Responses to Reviewers' Comments

We sincerely thank the reviewer for the constructive and thoughtful comments, which have significantly improved our manuscript. We have carefully considered all comments and revised the manuscript accordingly. Below are our detailed responses to your comments. Revised texts are marked red in the revised manuscript.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Major Comments

The study by Zhang et al. measured atmospheric GEM concentrations and isotopic compositions at long-term monitoring urban site and multiple short-term monitoring urban, suburban, rural sites in China and Pakistan. Based on the comprehensive observations, the authors show notable declines in GEM concentrations and clear changes in the isotopic compositions in the urban atmosphere. By using a Hg isotope mixing model, they quantify the relative contributions of anthropogenic emissions and evidence that the GEM declines are mainly driven by the control of anthropogenic emissions. This research is well designated. I broadly agree with the interpretations throughout the manuscript. I would suggest a minor revision of this manuscript (the manuscript is well written currently). Some of the minor suggestions are presented below.

RE: We sincerely appreciate the reviewer's constructive comments for improving the clarity and rigor of the manuscript. All comments have been carefully addressed in the revised version, as detailed below.

The source apportionment: in this study, the authors use a δ^{202}Hg and Δ^{199}Hg mixing model to quantify the source contributions. I would suggest to use the Δ^{199}Hg and Δ^{200}Hg mixing model as previous studies. This is because the GEM δ^{202}Hg in the boundary layer apt to modified by vegetation uptake process (particularly at the suburban and rural sites in this study), while the Hg-MIF signals are relative stable. As seen from Fig. 3a, some of the δ^{202}Hg and Δ^{199}Hg assemble especially in the background areas are not fully encompassed by δ^{202}Hg and Δ^{199}Hg of the three source endmembers. The authors argue that the Δ^{200}Hg of anthropogenic emissions overlap surface re-emissions. However, the Δ^{199}Hg of anthropogenic emissions and surface re-emissions are distinguishable, and this could enable the source quantification of these two sources. In addition, the Tianjin sampling site is close to seas, and the seawater re-emissions should be also considered. A recent study reported mean Δ^{199}Hg and Δ^{200}Hg values of -0.13‰ and 0.02‰, respectively, for ocean emissions (Fu et al., 2026, NSR). This oceanic signature is identical to soil re-emissions. Therefore, using the Δ^{199}Hg and Δ^{200}Hg mixing model would help to understand the contributions from the two most important natural emissions (e.g., soil + seawater reemission).

RE: Thank you for this important and insightful comment. We agree that MIF signatures are generally more conservative than δ^{202}Hg, and appreciate your suggestion to consider potential seawater re-emission. We now explicitly acknowledge the potential limitation of using δ^{202}Hg in boundary-layer environments, and adopted your proposed Δ^{199}Hg–Δ^{200}Hg model. We have also expanded the discussion of natural re-emission sources by considering the possible contribution of marine Hg re-emission. It is noted that the apportionment results from the new Δ^{199}Hg–Δ^{200}Hg

model are generally consistent with those of the original $\delta^{202}Hg$–$\Delta^{199}Hg$ model. Thus, our main conclusion (i.e., the observed decline in urban GEM was primarily driven by reduced anthropogenic emissions, while natural re-emission sources may become relatively more important under conditions of lower anthropogenic input) is still valid.

Revised text (Line 206): Given Tianjin's proximity to the Bohai Sea, seawater re-emissions are also expected to contribute to surface Hg fluxes. Recent field measurements report mean $\delta^{202}Hg$, $\Delta^{199}Hg$, and $\Delta^{200}Hg$ values of −1.04 ± 0.32‰, −0.13 ± 0.10‰ and 0.02 ± 0.02‰, respectively, for marine re-emissions (Fu et al., 2026). These values closely overlap with those reported for urban soil re-emissions (Zhu et al., 2022). As a result, marine and urban soil surface re-emissions are combined into a single surface re-emission endmember, with its Hg isotope values represented by those of urban soil re-emissions (Zhu et al., 2022).

Specific comments:

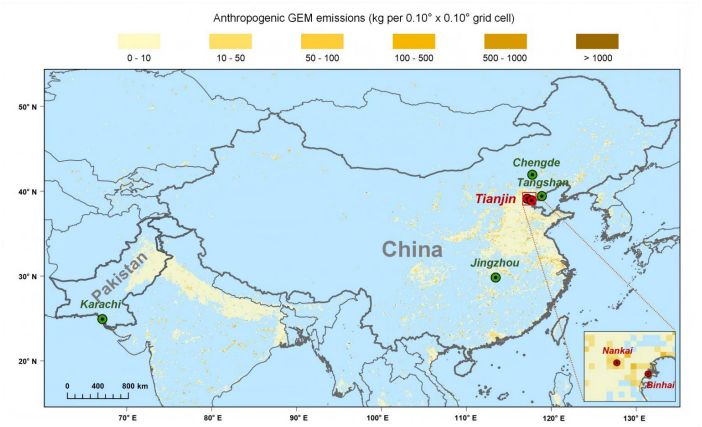
Line 13: please add 'mean' before GEM concentration.

RE: "mean" is now added before "GEM concentration".

