Response to Reviewer #1's Comments

The authors have addressed my questions and comments about their PMF application and a few specific comments.

1. However, I have a few remaining concerns. First, I don't find their response convincing, especially to one of the main questions I had: why did they estimate the oceanic evasion flux of GEM after removing the anthropogenic contribution? This estimation is after all the crux of the study. They replied that "(t)he purpose of this recalculation is to assess potential changes in the marine mercury flux under the scenario of reduced anthropogenic emissions". But does this imply that anthropogenic emissions should not be reduced simply because oceanic evasion might increase as a result? According to Eq. 1, removing all the anthropogenic contribution in Ca, while assuming that Cw remains unchanged, increases the concentration gradient and thereby inflates the estimated evasion flux. The issue is whether Cw would in fact remain constant if all anthropogenic contributions were eliminated. I suspect not—particularly in near-coastal regions, where reduced Ca would likely lead to less dry deposition to the ocean surface, and thus a decrease in Cw. Ignoring this positive feedback between anthropogenic emissions and oceanic evasion is, in my view, a major flaw in the study.

Response: Thank you for your insightful comments. We agree with the reviewer's opinion that it is unreasonable to recalculate the sea-air exchange flux by directly deducting the anthropogenic contribution. In the original submission, we aimed to evaluate the instantaneous mercury flux by changing the inputs into the model. However, we also realize that it will be a long-term process by cutting off all the anthropogenic mercury emissions. Thus, the Cw concentrations should gradually change along with the decreases of anthropogenic mercury emissions as you commented. In this regard, the variation of Cw under the scenario of anthropogenic mercury emissions reduction is hard to predict.

In the revised manuscript, we have removed the inappropriate discussions in Line 581-591 and added some discussions about the mercury sea-air exchange observed in different oceanic regions.

Line 572-577: This finding coincided with the discussions above that natural TGM exhibited much higher concentrations in the ECS and YS than in the BS.

Due to the higher concentrations and contributions of anthropogenic TGM in the BS, the release of mercury from the ocean was significantly suppressed, which likely explained the relatively low sea-air exchange flux of mercury there.

2. Second, the authors simply removed the TGM/BC ratio originally presented in the abstract in response to my question regarding its significance. However, the relevant content remains in the manuscript without further consideration. Even the original values were not updated despite the use of a different dataset and recalculated values (See Lines 428-436). BC concentrations

naturally decreased over the ocean as the distance from the coast increased. However, TGM concentrations are more complex, particularly given the authors' own finding of a significant oceanic source for GEM. The pattern of TGM/BC varied between region south and north of ~32.5 ºN: south of this latitude, the ratio increases with increasing distances from the coast, whereas the pattern to the north is more complicated. This suggests that the TGM/BC ratio is rather confounding, doesn't it? I have no objection to including this result in the manuscript, but the authors must provide a more meaningful discussion of its implications, which is currently lacking.

2.1 Even the original values were not updated despite the use of a different dataset and recalculated values (See Lines 428-436).

Response: Thank you for your comments. As you mentioned that "Even the original values were not updated despite the use of a different dataset and recalculated values (See Lines 428-436).", this refers to the TGM/BC ratios observed at DSL and HNI as presented in Figure 4d. Actually, the TGM/BC ratios at these two sites were calculated based on the observational data from October 14 to November 4, 2020, i.e., the time-series shown in Figure 4a. In the previous revision, the data used in Figure 4 was not changed. We guess that you meant the dataset that was used for PMF was changed, however, that didn't influence the calculation of the TGM/BC ratios. Thus, the TGM/BC ratios remained unchanged.

2.2 BC concentrations naturally decreased over the ocean as the distance from the coast increased. However, TGM concentrations are more complex, particularly given the authors' own finding of a significant oceanic source for GEM. The pattern of TGM/BC varied between region south and north of ~32.5 ºN: south of this latitude, the ratio increases with increasing distances from the coast, whereas the pattern to the north is more complicated. This suggests that the TGM/BC ratio is rather confounding, doesn't it? I have no objection to including this result in the manuscript, but the authors must provide a more meaningful discussion of its implications, which is currently lacking.

Response: We appreciate for your insightful comments and totally agree that the interpretation of the TGM/BC ratio indeed requires more in-depth discussions.

In Section 3.2, we aimed to emphasize the importance of marine emissions of TGM following Section 3.1. Ideally, the TGM/BC ratio should show an increasing trend with the increase of distances away from the coasts as the ocean emissions of TGM sustained while anthropogenic emissions over the ocean significantly diminished compared to the terrestrial areas. This could be verified by the significant difference of the TGM/BC ratio between DSL and HNI, even these two sites were only 150 km away. However, we only used the TGM/BC ratio as a qualitative index. That's why we further built a quantitative method to quantify the anthropogenic and natural contributions to TGM in the following sections.

