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Future mercury levels in fish: model vs. observational predictions under different policy scenarios

Henna Gull[1], Ju Hyeon Lee[2], Hoin Lee[1], Hélène Angot[3], Sae Yun Kwon[1]

[1]Division of Environmental Science and Engineering, Pohang University of Science and Technology, 77 Cheongam-Ro, Nam-Gu, Pohang 37673, South Korea
[2]Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, United States
[3]Univ. of Grenoble Alpes, CNRS, INRAE, IRD, Grenoble INP, IGE, 38400 Grenoble, France

Correspondence to: Sae Yun Kwon (saeyunk@postech.ac.kr)

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 in 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. Modeled 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, legacy emissions, meteorological drivers, methylation-demethylation kinetics, and food web dynamics, the linear trends persisted. This apparent linearity underscores robust large-scale cause–effect patterns but also calls for caution: do current models truly capture the complexity of real atmospheric and ecosystem processes, or might they oversimplify mercury's nonlinear cycling and ecological responses? These findings highlight the need to remain open to processes and interactions not yet fully represented in models, ensuring that future mercury assessments and policy strategies reflect the true complexity of natural systems.

1 Introduction

Mercury (Hg), identified among the top 10 chemicals by the World Health Organization (WHO 2024), occurs naturally in both inorganic and organic forms in the environment. Natural contributions include volcanic emissions, geothermal activity, and biomass burning (Kumar et al. 2018; Kumar and Wu 2019), whereas anthropogenic sources are dominated by gold mining, coal combustion, metal production, cement manufacturing, and waste incineration (Pacyna et al. 2006; Pirrone et al. 2010; Rafaj et al. 2013). In addition to these sources, surface re-emissions from ocean and land reservoirs, including legacy

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emissions, amplify present-day atmospheric Hg burdens (Gustin et al. 2008; Pirrone et al. 2010; Gworek et al. 2020). Accounting for both historical anthropogenic inputs and their continued re-emission, current atmospheric Hg concentrations are estimated to be approximately 450 % higher than pre-1450 (pre-anthropogenic) levels, accompanied by an average 300 % increase in global Hg deposition and a 230 % increase in surface ocean Hg concentrations (Outridge et al. 2018). This enrichment is spatially uneven, with atmospheric Hg levels increasing by up to 1600 % in the Northern Hemisphere and up to 400 % in the Southern Hemisphere relative to pre-anthropogenic conditions (Li et al. 2020). In the atmosphere, Hg primarily exists as gaseous elemental Hg (GEM or Hg0), gaseous oxidized Hg (GOM or HgII), and particulate-bound oxidized Hg (PHg) (Kim and Zoh 2012). GEM has a long atmospheric lifetime (0.5 to 1 year) due to its low solubility and relative chemical stability, enabling long-range transport (Selin 2009; Ariya et al. 2015), whereas GOM is more soluble and reactive, with a lifespan of days (Moore et al. 2013). GEM was traditionally thought to be removed mainly through oxidation to GOM, which, along with PHg, is deposited via wet and dry pathways (Kim and Zoh 2012). However, recent findings indicate that direct foliar and soil uptake of GEM accounts for 75 % of atmospheric Hg deposition to terrestrial surfaces (Obrist et al. 2018; Sonke et al. 2023).

Once deposited, inorganic Hg can be converted into methylmercury (MeHg), a potent neurotoxin (Gilmour et al. 2013). This process, facilitated by microbial activity, occurs in sediments (Dai et al. 2021; Chen et al. 2025; Zhou et al. 2025) and within the water column of lakes (Capo et al. 2023; Peterson et al. 2025), estuaries (Bradford et al. 2023; Liu et al. 2023), and the open ocean (Sun et al. 2020; Zhang et al. 2020). As MeHg moves through aquatic food webs, it bioaccumulates and biomagnifies, posing risks to fish and humans consuming fish (Al-Sulaiti et al. 2022; Basu et al. 2023). While aqueous MeHg concentrations are reliable predictors for fish Hg levels (Wu et al. 2021; Blanchfield et al. 2022), these concentrations are shaped by complex interactions beyond direct atmospheric Hg input. For example, fish MeHg levels may vary more strongly with watershed or riverine Hg inputs than with atmospheric deposition (Willacker et al. 2020; Emmerton et al. 2023). Parameters such as lake size, morphology, and physicochemistry of water (pH, conductivity, dissolved organic carbon (DOC), nutrients) can also alter MeHg production and biotic uptake (Bodaly et al. 1993; Kidd et al. 2012; Ahonen et al. 2018; Knott et al. 2020; Ogorek et al. 2021). Beyond local conditions, climatic and ecological processes, including permafrost thaw, sea ice loss, food web shifts, and changing primary production, can substantially modify MeHg exposure in aquatic food webs (Wang et al. 2019; Lepak et al. 2019; Schartup et al. 2019; Schaefer et al. 2020; Y. Zhang et al. 2021). These combined influences highlight the complex, multi-scale drivers of Hg bioaccumulation in aquatic ecosystems.

Recognition of Hg's global transport and multifaceted drivers has spurred international regulation. Early actions included the 1998 Convention on Long-Range Transboundary Air Pollution Protocol on Heavy Metals, which targeted reductions in industrial Hg, cadmium (Cd), and lead (Pb) emissions from major sources and combustion processes (Selin and Selin 2006). Reports by the Arctic Monitoring Assessment Program (AMAP) and the United Nations Environment Programme (UNEP) emphasized the need for global cooperation, complemented by European Union measures such as the 2005 Community Strategy Concerning Hg (Russ et al. 2009). The Minamata Convention on Hg, in force since 2017, now coordinates global efforts to reduce anthropogenic Hg emissions and releases across all sectors (Bank 2020).

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Over the years, projections of future Hg emission scenarios have been meticulously crafted through assessments of influencing factors (socio-economy, technology), and by employing diverse modeling techniques (Knightes et al. 2009; Streets et al. 2009; Rafaj et al. 2013; Giang et al. 2015; Pacyna et al. 2016; Chen et al. 2018; Perlinger et al. 2018; H. Zhang et al. 2021; Y. Zhang et al. 2021; Brocza et al. 2024; Geyman et al. 2024; 2025). Estimating future Hg emissions and their impacts on ecosystem and fish Hg levels requires consideration of evolving socio-economic and technological drivers, atmospheric chemistry, biogeochemical processes, climate variability, land-use change, and ongoing emissions and re-emissions from legacy sources. This complexity has led to differing research approaches and/or the development of simple to sophisticated models, with some studies omitting certain elements while others incorporating them to varying extents (Knightes et al. 2009; Streets et al. 2009; 2011; Corbitt et al. 2011; Sunderland and Selin 2013; Rafaj et al. 2013; Lei et al. 2014; Vijayaraghavan et al. 2014; Giang et al. 2015; Giang and Selin 2016; Pacyna et al. 2016; Angot et al. 2018; Chen et al. 2018; Perlinger et al. 2018; Streets 2019; H. Zhang et al. 2021; Y. Zhang et al. 2021; Schartup et al. 2022; Brocza et al. 2024; Geyman et al. 2024; 2025).

This study revisits previously developed Hg emission scenarios formulated under varying policy frameworks and their modeled deposition (atmospheric chemistry–transport models) and impacts on fish MeHg (bioaccumulation models) to evaluate how policy-driven emission changes influence projected 2050 deposition patterns and lake fish MeHg, and the extent to which lake-specific characteristics mediate these relationships. We hypothesize that future emission changes may produce non-linear responses in Hg deposition, reflecting differences among models in spatial resolution, emission inventories, meteorological drivers, and atmospheric chemistry. Deposition changes may further translate non-linearly to fish MeHg due to variability in bioaccumulation model structures and ecosystem-specific controls, which are often shaped by interacting environmental and climate-driven processes rather than by emissions alone (Burke et al. 2023; Gillies et al. 2024). Using published data and statistical analyses, we examine how prior future assessments simulate Hg deposition and bioaccumulation under varying policies and lake-specific effects.

2 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 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. Collectively, the selected studies evaluate future Hg emission scenarios and their impacts on global or regional Hg deposition, as well as the influence of projected deposition changes on fish MeHg concentrations, primarily within the United States (U.S.), using coupled atmospheric chemistry-transport and aquatic bioaccumulation models. The analysis focuses on lake and freshwater ecosystems, for which consistent model outputs of Hg deposition and fish MeHg concentrations were available across multiple scenarios, with most data derived

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from lake-based assessments. 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 3.

