#egusphere-2025-4018

Response to the reviewers

Reviewer 1

Reviewer Comment 1.1 — Gournia et al. present a modeling study aimed at assessing the contribution of uncertainty in anthropogenic Hg emissions to model errors with respect to surface observations. The goal of the study is to help identify regions where additional observations would be most effective in constraining anthropogenic emissions. The authors conclude that uncertainties in emissions are strongly influence surface concentrations in the northern hemisphere in the model, while uncertainties in chemistry dominate in the souther hemisphere. They identify eastern US, Greenland, and Russian Arctic as regions where observations could effectively constrain anthropogenic emissions.

Understanding and reducing uncertainties in emission inventories is important for scientific applications of Hg models and for informing policy decisions. This study address a critical aspect of Hg modeling.

Reply:

We appreciate the time and effort the reviewer has dedicated to providing valuable feedback on our manuscript. We are grateful to the reviewer for the insightful comments and summary of our study. We appreciate their recognition of the importance of quantifying anthropogenic Hg emission uncertainties and of our approach to identifying regions where observations can most effectively constrain emissions.

Reviewer Comment 1.2 — However I have significant concerns about the study's methods and conclusions, as outlined below:

One of the study's main conclusions is that observations in Greenland & the Arctic would help constrain Hg emission inventories. I find this conclusion difficult to accept. As shown by the authors in Fig 1, most of the uncertainty in Hg emissions is in Asia and S. America. It follows that more observations in these regions (and immediately downwind) would be most effective in reducing the emissions uncertainty, not observations in remote regions like the Arctic. This incorrect conclusion likely arises from their use of the "SNR" metric, which seems unsuitable for this purpose. The SNR values depend more on the day-to-day variation ("noise") in the modeled surface concentrations (Fig. 4), and less to the model uncertainties that the authors are tying to assess.

Reply: We thank the reviewer for this comment and for providing an opportunity to elaborate on the intent of the analysis and the basis for our conclusions. We agree with the general statement that observations in major source regions and their downwind areas are critically important for reducing anthropogenic Hg emission uncertainties. The largest absolute uncertainties in anthropogenic Hg emissions occur in Asia and South America, as shown in Fig. 1 and Fig. 2, and additional observations in these regions are important for improving emission inventories.

However, this study does not aim to determine which regions have the largest emission uncertainties. Instead, it focuses on where surface observations are most effective at constraining hemispheric-scale

anthropogenic emission uncertainties, given other sources of variability. For our purposes, temporal variability is a key indicator when assessing where observations are most effective. Variability represents a fundamental constraint on the information content of observations, rather than incidental noise. High temporal variability driven by meteorology, episodic emissions, and local processes directly limits the ability of surface measurements to isolate signals attributable to anthropogenic emission uncertainties. In this context, the Arctic and Greenland are highlighted as regions with high sensitivity to emission inventory differences relative to background variability.

This distinction between regions with large emission uncertainties and regions where those uncertainties are most detectable in observations is explicitly addressed through the signal-to-noise ratio (SNR) framework. In our equation, the "signal" corresponds to the propagated effect of differences among anthropogenic emission inventories on modeled concentrations, whereas intra-annual variability constitutes the "noise" that limits detectability of the signal. In high-emission regions (such as Asia and South America), although the emission uncertainty signal is large, it is often embedded within very large temporal variability driven by meteorology, episodic emissions, and local processes. As a result, the emission-driven signal is difficult to isolate using existing surface monitoring data, which is reflected in lower SNR values despite large absolute emissions.

By contrast, remote receptor regions such as Greenland and parts of the Arctic exhibit relatively low day-to-day variability and limited local source influence. In these regions, differences among emission inventories propagate into discernible differences in background Hg concentrations, resulting in high SNR values. This makes such locations particularly effective for constraining hemispheric-scale anthropogenic emissions, despite their distance from major source regions. We acknowledge that the SNR metric emphasizes detectability rather than the magnitude of emissions, and we have revised the manuscript text to more clearly distinguish between (i) regions where emissions are most uncertain and (ii) regions where observations are most effective for constraining those uncertainties. We highlight the complementary role of low-variability receptor sites in constraining large-scale emissions, alongside targeted observations in high-emission and downwind regions.

In addition, we also have edited the manuscript to emphasize that measurements in undersampled areas are very much useful and relevant for understanding sources, concentrations, and regional influences, and expansion of the monitoring network is needed. We hope that this clarifies our argument on where and when these analyses are most useful.

We added the following text to the Discussion section: "While the largest absolute uncertainties in anthropogenic Hg emissions occur in Asia and South America, our analysis identifies regions where background observations are most effective at isolating emission signals from other sources of variability, which is a distinct but complementary objective."

Reviewer Comment 1.3 — The study overlooks an important source of uncertainty in Hg modeling: the exchange of mercury between the atmosphere and land, ocean, and the biosphere. These exchanges are a significant source of uncertainty in Hg modeling and must be considered when assessing the relative importance of emission inventories compared to other processes in the model.

Reply:

We thank the reviewer for this clarification and agree that anthropogenic Hg emissions influence not only atmospheric concentrations directly, but also indirectly through their contribution to legacy Hg pools in land, snow, and ocean reservoirs, which in turn affect re-emissions to the atmosphere.

