Below, we provide a detailed, point-by-point response to the reviewer's comments and describe the corresponding revisions made to the manuscript. Reviewer comments are shown in pink, author responses and original manuscript text are shown in black, and revised manuscript text is shown in blue.

General overview of the manuscript:

This manuscript synthesizes outputs from ten modeling studies to estimate future mercury (Hg) emissions, deposition, and (in four studies) projected fish MeHg concentrations in 2050 under sixteen climate policy scenarios. The effort to integrate across models and policy scenarios is valuable and timely, particularly given ongoing international assessments of mercury mitigation effectiveness.

While the study provides useful comparative insights, several areas would benefit from clarification, particularly regarding model evaluation, geographic scope, and presentation of the tables and figure. While many of the models considered produce global outputs, the analysis focuses on a relatively small number of lakes in the United States due to the regional limitations of the fish MeHg bioaccumulation models. Additionally, comparisons between modelled and observed trends in fish MeHg are not presented for most of the select lakes, nor is there any comparison offered in the other environmental media considered in the bioaccumulation models. Finally, the compiled datasets are not provided in the Supplementary Information, which may limit their utility for future large-scale synthesis efforts (e.g., those associated with the international Minamata effectiveness evaluations).

Author response

Thank you for this thoughtful overview and for recognizing the value of integrating multiple modeling studies and policy scenarios to assess future Hg emissions, deposition, and fish MeHg responses. We appreciate the reviewer's constructive suggestions regarding model evaluation, geographic scope, and presentation of the compiled datasets.

We have made several clarifications in the revised manuscript to address these points. As noted in Lines 92 onward, the selected studies primarily focus on the United States because most of the available scenario-based fish MeHg data are derived from these regions and from lake-based assessments. To make this clearer, we have now further emphasized that the analysis of deposition–fish MeHg relationships is centered mainly on U.S. freshwater ecosystems because scenario-based projections linking atmospheric Hg deposition to fish MeHg are currently available for only a limited number of regions. A more detailed explanation of this geographic limitation is provided in our response to Lines 109–110 below.

In addition, we have expanded the discussion to place the modeled projections in the context of available observational datasets, particularly for atmospheric Hg deposition in North America and several U.S. regions, in order to provide observational benchmarks for the modeled outputs (response to Lines 243–257 and 258–266).

Comparisons with other environmental media (e.g., water and sediments) were not included because scenario-based projections linking Hg emissions and deposition to these compartments remain very limited in the current literature.

Finally, the compiled datasets are already presented in a structured form within the manuscript and Supplementary Information (Tables 1–2 and Tables S1, S2, and S4) and were therefore not provided separately in raw format. Overall, these revisions improve the clarity and transparency of the manuscript while maintaining its focus on scenario-based Hg projections.

Comments on the Abstract:

The abstract reads well. One particularly interesting result is the strength of the relationship between projected changes in fish Hg and atmospheric deposition. However, since all fish Hg concentrations are model-derived, it would be helpful to clarify to what extent this similarity reflects structural assumptions within the coupled atmospheric–bioaccumulation models (for example, assumptions regarding response times of fish MeHg to changes in atmospheric inputs). Without comparisons to observed deposition and fish MeHg relationships (e.g., using isotope approaches or long-term monitoring datasets), it is difficult to assess how independently this relationship emerges from the models versus being constrained by their structure. Some brief discussion of this point would strengthen the interpretation.

Additionally, the final two sentences adopt a cautionary tone regarding model limitations that feels somewhat abrupt. While it is important to acknowledge that models cannot capture the full complexity of atmospheric and ecosystem processes, the abstract does not specify which processes or interactions are currently missing or how future work might address these gaps. As such, the conclusion reads slightly general and somewhat disconnected from the specific findings of the study. The authors may wish to either briefly specify the key processes that require improved representation or reframe the concluding sentences to more directly emphasize the primary contribution of the study and avoid commenting on these limitations within the abstract. This would help the abstract end on a clearer and more cohesive note.

Author response

Thank you for this helpful and constructive feedback on the Abstract. We agree that the strong relationship between projected fish MeHg and atmospheric deposition requires clarification, particularly because the fish Hg values are derived from model outputs. To address this point, we revised the Abstract to note that the observed linear relationships may partly reflect structural assumptions within the modeling frameworks. We also refined the concluding sentences to more clearly identify specific processes that remain underrepresented in current models and to better connect these limitations to the broader implications of the study.

Revision made

Abstract

Mercury (Hg) poses a global threat due to its long-range transport and transformation into methylmercury (MeHg), a potent neurotoxin that bioaccumulates in aquatic food webs. While global and regional efforts to reduce anthropogenic Hg emissions are ongoing, the implications of these policies for future Hg deposition and consequent MeHg levels in fish remain uncertain. This study synthesizes published modeling studies to examine projected relationships among Hg emissions, atmospheric deposition, and lake fish MeHg concentrations by 2050 under various policy scenarios. While models reveal a strong linear relationship between emissions and deposition ($R^2 = 0.79$) and a moderate correlation between Hg deposition and fish MeHg ($R^2 = 0.63$), these trends contrast with observational data, which often show nonlinear or more complex responses. Our analysis suggests that these relationships are partly shaped by shared structural assumptions within current modeling frameworks, particularly the treatment of atmospheric deposition as the dominant driver of future fish MeHg change and the representation of relatively short response times between changing Hg inputs and fish exposure, with limited representation of the decadal-scale buffering provided by legacy Hg stores. Within these constraints, atmospheric deposition and lake area emerged as key predictors, with higher deposition and smaller lakes associated with higher modeled fish MeHg levels. Notably, despite wide variation in model structures, including differences in atmospheric chemistry, emission inventories, and food web dynamics, these linear trends persisted. By identifying these consistent patterns and the assumptions that support them, this study provides a benchmark for integrating currently underrepresented processes such as ecological complexity, and climate-driven ecosystem feedbacks, into future assessments supporting the Minamata Convention.