Fig.1: better to show the location of Karachi in this figure.

RE: The location of Karachi is now added in the updated Fig. 1.

Revised figure:



Line 127: please specify whether the sampling flow rate is operated under the standard temperature and air pressure.

RE: The reported flow rate of 4.0–5.5 L min^{-1} refers to the measured ambient flow rate during sampling. For concentration calculation, the cumulative sampled air volume was normalized to standard temperature and pressure (273.15 K and 101.325 kPa) using the recorded ambient temperature and pressure. We have clarified this point in the revised manuscript.

Revised text:

Line 128: Air was drawn through the traps at ambient flow rates of 4.0–5.5 L min^{-1}, with particulate matter removed upstream using a 47 mm quartz fiber filter housed in a Teflon filter pack.

Line 182: For active pump-trap sampling, GEM concentration was calculated as the blank-corrected Hg mass collected on each CLC trap divided by the sampled air volume normalized to standard temperature and pressure (273.15 K and 101.325 kPa).

Section 2.2: please provide the sampling interval or duration for each sample at the sampling sites, including the pump-trap and passive samples.

RE: We have provided the sampling durations for both active pump-trap and passive samples. Specifically, the deployment dates and sampling durations of passive samples are provided in Tables S2–S3, while the start/end times of active pump-trap samples are provided in Tables S5–S7. To improve clarity, we have further added sampling duration information in the Section 2.2.

Revised text (Line 135): At Tianjin (TJ-Nankai and TJ-Binhai), active sampling campaigns were conducted in November 2018 (Phase I, ~1 day per sample), from October 2021 to September 2022 (Phase II, 1–3 days per sample), and from December 2024 to January 2025 (Phase III, with separate day (07:00–18:00) - night (18:00–07:00) samples) at TJ-Nankai, but only from December 2024 to January 2025 (Phase III, ~1 day per sample) at TJ-Binhai (Tables S5–S7 of the Supplementary Material). To facilitate long-term monitoring and inter-city comparisons, paired MerPAS samplers were deployed at all study sites. For the long-term monitoring at TJ-Nankai, individual passive samples were deployed for 40–53 days in Phase II and 29–56 days in Phase III, yielding total monitoring periods of 192 and 343 days, respectively (Tables S2–S3 of the Supplementary Material).

Section 2.5: please add the method for the calculation of GEM concentrations using the pump-trap. Are all concentrations reported for the STP conditions?

RE: Yes, all reported concentrations were normalized to STP conditions.

Revised text (Line 182): For active pump-trap sampling, GEM concentration was calculated as the blank-corrected Hg mass collected on each CLC trap divided by the sampled air volume normalized to standard temperature and pressure (273.15 K and 101.325 kPa).

Line 221: add ‘mean±1sd’ after the values.

RE: To avoid repeatedly adding ‘mean±1sd’, we have clarified the data description at the beginning of the “Results and discussion” section: In the following, data is reported as mean ± 1SD uncertainty for each category of samples, unless otherwise indicated.

Line 223: please note ‘mean±1sd’ for the values reported in Beijing and Shijiazhuang.

RE: Please see our response to your comment at Line 225.

Line 235: add ‘mean,’ before n =35.

RE: Please see our response to your comment at Line 225.

Line 240: better to show the exact p value when it is higher than 0.05.

RE: We have shown the exact *p* value.

Revised text (Line 257): Relative to Phase I, GEM concentrations declined by ~68% in Phase II and ~66% in Phase III, with no statistically significant difference between Phases II and III (independent sample *t*-test, $p = 0.59$).

Line 245: add 'mean' before δ^{202}Hg.

RE: Please see our response to your comment at Line 225.

Line 246: change 'directly' to 'mainly'?

RE: We agree and have changed "directly" to "mainly".

Line 247-248: add 'mean±1sd' before n=16. Same in line 249, 251 and 255.

RE: Please see our response to your comment at Line 225.

Line 257: show the real value instead of >0.05.

RE: We have shown the exact p value.

Revised text: Hg isotopic compositions were broadly similar between Phases II and III, with no significant differences in δ^{202}Hg (independent sample *t*-test, $p = 0.75$) or Δ^{200}Hg (independent sample *t*-test, $p = 0.35$), although Δ^{199}Hg differed significantly (independent sample *t*-test, $p = 0.04$). Nevertheless, both Phase II and Phase III differed significantly (independent sample t-test, all $p < 0.01$) from Phase I in δ^{202}Hg, Δ^{199}Hg, and Δ^{200}Hg.

Line 262: add mean before GEM.

RE: Please see our response to your comment at Line 225.

Line 267-268: not note these are mean values.

RE: Please see our response to your comment at Line 225.

Line 277: add 'mean' before values.

RE: Please see our response to your comment at Line 225.

Line 303: please add 'mean±1sd' after the D200Hg values.

RE: Please see our response to your comment at Line 225.

Line 309: add 'mean' before GEM.

RE: Please see our response to your comment at Line 225.

Line 324: please add 'mean' before δ202Hg and D199Hg. please also check other place if the values are referred to mean values.

RE: Please see our response to your comment at Line 225.

Line 343: please add 'on average' before contributing.

RE: We have added "on average" before "contributing".