In the GEOS-Chem (v 12.8.01) for the Hg simulation, surface–atmosphere exchange processes (including soil, snow, and ocean re-emissions) are included but are not dynamically coupled to the anthropogenic emission inventories. As a result, higher anthropogenic emissions in one inventory do not lead to proportionally higher re-emissions in that simulation.

The objective of the study is to isolate the atmospheric signal of uncertainty arising from differences among anthropogenic emission inventories, independent of longer-term impacts associated with legacy Hg accumulation. We chose to keep re-emissions constant in order to better isolate the direct impact of anthropogenic emissions differences. Including inventory-specific legacy re-emissions, for example using a fully coupled, time-evolving multi-compartment Hg model, would complicate attribution of modeled atmospheric differences to specific sources of uncertainty, where differences in anthropogenic emissions would propagate into land and ocean reservoirs over decadal timescales.

However, we acknowledge that neglecting inventory-dependent re-emissions may lead to an underestimation of the full impact of anthropogenic emission differences, particularly in regions and seasons where surface re-emissions strongly influence atmospheric Hg. This limitation is now explicitly stated in the manuscript, and we emphasize that our results should be interpreted as a lower-bound estimate of the relative importance of anthropogenic emission uncertainties compared to chemistry and meteorology.

We added the following text to the Methods section: "Surface–atmosphere Hg exchange processes, including soil and ocean re-emissions, are included in GEOS-Chem but are prescribed independently of the anthropogenic emission inventory used in each simulation. As a result, differences among anthropogenic emission inventories do not propagate into inventory-specific legacy re-emissions. This modeling choice allows isolation of the atmospheric uncertainty attributable to inventories of direct anthropogenic emissions."

We added the following text to the Discussion section: "Because inventory-dependent re-emissions are not represented, the modeled response to anthropogenic emission differences likely underestimates the full influence of emissions on atmospheric Hg. Such an approach is beyond the scope of the present work and would complicate attribution of modeled atmospheric differences to specific sources of uncertainty. Therefore, our results should be interpreted as a lower-bound estimate of the relative importance of anthropogenic emission uncertainties compared to chemistry and meteorology."

Reviewer Comment 1.4 — The Hg chemistry in the model used in the study is outdated and the uncertainties in chemistry considered in the study do not reflect our current understanding. See Saiz-Lopez et al. 2020 (10.1073/pnas.1922486117) and related work. This affects the study's conclusion about the relative importance of emission uncertainty in comparison to uncertainty in chemistry.

Reply: Our intention with the chemistry group of simulations is to analyze differences in Hg cycling under two idealized yet incomplete representations of Hg chemistry. This allows us to identify which regions of the atmosphere would be more sensitive to drastic changes in the oxidation schemes. To

clarify this point, we have now added to the Methods section: "More advanced chemical mechanisms have since been developed which incorporate the oxidation of Hg^0 of both OH and Br, followed by oxidation by O_3 in a second-step (Saiz-Lopez et al., 2020; Shah et al., 2021; Castro et al., 2022; Saiz-Lopez et al., 2025). Nevertheless, by conducting two sets of simulations with radically different chemistry schemes (Br vs. OH/O_3), we can identify which regions of the atmosphere are most affected by chemical uncertainties".

We have also added the following text to the Discussion section: "Future studies could conduct similar analysis using other global mercury models (Dastoor et al., 2025), as well as updated Hg chemistry schemes (Saiz-Lopez et al., 2025; Shah et al., 2021)."

Reviewer Comment 1.5 — Minor and technical comments:

1) Table 1: Please clarify which quantity the uncertainty ranges refer to. Do they represent global emissions or emissions at the grid-point level?

2) Fig 1(B): Which species' emission range is depicted—TGM or GEM?

3) Fig 2: Please specify the years of the emission inventories being compared.

4) Table 2: The correct name of the meteorological dataset is MERRA-2, not MERRA.

5) Line 145: MERRA 2 & GEOS-FP are products of the same weather model (GEOS) and therefore do not represent meteorological uncertainty in any meaningful way.

6) Fig 4(A) and (B): Units for the color bars are missing.

Reply: We thank the reviewer for these helpful clarifications. We have addressed each point as detailed below.

1) The uncertainty ranges refer to global total anthropogenic Hg emissions as reported by the respective inventory sources. We have clarified this in the Table 1 caption.

2) Fig. 1B depicts the range of total anthropogenic Hg TGM emissions across inventories. We have clarified this in the figure caption.

3) We have added the inventory years corresponding to each dataset used in Fig. 2 to the caption for clarity.

4) Corrected. All references to "MERRA" have been updated to MERRA-2 throughout the manuscript, including Table 2 and Section 2.2.

5) We intended to assess the sensitivity of Hg simulations to differences in assimilated meteorological fields commonly used in GEOS-Chem applications. We have clarified this limitation in the discussion section, as follows: "The Meteo simulations are intended to assess sensitivity to commonly used assimilated meteorological datasets within the GEOS framework rather than to represent the full range of meteorological model uncertainty."

6) Units have been added to the Fig. 4 caption to clarify the quantities shown in Fig. 4A and 4B.

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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.