We totally agree with you that the observed TGM/BC ratio exhibited distinct patterns in the East China Sea and the Yellow Sea. In the East China Sea, the TGM/BC ratio increased with distance from the coast, aligning with our expectations as anthropogenic influences diminished

farther offshore. However, the Yellow Sea showed a more complex situation, lacking the consistent trend observed in the East China Sea. This should be attributed to the Yellow Sea being a comparatively enclosed basin with more complex sources of TGM and BC, simultaneously influenced by emissions from mainland China and the Korean Peninsula. In details, as visualized in Figure 4e, the very northern, western, and eastern cruise legs in the Yellow Sea showed relatively low TGM/BC ratios compared to the other cruise periods. These cruise legs above were geographically close to Liaoning province in northeast China, the North China Plain, and the Korean Peninsula. Thus, more influences from the terrestrial emissions explained the low TGM/BC ratios. In contrast, the spatial distribution trend of TGM/BC ratios in the East China Sea was transparent. Overall, we think the application of the TGM/BC ratio is still doable. But the usage of this index should consider various factors as you commented, thus we emphasize that this index is only used as a qualitative index.

In the revision, we have made the following changes.

Line 401 - 405:

To evaluate the relative importance of anthropogenic and natural sources to TGM, the ratio of TGM/BC was introduced as a qualitative index. Since TGM and BC shared common anthropogenic sources, and TGM had additional natural sources, an increase in the TGM/BC ratio may indicate the growing importance of natural source contributions, and vice versa.

Line 410 - 421:

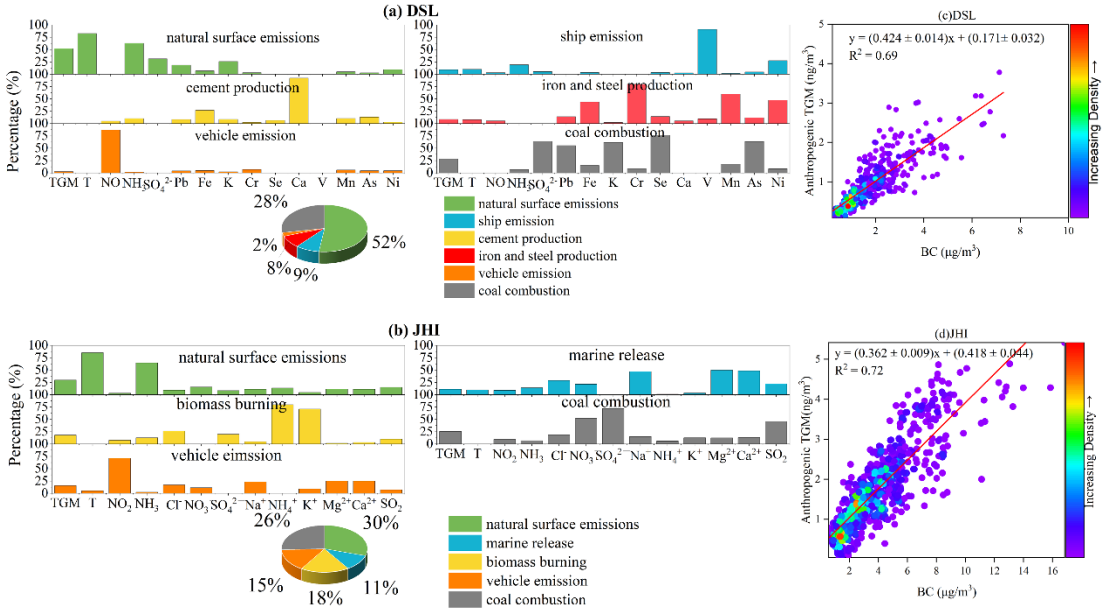
The cruise measurement illustrated the spatial distribution of the TGM/BC ratio over YS/ECS (Figure 4e). In the East China Sea, the TGM/BC ratio increased with increasing distances away from the coasts. For instance, the TGM/BC ratio near the coasts typically ranged from 0.3 to 5.2 ng μg^{-1}, while offshore values generally fluctuated between 8.6 and 22.9 ng μg^{-1}. This indicated the contribution of natural sources to TGM obviously increased over the open ocean waters. However, this spatial trend was not observed in the Yellow Sea. As depicted in Figure 4e, the very northern, western, and eastern cruise legs in the Yellow Sea showed relatively low TGM/BC ratios compared to the other cruise periods. This phenomenon should be due to the Yellow Sea being a comparatively enclosed basin, as these cruise legs above were geographically close to Liaoning province in northeast China, the North China Plain, and the Korean Peninsula. Thus, more influences from the terrestrial emissions induced the low TGM/BC ratios.