Comments on the Introduction:

The Introduction reads clearly and covers much of the relevant literature. However, additional details regarding the atmospheric modeling frameworks would be helpful. For example, do the emissions inventories primarily reflect anthropogenic sources, or do they also incorporate natural re-emissions under changing temperature and precipitation conditions in the climate scenarios? A brief clarification would improve transparency for readers less familiar with these modeling systems.

Author response

Thank you for this helpful suggestion. To clarify how atmospheric Hg modeling frameworks treat emission inventories and natural re-emissions, we have added a brief explanatory paragraph in the Introduction (paragraph 4), as suggested.

Revision made

In many atmospheric Hg modeling frameworks, emission inventories primarily represent anthropogenic sources, while natural and legacy re-emissions from land and ocean reservoirs are treated as separate fluxes within the model system (Corbitt et al., 2011.; Perlinger et al. 2019). In several studies, these natural and legacy fluxes are prescribed as constant emissions based on historical deposition rather than responding dynamically to climate-driven changes (Corbitt et al., 2011; Giang et al. 2015). However, some recent frameworks couple atmosphere–ocean–land Hg cycling and allow legacy Hg in soils and oceans to respond to climate-driven changes in temperature, precipitation, and ocean circulation, thereby linking anthropogenic emission trajectories with climate-sensitive re-emissions in future projections (Zhang et al. 2021).

References

Corbitt, Bess Sturges, Chris Holmes, Elsie Sunderland, et al. 2011. *Global Biogeochemical Modeling of Mercury in GEOS-Chem*.

Giang, Amanda, Leah C. Stokes, David G. Streets, Elizabeth S. Corbitt, and Noelle E. Selin. 2015. “Impacts of the Minamata Convention on Mercury Emissions and Global Deposition from Coal-Fired Power Generation in Asia.” *Environmental Science & Technology* 49 (9): 5326–35. https://doi.org/10.1021/acs.est.5b00074.

Zhang, Yanxu, Zhengcheng Song, Shaojian Huang, et al. 2021. “Global Health Effects of Future Atmospheric Mercury Emissions.” *Nature Communications* 12 (1): 3035. https://doi.org/10.1038/s41467-021-23391-7.

Major comments on the body of the text:

Lines 109–110:

The authors note that fish MeHg projections were geographically restricted. It would be helpful to clarify whether this restriction reflects limitations in available published model outputs or inherent constraints of the bioaccumulation models themselves. My understanding is that models such as BASS and SERAFM can be parameterized for different regions if sufficient local ecological and biogeochemical data are available. A brief explanation of why projections were limited to these specific regions would improve clarity.

Author response:

Thank you for this helpful comment. We clarify that the geographic restriction reflects the availability of published model outputs rather than inherent constraints of the bioaccumulation models. While models such as BASS and SERAFM can be applied to other regions, their implementation requires detailed local ecological, hydrological, and biogeochemical input data, which are not widely available. As such, published applications of these models have been limited to specific regions, leading to the geographic scope reflected in our synthesis.

Revision made

Deposition estimates spanned both global and regional scales, whereas fish MeHg projections were geographically restricted, primarily to several U.S. states (Maine, New Hampshire, Michigan, Florida, North Carolina, South Dakota, and Georgia), as mapped in Figure 1 and summarized in Table 2. This limitation reflects the availability of published model outputs rather than inherent constraints of the bioaccumulation models themselves; although these models can be parameterized for other regions, their application requires detailed local ecological, hydrological and biogeochemical input data that are not widely available.

Line 111:

The manuscript refers to a “detailed evaluation” of the models used to simulate Hg deposition and fish MeHg bioaccumulation. Could the authors elaborate on what this evaluation involved beyond generating the summary values presented in Tables 1–3? For example, were any statistical comparisons conducted between model outputs and historical observational datasets (atmospheric Hg, deposition, soil/sediment Hg, or fish MeHg)? If such comparisons were not possible due to the forward-looking 2050 projections, explicitly stating this limitation would be helpful.

Related to this, Lines 121–122 indicate that multiple linear regression models were used to test whether lake characteristics explain variation in projected fish MeHg changes. Are these lake characteristics projected for 2050 as well? If so, this suggests that the regression examines relationships among modeled 2050 variables. Clarifying this would help readers interpret the independence and robustness of statistical analysis.

Author response

Thank you for this insightful comment. We clarify that our "evaluation" refers to a comparative synthesis of previously published model frameworks and outputs rather than a direct validation against observational datasets. Because the study compiles forward-looking projections for 2050 from the literature, direct statistical comparisons with historical observational datasets were not possible; this limitation has now been stated in the manuscript. We also clarify that the lake characteristics used in the regression analysis are present-day physical attributes reported in the original studies and were not projected for 2050.

Revision made

A detailed evaluation was also conducted of the models used to simulate Hg deposition and fish MeHg bioaccumulation, with emphasis on emission inventories, atmospheric transport mechanisms, biogeochemical processes, and methylation dynamics. Key variables, including emission scenarios, base years, global anthropogenic Hg emissions in the base year and in 2050, percentage change in deposition by 2050, atmospheric models, percentage change in fish MeHg by 2050, bioaccumulation model, study region, fish species, and references, were systematically compiled using a pre-designed Excel spreadsheet. Since this study synthesizes previously published projections for 2050, the evaluation represents a comparative assessment of model frameworks and outputs rather than a direct statistical comparison with observational datasets, which is therefore a limitation of the present analysis.

Multiple linear regression models (MLRM) were used to test whether lake characteristics, including average depth, lake surface area, watershed area, and wetland percentage, could explain variation in projected changes in fish MeHg. These lake characteristics represent present-day physical attributes reported in the original studies and were not projected for 2050.

Table 2 (Policy-in-action scenario):

There are two rows for Walleye from the New York Adirondacks with different % changes in fish MeHg in 2050, despite identical base years, references, region, and species. Could the authors clarify what distinguishes these two rows? Additionally, deposition change is listed as

−29% for the Adirondacks overall, yet a value of +12% appears in the bottom two rows for Walleye. Some clarification of how these values relate would improve interpretability.

