Dear editor and dear reviewers,

Thank you for your letter and the reviews' comments concerning our manuscript entitled "A high-resolution marine mercury model MITgcm-ECCO2-Hg with online biogeochemistry" (GMD-2023-89). These thoughtful comments are valuable and helpful for improving our paper. We have studied the comments carefully and responded to the referee's comments below (in blue).

Response to Referee #1:

This manuscript presents the development and evaluation of a high-resolution online biogeochemical ocean mercury model. The model improves the resolution of mercury modeling to resolve better the influences including rivers and coastal areas, ocean turbulence, and the horizontal and vertical transport of substances in the ocean. The study is significant, as it fills the gap of only coupling offline biogeochemical models for ocean mercury research. The workload and computational cost are both considerable and impressive. I appreciate what this research group has done. It's an overall well-written paper that deserves publication. I have some comments but they are all minor.

We extend our heartfelt appreciation for your professional review of our article. Your constructive and insightful feedback and suggestions are truly valued. To effectively address the issues you raised, we have undertaken extensive revisions to our initial draft.

1. Comment: introduction, "Finally, we study the fate of the riverine discharge of Hg and the impact of nutrients over coastal waters near big river mouths on the transport of coastal Hg." The generalization doesn't seem to be entirely accurate. It is better to explain the specific influence of nutrients transported by rivers on marine mercury.

Thank you for pointing this out. Nutrients transported by rivers do not affect the mixing and transport of Hg in estuaries; instead, they pass through estuaries and participate in Hg-related biochemical processes after entering the oceans. This affects ocean biochemistry and, consequently, the biogeochemical cycling of marine Hg in a short period.

We made it clear by modifying the sentences in lines 76-77 as:

"Finally, we study the fate of the riverine discharge of Hg and the impact of riverine nutrients on the biogeochemistry of marine Hg."

2. Comment: Model description, can you explain why refractory particulate oxidized Hg is chosen as the tracer to characterize the fate of riverine discharge of Hg?

Zhang et al. (2015) explained that the amount of riverine Hg that settles in estuaries and on the shelf is highly sensitive to the fraction of the HgP pool specified as refractory. In their simulation, when the Hg from the river is specified as refractory, a greater portion of them is burial in the estuaries and less export to the open ocean. The actual fate of HgP discharges from rivers depends on the size of the refractory POC pool (Blair and Aller, 2012). Therefore, in lines 152-153, we state, "A separate refractory HgP tracer is used to simulate the particulate Hg from rivers (HgPR), reflecting its strong combination with terrestrial source POC." Furthermore, observational constraints on

seawater Hg concentrations and air-sea exchange suggest that the majority of Hg from rivers in the global ocean is refractory (Zhang et al., 2015).

Thus, we have modified the sentences in lines 152-153 as follows: With the observational constraints on seawater Hg concentrations and air-sea exchange, we choose a separate refractory HgP tracer (refractory particulate oxidized Hg or HgPR) to simulate the fate of Hg from the river, which also reflects its combination with terrestrial source POC.

3. Comment: line 236-238, "We find that riverine inputs of HgPR to the coastal ocean have a limited impact on Hg concentrations in surface seawater beyond the shelf region. HgPR is transported to the left or right along the coast by the Coriolis force, and the influence of currents and eddies leads to dispersion slightly further away from the coast." To provide a more accurate depiction, it would be better to check the logical order of the sentences, and give more description about "slightly further away from the coast."

4. Comment: line 251-253, "Hg exports from rivers are important to specific coastal areas, Liu et al. (2021) have raised further concerns about riverine Hg and a deeper understanding of the role of rivers in the global Hg cycle. The discharge of HgPR from rivers greatly impacts environmental pollution in coastal and shelf regions." This topic sentence does not correspond to the following.