3. Third, I still do not fully understand the authors' choice of the time period for the DSL data. In the original manuscript, I pointed out a mismatch in time periods: DSL data from 2015-2019 and HNI from 10/14/2020-11/4/2020. This was a critical concern, as the authors used the anthropogenic portion of ambient TGM, regressed as a function of BC concentrations from DSL, to further estimate the oceanic flux at HNI. In the revised version, they used DSL data from Oct-Dec 2020, still not a match with the HNI period. Wouldn't the author want to justify their selection of the DSL time frame? Additionally, to separate anthropogenic TGM from TGM in the cruise measurements over the ECS and YS, the authors used the anthropogenic

TGM-BC relationship derived from DSL data when backward trajectories originated from DSL, and used a relationship from JHI when backward trajectories were northerly. However, the JHI period also mismatched the cruise measurement periods. In doing so, the authors seemed to assume that the anthropogenic GEM-BC relationship remains constant over time. If this is indeed their underlying assumption, shouldn't they make an effort to demonstrate its validity?

3.1 In the revised version, they used DSL data from Oct-Dec 2020, still not a match with the HNI period. Wouldn't the author want to justify their selection of the DSL time frame?

Response: Thanks for your comments. In the previous revision, we used DSL data from Oct-Dec 2020 with the purpose of incorporating more data points. As suggested, we have now exactly matched the time period of the DSL data used in our analysis with the observation period at Huaniao Island during October 14 to November 4, 2020. Consequently, Figure 5 in the manuscript has been revised as shown below. Related calculations throughout the text have also been updated accordingly.



3.2 Additionally, to separate anthropogenic TGM from TGM in the cruise measurements over the ECS and YS, the authors used the anthropogenic TGM-BC relationship derived from DSL data when backward trajectories originated from DSL, and used a relationship from JHI when backward trajectories were northerly. However, the JHI period also mismatched the cruise measurement periods. In doing so, the authors seemed to assume that the anthropogenic GEM-BC relationship remains constant over time. If this is indeed their underlying assumption, shouldn't they make an effort to demonstrate its validity?

Response: Thanks for your valuable comments and this is critical for this study. The fact was that due to the incapability of instruments and logistics, the JHI field campaign and the cruise measurements cannot be conducted during the same period. However, the JHI field campaign

and the cruise measurements occurred in the same season in the neighboring years. We think that the anthropogenic TGM-BC relationship should vary little within several years.

In order to validate this, we investigated the relationship between anthropogenic GEM and BC concentrations during winter periods of multiple years before 2020 at Dianshan Lake as shown in the figure below. It can be found that the regression equation (y=0.401x+0.394) during the winter of 2015-2018 was close to the regression equation (y=0.424x+0.171) obtained during this study period (see the revised Figure 5c). However, since there were no long-term measurements of TGM at JHI available, we cannot perform the similar validation as above.

Furthermore, we checked the mercury emission inventory (Feng et al., 2024). China's anthropogenic GEM emissions in 2019 and 2020 were 194.2 tonnes and 191.8 tonnes, respectively, showing negligible changes. Similarly, BC emission inventories in 2019 and 2020 obtained from the Multi-resolution Emission Inventory for China (MEIC, http://meicmodel.org.cn) also exhibited little variation (Geng et al., 2024).

Overall, we think that the relationship between anthropogenic TGM and BC should remain quite constant, especially in neighboring years of 2019 and 2020.

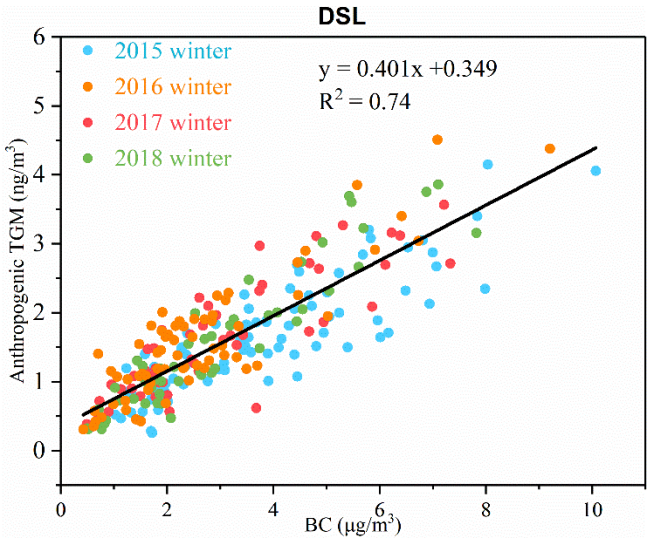


Figure. The relationship between anthropogenic GEM and BC concentrations (daily average) during winter periods of 2015-2018 at Dianshan Lake

In the revision, we have made the following changes.