Also related to Tables 1–3: some models listed have geographic coverage outside the primary U.S. study area (e.g., India, China, South Pacific, Arctic). It would be helpful to explain how outputs from these regions were incorporated into the present analysis and why they are relevant to the study's geographic scope.

Author response

Thank you for pointing out this ambiguity. We apologize for the confusion caused by these entries. The identical rows were not duplicates but reflected two distinct virtual experiments reported by Perlinger et al. (2018), in which the same Policy-in-Action deposition scenario was applied under different watershed configurations. Specifically, the overall deposition change for the Adirondacks under the Policy-in-Action scenario is −29%; however, this case did not include corresponding fish MeHg projections in the original study, and therefore this row has now been removed to avoid confusion. The two remaining rows (previously appearing as identical) correspond to simulations with identical deposition conditions (+12%) but differing watershed characteristics. In one case, Michigan Upper Peninsula watershed characteristics were applied, resulting in an increase in modeled fish MeHg (+19%). In the second case, Adirondack watershed characteristics (characterized by lower wetland percentage and reduced forest runoff) were applied under the same deposition conditions, leading to a decrease in modeled fish MeHg (−38%). These differences highlight the strong influence of watershed properties on fish MeHg responses, even under identical atmospheric deposition scenarios. To improve clarity and avoid potential confusion, the table has been revised by removing incomplete entries and restructuring the presentation of these cases.

Regarding these tables 1-3, the objective of our synthesis was to include all available studies (published between 2008 and 2025) that projected Hg emissions, atmospheric deposition, and fish MeHg responses under future scenarios. Because relatively few studies provide projections linking Hg deposition to fish MeHg, most of the available datasets are limited to U.S. freshwater ecosystems. However, studies examining the relationship between Hg emissions and atmospheric deposition are available for multiple geographic regions (e.g., global, Arctic, India, China, and the

South Pacific). These datasets were included to capture the broader range of modeled emission–deposition responses across different regions and modeling frameworks. Importantly, the inclusion of multiple regions helps demonstrate that the relationship between emission changes and deposition responses remains consistently linear across different geographic scales in the model-based projections.

Revision made

The revised version of the table is provided on page 20-21 below.

Methodology

A total of ten peer-reviewed studies were selected for analysis of projected anthropogenic Hg emissions, atmospheric deposition, and fish MeHg concentrations under 16 policy scenarios extending to 2050. These studies were identified (published between 2008 and 2025) through a targeted literature review and selected based on their ability to quantify at least two linked components of the Hg pathway: emissions and deposition, deposition and bioaccumulation, or all three using scenario-based modeling frameworks. Studies addressing only a single component were excluded. Studies examining the relationship between Hg emissions and atmospheric deposition are available across multiple geographic scales (e.g., global, regional, or local), and these were included to capture the broader range of modeled emission–deposition responses across different regions and modeling frameworks. In contrast, studies linking scenario-based Hg deposition projections to fish MeHg responses, using coupled atmospheric chemistry–transport and aquatic bioaccumulation models, are more limited and are primarily available for U.S. freshwater ecosystems, particularly lake systems. As a result, the analysis of deposition–MeHg relationships focuses mainly on several U.S. states where such projections have been reported. The studies employ distinct emission inventories, policy frameworks, and modeling approaches to simulate Hg pathways from emissions to ecosystem response. Detailed descriptions of the emission scenarios and modeling frameworks are provided in the Results and Discussion (Sect. 3.1 and 3.2) to support interpretation of results. All extracted data and references are summarized in Tables 1–2 and Tables S1, S2 and S4.

Lines 157–164:

The number of references listed for the different policy scenarios somewhat interrupts the flow of the paragraph. It may improve readability to refer readers primarily to Tables 1–3 and streamline the in-text citations. However, some references cited here (e.g., Sunderland and Selin 2013; Geyman et al. 2024, 2025) do not appear in the tables. If these are particularly important for contextual interpretation, perhaps a brief explanation of their specific relevance would clarify their inclusion.

Author response

Thank you for this helpful suggestion. We will refer to Tables 1-3 where necessary, instead of citing the reference. The references Sunderland and Selin (2013) and Geyman et al. (2024, 2025) were retained in the text because they provide important contextual insight into future mercury emission and deposition trajectories, even though they were not included in the compiled tables. Sunderland and Selin (2013) is a commentary synthesizing projected trends in environmental mercury and highlighting the role of legacy emissions in shaping future deposition and exposure patterns. Similarly, recent studies by Geyman et al. present updated projections of global anthropogenic Hg emissions and deposition under Shared Socioeconomic Pathways (SSPs) and represent some of the most recent modeling work on future Hg emissions.

Lines 243–257:

This is a strong section, and it is helpful to see discussion of discrepancies between modeled and observed values. For the specific U.S. regions examined in this study, are there atmospheric deposition or elemental Hg observations available that could be used for comparison? Even brief contextual comparison of observed vs. model predicted values would strengthen confidence in the modeled outputs and their interpretations.

Author response

Thank you for this helpful suggestion. To provide observational context for the selected U.S. regions examined in this study, we have added a brief comparison between modeled and observed atmospheric Hg deposition.

Revision made

(Holloway et al. 2012) found that across 31 Mercury Deposition Network (MDN) sites in the Great Lakes region, the CMAQ-Hg model underestimated annual Hg wet deposition by about 21% on average, with seasonal biases showing underprediction in spring, summer, and fall and overprediction in winter by about 70%. Similarly, observations from the MDN show that Hg wet deposition at southeastern U.S. sites (Georgia, Alabama, Mississippi, South Carolina, and Florida) averaged 6.25 ± 1.48 µg m^{-2}, whereas the global GEOS-Chem model (4° × 5°) simulated only 3.33 µg m^{-2}, underestimating deposition by ~46%, largely because coarse-resolution models cannot resolve deep convection, a key driver of Hg wet deposition in the region (Xu et al. 2022)

References:

Holloway, T., C. Voigt, J. Morton, S. N. Spak, A. P. Rutter, and J. J. Schauer. 2012. "An Assessment of Atmospheric Mercury in the Community Multiscale Air Quality (CMAQ) Model at an Urban Site and a Rural Site in the Great Lakes Region of North America." *Atmospheric Chemistry and Physics* 12 (15): 7117–33. https://doi.org/10.5194/acp-12-7117-2012.