Thanks for the two valuable pieces of advice. The sentences in lines 251-253 are a reasonable extension and explanation of the presentation of the results in Figure 5, so they are added to this paragraph in lines 236-238. To express the results more precise and logically, we have rewritten this paragraph in lines 236-238 and added explanation summarized from line 251-253:

"We find that when HgPR is transported from river mouths, it flows along the coast in an eddy shape and in the direction in which the Coriolis force works. However, most of them are burial in estuaries and less exported to the open ocean. Thus, HgPR has a limited impact on Hg concentrations in surface seawater beyond the shelf region and greatly impacts environmental pollution in coastal and shelf regions. Hg exports from rivers are important to specific coastal areas, Liu et al. (2021) have raised further concerns about riverine Hg and a deeper understanding of the role of rivers in the global Hg cycle."

The words in lines 253-255 as the new topic sentence for the paragraph, "Our model results show that HgPR is transported from the coast to the ocean in an outwardly extending eddy shape, indicating the influence of turbulence mixing and the transmission of ocean eddy energy (Wyrtki et al., 1976)."

5. Comment: line 259-263, "The results show that HgPR spreads outward in an eddy shape, driven by HgPR concentrations and influenced by the kinetic energy transmission by ocean eddies and the mixing of turbulence. This further emphasizes the important role of eddy-driven processes and turbulence mixing, in the transport and distribution of riverine Hg in coastal and shelf regions." It can be added that in comparison with previous models, your model results are more innovative and evolving.

Appreciate your advice. The modified expression is:

The results show that HgPR spreads outward in an eddy shape, driven by HgPR concentrations and influenced by the kinetic energy transmission by ocean eddies and

the mixing of turbulence. Compared with the model results in Figure 3 by Zhang et al. (2015), the distinct depiction of ocean eddies achieved through our high-resolution models is evident. This notably underscores the pivotal role played by eddy-driven processes and turbulence-induced mixing in the transportation and dispersion of river-derived Hg within coastal and shelf regions.