Across the selected studies, scenario-based Hg emissions were first projected using global and/or regional inventories that differed in baseline year, emission source coverage, and assumed policy actions. These emissions served as inputs to atmospheric chemical transport models including GEOS-Chem, GLEMOS, ECHMERIT, and CAM-Chem/Hg (Lei et al. 2014; Pacyna et al. 2016; Angot et al. 2018; Chen et al. 2018; Perlinger et al. 2018; H. Zhang et al. 2021; Y. Zhang et al. 2021), to estimate spatial patterns of future Hg deposition. The modeled deposition outputs were subsequently coupled with aquatic bioaccumulation models such as BASS, D-MCM, and SERAFM (Knightes et al. 2009; Vijayaraghavan et al. 2014; Angot et al. 2018; Perlinger et al. 2018), to predict MeHg concentrations in fish. This sequential framework, from emissions to deposition to bioaccumulation, formed the foundation for the correlation and regression analyses conducted in this study.

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 summarized in Table 1 and mapped in Figure 1. 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.

Our study employed Pearson correlation and regression analyses to investigate how changes in global anthropogenic Hg emissions under various policy scenarios relate to variations in Hg deposition by 2050. Statistical significance was assessed at the 95 % confidence level ($p < 0.05$). Changes in Hg deposition (% yr^{-1}) and changes in fish MeHg (% yr^{-1}) were evaluated for a subset of emission scenarios for which MeHg data were available. Multiple linear regression models (MLRM) were first 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. To explore multivariate relationships among predictor variables, Principal Component Analysis (PCA) was conducted on standardized data using Python. The analysis included lake characteristics as used in the MLRM, projected changes in Hg deposition (% yr^{-1}), and projected changes in fish MeHg (% yr^{-1}) for 2050, all within the framework of Hg emission scenarios. PCA reduced dimensionality by consolidating explanatory variables into principal component (PC) scores. A biplot was generated to visualize variable contributions and correlations to the first two PCs, with color intensity indicating relative influence. Scree and contribution plots were also used to assess the relative importance of each component and to identify the most influential drivers in the dataset. In one case, results from (Knightes et al. 2009) were interpolated to estimate fish MeHg levels for 2050 under a simulated 50 % reduction in Hg deposition. Study locations were georeferenced and mapped using QGIS 3.28.5 to depict spatial coverage.

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Table 1. Projections of global Hg emissions, deposition, and fish MeHg levels under minimal-control with high-growth futures (2050 vs. Base Year).

Scenario	Base year	Global anthropogenic Hg emissions in base year (Mg yr^{-1})	Global anthropogenic Hg emissions in 2050 (Mg yr^{-1})	Change in Hg deposition in 2050 w.r.t. base year (%)	Model used for deposition	Change in fish MeHg in 2050 w.r.t. base year (%)	Models used for fish MeHg	Regions analyzed	Fish type	Reference
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]										
SRES A1B	2000	2190	4856	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.										
SRES A2	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	5	D-MCM	Mendums Pond, NH	LMB and yellow perch	(Vijayaraghavan et al. 2014)
A1F1: subset of A1 ("Fossil-intensive") where future energy use remains heavily dependent on fossil fuels.										
SRES A1F1	2000	2189.9	5984	100 75	CAM-Chem/Hg			Eastern U.S. Western U.S.		(Lei et al. 2014)
No policy: Assumes no new Hg or air-quality controls beyond those implemented by 2010, with continued coal combustion and limited emission-control expansion.										

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No policy	2005 - 2006	4140	30	GEOS-Chem	U.S.		(Giang and Selin 2016)
Minimal Regulation: Limited global progress with weak Hg controls and continued coal dependence, nearly doubling emissions by 2050.							
Minimal-Regulation	2005-2006		35 34	Geos-Chem	Great Lakes Lakes in Michigan's Upper Peninsula		(Perlinger et al. 2018)

135 **Table 2.** Projections of global Hg emissions, deposition, and fish MeHg levels under intermediate-policy pathways (2050 vs. Base Year).

Scenario	Base year	Global anthropogenic Hg emissions in base year (Mg yr^{-1})	Global anthropogenic Hg emissions in 2050 (Mg yr^{-1})	Change in Hg deposition in 2050 w.r.t. base year (%)	Model used for deposition	Change in fish MeHg in 2050 w.r.t. base year (%)	Model used for fish MeHg	Regions analyzed	Fish type	Reference
B1: An environmentally sustainable, convergent world with service-based economy and clean, efficient technologies.										
SRES B1	2000	2190	2386	13 0	CAM-Chem/Hg			Eastern U.S. Western U.S.		(Lei et al. 2014)
	2005	1900	1900	1 -10 -22	GEOS-Chem			Global U.S. Northeast U.S.		(Corbitt et al. 2011)
	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.										
SRES B2	2005	1900	2200	7	GEOS-Chem			Global		(Corbitt et al. 2011)
	2006	2480	2630	-15	CTM-Hg, TEAM, AERMOD	-14	D-MCM	Mendums Pond, NH	LMB and yellow perch	(Vijayaraghavan et al. 2014)
Minamata: Global treaty scenario achieving major emission cuts via best available technologies and control measures across key Hg sources.										
Minamata Convention	2005 - 2006		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.]										
NPS	2010	1960	0	-14	GEOS-Chem +			Global		

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					GBC Model					(Angot et al. 2018)
				26				Ahmedabad (India)		
				-55				Shanghai (China)		
				-13				South Pacific		
				-15		-11	Hendrick's LMB model	Eastern lakes, Maine	Brook trout, brown trout, burbot, Landlocked salmon, and smallmouth bass	
NPS - delayed	2010	1890	1020	-28	GEOS-Chem + MITgcm + GTMM			Global		(Y. Zhang et al. 2021)
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.										
Policy-in-action	2005 - 2006			-20	GEOS-Chem			Great Lakes		(Perlinger et al. 2018)
				-15		-11	Hendrick's LMB model	Lakes in Michigan's UP	Northern pike, LMB, Yellow perch, and Pickerel	
				-29				New York Adirondacks		
				12		19		New York Adirondacks	Walleye	
				12		-38		New York Adirondacks	Walleye	
Constant emissions: Assumes no change in anthropogenic Hg emissions by 2050 with respect to 2015										
Constant emissions	2015	2500	2500	12	5-box geochemical model for Arctic + Global Box Model			Arctic		(Chen et al. 2018; AMAP/UNEP, 2018)

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Table 3. Projections of global Hg emissions, deposition, and fish MeHg levels under maximum-mitigation futures (2050 vs. Base Year).

Scenario	Base year	Global anthropogenic Hg emissions in base year (Mg yr^{-1})	Global anthropogenic Hg emissions in 2050 (Mg yr^{-1})	Change in Hg deposition in 2050 w.r.t. base year (%)	Model used for deposition	Change in fish MeHg in 2050 w.r.t. base year (%)	Model used for fish MeHg	Regions analyzed	Fish type	Reference
MFR: Most optimistic case with universal adoption of best available technologies for maximum emission reduction.										
MFR	2010	1890	300	-48	GEOS-Chem + MITgcm + GTMM			Global		(Y. Zhang et al. 2021)
		2000	0	-24	GEOS-Chem + GBC Model			Global		(Angot et al. 2018)
				-38				Ahmedabad (India)		
				-68				Shanghai (China)		
				-22				South Pacific		
				-26		-22	Hendrick's LMB model	Eastern lakes, Maine	Brook trout, brown trout, burbot, Landlocked salmon, and smallmouth bass	
Aspirational: Assumes complete elimination of anthropogenic Hg emissions by 2050 through a global transition to Hg-free technologies and renewable energy sources.										
Aspirational Scenario	2005 - 2006			-70	GEOS-Chem		Hendrick's LMB model	Great Lakes		(Perlinger et al. 2018)
				-78				New York Adirondacks		
				-65		-65		Lakes in Michigan's UP	Northern pike, LMB, Yellow perch, and Pickerel	
Hg Controls: Assumes 50 % reduction in primary anthropogenic Hg emissions by 2050 via widespread Hg-specific controls.										
Hg Controls	2015	2500	1250	-16	5-box geochemical model for Arctic + Global Box Model			Arctic		(Chen et al. 2018; AMAP/UNEP, 2018)