Xu, Xiaotian, Xu Feng, Haipeng Lin, et al. 2022. "Modeling the High-Mercury Wet Deposition in the Southeastern US with WRF-GC-Hg v1.0." *Geoscientific Model Development* 15 (9): 3845–59. https://doi.org/10.5194/gmd-15-3845-2022.

Lines 258–266:

The discussion of hemispheric discrepancies appears to rely largely on a single Southern Hemisphere example. If observational data from the U.S. or broader Northern Hemisphere are available, incorporating them (even briefly) would provide a more balanced perspective to this section.

Author response

Thank for this helpful suggestion. To provide a more balanced perspective between hemispheres, we have added a brief discussion of observational comparisons from the Northern Hemisphere too.

Revision made

(Travnikov et al. 2017) evaluated four global Hg models (GLEMOS, GEOS-Chem, GEM-MACH-Hg, and ECHMERIT) against observational deposition data and found that model performance depends strongly on oxidation chemistry and precipitation scavenging. Br-based schemes shift deposition maxima to spring, whereas OH-based schemes better reproduce the observed summer peaks in North America and Europe, indicating that no single oxidation pathway fully explains observed Hg deposition patterns.

References:

Travnikov, Oleg, Hélène Angot, Paulo Artaxo, et al. 2017. "Multi-Model Study of Mercury Dispersion in the Atmosphere: Atmospheric Processes and Model Evaluation." *Atmospheric Chemistry and Physics* 17 (8): 5271–95. https://doi.org/10.5194/acp-17-5271-2017.

Lines 267 and 279:

Section titles 3.2 and 3.3 are somewhat non-descriptive, and the placement of ", 2050" at the end reads awkwardly. Revising the titles for clarity and flow would improve readability.

Author response

Thank you for this helpful suggestion. We agree that the original section titles were somewhat non-descriptive and that placing "2050" at the end of the titles disrupted the flow. The section titles have therefore been revised to improve clarity and readability.

Revision made

The section titles have been revised as follows:

- 3.1 Global Hg emission scenarios vs deposition, 2050 → Global anthropogenic Hg emission scenarios and atmospheric deposition
- 3.2 Ecosystem deposition vs MeHg in fish, 2050 → Relationship between Hg deposition and fish MeHg under future scenarios

- 3.3 Lake characteristics vs MeHg in fish, 2050 → Influence of lake characteristics on projected fish MeHg responses

Line 334:

The phrase "two integrated data points" is unclear. Are these integrated data points associated with specific lakes or regions? A brief clarification would help readers who are unfamiliar with the cited study (Perlinger 2018) to understand the distinction. Related to this sentence, the mention of an additional five integrated data points on line 226 under the high deposition reduction scenario do not have a citation – are these also from the same Perlinger study or from the Knightes 2009 study that is referenced in the sentence following this statement?

Author response

Thank you for this helpful comment. We clarify that the term "integrated data points" refers to scenario-based model outputs representing individual lake simulations under specific combinations of atmospheric deposition and watershed characteristics. In Perlinger et al. (2018), these correspond to virtual experiments designed to isolate the effects of deposition and watershed sensitivity. In contrast, the five integrated data points under the high deposition-reduction scenario are derived from multiple lake simulations reported by Knightes et al. (2009), which has now been explicitly cited. The text has been revised to clarify the meaning of "integrated data points" and to explicitly cite Knightes et al. (2009) for the high deposition-reduction scenario. The description of the underlying experiments has been simplified to improve clarity and conciseness.

Revision made

For example, under the intermediate Policy-in-Action scenario with moderate deposition reductions, the modeled decline in fish MeHg for lakes in Michigan's Upper Peninsula (UP) is moderate (-0.250 % yr^{-1}). However, two virtual experiments reported by Perlinger et al. (2018) separate the effects of scenario-based atmospheric deposition and watershed sensitivity on fish MeHg responses (Ellipse A; symbol ∇). Applying Adirondack deposition to the Michigan UP lake resulted in increased modeled fish MeHg, whereas applying Adirondack watershed characteristics under the same deposition conditions led to a decrease, highlighting the influence of watershed properties. Similarly, under the high deposition-reduction scenario involving a 50 % decrease in Hg deposition (Ellipse B; symbol Δ), five integrated data points show substantial variability in fish MeHg responses under the same policy scenario (Knightes et al., 2009).

Lines 357–358 and Line 408:

Line 408 indicates that systems with larger lake area and higher wetland coverage show greater declines in fish MeHg. Meanwhile, lines 357–358 suggest that fish from smaller lakes may respond more rapidly to atmospheric Hg changes. These statements appear somewhat at odds. Clarifying the reason for these discrepancies would help with the interpretation, as well as some evaluation of how well robust these PCA results are.

Author response

Thank you for carefully pointing out this inconsistency. We believe the reviewer may have intended to refer to lines 337–338 rather than lines 357–358, as the statement mentioned in the comment corresponds to the discussion presented in lines 337–338. We have reviewed the section and clarified the wording to ensure that the relationship between lake characteristics and fish MeHg responses is described consistently. The revised text now better explains how ecosystem characteristics influence the variability in modeled fish MeHg responses.

Revision made

Similarly, under the high deposition-reduction scenario involving a 50 % decrease in Hg deposition (Ellipse B; symbol Δ), five integrated data points show substantial variability in fish MeHg responses under the same policy scenario (Knightes et al., 2009). Lakes with large surface areas—indicating greater direct atmospheric exposure—together with large watersheds and higher wetland percentages (e.g., bay lakes and coastal rivers) showed the greatest declines in fish MeHg. Intermediate declines occurred in seepage and stratified drainage lakes, whereas systems with lower wetland influence and weaker watershed-mediated Hg inputs (e.g., farm ponds) exhibited slower declines in fish MeHg (Knightes et al., 2009).