Line 464 - 468: Furthermore, correlation analysis was conducted between the absolute contribution of anthropogenic sources to GEM and BC, yielding strong correlations at both DSL (Anthropogenic TGM = (0.424 ± 0.014)*BC + (0.171± 0.032), R^2 = 0.88, Figure 5c) and JHI (Anthropogenic TGM = 0.362 ± 0.009)*BC + (0.431 ± 0.044), R^2 = 0.86, Figure 5d), respectively.

Line 472 - 479: To validate the robustness of this relationship in different years, we derived the relationship between anthropogenic GEM and BC at DSL before 2020 based on the same methodology. It can be found that the regression equation during the winter of previous years

was close to that obtained during this study period (Figure S5). In fact, the mercury emissions (Feng et al., 2024) and black carbon emissions (Geng et al., 2024) were quite stable in the neighboring years of this study period. For instance, China's anthropogenic GEM emissions in 2019 and 2020 were 194.2 tonnes and 191.8 tonnes, respectively, showing negligible changes. Thus, it can be assumed that the relationship between anthropogenic GEM and BC remained relatively constant.

4. Fourth, one of my specific comments on the original version was about their explanation for the effect of temperature on seasonal variation in DGM. What they added in lines 557 – 560 merely stated the opposing effects of temperature and didn't really explain how such opposing effects resulted in the seasonal variation their data showed.

Response: Thanks for your comments. The seasonal variation in DGM in this study indicates that the DGM concentrations observed during our winter cruise were significantly lower than those reported in the literature from the summer and autumn cruises in the similar region. The warmer temperature in summer and autumn favored the DGM production in seawater but also promoted the release of DGM from seawater. In principle, the reduction of Hg^{2+} to DGM and the oxidation of DGM back to Hg^{2+} in seawater were simultaneously competing processes (O'driscoll et al., 2006). The increased release of DGM due to higher temperature pushes this reaction equilibrium towards reduction, thereby generating more DGM to maintain the high concentrations of DGM in seawater under warmer summer and autumn conditions. As reported extensively in the literature, both dissolved gaseous mercury concentrations and air-sea exchange fluxes were generally higher under warmer conditions (Huang and Zhang, 2021; Kuss et al., 2011; Wang et al., 2019).

In the revised manuscript, we have added the following discussions.

Line 543 - 549: This seasonal variation pattern of seawater DGM, with lower levels in winter compared to summer and autumn, can be attributed to the dynamic equilibrium between competing redox processes. This equilibrium can be represented as: Hg^{2+} + photo-reductants ⇌ DGM + photo-oxidants (O'driscoll et al., 2006). During warmer seasons, higher temperature accelerated the volatilization of DGM from seawater, and also drove the equilibrium toward Hg^{2+} reduction to replenish the lost DGM. Therefore, DGM concentrations in seawater were usually lower in winter due to suppressed redox processes.

5. Lines 374 – 376: I don't recall Lin et al. (2010b) and Soerensen et al. (2013) suggested that "wetting processes may promote the reduction of Hg to Hg0 in surface seawater". Through what mechanism?

Response: We thank the reviewer for their comments and apologize for the citation error. The reference "Lin et al. (2010b)" should in fact be "Lin et al. (2010a)" from the paper titled Empirical Models for Estimating Mercury Flux from Soils, which reported a positive correlation between Hg flux and soil moisture. Additionally, Soerensen et al. (2013) found that high Hg^0 concentrations (leading to high evasion) in near-coastal regions during the August 2008 shelf cruise were attributable to elevated total Hg inputs, partly influenced by rainfall.

However, neither study provides direct evidence supporting the claim that "wetting processes may promote the reduction of Hg(II) to Hg^0 in surface seawater." Therefore, in the revised manuscript, we have deleted the sentence "wetting processes may promote the reduction of Hg to Hg0 in surface seawater", and the original phrasing has been revised as follows.

Line 351 - 356: The positive correlation between humidity and TGM may be due to the fact that high humidity is typically associated with the stable atmospheric stratification, which promoted the accumulation of TGM. As for wind speed, it is a key parameter influencing air-sea exchange in the double-membrane theory model (Wanninkhof, 1992). For example, Soerensen et al. (2014) found a 2–4 times greater Hg^0 flux due to the high wind speed in the Intertropical Convergence Zone (ITCZ) region.

6. Line 499: How was that 5% uncertainty estimated?

Response: When performing the linear regression based on all input data, both the obtained slope and intercept carry uncertainties. The resulting 5% uncertainty (±5%) arises from the propagation of these fitting parameter errors.

7. Lines 588 – 590: I disagree that what they did underscored "the potential effects of diminished anthropogenic emissions on oceanic mercury cycling". See my first comment above.

Response: As shown in the response to the first question, we have removed the related discussions. Thanks for your suggestion.

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