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Zero Emissions: Idealized case where all primary anthropogenic Hg emissions cease after 2015, representing the maximum achievable reduction.										
Zero emissions	2015	2500	0	-50	5-box geochemical model for Arctic + Global Box Model			Arctic		(Chen et al. 2018; AMAP/UNEP, 2018)
Reduction in deposition, 50 %: A scenario applying a uniform 50 % reduction to locally observed Hg deposition (without specifying global emissions).										
Reduction in deposition, 50 %	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	

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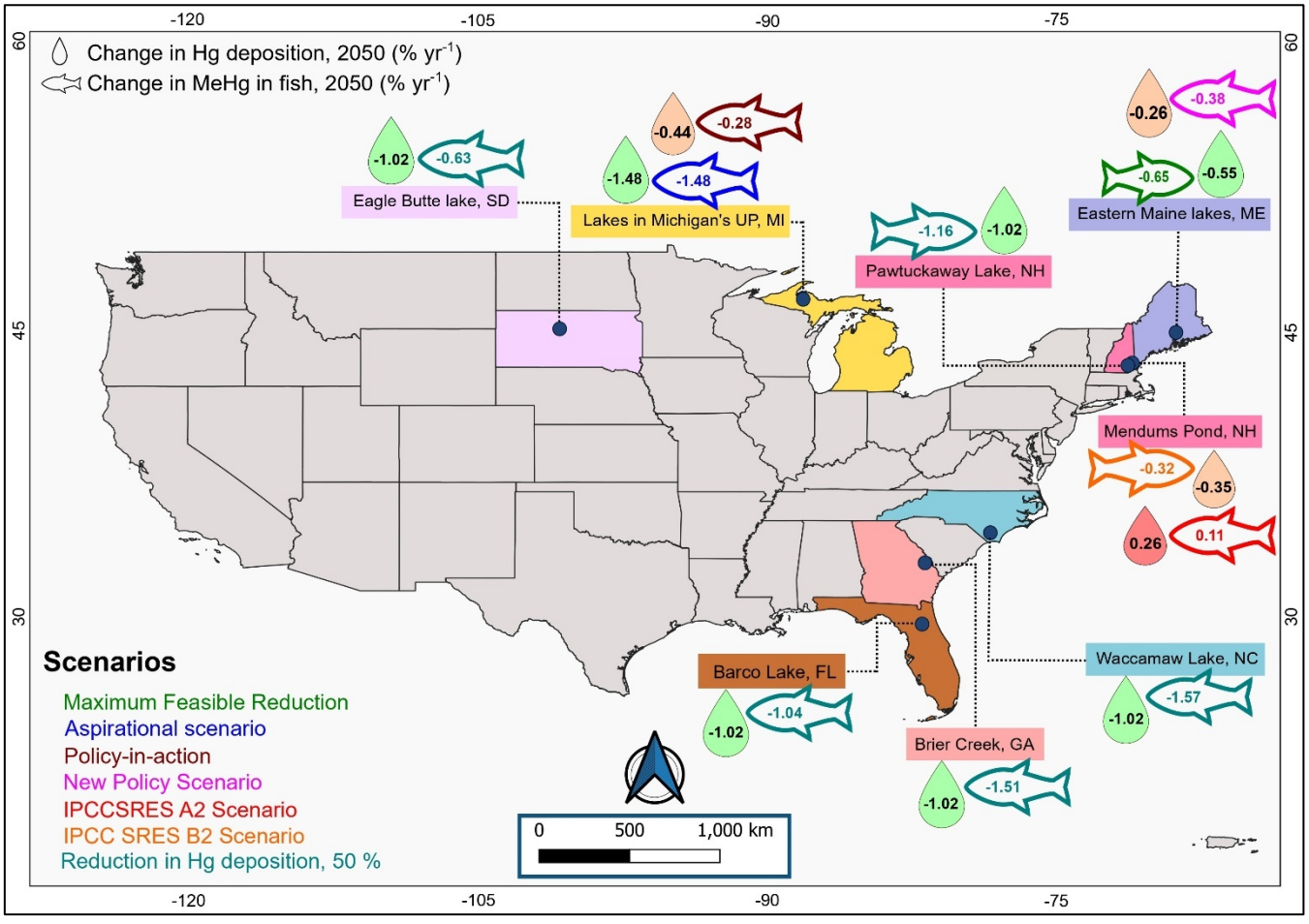

Figure 1: Geospatial distribution of projected changes in atmospheric Hg deposition and fish MeHg concentrations across the United States by 2050. Blue dots indicate study locations. Droplet symbols represent scenario-based changes in Hg deposition, while fish symbols show corresponding changes in fish 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).

3 Results and discussion

3.1 Global Hg emission scenarios vs deposition, 2050

Understanding the relationship between Hg emissions and deposition is essential for assessing future environmental risks and exposure pathways. Researchers have employed various atmospheric Hg models to project Hg deposition patterns under multiple emission scenarios, each with distinct atmospheric chemistry mechanisms, meteorological drivers, and varying assumptions regarding legacy emissions. In particular, numerous studies have projected global anthropogenic Hg emissions up to 2050, using different scenario frameworks (Tables 1–3) and emission inventories (Supporting Information: Table

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S1). The main differences between scenarios lie in the stringency of Hg control measures and the underlying socio-economic–energy trajectories—ranging from *minimal-control, high-growth futures* (A1F1, A1B, A2, CPS, Minimal-regulation, SSP5-8.5) (Knightes et al. 2009; Sunderland and Selin 2013; Vijayaraghavan et al. 2014; Lei et al. 2014; Giang et al. 2015; Chen et al. 2018; Perlinger et al. 2018; H. Zhang et al. 2021; Schartup et al. 2022; Brocza et al. 2024; Geyman et al. 2024; 2025) with rising emissions (Table 1), through *intermediate-policy pathways* (B1, B2, PIA, NPS, CLIM, Minamata, SSP5-3.4, SSP2-4.5) (Streets et al. 2009; Sunderland and Selin 2013; Rafaj et al. 2013; Vijayaraghavan et al. 2014; Lei et al. 2014; Giang and Selin 2016; Angot et al. 2018; Perlinger et al. 2018; H. Zhang et al. 2021; Y. Zhang et al. 2021; Geyman et al. 2024; 2025) that stabilize or moderately reduce emissions (Table 2), to *maximum-mitigation futures* (50 % Reduction, MFR, CLIM + MFR, Aspirational, Zero emissions, Net Zero + MFR, SSP1-2.6) (Pacyna et al. 2016; Angot et al. 2018; Chen et al. 2018; Perlinger et al. 2018; Y. Zhang et al. 2021; Brocza et al. 2024; Geyman et al. 2024; 2025) achieving the deepest cuts (Table 3). In some studies, e.g., (Giang and Selin 2016), Minamata and No-Policy scenarios adopt B1- and A1B-type socio-economic assumptions, respectively. To illustrate their origins and policy focus, Figure 2 organizes these scenarios into four thematic families: Hg-focused policy scenarios, SRES-based frameworks, shared socioeconomic pathways, climate-oriented pathways, and hybrid syntheses. Collectively, these studies span the full spectrum from continued growth to deep global reductions in anthropogenic Hg emissions. The acronyms for Hg-focused and SRES scenarios are defined in Tables 1–3, while climate-oriented and hybrid frameworks, often reported only as emission trajectories and, in some cases, not extending to 2050 or lacking deposition outputs—are listed separately in Table S2.