Lines 431–434 (and Line 408):

The negative relationship between wetland extent and predicted fish MeHg change is intriguing and contrasts with many field observations that associate wetlands with enhanced methylation. Given that this relationship appears consistently across multiple models, it would be valuable to explore potential mechanistic explanations in greater detail. For example, do model assumptions regarding hydrology, dilution, or loading pathways drive this outcome? Additional discussion and statistical analysis would strengthen confidence in the PCA results and clarify how they should be interpreted relative to observed trends in empirical studies.

Author response

Thank you for this insightful observation. We agree that the negative relationship between wetland extent and projected fish MeHg change requires clearer mechanistic explanation, particularly given contrasting empirical observations.

We revised the manuscript to clarify that this relationship arises from how the models represent Hg loading pathways and response times. Specifically, these models assume that atmospheric deposition to the watershed is rapidly transferred to the lake, creating a strong coupling between deposition and lake Hg loading. Consequently, wetland-dominated systems, which receive greater atmospheric inputs, are represented as more sensitive to changes in deposition.

In addition, fish MeHg is estimated using steady-state approaches that do not account for ecological complexity or legacy Hg storage lags. As a result, reductions in atmospheric deposition are translated into relatively rapid declines in modeled fish MeHg.

To further support this interpretation, we also added a supplemental regression analysis demonstrating the strong statistical relationship between wetland area, watershed area, and total Hg loading in the modeled systems, which helps explain the PCA outcome. Together, these revisions clarify that the observed negative relationship reflects the relative responsiveness of wetland-dominated systems within the models, rather than lower absolute MeHg production.

Revision made

The negative association between wetland extent and projected declines in fish MeHg observed in the PCA contrasts with many field studies that report elevated absolute fish MeHg with increasing wetland coverage, although the magnitude and direction of this relationship vary with sulfate availability, dissolved organic matter, and hydrologic setting (Watras et al., 2005; Ackerman et al., 2019; Poulin et al., 2025). This apparent discrepancy likely reflects model assumptions regarding Hg loading pathways and response kinetics. First, modeling frameworks derived from or conceptually similar to SERAFM assume relatively direct transfer of atmospherically deposited Hg from the catchment to the lake, with limited representation of hydrologic retention and delayed release from soils and wetlands (Knightes et al., 2008). Under this structure, wetlands and large watersheds act as highly sensitive receptors of atmospheric Hg inputs, strengthening atmospheric–catchment coupling in wetland-dominated systems. Our supplemental regression analysis supports this interpretation, showing that wetland area was a strong predictor of both watershed Hg loading and total Hg loading to the lake ($R^2 = 0.89$ and 0.87, respectively; $p < 0.001$). Second, fish MeHg concentrations are estimated using a steady-state Bioaccumulation Factor (BAF) approach, which does not capture the biological and ecological lag times required for fish populations to fully adjust to changes in water-column Hg exposure (Knightes et al., 2008; Angot et al., 2018). Consequently, declines in modeled water-column Hg are translated into relatively rapid declines in fish MeHg, whereas in natural systems, Hg stored in wetland soils and catchment reservoirs can delay responses to emission reductions. Thus, although wetland-dominated systems often support higher absolute MeHg concentrations in empirical studies, they emerge in these simulations as the most dynamic responders to declining atmospheric inputs because legacy storage and delayed ecosystem adjustment are only partially represented. Biogeochemical reaction rates and lake-chemistry variables (e.g., DOC, alkalinity, sulfur) could not be evaluated here due to data limitations but incorporating them in future work will be essential for linking model behavior with real-world ecosystem responses.

References

Angot, Hélène, Nicholas Hoffman, Amanda Giang, et al. 2018. “Global and Local Impacts of Delayed Mercury Mitigation Efforts.” *Environmental Science & Technology* 52 (22): 12968–77. https://doi.org/10.1021/acs.est.8b04542.

Knightes, Christopher D., Elsie M. Sunderland, M. Craig Barber, John M. Johnston, and Robert B. Ambrose. 2009. “Application of Ecosystem-Scale Fate and

Bioaccumulation Models to Predict Fish Mercury Response Times to Changes in Atmospheric Deposition.” *Environmental Toxicology and Chemistry* 28 (4): 881–93. https://doi.org/10.1897/08-242R.1.

Perlinger, J. A., N. R. Urban, A. Giang, et al. 2018. “Responses of Deposition and Bioaccumulation in the Great Lakes Region to Policy and Other Large-Scale Drivers of Mercury Emissions.” *Environmental Science: Processes & Impacts* 20 (1): 195–209. https://doi.org/10.1039/C7EM00547D.

Notes on Tables and Figures

Tables 1–3:

These tables contain important compiled information but are currently difficult to interpret.

Suggestions:

Define SRES in each table caption, as opposed to a row header in Table 1 only.

Consider streamlining columns to improve readability (for example, removing the “Scenario” column since scenarios are already defined in row headers).

Instead of grouping the model projections by policy pathways (minimal control, intermediate mitigation, maximum mitigation), I suggest that the projections are separated into two tables, with one table containing models that provide deposition/ emission only outputs and another table containing models with both deposition and fish MeHg outputs. All policy pathways with projected data should be included in each of the two tables, respectively.

I am unsure whether formatting changes occurred during manuscript upload, but I am having difficulty interpreting rows where a single reference appears to correspond to both a deposition model and one or more fish MeHg models. For example, in Table 3 under the *Aspirational scenario*, “Hendrick’s LMB model” is listed in the first row corresponding to the Great Lakes region; however, no species or % change in MeHg values are listed within that row. All fish species and associated % MeHg changes appear to be listed only for the Michigan Upper Peninsula lakes. Is it correct to interpret this as meaning that the Perlinger et al. (2018) dataset does not include fish MeHg outputs for the broader Great Lakes or New York Adirondacks regions? If so, this could be clarified explicitly in the table. If not, additional explanation is needed to distinguish how the deposition model output relates to the fish MeHg outputs presented below.

