: We thank the reviewer for this important clarification. We agree that the terminology in the original manuscript was inconsistent regarding the species used in the model–observation comparison.

In the revised manuscript, all model–observation comparisons have been recalculated using modeled GEM only, consistent with the fact that the observational datasets represent gaseous elemental mercury. All relevant figures and metrics have been updated accordingly.

Reviewer Comment 2.3 — Need for a regional map and station overview

A map showing the boundaries of the defined regions (e.g., Arctic, Asia, Southern Hemisphere) and the location of the monitoring stations is essential. This would help readers understand which datasets are used and how the "regional" results are aggregated. For example, the observed Arctic

signal appears unusual, with surprisingly high concentrations in winter and spring. Which stations were included? The authors should verify that the seasonal cycles used are consistent with established patterns (see e.g., Angot et al., (2016a)).

Reply: We thank the reviewer for this important suggestion. In response, we have added a new figure (Fig. S5) showing the geographic locations of the monitoring stations used in this study, together with their regional classification (Arctic, Europe, Asia, USA, Southern Hemisphere midlatitudes, and Antarctic).

Reviewer Comment 2.4 — Regional aggregation and observed variability

The statement that "The discrepancy in modeled TGM in Asia cannot be easily constrained by model-observation comparisons using the current observation sites. The reason is that, in this case, the range of the model results falls within the range of TGM measurement variability" should be interpreted with caution. Part of this large observational variability may arise from spatial aggregation of heterogeneous sites across a vast region. Focusing on smaller, more homogeneous subregions could reduce the apparent observational noise and improve the ability to constrain emission-related uncertainties.

Reply: We thank the reviewer for this insightful comment. We have revised the Discussion section (Lines 270–276) to clarify that this pattern primarily reflects regional aggregation across heterogeneous monitoring sites in Asia. Additionally, the revised text now highlights subregional differences in SNR, as shown in Fig. 4C, where some areas (e.g., central China and Central Asia) exhibit higher signal of emission-inventory differences, while others (e.g., eastern China) show lower SNR.

Reviewer Comment 2.5 — Southern Hemisphere (SH) emissions and SNR interpretation

The authors conclude that SH monitoring networks are not ideal for constraining anthropogenic emissions uncertainties. While this may be true, the low SNR in the SH could also stem from poorly constrained emission inventories in this region. The apparent weak emissions signal may reflect limitations in the input data, not just a true physical insensitivity. This should be explicitly discussed.

Reply: We thank the reviewer for this important clarification. We agree that the low SNR in the Southern Hemisphere should not be interpreted solely as evidence of physical insensitivity to anthropogenic emissions. As the reviewer notes, the weak emissions signal may also reflect substantial uncertainties and potential biases in currently available emission inventories for the Southern Hemisphere, where anthropogenic sources—particularly ASGM and diffuse industrial activities—are known to be poorly constrained. We have revised the Discussion to explicitly acknowledge that the low SNR in the Southern Hemisphere likely arises from a combination of factors, including (i) the smaller fraction of global anthropogenic Hg emissions located in the Southern Hemisphere, (ii) strong influence of chemical processing and ocean–atmosphere exchange on atmospheric Hg variability, and (iii) limitations and uncertainties in the underlying emission inventories themselves.

Reviewer Comment 2.6 — Real-world feasibility of proposed monitoring strategies

Some of the recommendations in the Discussion appear unrealistic given logistical constraints. For example:

The suggestion that the Arctic's high winter SNR makes it "an excellent location" for studying background Hg ignores practical challenges (cold, darkness, snow/ice cover). The proposal to enhance wet deposition monitoring in South Africa overlooks that it is a dry region, often under drought conditions. Similarly, strengthening monitoring in Greenland and Arctic Russia is operationally difficult, especially in the current geopolitical context. These limitations should be acknowledged to ensure recommendations remain grounded in real-world feasibility.