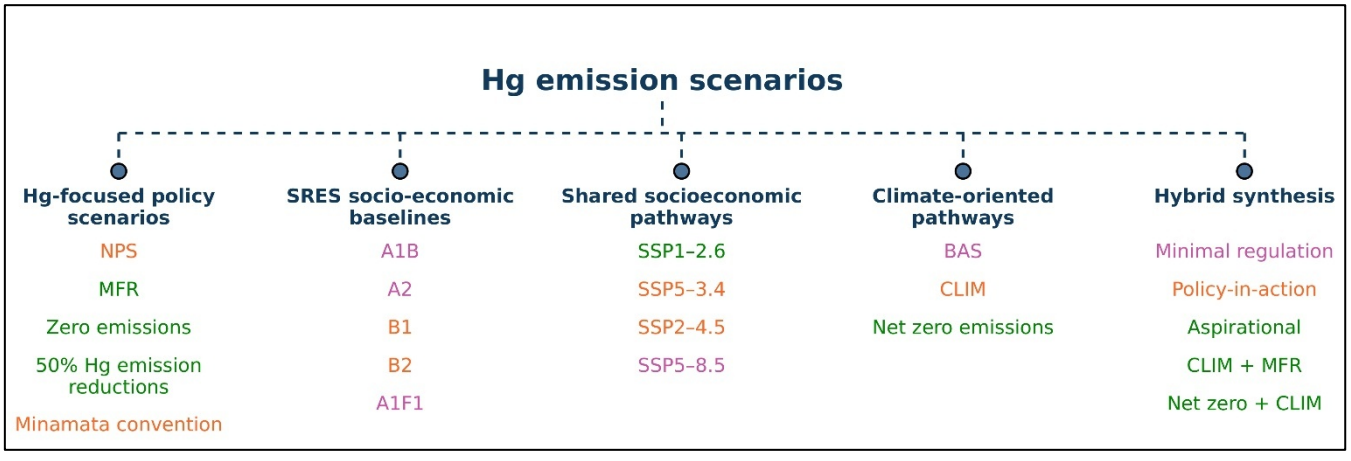

Figure 2: Classification of future Hg emission scenarios by policy focus and emission level. Emission levels are categorized as high (pink), medium (orange), and low (green).

Table 4. Characteristics of atmospheric Hg models, including resolution, primary oxidation pathways, and meteorological drivers.

Model	Spatial resolution	Vertical resolution	Primary Hg^0 oxidation pathways	Meteorological data sources (type)	References

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GEOS-Chem	4°×5°	47 levels	Br	GEOS-5 assimilated meteorology (Offline)	(Corbitt et al. 2011; Giang and Selin 2016; Chen et al. 2018)
	2°×2.5°	40 levels	Br (1st - stage)	MERRA-2 by NASA's GMAO (Offline);	(Angot et al. 2018; Y. Zhang et al. 2021; H. Zhang et al. 2021)
			NO_2 and HO_2 (2nd - stage)	NASA GISS GCM ModelE2 (Online)	
GLEMOS	1°×1°	20 levels	O_3 and OH	WRF modelling system, based on ECMWF (Offline)	(Pacyna et al. 2016; De Simone et al. 2017; Travnikov et al. 2017)
			Br (Polar regions)		
ECHMERIT	2.8°×2.8°	19 levels	O_3 and OH	ECHAM5 based on ECMWF (Online)	
HYSPLIT-Hg	2.5°×2.5°	17 levels	O_3 and OH	NCEP/NCAR reanalysis model (Offline)	(Cohen et al. 2016)
GEM-MACH-Hg	1°×1°	58 levels	OH	GEM model, based on ECCC weather prediction model (Online)	(Travnikov et al. 2017)
			Br (Polar regions)		
CAM-Chem/Hg	1.9°×2.5°	26 levels	O_3 and OH	CCSM, V3 (Offline)	(Lei et al. 2014)
CTM-Hg	8°×10°	9 levels	O_3 and OH	GCM (Offline)	(Bullock Jr. et al. 2009; Vijayaraghavan et al. 2014)
TEAM	10–20 km	6 layers	O_3 and OH	NCAR-MM5 (Offline)	
AEROMOD	1 km	-	O_3 and OH	NOAA and NCDC (Offline)	

Full forms of meteorological abbreviations used in this table are provided in Table S3.

These emission scenarios have been applied in a variety of atmospheric models, which differ in spatial resolution, redox chemistry and meteorological drivers (Table 4). The oxidation of Hg^0, primarily by ozone (O_3) and hydroxyl radicals (OH), is incorporated in several models, including HYSPLIT-Hg (Cohen et al. 2016), CAM-Chem/Hg (Lei et al. 2014), CTM-Hg (Vijayaraghavan et al. 2014), GLEMOS and ECHMERIT (Pacyna et al. 2016; De Simone et al. 2017; Travnikov et al. 2017). Some models, such as GLEMOS and GEM-MACH-Hg, also incorporate Br oxidation in polar regions, although GEM-MACH-Hg mainly relies on OH chemistry (Travnikov et al. 2017). The older version of GEOS-Chem simulated Hg^0 oxidation by Br and HgII photoreduction in clouds, following the mechanism proposed by (Holmes et al. 2010). (Horowitz et al. 2017) later refined this chemistry by introducing second-stage oxidation of HgBr by HO_2 and NO_2, based on the mechanisms of (Dibble et al. 2012) and (Shah et al. 2016). More recently, (Shah et al. 2021) proposed a revised mechanism where Hg^0 oxidation is driven by both Br and OH radicals, with O_3 playing a secondary role. HgII is mainly reduced via photolysis of HgII-organic complexes and gas-phase HgI species. In addition, models employ distinct meteorological drivers, ranging from offline simulations forced by reanalysis or prescribed meteorological fields (e.g., GEOS-5, MERRA-2, ECMWF, CCSM in specified-dynamics mode, and NCEP/NCAR) (Bullock Jr. et al. 2009; Corbitt et al. 2011;

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Vijayaraghavan et al. 2014; Lei et al. 2014; Pacyna et al. 2016; Cohen et al. 2016; Travnikov et al. 2017; Angot et al. 2018) to fully online coupling with general circulation or weather prediction models (e.g., NASA GISS GCM ModelE2, ECHAM5, and GEM) (Travnikov et al. 2017; Y. Zhang et al. 2021; H. Zhang et al. 2021) (Table 4). These differences introduce additional variability in simulated atmospheric transport, oxidation, and deposition patterns. Models also rely on different anthropogenic and natural Hg emission inventories, further contributing to variability in projected deposition. Details of the inventories used in the models and the scenarios to which they are coupled are provided in Table S1, while abbreviations for the meteorological drivers are listed in Table S3.

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. The relationship is strikingly linear ($R^2 = 0.79$, slope = 0.41, $p < 0.001$), meaning that 79 % of the observed variability in Hg deposition is explained by changes in emissions. Yet the slope indicates that a 1 % change in emissions yields about a 0.4 % change in deposition, implying that a substantial fraction of emitted Hg undergoes chemical transformation and/or transport.