Author response

Thank you for these thoughtful and constructive suggestions regarding the presentation of Tables 1–3. We have revised the tables to improve clarity and readability. Specifically, the acronym SRES (Special Report on Emissions Scenarios) is now defined directly in the captions of the relevant tables. We have also streamlined the table structure by removing redundant columns where scenarios were already defined in the row headers.

In addition, we clarified the presentation of deposition and fish MeHg outputs. In the case highlighted by the reviewer, the dataset from Perlinger et al. (2018) does not provide projected fish MeHg outputs for the broader Great Lakes region. To avoid confusion, rows corresponding to regions without fish MeHg projections have been removed, and the tables have been revised accordingly to clearly indicate where fish MeHg outputs are available.

Revision made

Tables 1–3 have been revised and consolidated into two tables to improve readability: one table presents emission → deposition projections, while the second presents deposition → fish MeHg projections. The acronym SRES is now defined in the table captions, redundant columns have been removed, and rows corresponding to regions without available fish MeHg projections (e.g., the broader Great Lakes region in Perlinger et al., 2018) have been removed.

Table 1. Projected changes in global anthropogenic mercury emissions and atmospheric deposition by 2050 under different policy scenarios and modeling frameworks.

Base year	Global anthropogenic Hg emissions in base year (Mg yr^{-1})	Global anthropogenic Hg emissions in 2050 (Mg yr^{-1})	Change in total Hg deposition in 2050 w.r.t. base year (%)	Model used for deposition simulations	Regions analyzed	References
SRES: Special Report on Emissions Scenarios—an Intergovernmental Panel on Climate Change (IPCC, 2000) framework that projects future emissions based on trajectories of energy use, fuel consumption, economic development, and technological change. The scenarios are grouped into four main families: A1, A2, B1, and B2.						
A1B: A subset of A1 where all energy sources are balanced (neither high fossil dominance nor full non-fossil). [A1: A future characterized by rapid economic and technological growth, with global population peaking mid-century]						
2000	2190	4856	75 75	CAM-Chem/Hg	Eastern U.S. Western U.S.	(Lei et al. 2014)
2005	1900	4300	21 30	GEOS-Chem	Global US	(Corbitt et al. 2011)

2010	1890	4900	87	GEOS-Chem + MITgcm + GTMM	Global	(Y. Zhang et al. 2021)
2015	2500	4900	40	5-box geochemical model for Arctic + Global Box Model	Arctic	(Chen et al. 2018; AMAP/UNEP, 2018)
2005	1900	4300	37	GEOS-Chem	Great Lakes	(H. Zhang et al. 2021)
A2: A fragmented, self-reliant world with high population growth, slow technological change, and regionally oriented development.						
2005	1900	3400	25	GEOS-Chem	Global	(Corbitt et al. 2011)
2010	1890	3900	59	GEOS-Chem + MITgcm + GTMM	Global	(Y. Zhang et al. 2021)
2006	2480	3900	12	CTM-Hg, TEAM, AERMOD	Mendums Pond, NH	(Vijayaraghavan et al. 2014)
A1F1: subset of A1 ("Fossil-intensive") where future energy use remains heavily dependent on fossil fuels.						
2000	2190	5984	100	CAM-Chem/Hg	Eastern U.S.	(Lei et al. 2014)
			75		Western U.S.	
No policy: Assumes no new Hg or air-quality controls beyond those implemented by 2010, with continued coal combustion and limited emission-control expansion.						
2005 - 2006	2000	4140	30	GEOS-Chem	U.S.	(Giang and Selin 2016)
B1: An environmentally sustainable, convergent world with service-based economy and clean, efficient technologies.						
2000	2190	2386	13	CAM-Chem/Hg	Eastern U.S.	(Lei et al. 2014)
			0		Western U.S.	
2005	1900	1900	1	GEOS-Chem	Global	(Corbitt et al. 2011)
			-10		U.S.	
			-22		Northeast U.S.	
2005	1900	1900	-13	GEOS-Chem	Great Lakes	(H. Zhang et al. 2021)
B2: A local sustainability-focused scenario with moderate growth and slower, diverse technological change.						
2005	1900	2200	7	GEOS-Chem	Global	(Corbitt et al. 2011)
2006	2480	2630	-15	CTM-Hg, TEAM, AERMOD	Mendums Pond, NH	(Vijayaraghavan et al. 2014)
Minamata: Global treaty scenario achieving major emission cuts via best available technologies and control measures across key Hg sources.						
2005 - 2006	2000	2270	5	GEOS-Chem	U.S.	(Giang and Selin 2016)
NPS: Implementation of pledged global actions (e.g., Minamata, fossil-fuel phase-outs) achieving notable emission cuts by 2035. [CPS: Continuation of 2010 policies and controls without new Hg-specific or climate initiatives.]						
2010	1960	0	-14	GEOS-Chem + GBC Model	Global	(Angot et al. 2018)
			26		Ahmedabad (India)	
			-55		Shanghai (China)	
			-13		South Pacific	

			-15		Eastern lakes, Maine (U.S.)	
2010	1890	1020	-28	GEOS-Chem + MITgcm + GTMM	Global	(Y. Zhang et al. 2021)
Constant emissions: Assumes no change in anthropogenic Hg emissions by 2050 with respect to 2015						
2015	2500	2500	12	5-box geochemical model for Arctic + Global Box Model	Arctic	(Chen et al. 2018; AMAP/UNEP, 2018)
MFR: Most optimistic case with universal adoption of best available technologies for maximum emission reduction.						
2010	1890	300	-48	GEOS-Chem + MITgcm + GTMM	Global	(Y. Zhang et al. 2021)
2010	2000	0	-24 -38 -68 -22 -26	GEOS-Chem + GBC Model	Global Ahmedabad (India) Shanghai (China) South Pacific Eastern lakes, Maine (U.S.)	(Angot et al. 2018)
Hg Controls: Assumes 50% reduction in primary anthropogenic Hg emissions by 2050 via widespread Hg-specific controls.						
2015	2500	1250	-16	5-box geochemical model for Arctic + Global Box Model	Arctic	(Chen et al. 2018; AMAP/UNEP, 2018)
Zero Emissions: Idealized case where all primary anthropogenic Hg emissions cease after 2015, representing the maximum achievable reduction.						
2015	2500	0	-50	5-box geochemical model for Arctic + Global Box Model	Arctic	(Chen et al. 2018; AMAP/UNEP, 2018)

Table 2. Projected changes in atmospheric Hg deposition and corresponding fish MeHg concentrations by 2050 across different scenarios and modeling frameworks.