Reply: We thank the reviewer for this important comment, highlighting the practical and logistical constraints associated with atmospheric mercury monitoring in remote and geopolitically sensitive regions. We agree that our original wording could be interpreted as underestimating real-world challenges related to climate, accessibility, infrastructure, and geopolitical context.
We have revised the Discussion to explicitly acknowledge these limitations. Our intent is to identify regions where observations would be most informative from a signal-to-noise perspective, recognizing that feasibility, cost, safety, and political considerations ultimately govern implementation.

We added the following text to the Discussion section:

"While the SNR analysis identifies regions such as the Arctic and specifically Arctic Russia, Greenland, as theoretically optimal for isolating anthropogenic Hg emission signals—particularly during winter—these findings should be interpreted in the context of substantial real-world constraints. Harsh environmental conditions, including extreme cold, prolonged darkness, snow and ice cover, and limited accessibility, pose significant operational challenges for sustained monitoring in the Arctic. In addition, strengthening monitoring coverage in remote areas such as Greenland and Arctic Russia faces logistical, infrastructural, and geopolitical constraints."

"Although enhanced wet deposition monitoring in regions such as South Africa may be informative from a modeling perspective, such recommendations must be considered alongside regional climatology. Much of southern Africa is characterized by arid or semi-arid conditions and recurrent droughts, limiting the feasibility and interpretability of continuous wet deposition measurements."

Reviewer Comment 2.7 — Minor Comments

1) Line 42: Dastoor et al., 2024 should be Dastoor et al., 2025.

2) Figure 1: The emission inventories cover different time windows. Which years are compared? Please clarify this to avoid comparing "apples to oranges." Similarly, why did the authors use different emission years in their "inventories" simulations (e.g., 2015 for AMAP/GMA, 2012 for EDGAR, 2013–2015 for STREETS, and 2010 for WHET)?

3) Methods: Note that the GEOS-Chem version used is outdated and does not include the most recent Hg chemistry (Shah et al., 2021). The EDGAR inventory is also not the latest version, and another recent global inventory (Qiu et al., 2025) could be mentioned. The authors need not rerun simulations, but these limitations should be acknowledged in the Discussion.

4) Line 149: "GOM wet deposition flux", replace with "wet deposition flux."

5) Lines 196–197: Please consider citing recent studies describing the summertime phenomenon more accurately (Angot et al., 2016a; Araujo et al., 2022; Huang et al., 2025; Yue et al., 2023). Ahmed et al. (2023) refers to springtime events, and the Dastoor papers do not specifically address this point.

6) Lines 217–218: Please consider citing Angot et al., (2016a, c, b), which provide multi-year Antarctic Hg observations directly relevant to this discussion. I mention these papers not because

I am the author, but because they contain the most comprehensive long-term Antarctic datasets available to date.

7) Line 242: Typo: "MEteo."

8) Lines 329–330: The statement is valid only in winter; please clarify.

Reply: We thank the reviewer for the careful reading of the manuscript and for these helpful minor comments. We address each point below.

1) Corrected. The reference has been updated to Dastoor et al. (2025) in the revised manuscript.

2) We have clarified in the caption of Figure 1 that the inventories correspond to different representative years (EDGAR: 2012; AMAP/GMA: 2015; STREETS: 2013–2015; WHET: 2010). We were limited by which years are available from the different inventories. The EDGARv4.tox2 inventory only continued until 2012, only the year 2015 was available from the GMA inventory, and WHET only provided emissions for the year 2010.

3) We have addressed this comment through additions to the Methods and Discussion, as detailed in the response to Comment 1.4. We have also added in the discussion references to global inventories that were published after these simulations were conducted: "Future studies could address this point using new emissions inventories that have been published after these simulations were completed (Qiu et al., 2025; Muntean et al., 2024; Cui et al. 2024; Macfarlane et al. 2022.)

4) Corrected. The phrase has been replaced with "wet deposition flux."

5) We thank the reviewer for these helpful suggestions. This comment has been addressed in the revised manuscript. We very much appreciate these suggestions, which improved the scientific context of the manuscript.

6) We appreciate very much the reviewer drawing our attention to these comprehensive long-term Antarctic datasets.

7) Corrected to "Meteo".

8) We agree with this comment and thank the reviewer for the clarification.