



*Supplement of*

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

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**Section S1.** Percentage annual change for each variable (X = Hg emissions, Hg deposition, or fish MeHg) was calculated as:

$$\% \text{ annual change in } X = \frac{\left( \frac{X_{2050, \text{scenario}}}{X_{\text{base year, scenario}}} - 1 \right)}{n} * 100$$

5 Where:

$X_{2050, \text{scenario}}$  = modeled value under a given scenario in 2050

$X_{\text{base year, scenario}}$  = modeled value in the base year used in that study

$n$  = number of years between the base year and future year

10 **Table S1.** Anthropogenic and natural mercury emission inventories applied in atmospheric Models.

| <b>References (models)</b>   | <b>Anthropogenic</b>  | <b>Natural</b>   |
|--|---|--|
| <b>↓ Scenarios</b>   |   |  |
| (Corbitt et al. 2011) (Geos-Chem)<br>A1B, A2, B1, and B2   | (Pacyna et al. 2010) – base year emissions<br>(Streets et al. 2009) – future emission scenarios   | GEOS-Chem parameterization – geogenic/ legacy emissions  |
| (Lei et al. 2014) (CAM-Chem/Hg)<br>A1B, B1, and A1F1   | (Pacyna et al. 2006) – base year emissions<br>(Streets et al. 2009) – future emission scenarios   | (Lei et al. 2013) – natural/legacy emissions<br>(Witt et al. 2008; Aiuppa et al. 2007) – volcanic emissions<br>(IPCC 2001) – forest & grassland burning<br>(Smith-Downey et al. 2010) – terrestrial Hg storage |
| (Vijayaraghavan et al. 2014) (CTM)<br>A2 and B2  | (EPRI 2006) – base year emissions<br>(Streets et al. 2009) – future emission scenarios  | (Seigneur et al. 2004) – natural/legacy re-emissions   |
| (Giang and Selin 2016) (Geos-Chem)<br>No policy and Minamata   | (Pacyna et al. 2006) – base year emissions<br>(Streets et al. 2009) – future emissions  | GEOS-Chem parameterization   |
| (Pacyna et al. 2016) (GLEMOS and ECHMERIT)<br>CPS, NPS, and MFR  | (AMAP/UNEP, 2013) – baseline emissions<br>(Streets et al. 2009; 2011; 2017) – historical and future scenario emissions<br>(Pacyna et al. 2006; 2010) – earlier global anthropogenic inventories | (Pirrone et al. 2010) – volcanic / geogenic emissions<br>(Smith-Downey et al. 2010) – terrestrial storage and natural re-emissions   |
| (Angot et al. 2018) (Geos-Chem)<br>NPS and MFR   | (AMAP/UNEP, 2013) – base year emissions<br><br>(Streets et al. 2011) – historical emissions (2000 BCE–2008 CE)<br>(Pacyna et al. 2016) – future scenarios                                       | (Pirrone et al. 2010; Bagnato et al. 2011) – geogenic emissions  |
| (Perlanger et al. 2018) (Geos-Chem)<br>Minimal regulation, Policy-in-action, and aspirational scenario | (AMAP/UNEP, 2013) – baseline emissions<br>(Streets et al. 2009; 2011; 2017) – historical and future scenario emissions  | (Pirrone et al. 2010; Bagnato et al. 2011) – geogenic<br>GEOS-Chem parameterization – natural/legacy emissions   |

|   |  |  |
|---|--|--|
| (Chen et al. 2018) (Geos-Chem)<br>A1B, constant emissions, Hg control, and zero emissions | (Streets et al. 2011) – emissions (2000 BCE–2008 CE)<br>(Horowitz et al. 2014) – commercial-product emissions (1850–2008)<br>AMAP/UNEP (2018) – baseline emissions<br>(Streets et al. 2011) – emissions (1850–1970) by region<br>(Streets et al. 2009; Amos et al. 2013) – future scenario emissions | (Pirrone et al. 2010; Bagnato et al. 2011) – geogenic emissions  |
| (Y. Zhang et al. 2021) (Geos-Chem)<br>A1B, NPS-delayed, and MFR                           | AMAP/UNEP (2013) – base year emissions<br>(Streets et al. 2009; Pacyna et al. 2016) – future scenario emissions  | Default GEOS-Chem datasets   |
| (H. Zhang et al. 2021) (Geos-Chem)<br>A1B, B1   | (Pacyna et al. 2010; Zhang et al. 2012) – base year emissions<br>(Streets et al. 2009; Corbitt et al. 2011) – future scenario emissions  | (Mason 2009; Bagnato et al. 2011) – geogenic emissions<br>(Holmes et al. 2010) – biomass burning and soil evasion<br>(Selin et al. 2008) – prompt terrestrial re-emission<br>(Holmes et al. 2010; Zhang et al. 2016) – ocean evasion |