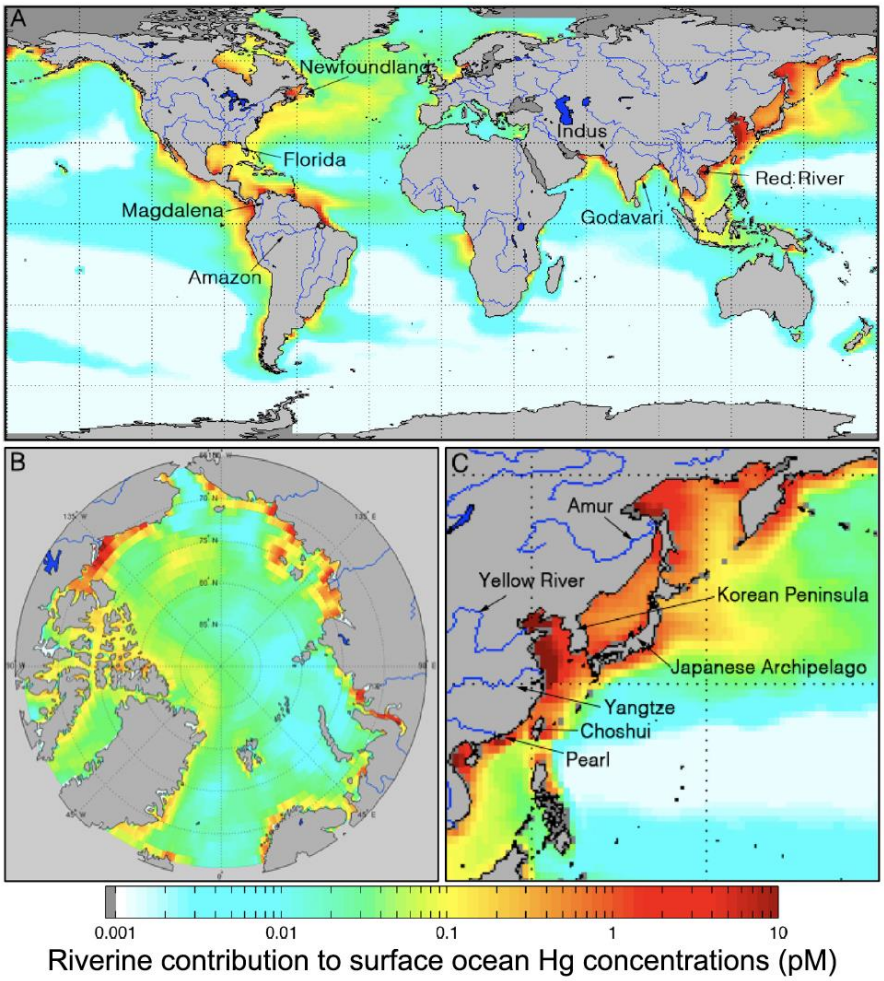

Figure 3. Contributions of riverine discharges to modeled annual total Hg concentrations (pM) at 0–10 m depth. (a) Global ocean simulation, (b) Arctic Ocean simulation, and (c) zoom for the western Pacific Rim.

6. Comment: The expression can be more concise:
   in lines 20-21, it would be better to replace "the inclusion of" with "including";
   in line 169, replace "takes into account" with "considers";
   in line 194, replace "at the same time" with "simultaneously";
   in line 205, replace "is the area with" with "has";
   in lines 210-211, replace "as compare to" with "more";
   in lines 233-234, replace "To gain a clearer understanding" with "To understand better";
   in line 285, replace "which can lead" with "leading";

Thank you immensely for providing these valuable and practical suggestions regarding my writing. We have diligently incorporated the comments (6-16) to enhance the precision and organization of the manuscript as advised.

7. Comment: in line 243, it is advisable to label which legend corresponds to the description;

Revised as suggested.

8. Comment: in line 250, punctuation is lost at the end.

Revised as suggested.

9. Comment: Empty filler words and phrases will make your writing not precise enough: in line 155, it is advisable to delete the "which is" in "…, which is a global, spatially explicit…" and in line 169, delete the "of which" in "all of which contribute to differences in oceanic Hg distribution"; in line 203, delete the "the effect of" in "the effect of turbulence across the ocean causes the spread of Hg in a vortical shape."

Revised as suggested.

10. Comment: in line 62, replacing "couldn't" with " could not " may be suitable.

Revised as suggested.

11. Comment: in line 269, it is advisable to modify "And" at the beginning of the sentence with "Moreover" will be more formal.

Revised as suggested.

12. Comment: in lines 314-315, "This suggests that the sinking of particle-bound Hg is influenced by ecological changes in seawater." It will be better to use active voice.

Revised as suggested.

13. Comment: lines 318-319, "which promptly reflected the abrupt fluctuations in phytoplankton and enabled us to quickly observe changes in the sinking of Hg particles." It will be better to replace it with "which promptly reflected the abrupt fluctuations in phytoplankton and enabled us to observe changes in the sinking of Hg particles quickly."

Revised as suggested.

14. Comment: in line 329, you'd better delete "the" in front of "turbulence."

Revised as suggested.

15. Comment: in line 150, you'd better add "that" in later of "infer."

Revised as suggested.

16. Comment: in line 282, "undergo" should be written with "undergoes."

Revised as suggested.

Response to Referee #2:

This manuscript introduces a high-resolution marine mercury model with concurrently model simulation of biogeological cycling. The higher resolution of the model makes it able to capture turbulence that can influence the mercury cycling of some local region. The application of concurrently simulated biogeochemistry or the cycling of phytoplankton enables the model to capture the particle-bound mercury sinking due to some ecological related and climate driven event such as increased effects of bio pump. The model also aims to capture the eddy or fine scale ocean current in the estuary region with riverine mercury input. Overall, the paper is well prepared and adds new knowledge and insights into the literature. For the authors to polish the paper, I have a few questions and comments, which I think also represent those from the general readers of GMD. After the authors address those points and include in the revision, I will enthusiastically support the publication of the paper in GMD.

We express our gratitude for your decision and constructive comments on our manuscript. Your feedback holds significant value as it aids in the revision and enhancement of our paper, providing important guidance for our research. We have meticulously considered your suggestions and worked diligently to implement improvements and make necessary changes to the manuscript.