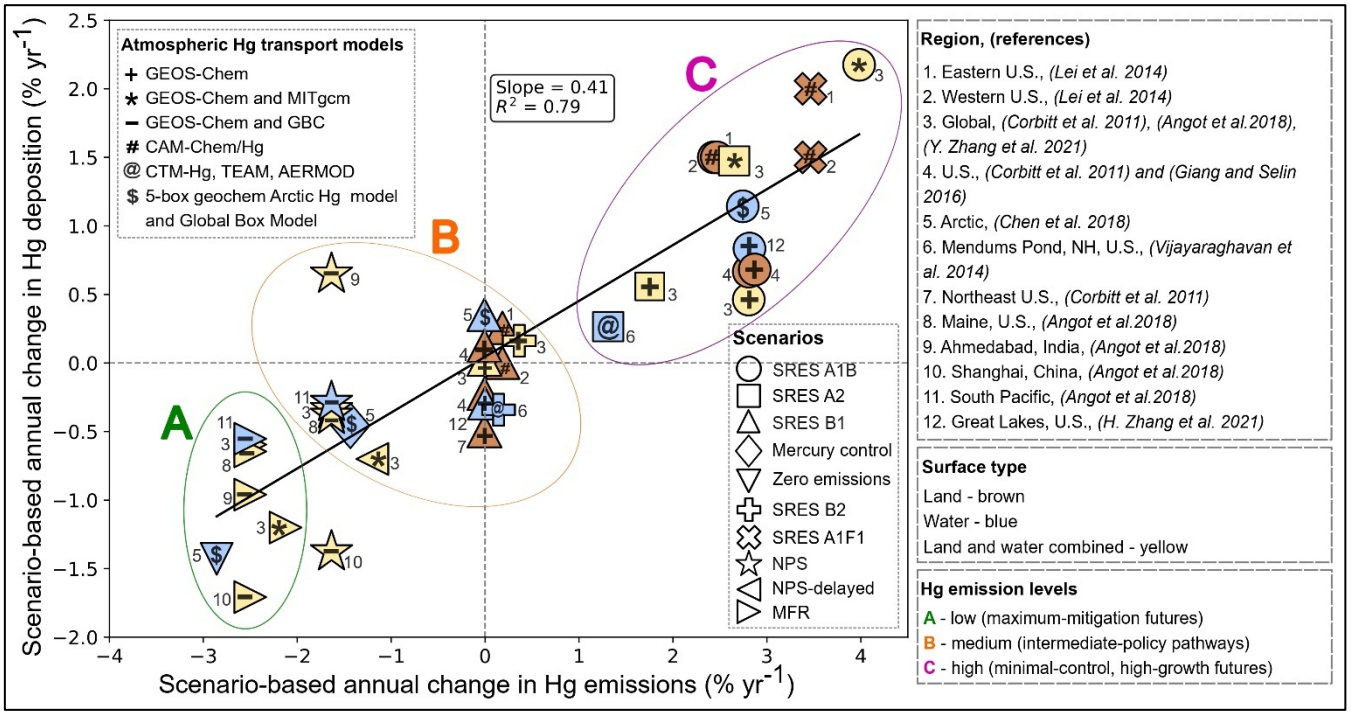

Figure 3: Relationship between scenario-based annual changes in global anthropogenic Hg emissions (% yr^{-1}) and corresponding changes in Hg deposition (% yr^{-1}) projected for 2050 across multiple models, regions, and policy scenarios. Marker shapes represent emission scenarios, while symbols indicate deposition models. Numbers correspond to regions and references (top right panel). Marker fill colors indicate surface types: land (brown), water (blue), and combined land–water (yellow).

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Ellipses indicate emission levels, with A (green) representing low emissions, B (orange) medium emissions, and C (pink) high emissions.

While these emission projections largely explain future Hg deposition trends, Figure 3 also illustrates variability in annual changes (% yr^{-1}) in Hg deposition under the same scenarios. These differences, although relatively small, arise from variations in model configurations, including redox chemistry, spatial resolution, meteorological drivers, and treatment of legacy reservoirs. For instance, under the A1B scenario, (Corbitt et al. 2011) applied a coarse-resolution GEOS-Chem configuration with a simplified one-step Br oxidation scheme and static legacy pools, resulting in only a 0.46 % yr^{-1} increase in global Hg deposition (ellipse C, region 3). In contrast, (Y. Zhang et al. 2021) projected a much stronger 2.18 % yr^{-1} increase, reflecting the use of finer spatial grids, updated anthropogenic inventories, a two-step Br oxidation pathway, and fully coupled ocean-terrestrial reservoirs. Even in the relatively stable B1 scenario, where global anthropogenic emissions remain nearly constant, deposition projections for the U.S. diverge slightly. (Corbitt et al. 2011) projected a modest decrease in deposition, while (Giang and Selin 2016) projected a slight increase (ellipse B, region 4). This contrast can be attributed to differences in model setup: (Corbitt et al. 2011) used a coarse 4° × 5° global grid with fixed meteorology and treated biomass burning as part of the legacy pool, whereas (Giang and Selin 2016) applied a nested 0.5° × 0.667° North American domain with updated emission inventories.

In addition to these differences, the treatment of historical emission inventories further contributes to uncertainty in modeled Hg deposition. While recent work by (Guerrero and Schneider 2023) challenged previous overestimates of pre-industrial emissions by (Nriagu 1993; 1994; Streets et al. 2009; 2011; 2017; Streets 2019), they demonstrated that chemically sequestered forms of Hg (e.g., calomel, vermilion) were previously omitted or underestimated in historical emission inventories. Their findings, supported by (Engstrom et al. 2014) and (Outridge et al. 2018), indicate that pre-industrial emissions may have been 2–5 times lower than earlier estimates. However, (Angot et al. 2018) demonstrated that halving historical (1850–1920 CE) mining emissions had no significant impact on the projected benefits of delayed policy action, reinforcing the finding that atmospheric deposition is most sensitive to recent anthropogenic emissions. (Médieu et al. 2024) also emphasized the rapid responsiveness of the atmospheric Hg reservoir to emission changes, in contrast to the slower response of legacy reservoirs like the ocean.

Taken together, these results indicate that contemporary anthropogenic emissions overwhelmingly drive global Hg deposition in the model-simulated world, with diverse modeling frameworks converging on this conclusion despite differences in atmospheric chemistry, spatial resolution, treatment of legacy reservoirs, meteorological drivers, and uncertainties in historical emission inventories. However, an important question remains as to whether real-world systems respond as linearly as suggested by current models.

Model–observation inconsistencies are evident at both national and regional scales. Recent observational analyses by (Feng et al. 2024) show that ground-level GEM concentrations in China declined substantially faster than predicted by earlier model-based assessments. While (Liu et al. 2019) projected an 11 % decrease in GEM between 2013 and 2017, long-term observations indicate an approximately 19 % reduction over the same period, with continued declines of 1.8–6.1 %

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yr^{-1} through 2022 (Feng et al. 2024). Similar discrepancies emerge at the regional scale. Chongqing, a highly industrialized and coal-dependent megacity, exhibits a pronounced 7 % yr^{-1} decline in Hg deposition between 2010 and 2021 (Zhan et al. 2025), far exceeding the 1.4–1.7 % yr^{-1} decline projected for Shanghai under the NPS and MFR scenarios for 2010–2050 (Angot et al. 2018) (Figure 3, ellipse A). Chongqing (inland) and Shanghai (coastal) differ in geographic setting and meteorology yet both serve as key indicators of China's emission control effectiveness. Declines in ambient GEM at Chongming Island in Shanghai (Tang et al. 2018) and in Hg deposition in Chongqing (Zhan et al. 2025) provide independent evidence of rapid environmental responses to emission reductions. Although these indicators are not physically equivalent, their consistent downward trends, highlighted by a national 38.6 ± 12.7 % reduction in GEM between 2013 and 2022, particularly in eastern and southwestern China—underscore the strong sensitivity of regional Hg responses to emission controls and suggest that observational records may capture steeper declines than those projected by current scenario-oriented models (Feng et al. 2024).

Model-observation discrepancies are also apparent at the hemispheric scale. In the Southern Hemisphere, Hg emissions under these same scenarios (NPS and MFR) are projected to decline by 0.4–2.3 % yr^{-1} (2010–2035) (Rafaj et al. 2013). Yet GEOS-Chem and MITgcm overestimate wet deposition (15–20 μg m^{-2} yr^{-1} vs. observed 1–5 μg m^{-2} yr^{-1}; approximately 5-fold higher) and underestimate dry deposition (0.5–5 μg m^{-2} yr^{-1} vs. observed 10 μg m^{-2} yr^{-1}; approximately 3.5-fold lower), largely due to simplified treatments of convective uplift and precipitation scavenging (Leiva González et al. 2022).

Together, these discrepancies highlight the need for more routine and systematic comparisons between modeled and observed deposition trends under evolving policy conditions. Strengthening long-term monitoring networks and further harmonizing emission inventories will be essential to validate model predictions and to assess the real-world effectiveness of Hg control measures under the Minamata Convention.

3.2 Ecosystem deposition vs MeHg in fish, 2050

Atmospheric deposition plays a pivotal role in biogeochemical cycling and fate of Hg in aquatic ecosystems. Studies have shown that atmospherically supplied Hg can rapidly be converted to MeHg in lake ecosystems and enter the base of the food chain, elevating fish MeHg levels within a short period (Harris et al. 2007; Ogorek et al. 2021). Understanding the relationship between atmospheric Hg deposition and MeHg concentrations in fish is essential for predicting future risks under varying emission scenarios, especially in the context of evolving policies.

To estimate MeHg concentrations in fish under future deposition scenarios, researchers have used multiple bioaccumulation models, including SERAFM, Hendrick's Lake Mass Balance Model, D-MCM, BASS, and WASP, as described below.

(a) **SERAFM**: Spreadsheet-based Ecological Risk Assessment for the Fate of Hg model operates under steady-state conditions, estimating Hg concentrations in fish using bioaccumulation factors (BAFs). It employs a three-box system (epilimnion, hypolimnion, and sediments) to model Hg dynamics, incorporating atmospheric deposition of Hg0, MeHg, and HgII with same wet and dry deposition fluxes for lakes and watersheds. However, it does not account for Hg accumulation during ice cover and subsequent release during spring melt. Watershed inputs rely on constant runoff coefficients without

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seasonal variability, and erosion is modeled using the Revised Universal Soil Loss Equation (RUSLE). Seasonal changes in temperature, stratification, ice cover, and DOC inputs are not simulated, and rate constants for oxidation-reduction, photodegradation, and other processes remain fixed throughout the simulation (Knightes 2008; Knightes et al. 2009).