Base year	**Change in total Hg deposition in 2050 w.r.t. base year (%)**	**Model used for deposition simulations**	**Change in MeHg in fish in 2050 w.r.t. base year (%)**	**Models used for MeHg in fish simulations**	**Regions analyzed**	**Fish type**	**References**
SRES: Special Report on Emissions Scenarios—an Intergovernmental Panel on Climate Change (IPCC, 2000) framework that projects future emissions based on trajectories of energy use, fuel consumption, economic development, and technological change. The scenarios are grouped into four main families: A1, A2, B1, and B2.							
SRES A2: A fragmented, self-reliant world with high population growth, slow technological change, and regionally oriented development.							
2006	12	CTM-Hg, TEAM, AERMOD	5	D-MCM	Mendums Pond, NH	LMB and yellow perch	(Vijayaraghavan et al. 2014)
SRES B2: A local sustainability-focused scenario with moderate growth and slower, diverse technological change.							
2006	-15	CTM-Hg, TEAM, AERMOD	-14	D-MCM	Mendums Pond, NH	LMB and yellow perch	(Vijayaraghavan et al. 2014)
NPS: Implementation of pledged global actions (e.g., Minamata, fossil-fuel phase-outs) achieving notable emission cuts by 2035. [CPS: Continuation of 2010 policies and controls without new Hg-specific or climate initiatives.]							

2010	-15	GEOS-Chem + GBC Model	-11	Hendrick's LMB model	Eastern lakes, Maine	Brook trout, brown trout, burbot, Landlocked salmon, and smallmouth bass	(Angot et al. 2018)
Policy-in-action: Full implementation of existing Hg-control measures, including Minamata Convention and U.S. Clean Air Act rules, stabilizing emissions near current levels by 2050.							
2005-2006	-15	GEOS-Chem	-11	Hendrick's LMB model	Lakes in Michigan's UP	Northern pike, LMB, Yellow perch, and Pickerel	(Perlinger et al. 2018)
	12		19		Lake in Michigan's UP with Adirondack's deposition	Walleye	
	12		-38		Lake in Michigan's UP with Adirondack's deposition and watershed features	Walleye	
MFR: Most optimistic case with universal adoption of best available technologies for maximum emission reduction.							
2010	-26	GEOS-Chem + GBC Model	-22	Hendrick's LMB model	Eastern lakes, Maine	Brook trout, brown trout, burbot, Landlocked salmon, and smallmouth bass	(Angot et al. 2018)
Aspirational: Assumes complete elimination of anthropogenic Hg emissions by 2050 through a global transition to Hg-free technologies and renewable energy sources.							
2005-2006	-65	GEOS-Chem	-65	Hendrick's LMB model	Lakes in Michigan's UP	Northern pike, LMB, Yellow perch, and Pickerel	(Perlinger et al. 2018)
Reduction in deposition, 50%: A scenario applying a uniform 50% reduction to locally observed Hg deposition (without specifying global emissions).							
2001	-50	Community Multi-scale Air Quality Model (CMAQ)	-31	WASP, SERAFM, BASS	Eagle Butte Lake, SD	Northern pike	(Knightes et al. 2009)
			-51		Lake Barco, FL	LMB	
			-57		Pawtuckaway Lake, NH	yellow perch	
			-77		Lake Waccamaw, NC	LMB	
			-74		Brier Creek, GA	Pickerel	

Figure 1:

Clear and informative. It may improve flow if presented before the tables so readers can orient themselves spatially prior to reviewing tabulated data. Aligning legend order with caption order (e.g. placing NPS above MFR in the figure legend) may also improve clarity.

Author response

Thank you for this helpful suggestion. We agree that presenting Figure 1 before the tables improves the flow of the manuscript and helps readers spatially orient themselves prior to reviewing the tabulated data. We have therefore repositioned Figure 1 earlier in the manuscript. In addition, the legend order in the figure has been adjusted to match the order used in the caption (e.g., NPS now appears above MFR) to improve clarity.

Revision made

Figure 1 has been repositioned earlier in the manuscript before the tables, and the legend order has been revised to align with the caption.