1. Comment: The meaning of the term "online biogeochemistry" as used in the title is not intuitive for general readers who may think online means on the internet. To be friendly for general readers, I suggest authors using another term (e.g. concurrently simulated biogeochemistry processes) or explain the meaning in the abstract as early as possible.

I am sorry for any misunderstanding, and I genuinely appreciate your helpful comment. In response to your suggestion, we have clarified the term 'online biogeochemistry' in the abstract. In our approach, we utilize a high-resolution ocean model (MITgcm-ECCO2, referred to as high-resolution-MITgcm) coupled with the concurrent simulation of biogeochemistry processes from the Darwin project (referred to as 'online'). This integration enables us to comprehensively simulate the global biogeochemical cycle of Hg with a horizontal resolution of 1/5°.

2. Comment: Line 48, reference format

Thanks for pointing out the problem; we have corrected it in the manuscript!

3. Comment: L 50, although this limitation is related to the contribution of this presented study, it would be better to point out all limitations of earlier models and then explain why overcoming this limitation is more important.

Thanks for the helpful advice. Limitations of earlier models include less description of oceanic vertical transport, lack of description of ocean biogeochemistry from a microscopic perspective, and insufficient model resolution. In lines 55-60, we have elucidated the significance of resolution in ocean models for effectively simulating oceanic material transport and nearshore mixing processes. Therefore, addressing and overcoming the limitations related to model resolution hold particular importance.

4. Comment: L58-59, "anthropogenic disturbances and climate-induced changes" is too general to make reader understand how the processes need to be characterized with

high-resolution model.

Appreciate your suggestion. To provide a clearer illustration of the concept, we have rephrased the sentence from lines 58 to 59 as follows: High-resolution models are capable of accurately portraying transport and mixing dynamics within estuaries and continental shelves. This capacity enables them to promptly capture shifts in Hg cycling within coastal waters, thereby highlighting their exceptional sensitivity to specific factors like human-driven disruptions and alterations induced by climate variations.

5. Comment: L150, “Most” is unclear. Is it 50% or 90%? Better to give a the ranges quantitatively.

Thanks for the helpful advice. A large fraction (typically >80%) of the Hg in rivers is in the particulate phase (Emmerton et al., 2013; Schuster et al., 2011); Zhang et al. (2015) use observational constraints on seawater Hg concentrations and evasion to infer that most Hg from rivers is sorbed to refractory organic carbon and preferentially buried (~93%).

6. Comment: L192-193, Better to give quantitative information on how much closer to the observations can be brought in by the high-resolution model compared with the lower resolution model.

Thanks for your thoughtful response. To quantitatively give more information on how much closer to the observation the high-resolution model (HR) can be brought than the lower resolution model (LLC90), we calculated the disparity between the observation and the model simulated results (The values at the position consistent with the observation).

We have employed two indicators for comparison: firstly, the sum of the absolute between the simulated values and the observation. A smaller value indicates a smaller simulation-observation disparity (simulation is closer). Our findings reveal that for HR, indicator 1 is 23.11 (Fig2.a) and 10.11 (Fig2.c), while for LLC90, it is 30.11 (Fig2.b) and 11.41 (Fig2.d).

Secondly, we utilized the absolute value of the covariance between the simulated outcomes and the observation. Larger values denote a stronger correlation, indicating a closer alignment between the simulation and observation. Our results indicate that for HR, indicator 2 is 0.012 (Fig2.a) and 0.010 (Fig2.c), and for LLC90, it is 0.001 (Fig2.b) and 0.009 (Fig2.d).

Both of these indicators collectively underscore the quantified closeness of our high-resolution model's simulation outcomes to the observed data.