(b) **Hendrick's Lake Mass Balance Model**: A modified version of SERAFM, operates under non-steady-state conditions while still using BAFs for fish Hg estimation (Perlinger et al. 2018). It also employs a three-box reservoir system but distinguishes deposition between lakes and watersheds, incorporating higher friction velocities in watersheds (Angot et al. 2018). Unlike SERAFM, it accounts for accumulated deposition during ice cover and its release in spring melt. Seasonal dynamics in temperature, stratification, and ice cover are simulated, and DOC inputs follow a sine function to capture seasonal fluctuations. Rate constants for redox reactions, methylation, demethylation, and photodemethylation adjust dynamically based on lake temperature and light attenuation, while thermocline dispersion rates vary seasonally to reflect mixing and stratification (Hendricks 2018).

(c) **D-MCM** – The Dynamic Mercury Cycling Mode predicts time-dependent concentrations of Hg0, MeHg, and HgII across the water column, sediments, and a six-level food web, including phytoplankton, zooplankton, benthos, and three fish trophic levels (Zhu et al. 2018). It models dry and wet atmospheric deposition of HgII and MeHg, while incorporating processes such as adsorption, desorption, particulate settling, sediment-water exchange, resuspension, burial, air-water gaseous exchange, and in-situ transformations in lakes (Massoudieh et al. 2010). Bioenergetics influence MeHg accumulation by linking fish metabolism, feeding, and growth to Hg uptake and retention. In the model, higher activity costs or lower growth efficiency result in higher MeHg concentrations, highlighting how fish allocate consumed energy between growth and activity and its effect on Hg burden (Harris and Bodaly 1998; Trudel and Rasmussen 2006). However, like SERAFM, D-MCM does not account for Hg accumulation during ice cover and its subsequent release. Watershed inputs rely on time series data of precipitation and Hg concentrations, with constant MeHg assumptions in precipitation. Surface flow rates are modeled using constant runoff coefficients (Vijayaraghavan et al. 2014).

(d) **BASS** – The Bioaccumulation and Aquatic System Simulator explicitly models Hg uptake in fish through dietary ingestion and gill exchange, rather than relying on BAFs. Hg elimination occurs via metabolism and growth dilution (Barber 2008). BASS incorporates age-structured fish populations, covering both small, short-lived species and large, long-lived species, while allowing simulation of diverse fish communities based on species-specific physiological and ecological parameters, including dietary composition and growth patterns. It also models food web interactions by simulating biomass changes in phytoplankton, zooplankton, and benthos due to consumption, respiration, and mortality (Barber 2018). However, BASS does not simulate external Hg inputs, such as those from atmospheric deposition, watershed runoff, or upstream river inflows. Instead, it requires the user to provide initial Hg concentrations in water and biotic compartments (e.g., plankton, benthos) (Knightes et al. 2009; Barber 2018). These concentrations are typically estimated from field measurements or generated by external models such as the Watershed Characterization System Hg Loading Model (WCS-MLM) (Greenfield et al. 2002) and SERAFM. Additionally, BASS lacks capabilities for modeling Hg deposition, transport, transformation, and fate within aquatic systems, requiring integration with models such as SERAFM and WASP (Knightes et al. 2009).

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(e) **WASP** – The Water Analysis Simulation Program (WASP) supports both steady-state and non-steady-state simulations, utilizing a four-layer system that includes the water column, mixed sediment, methylation layer, and deep sediments (Vuksanovic et al. 1996). The model receives atmospheric deposition inputs of Hg0, HgII, and MeHg but focuses solely on in-waterbody processes, relying on external models for watershed Hg fluxes (Lindenschmidt 2006). Hg transformations in WASP are compartment-specific: methylation occurs primarily in the anaerobic sediment methylation layer, while demethylation, oxidation, and reduction occur in both the surface water and in the aerobic mixed sediment layer (Gidley et al. 2017). WASP does not simulate sediment resuspension processes directly; these must be estimated separately. Seasonal drivers like temperature variation, stratification, and ice cover are not incorporated into the model (Knightes et al. 2009).

To assess how changes in atmospheric Hg deposition influence fish MeHg concentrations, we analyzed the relationship between annual changes (% yr^{-1}) in atmospheric Hg deposition and corresponding changes (% yr^{-1}) in fish MeHg projected for U.S. ecosystems by 2050 (Figure 4). Each point integrates atmospheric and bioaccumulation models, fish trophic levels, sites, and emission scenario-based deposition. Despite the diversity of atmospheric models, bioaccumulation frameworks, fish species, and ecosystem types represented, the relationship is strongly linear ($R^2 = 0.63$, slope = 0.87, $p < 0.01$), indicating that roughly 63 % of the variance in fish MeHg is explained by atmospheric deposition. The slope shows that every 1 % change in deposition yields an approximately 0.87 % change in fish MeHg, a far steeper response than the 0.4 slope found for emissions versus deposition (Figure 3). This suggests that fish incorporate nearly all deposited Hg into food webs in the model-simulated world. However, the correlation is weaker than that observed between emissions and deposition (Sect. 3.1), reflecting the added complexity of lake-specific characteristics and ecological factors. For example, under the intermediate Policy-in-action scenario with moderate deposition reductions (Ellipse A), two integrated data points exhibit markedly different annual changes in fish MeHg, suggesting strong landscape sensitivity, with watershed characteristics, particularly wetland percentage and forest runoff, exerting dominant control on fish MeHg compared to direct atmospheric deposition (Perlinger et al. 2018). Similarly, under the high deposition reduction scenario involving a 50 % decrease in Hg deposition (Ellipse B), five integrated data points show substantial variability in fish MeHg responses under the same policy scenario. Lakes with small watersheds and high direct atmospheric exposure (e.g., drainage and seepage lakes) respond rapidly, whereas larger or stratified systems with extensive watersheds (e.g., farm ponds, coastal-plain rivers, stratified lakes) exhibit slower declines in fish MeHg (Knightes et al. 2009). Together, these findings highlight that while deposition strongly governs fish MeHg, lake characteristics can result in site-specific variability, which we elaborate further in Sect. 3.3.

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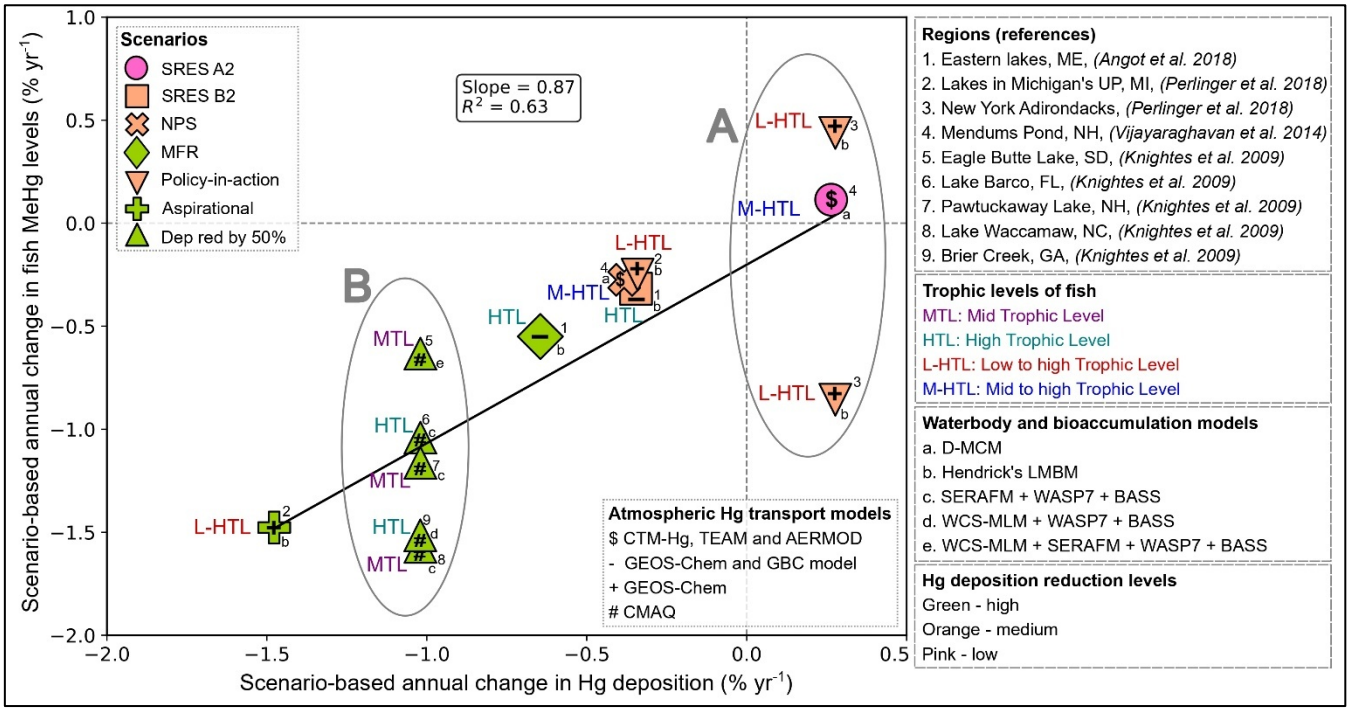

