Response to Reviewer #2 (RC2)

We would like to thank Reviewer 2 for taking the time to carefully read our manuscript and provide feedback. We sincerely appreciate the detailed comments, which have helped us improve the manuscript. Text in blue denotes the reviewer's comments, while text in black denotes our responses. Text in red denotes excerpts from the revised manuscript. The corresponding revisions are shown in the revised manuscript using Track Changes.

Comments (in blue): This manuscript presents POP2/Hg v1.0, a new global ocean mercury model embedded in the ocean component of CESM2 (POP2) and coupled to MARBL. The work is a worthwhile contribution: coupling mercury chemistry to a prognostic biogeochemistry library with separate soft and hard POC pools is a genuine advance over offline or climatology-driven predecessors, and the model reproduces several large-scale features of marine mercury. However, several issues must be resolved before the paper can be accepted. Most of these require clarification, correction, and modest added analysis rather than new simulations.

Response (in black): We thank the reviewer for the careful and constructive assessment. We have corrected the inconsistent units and reported values, clarified the initial conditions and the interpretation of the deep-ocean results, added a compact global-budget comparison and quantitative model-observation statistics, and corrected the notation, figure units, duplicated text, and typographical errors identified by the reviewer. Our point-by-point responses are provided below.

1. Lines 350 and 450: Section 4.1 reports a global Hg^0 net evasion of ~4.43 Mg yr^{-1} against an HgII deposition of 4.20 × 10^3 t yr^{-1}, then calls this "near-equilibrium exchange." Taken literally, evasion (4.43 t) is ~1000× smaller than deposition (4200 t), which is the opposite of equilibrium. The Conclusion instead gives a net evasion of ~4.5 × 10^3 t yr^{-1}, differing from Section 4.1 by three orders of magnitude.

Published global ocean Hg0 evasion is on the order of thousands of Mg yr^{-1}, so "4.43 Mg yr^{-1}" appears to be a unit error. Please reconcile these numbers and re-examine the equilibrium statement.

Thank you for identifying this inconsistency. The reported value of 4.43 Mg yr^{-1} resulted from an incorrect unit conversion and an inadvertent mismatch in the simulation output used for the calculation. We have corrected the global Hg^0 net evasion to 4.52×10^3 Mg yr^{-1}, and the combined gaseous Hg evasion including DMHg is 4.61×10^3 Mg yr^{-1}. We also harmonized this value throughout the manuscript, including the Conclusion. With this correction, Hg^0 evasion (4.52×10^3 Mg yr^{-1}) and atmospheric Hg^{II} deposition (4.20×10^3 Mg yr^{-1}) are comparable in magnitude, indicating a near balance between these two atmospheric exchange terms. Accordingly, we revised the wording in Sect. 4.1 from "near-equilibrium exchange" to "a near balance between atmospheric deposition and oceanic evasion" to avoid implying that the ocean Hg system has reached equilibrium.

2. Lines 263–265 and 334: Figure 11 is described as "year 10 (balanced state)", yet deep-ocean ventilation timescales are centuries to over a millennium. Consistent with this, the authors state that the elevated Hg0 at ~1000 m (up to 0.8 pM) is contributed by "the initial condition of the model". This undermines confidence in all 985/1000 m results (Hg0, HgII, Hgp, MeHg, HgT), which may reflect the spin-up initial field rather than the model's biogeochemistry. Please justify the spin-up duration for the deep ocean or explicitly qualify that deep-ocean fields are not equilibrated and interpret them accordingly.

Thank you for this important comment. We agree that a 10-year integration is insufficient to equilibrate the deep ocean, where ventilation and tracer adjustment occur on centennial to millennial timescales. We have revised the manuscript accordingly, and the phrase "year 10 (balanced state)" was also removed from the Fig. 11 caption. We now explicitly state that the deep-ocean Hg fields were not fully equilibrated and remained influenced by the prescribed initial conditions:

Section 2.4: "*For the simulations in this study, the five ocean Hg tracers were initialized using externally prescribed three-dimensional fields prepared for the MITgcm-Hg*

configuration described by Zhang et al. (2020). These fields were interpolated onto the POP2 grid and used to initialize the corresponding POP2/Hg tracers. Starting from the prescribed initial fields, POP2/Hg was integrated for 10 years. By the end of the 10-year integration, year-to-year changes in the surface Hg fields had decreased to below 0.1%, suggesting the surface and upper-ocean Hg fields have adjusted to the POP2 circulation and MARBL biogeochemistry. In contrast, the deep-ocean Hg fields were not considered fully equilibrated and remained influenced by the prescribed initial conditions."