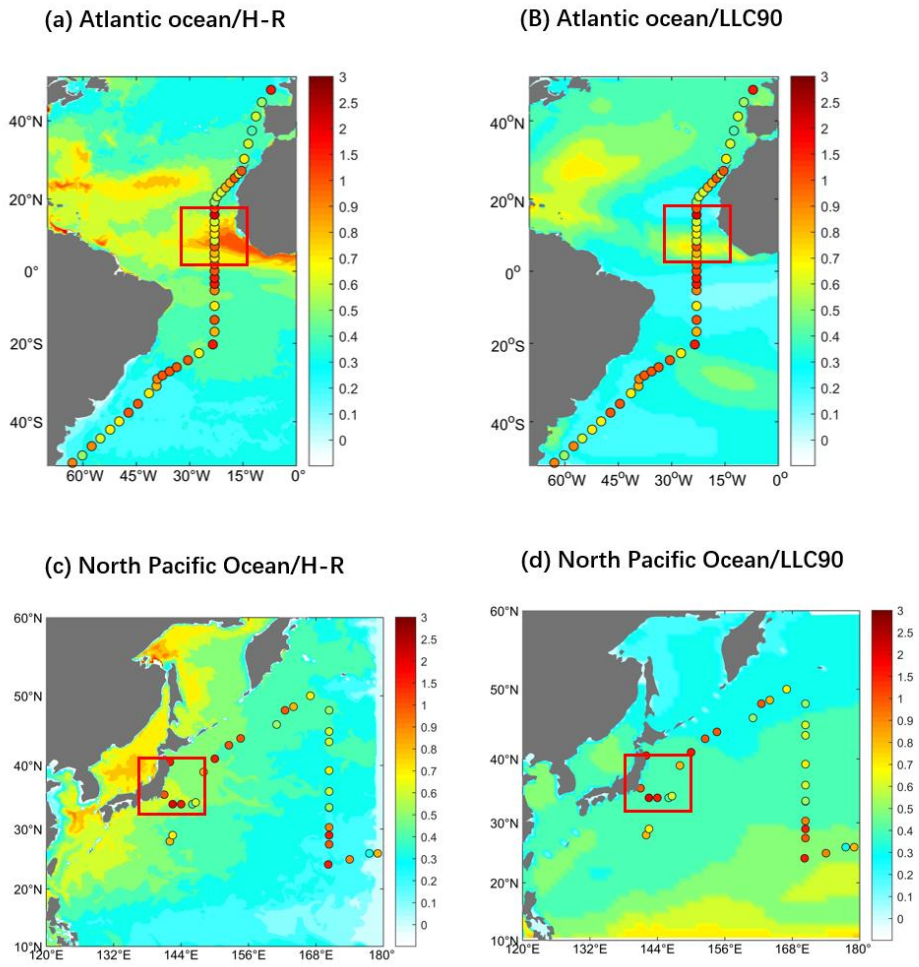

Figure 2. MITgcm simulation for total inorganic Hg results of high resolution (1/5°×1/5° horizontal) ECCO2 (H-R) and coarser (1°×1° horizontal) resolution ECCO v4 (LLC90) (0-10 m depth; color, unit pM, (Huang and Zhang, 2021)). The scatter is observations from two single high spatial precision cruises, (a,b) from Kuss et al. (2011) Atlantic Ocean observations in a month, and (c,d) from Laurier et al. (2004) North Pacific observations in a day(0-10 m depth; scatter, unit pM).

7. Comment: L206, "significant" is normally associated with statistical meaning. If no statistical test was conducted here, better to use the term "more apparent".

That's a valuable suggestion! We have incorporated a more suitable expression into the manuscript.

8. Comment: L213, the figure caption is not intuitive.

Thanks for the excellent advice. The original figure caption, "Total inorganic Hg Simulation of regions with ocean currents," is not intuitive. We have modified the figure caption: Distribution of inorganic mercury at the sea surface within regions influenced by various global ocean currents (0-10m depth; color, unit pM). Zoom for (a) the Northwest Pacific Ocean current-affected area, (b) the Northwest Atlantic Ocean current-affected area, (c) the North Indian Ocean current-affected area, and (d) the South Atlantic Ocean current-affected area.

9. Comment: L217, since the model is in higher resolution, river discharge being transferred along the seashores can be resolved, it is essential to mention the spatial

resolution of the riverine discharge as the model input.