Figure 4: Relationship between scenario-based projected annual changes in atmospheric Hg deposition (% yr⁻¹) and corresponding changes in fish MeHg concentrations (% yr⁻¹) by 2050. Marker shapes (top left panel) denote Hg deposition scenarios, while special characters (bottom right panel) represent atmospheric Hg deposition models. Numbers shown in the top right panel correspond to study regions and references. Fish trophic levels are color-coded in the lower panel, and the bottom right panel also indicates waterbody and watershed type together with the bioaccumulation models used. Marker fill colors indicate the magnitude of Hg deposition reduction, classified as high (green), medium (orange), and low (pink).

Apart from the lake-specific characteristics, additional ecological and climate-related factors can modulate fish MeHg levels. These have been evident through many individual studies. In particular, the changes in fish MeHg concentrations documented at various lakes in Michigan's Upper Peninsula (UP), U.S. provides a key case study. Figure 5 compares modeled and observed changes in lake trout MeHg, revealing notable discrepancies across different time periods.

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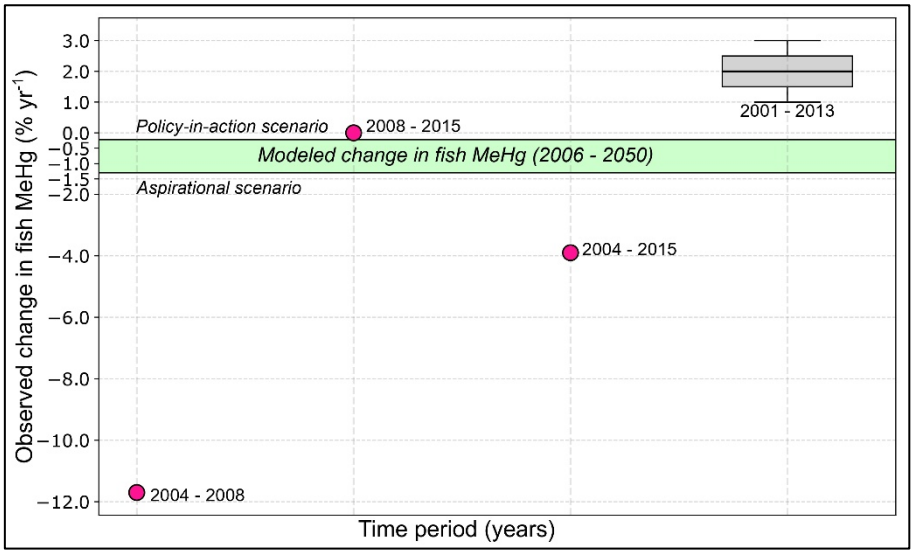

Figure 5: Observed annual changes in lake trout MeHg in lakes of Michigan's UP, U.S. (Zhou et al. 2017; Kerfoot et al. 2018). The green shaded bar indicates the range of modeled results under two policy scenarios simulated by (Perlinger et al. 2018).

Model simulations for lakes in Michigan's UP estimated modest annual fish MeHg declines (-0.25 to -1.48 % yr^{-1}) during 2006–2050 under policy scenarios (Perlinger et al. 2018), whereas, observed lake trout data from lakes in Michigan's UP showed a sharper decline of 3.9 % yr^{-1} (2004–2015) (Zhou et al. 2017). The reduction was largely driven by a steep drop in fish MeHg before 2008 (-11.7 % yr^{-1}), followed by a subsequent plateau, which was attributed to climate-relevant changes in food web structure and amount of wet Hg deposition (Zhou et al. 2017). More recent work by (Lepak et al. 2025) also reported counterintuitive increases in fish MeHg despite declining local-regional Hg emissions and atmospheric deposition, attributing them to invasive dreissenid mussels that redirected trout toward lower-calorie, benthic prey, coupled with reduced lake productivity and slower fish growth. Stable isotope analyses of Hg (Δ^{199}Hg), together with carbon and nitrogen isotopes (δ^{13}C, δ^{15}N), confirmed these dietary and energy-flow changes (Lepak et al. 2019; 2025). Similarly, (Kerfoot et al. 2018) observed rising fish MeHg (1–3 % yr^{-1}, 2001–2013) despite an 81 % regional emission decline, driven by wetland recovery and renewed methylation of legacy Hg.

Despite these ecological complexities, isotopic evidence provides further insight into system responsiveness. Stable-isotope evidence from the Great Lakes indicates that fish Hg isotopic signatures respond more rapidly to changes in atmospheric Hg concentrations than sedimentary records, suggesting that fish act as sensitive biosentinels of recent deposition trends (Lepak et al. 2025). Long-term whole-ecosystem isotope-labeled experiments in a boreal lake support this view, showing that reducing direct atmospheric inputs drives rapid declines in fish MeHg through food webs, even if watershed Hg is retained and mobilized more slowly (Blanchfield et al. 2022). Yet interacting stressors, such as invasive species, climate change, and enhanced wetland inflows, can amplify methylation and delay Hg recovery (Hall et al. 2023; Thompson et al. 2023; Rodríguez 2023; McCarter et al. 2024; Lepak et al. 2025; Olson et al. 2025).

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Collectively, these findings highlight the constraints of assuming that fish MeHg levels will decline in direct proportion to atmospheric deposition. They raise an important question for future assessments: do lake ecosystems truly follow linear dynamics, or do current models oversimplify the complex ecological, biogeochemical, and climate-driven interactions that regulate Hg uptake and bioaccumulation in natural ecosystems?

3.3 Lake characteristics vs MeHg in fish, 2050

This section examines how lake-specific characteristics are represented across models when simulating future fish MeHg levels. To evaluate how these characteristics are treated within the model-simulated framework, we incorporated key environmental predictors used in the same studies analyzed in Sect. 3.2. These include lake area, watershed area, wetland percentage, and average depth, together with scenario-specific changes in Hg deposition as well as the corresponding changes in fish MeHg, into our statistical analyses (Knightes et al. 2009; Vijayaraghavan et al. 2014; Angot et al. 2018; Perlinger et al. 2018). The full list of used variables is provided in Table S4. Extensive empirical work has shown that fish MeHg levels respond to lake morphometry (area, depth, wetland extent) (Bodaly et al. 1993; Choy et al. 2009; Ackerman et al. 2019; Knott et al. 2020), watershed size and composition (Sonesten 2003; Evans et al. 2005; Rypel 2010; Marusczak et al. 2011; Eagles-Smith et al. 2016; Backstrom et al. 2020), water chemistry (pH, hardness, alkalinity) (McMurtry et al. 1989; Backstrom et al. 2020), hydrologic conditions (drainage pattern) (Grieb et al. 1990), geographical gradient (Morel et al. 1998), seasonal variability (Barletta et al. 2012; Keva et al. 2017; Mills et al. 2018), and fish-specific traits (age, size, trophic level) (Cizdziel et al. 2002; Evans et al. 2005; Marusczak et al. 2011; Backstrom et al. 2020). Together, these studies illustrate that multiple environmental processes regulate MeHg bioaccumulation, providing a basis for examining how Hg bioaccumulation models prioritize these same factors.

We first evaluated whether lake characteristics alone could explain variability in projected changes in fish MeHg across models using multiple linear regression (MLRM). No strong relationships were identified (Table S5). When scenario-specific changes in Hg deposition were incorporated, as described in Sect. 3.2, only deposition change ($p = 0.007$) and lake area ($p = 0.04$) emerged as significant predictors, while other variables contributed weakly (Table S6). These results motivated subsequent PCA analyses to examine how deposition and lake characteristics interact collectively within Hg bioaccumulation model simulations.