The MITgcm-Hg fields used to initialize POP2/Hg ultimately trace back to the steady-state OFFTRAC simulation of Zhang et al. (2014). In that study, ocean Hg tracers were initialized at zero and integrated for 10,000 years under repeated monthly climatological circulation and boundary forcing. Steady state was defined by a repeating annual cycle in surface Hg concentrations together with changes of less than 5% in deep-ocean Hg concentrations over 1000 years. In contrast, the 10-year POP2/Hg integration used in this study is too short for surface forcing and upper-ocean adjustments to propagate throughout the deep ocean. Consequently, the simulated Hg distributions in the deep ocean are interpreted as transient fields that remain strongly influenced by the prescribed initial conditions rather than as equilibrated deep-ocean states. We have therefore revised the discussion of the concentrations near 1000 m to explicitly acknowledge the contribution of the inherited initial field and have qualified the interpretation of deep-ocean Hg^0, Hg^{II}, Hg^p, MeHg, and Hg^T throughout the revised manuscript.

3. Lines 365, 371, 375 versus 425–430: Section 4.2 reports surface HgII reduction at 1.53 × 105 t yr^{-1}, intermediate net reduction at 1.74 × 102 t yr^{-1}, and deep net oxidation at 3.74 × 103 t yr^{-1}. The sensitivity section then reports surface, mid-depth, and deep net reduction in "Mg yr^{-1}" with values of 8.22, 3.62/1.53, and 1.11/3.69. These cannot be reconciled with the Section 4.2 values (e.g., mid-depth 1.74 × 102 t yr^{-1} vs. 3.62 Mg yr^{-1}). Please harmonize units and magnitudes throughout, ideally with a single flux-budget table.

Thank you for identifying these inconsistencies. We carefully rechecked all quantitative values reported throughout the manuscript against the model outputs and corrected the

corresponding inconsistencies. During this review, we also identified and corrected an editing-related transcription error in Sect. 4.2 that resulted in several values being interchanged. These values have now been corrected.

All redox fluxes are now consistently reported in Mg yr^{-1}, with net redox flux defined as reduction minus oxidation. The corrected sensitivity-experiment values are now summarized consistently in Table 3:

Budgets	***Default-rate case***	***Reduced-rate case***
Global net redox flux	1.65×10^5	1.07×10^4
Surface ocean net redox flux	1.67×10^5	8.22×10^3
Intermediate ocean net redox flux	1.53×10^3	3.62×10^3
Deep ocean net redox flux	-3.69×10^3	-1.11×10^3
$Hg^{II} \rightarrow$ *MMHg methylation*	5.16×10^3	8.46×10^2
MMHg $\rightarrow$ Hg^0 *reductive demethylation*	4.86×10^3	4.41×10^3
Hg^0 *net evasion*	−4432	−4860
DMHg net evasion	−67.48	−35.08
Global Hg inventory	360.4	346.4

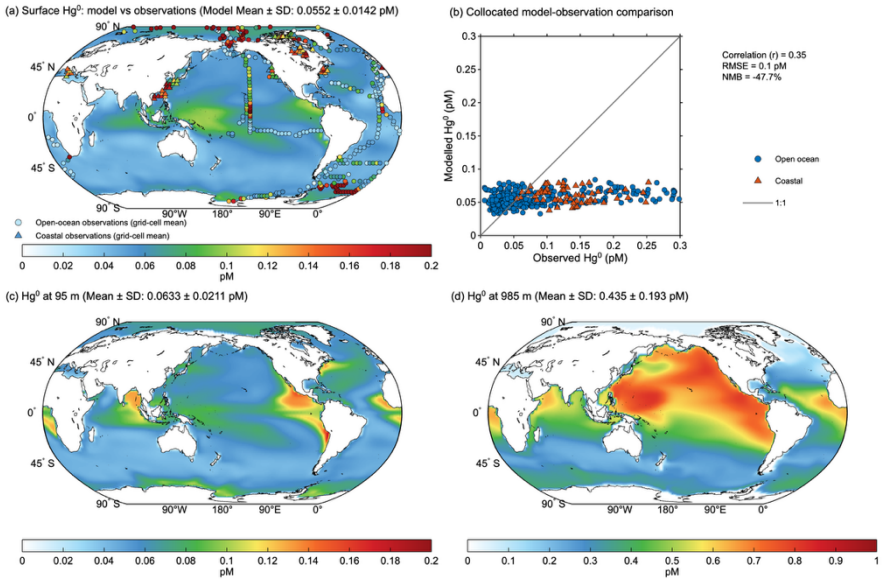
Fluxes are reported in Mg yr^{-1}, except for global Hg inventory, which is reported in Gg. Net redox flux is defined as reduction minus oxidation; positive values indicate net reduction, and negative values indicate net oxidation. Negative air-sea fluxes indicate net evasion from the ocean to the atmosphere. For these depth-integrated diagnostics, 50 and 1000 m denote nominal layer boundaries. A fixed 50 m boundary is used because MLD was not tracked in the budget diagnostics, and the nearest POP2 model level at 985 m is used to represent the nominal 1000 m boundary in the calculations.