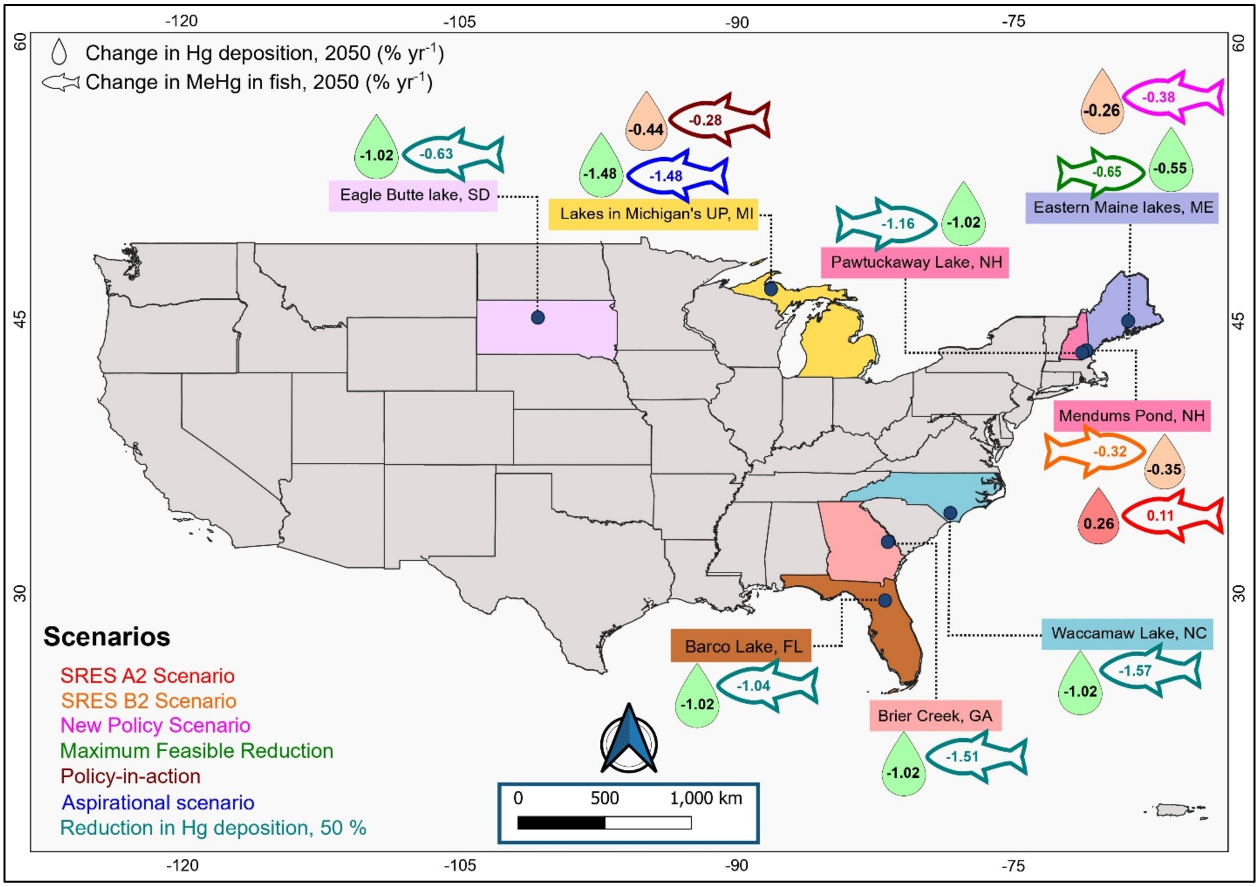

Figure 1: Geospatial distribution of projected changes in atmospheric Hg deposition and fish MeHg concentrations across the United States by 2050. Blue dots indicate study locations. Droplet symbols represent scenario-based changes in Hg deposition, while fish symbols show corresponding changes in fish MeHg. Fish colors denote emission policy scenarios: MFR and NPS (Angot et al. 2018), SRES A2 and B2 (Vijayaraghavan et al. 2014), Policy-in-action and Aspirational (Perlinger et al. 2018), and a

uniform 50 % reduction in Hg deposition (Knightes et al. 2009). Droplet fill colors indicate relative emission (or deposition) levels across scenarios, classified as low (green), medium (orange), and high (pink).

Figure 2:

Interesting figure, though its centrality to the manuscript could be reconsidered. The authors may wish to consider whether it belongs to the main text or Supplementary Material.

Author response

Thank you for this helpful suggestion. We agree that Figure 2 provides a conceptual overview and may not be essential to the main results of the manuscript. Therefore, to improve the overall focus and readability of the paper, we have moved Figure 2 to the Supplementary Material.

Revision made

Figure 2 has been moved from the main manuscript to the Supplementary Material.

Table 4:

The technical comparison of model structures is valuable but may be more appropriate for Supplementary Material, particularly since the main text (Lines 180–195) summarizes the most relevant distinctions.

Author response

Thank you for this helpful suggestion. We agree that the detailed technical comparison of atmospheric model structures is better suited to the Supplementary Material, as the main text already summarizes the most relevant distinctions. Therefore, to improve the flow and readability of the manuscript, we have moved Table 4 to the supplementary Material.

Revision made

Table 4 has been moved from the main manuscript to the Supplementary Material.

Figure 5:

This figure is somewhat difficult to interpret in its current form, and the caption does not provide enough detail for it to stand alone. The associated text (Lines 356–361) communicates the key message clearly without needing to reference the figure. The authors may wish to reconsider whether this figure adds sufficient clarity in its present format. If the exact values plotted in Figure 5 are captured in Tables 1-3, the authors may want to reference the appropriate table within the associated text.

Author response

Thank you for this helpful suggestion. We agree that Figure 5 does not substantially improve the clarity of the results in its current format. The key information shown in this figure is already clearly described in the associated text (Lines 356–361) and summarized in Table 2. Therefore, to avoid redundancy and improve the clarity of the manuscript, we have removed Figure 5.

Revision made

Figure 5 has been removed from the manuscript.

Very Minor Notes

Lines 200–203:

It may help to very briefly clarify that emissions and deposition estimates are derived from the same reference for each region.

Author response

Thank you for this helpful suggestion. We agree that clarifying the origin of the emission and deposition estimates improves transparency. We have therefore added a short sentence to specify that the emission and deposition values for each data point are derived from the same study.

Revision made

To assess the influence of changes in anthropogenic Hg emissions on future deposition, we analyzed the relationship between annual changes (% yr^{-1}) in global anthropogenic Hg emissions and corresponding annual changes (% yr^{-1}) in Hg deposition projected for 2050 (Figure 3). Each data point represents a specific combination of atmospheric model, emission scenario, surface type, and region. For each case, the emission and deposition estimates are derived from the same study to maintain internal consistency within each regional scenario.

Line 290:

“Dynamic Mercury Cycling Mode” should likely read “Model.”

Author response

Thank you for noting this typographical error. The text has been corrected accordingly.

Revision made

Dynamic Mercury Cycling Model