It's a reasonable suggestion! We have added explanatory notes on river inputs: Before using the river discharge as the model input, we interpolated the river discharge so that its spatial resolution is consistent with the high-resolution MITgcm model.

10. Comment: L280, "shorter time scales" How short? Give the specific time scale.

Sorry for this problem. We have modified the expression with "Online biogeochemistry makes Hg more sensitive to environmental changes within at least one time step of the model (1h)."

11. Comment: L293, "a certain range of time" what is the time range?

This sentence needs to be more accurate due to the repetition of the expression. The time range is precisely the simulated period. Thus, we delete "a certain range of time" in the definition of bloom events to enhance the clarity of expression. The correct term is: "Hence, we can devise an algorithm to identify potential algal bloom events: within the simulated period, an algal bloom event is recognized if it exhibits the most deviation from the average." Additionally, we provide a more precise definition of these blooms by prefixing them with 'potential,' which is also part of the answer for Comment 13.

12. Comment: L297 and Fig 7: The description of "peak day" as shown in the figure by the vertical line should be added to the figure caption.

Thank you very much for the valuable suggestion. We have followed the comment to add it to the figure caption.

13. Comment: L300-301, it is indeed that nutrient levels during the peak days are above the average but it is unclear how can this suggests the occurrence of algal blooms here.

Yes, relying only on plankton levels, nitrogen, and phosphorus nutrient levels at a site can only indicate that a bloom event may have occurred. Certainty regarding the occurrence of a bloom requires additional supporting data, including physicochemical properties of seawater during the same period and remotely sensed imagery demonstrating bloom presence. Nevertheless, our study just aims to exhibit the swift response of Hg behavior within our model to ecosystem changes within a one-hour timeframe. Consequently, an exhaustive validation of an event causing a sudden plankton shift as an algal bloom event is not our primary focus. As a result, the definition of a bloom event in this paper is presented as a potential scenario for modeling, and we have incorporated the qualifier 'potentially' into this definition.

Thus, in the subsequent statement, we emphasized that the bloom event is conceptualized as a potential occurrence characterized by elevated plankton biomass alongside increased nitrogen and phosphorus nutrients. While this description may not match the precision of an exact bloom event, it does serve to indicate that the biochemical conditions of the ocean are altered when a bloom-like event triggers a surge in phytoplankton. This process generates more particulate matter capable of binding to Hg, thereby expediting the sinking process of particulate Hg and its removal through remineralization. Our model effectively illustrates Hg's responsiveness to ecological changes, manifested within a one-hour timeframe.

14. Comment: L325, elemental and divalent Hg

Sorry, the error has been corrected.

15. Comment: L329, besides turbulent mixing and shear effects, is there any other

factors whose spatial variability is not capture by the model simulation? For example, continental input to the coastal sea. Is the emission at a resolution that match the resolution of 18 km. It is recommended that the authors put some discussions on factors that can influence the fine-scale processes and levels of Hg that are still not solved by this study.

Your question is greatly appreciated. The physical transport and biogeochemical changes of Hg in the ocean are complex and have been the subject of prolonged study. However, our high-resolution model optimizes this simulation by capturing the small- to medium-scale dynamics of Hg processes in the ocean, simulating mixed turbulence effects in the nearshore and estuaries. Our improved simulation of biogeochemical conditions also makes it possible to provide help in the rapid response to ecological changes in Hg.

Nevertheless, our model does have its limitations. Firstly, high-resolution models with short time steps encounter challenges in comprehensively depicting climate-scale changes. It is difficult to simulate climate change-induced alterations in the oceanic Hg cycle. Secondly, closer integration with ecological models is necessary to characterize processes occurring at finer time scales, including migrations and changes in the structure of phytoplankton communities and the weakening or enhancement of biological pumps.

Our coupled high-resolution model with advanced biogeochemistry holds promise in offering fresh insights into Hg's methylation and demethylation processes. This aspect will be delved into further in our upcoming work. Additionally, our enthusiasm lies in the anticipation of refining existing models to address a broader range of issues beyond those previously mentioned.

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