PCA was conducted within a scenario-based modeling framework consistent with Sect. 3.2, using lake area, watershed area, wetland percentage, average depth, and projected changes in Hg deposition as explanatory variables, with projected changes in fish MeHg included as a supplementary variable. The resulting biplot (Figure 6) summarizes dominant patterns of covariation, with the first two principal components explaining 70.82 % of the total variance (PC1 = 38.13 %, PC2 = 32.69 %). PC1 was primarily defined by lake area and wetland percentage, whereas PC2 was driven by average depth and Hg deposition change, with a moderate contribution from watershed area. Consistent with this structure, a MLRM using PC1 and PC2 explained a substantial portion of the variability in the simulated response variable (Change in fish MeHg) and was statistically significant ($R^2 = 0.57$; $F = 6.74$, $p = 0.01$). PC1 exhibited a significant negative association ($\beta = -0.26$, $p =$

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0.02), indicating that systems with larger lake area and higher wetland coverage tend to show greater declines in fish MeHg in the model simulations. In contrast, PC2 showed a significant positive relationship ($\beta = 0.24$, $p = 0.04$). This component reflects a trade-off between atmospheric Hg loading and lake depth versus watershed influence (Figure 6), such that the bioaccumulation models predict the greatest simulated MeHg increases (or smallest reductions) in deep, atmospherically impacted systems with relatively small contributing catchments. The proximity of the fish MeHg vector in Figure 6 confirms that modeled future fish MeHg responses are structured along gradients of lake morphometry and atmospheric Hg loading.

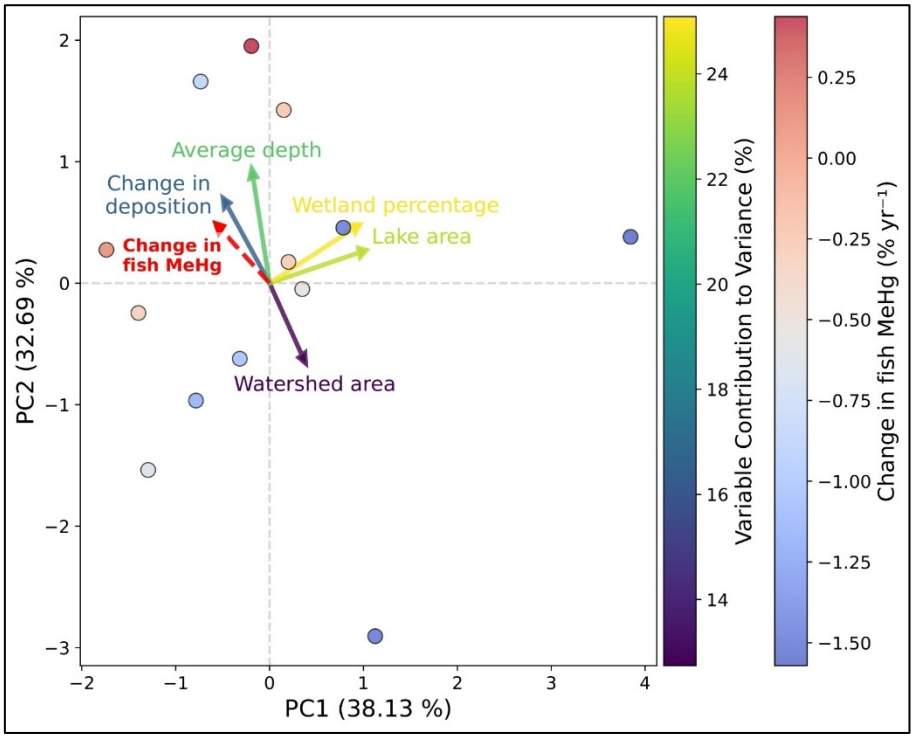

Figure 6: PCA biplot of environmental predictors and scenario-based Hg deposition. Points represent individual model realizations positioned by their PC1 and PC2 scores and colored by projected change in fish MeHg (% yr^{-1}). Solid arrows show predictor loadings (length and color indicate contribution to variance), and the dashed red arrow represents the supplementary projection of fish MeHg.

Collectively, results from the MLRM and PCA analyses indicate that, within the modeling framework, scenario-specific changes in atmospheric Hg deposition consistently emerge as the dominant predictor of future fish MeHg responses, with lake area exerting a secondary influence and wetland extent and average depth contributing primarily through multivariate structure in the PCA. Across these statistical approaches, deposition change explains the largest share of variance in modeled fish MeHg, reinforcing the central role of Hg-emission policies in shaping projected bioaccumulation outcomes in the model-simulated world. This finding mirrors the pattern of whole-ecosystem experimental studies showing declines in fish MeHg following reductions in Hg deposition (Blanchfield et al. 2022). However, comparisons between modeled projections and long-term observational records reveal a more complex reality. Case studies from lakes in Michigan's UP demonstrate that

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fish MeHg concentrations may increase, decrease, or respond nonlinearly despite substantial declines in atmospheric Hg deposition, reflecting the influence of ecological and biogeochemical processes not fully resolved by current models. These include food-web restructuring, invasive species, productivity shifts, wetland-mediated methylation, internal Hg cycling, and climate-driven changes in hydrology and redox conditions. Moreover, the negative association between wetland extent and projected fish MeHg observed here in the PCA differs from many field studies, which commonly report elevated 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). Similarly, (Perlinger et al. 2018) found that under a Policy-in-action scenario, wetland extent and DOC exerted stronger controls on fish MeHg outcomes than deposition alone. Together, these contrasts underscore that the dominant predictors identified here primarily reflect the processes emphasized within model structures, rather than a complete representation of the mechanisms governing MeHg dynamics in natural ecosystems. 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.

4 Conclusion

In summary, despite substantial differences among atmospheric models in spatial resolution, emission inventories, meteorological drivers, and redox chemistry, and despite variability in bioaccumulation models and ecosystem complexity, our study shows that they consistently predict strong linear relationships between emissions, deposition, and fish MeHg. Within the model world, deposition change and lake morphometry emerge as dominant predictors of future fish MeHg responses.

Yet comparisons with observed data reveal important departures from these idealized trends across both steps of the causal chain: emissions → deposition, and deposition → fish MeHg. These comparisons show that while models generally capture the direction of change under emission controls, they substantially underrepresent the ecological, biogeochemical, and regional heterogeneity that governs how rapidly deposition and fish MeHg respond in real ecosystems. Empirical evidence shows that fish MeHg responses can be nonlinear, delayed, or even decoupled from declining deposition due to interacting ecological and biogeochemical processes, including food-web restructuring, invasive species, wetland-mediated methylation, internal Hg cycling, and climate-driven changes in productivity and hydrology. These findings underscore the need to pair global modeling with long-term atmospheric, deposition, and ecological measurements, and to improve representation of watershed processes, food-web dynamics, and climate feedbacks to more accurately predict the benefits of emission reductions under the Minamata Convention.

Thus, although current models are powerful and indispensable tools, their orderly linearity invites reflection: do we truly capture the intricate feedbacks of the real atmosphere and natural ecosystems? To strengthen future assessments and policy strategies, we should enhance model complexity by incorporating nonlinear processes and bridge gaps with observations

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through prioritized field studies and long-term monitoring. By doing so, we can better validate projections, uncover unaccounted-for interactions, and ensure models evolve in step with the complexity of Hg cycling.

Code, data, or code and data availability

Custom scripts used for statistical analyses are available from the corresponding author upon reasonable request. All data analysed in this study are available from the cited publications. Any compiled datasets are available from the corresponding author upon reasonable request.

Supplement link

The link to the supplement will be included by Copernicus.

Author contributions

Henna Gull compiled and curated the data, conducted the analyses, and led the writing of the manuscript. Hoin Lee cross-checked and validated the compiled data. Ju Hyeon Lee provided critical feedback and contributed to manuscript revision. Hélène Angot provided scientific feedback and suggestions that helped improve the manuscript. Sae Yun Kwon supervised the study and contributed to conceptualization and manuscript revision. All authors reviewed and approved the final manuscript.

Competing interests

The authors declare that they have no conflict of interest.

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Review statement

The review statement will be added by Copernicus Publications listing the handling editor as well as all contributing referees according to their status anonymous or identified.

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