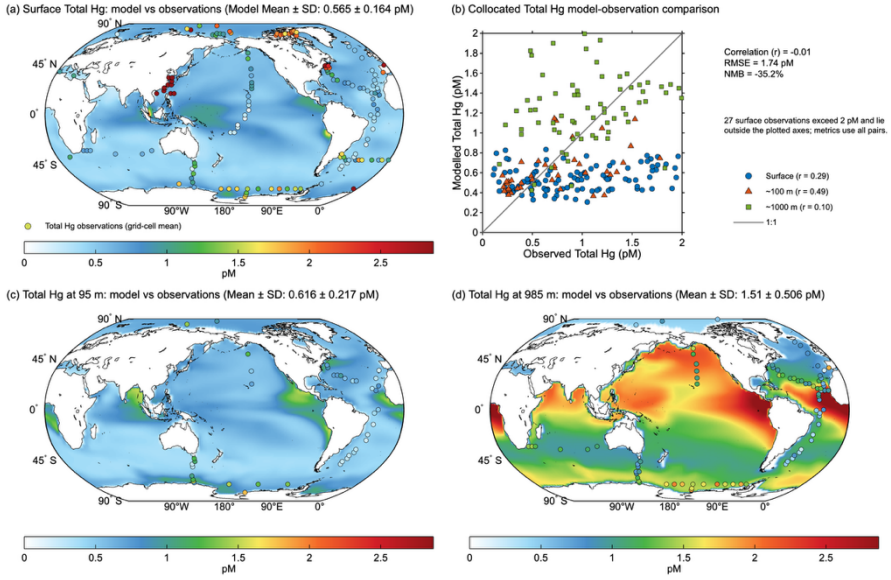
We also clarified that Sect. 4.2 reports the main simulation, whereas Sect. 4.4 compares two separate 20-year sensitivity experiments. The default-rate sensitivity experiment is therefore not identical to the main simulation, and small differences between their diagnosed budgets are expected.

4. Sections 3.1–3.4: The model-observation evaluation is entirely qualitative; no

quantitative skill metrics are reported. The Results overlay pointwise observations on maps (Figs. 7, 10, 11) and describe agreement in words ("reasonably well," "good agreement"), but no correlation, RMSE, bias, or model-vs-observation scatter/statistics are given anywhere. For a model-description-and-evaluation paper, at least basic quantitative skill statistics against the compiled datasets (cited in Sect. 2.4) are expected and are computable from data already in hand. I recommend adding them.

Thank you for this suggestion. We have added correlation coefficients, RMSE, and normalized mean bias to the corresponding model-observation evaluation in Sects. 3.1, 3.3, and 3.4. Quantitative statistics for the MeHg comparison in Fig. 10 are reported in Sect. 3.3, while model-observation scatter plots with quantitative statistics have additionally been added to Figs. 7 and 11:

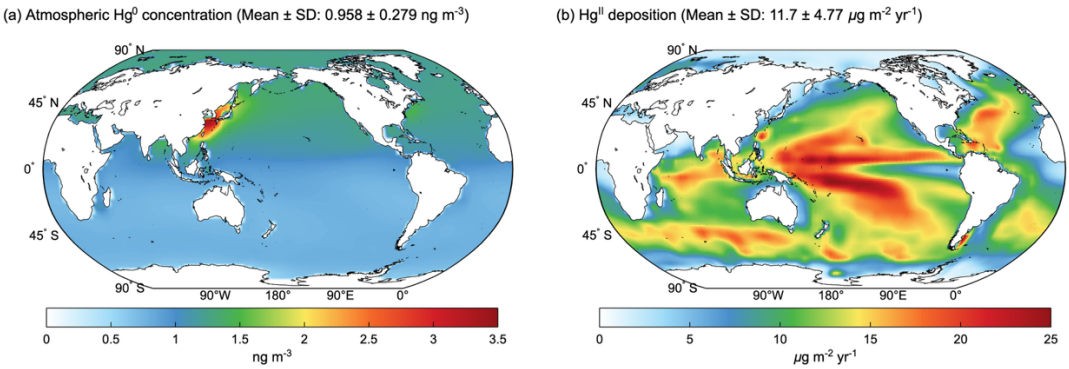




5. Table 1 and Figure 6 caption: In Table 1, the symbols "OCRR," "hv," and "T_c/T_k" (reaction 7) are used but never defined in the text. In Figure 6, panel (a) atmospheric Hg0 "concentration" is labelled ng m^{-2} d^{-1} (a flux unit; concentration should be ng m^{-3}), and panel (b) HgII deposition is captioned mol m^{-2} s^{-1} while the colorbar reads pg m^{-2} s^{-1}. Please define all symbols and correct the units.

   Thank you for pointing out these inconsistencies. We have defined OCRR, hv, and T_c /T_k in the revised text and clarified the symbols used in Table 1.

   We have also corrected the units in Fig. 6 so that the atmospheric Hg^0 concentration and HgII deposition flux are reported with the appropriate and consistent units:



6. Lines 187–189, 249, 320 (plus Eqs. 5–7): Duplicated text, typographical errors, and a POC/bioC notation mismatch. Lines 187–189 contain a near-verbatim duplicated

sentence about Hgp gravitational settling. Typos include "Hg0remain" and "below 0.009o4 pM". In addition, sorption (Eq. 5) partitions HgII onto POC only, whereas the sinking-velocity formulation (Eqs. 6–7) uses POC + bioC (living biomass); please clarify whether Hgp is carried by living biomass and reconcile the notation. A careful language and consistency edit throughout is advised.

Thank you for identifying these issues. We performed a careful language and consistency check throughout the manuscript and corrected the duplicated text, typographical errors, grammatical problems, and inconsistent terminology. For example, we (1) removed the duplicated sentence describing gravitational settling of Hgp in Sect. 2.3.3; (2) corrected "Hg0remain" to "Hg^0 remains"; (3) corrected the malformed numerical expression "below 0.009o4 pM" in the MeHg discussion; (4) revised phrases such as "Hgp concentrations get lower" to "Hgp concentrations decrease"; and (5) replaced grammatically incorrect expressions such as "reactions remain to be oxidation" with "net oxidation persists." Similar language and notation corrections were made throughout the manuscript.

We also revised Sects. 2.3.3–2.3.4 and Eqs. (5)–(8) to clarify the particulate carbon pools involved in Hg^{II} partitioning and Hg^{p} transport. In the implemented model formulation, Hg^{II} partitions onto the total organic particulate carbon pool represented by non-living detrital POC plus living particulate biomass carbon (C_{bio}). We therefore revised Eq. (5) to include both $[POC]$ and $[C_{bio}]$, consistent with the particulate-carbon pool used in the sinking-velocity formulation. We now explicitly define C_{bio} as the summed carbon biomass of small phytoplankton, diatoms, diazotrophs, and zooplankton.

To simplify the presentation, the separate soft- and hard-POC expressions previously given in Eqs. (6)–(7) and (8)–(9) have each been consolidated into a single general expression (now Eqs. (6) and (8), respectively), with the text clarifying that the diagnostics are evaluated separately for the soft- and hard-POC pools. The corresponding biomass sinking flux is now explicitly defined in the newly added Eq. (7) $F_{bioC} = \sum_p w_p C_p$. Following the conceptual treatment of living particulate biomass in Zhang et al. (2020), all living particulate groups contribute to the particulate-carbon concentration available for Hg^{II} partitioning, whereas only groups assigned non-zero settling velocities contribute to the

biomass sinking flux. Thus, Hgp in POP2/Hg can be associated with both detrital POC and living particulate biomass, and the notation is now consistent between the sorption and biological-pump formulations.