**Table S2.** Acronyms and descriptions for climate-oriented and hybrid mercury emission scenarios.

| <b>Scenarios</b>             | <b>Descriptions</b>   |
|------------------------------|---|
| BAS<br>(Baseline)            | Assumes continuation of air-quality and climate policies in force up to 2012, without introducing additional mercury or emission-control measures beyond 2030.                                    |
| CLIM<br>(Climate Mitigation) | Incorporates a 2 °C global climate policy, achieving substantial reductions in fossil-fuel use and associated mercury emissions by 2050.  |
| CLIM+MFR                     | Climate mitigation scenario with maximum feasible mercury reduction measures, representing the lowest achievable global Hg emissions.   |
| Net-Zero                     | Net-zero energy pathway (< 1.5 °C by 2100) combined with current-legislation Hg and air-quality policies.   |
| Net-Zero + Hg MFR            | Combines net-zero climate policy with maximum Hg-specific controls, yielding the lowest feasible Hg emissions by 2050.  |
| SSP1-2.6                     | A lower-bound emissions scenario reflecting strong emphasis on sustainability and intensive control of climate-forcing agents.  |
| SSP2-4.5                     | A middle-of-the-road scenario reflecting moderate socioeconomic development and continuation of current trends without strong climate mitigation.   |
| SSP5-3.4                     | An overshoot scenario characterized by short-term growth in fossil fuel use and minimal consideration of climate control measures until 2040 followed by aggressive mitigation after mid-century. |
| SSP5-8.5                     | An upper-bound emissions scenario in which fossil-fuel use continues with little consideration of climate mitigation or transition to clean technologies.   |

**Table S3.** Full forms of abbreviations used for meteorological datasets and drivers in atmospheric Hg models.

| <b>Acronym / Model</b> | <b>Descriptions</b>  |
|------------------------|--|
| GEOS                   | Goddard Earth Observing System   |
| ECHAM5                 | The fifth-generation atmospheric general circulation model developed at the Max Planck Institute for Meteorology in Hamburg, based on the operational forecast model framework from ECMWF. |
| WRF                    | Weather Research and Forecasting   |
| ECMWF                  | European Centre for Medium-Range Weather Forecasts   |
| NCEP                   | National Centers for Environmental Prediction  |
| NCAR                   | National Center for Atmospheric Research   |
| ECCC                   | Environment and Climate Change Canada's  |
| GEM                    | Global Environmental Multiscale  |
| GCM                    | General circulation model  |
| CCSM                   | Community Climate System Model   |
| NCAR-MM                | National Center for Atmospheric Research - Mesoscale Model   |
| NOAA                   | National Oceanic and Atmospheric Administration  |
| NCDC                   | National Climatic Data Center  |
| MERRA-2                | Modern-Era Retrospective analysis for Research and Applications, Version 2 by NASA's Global Modeling and Assimilation Office (GMAO)  |
| GISS-GCM               | Goddard Institute for Space Studies - general circulation model  |

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**Table S4.** Summary of variables used in PCA and MLRM analyses.

| <b>Scenarios</b> | <b>Change in fish MeHg (% yr<sup>-1</sup>)</b> | <b>Change of Hg deposition (% yr<sup>-1</sup>)</b> | <b>Watershed (km<sup>2</sup>)</b> | <b>Lake Area (km<sup>2</sup>)</b> | <b>Wetland (%)</b> | <b>Average depth (m)</b> | <b>Waterbodies</b>         |
|------------------|--|--|-----------------------------------|-----------------------------------|--------------------|--------------------------|----------------------------|
| SRES A2          | 0.114  | 0.264  | 17.040                            | 1.070                             | 4.030              | 6.400                    | Mendums Pond, NH           |
| SRES B2          | -0.318   | -0.346   | 17.040                            | 1.070                             | 4.030              | 6.400                    | Mendums Pond, NH           |
| NPS              | -0.275   | -0.383   | 183.100                           | 7.148                             | 14.285             | 5.570                    | Lakes in Maine, ME         |
| MFR              | -0.550   | -0.645   | 183.100                           | 7.148                             | 14.285             | 5.570                    | Lakes in Maine, ME         |
| Policy-in-action | -0.250   | -0.341   | 190.000                           | 9.700                             | 14.210             | 15.000                   | Lakes in Michigan's UP, MI |

|                               |        |        |          |        |        |        |   |
|-------------------------------|--------|--------|----------|--------|--------|--------|---|
| Aspirational                  | -1.477 | -1.477 | 190.000  | 9.700  | 14.210 | 15.000 | Lakes in Michigan's UP, MI  |
| Policy-in-action              | 0.438  | 0.277  | 190.000  | 9.700  | 14.210 | 15.000 | Lake in Michigan's UP with Adirondack's deposition                        |
| Policy-in-action              | -0.857 | 0.277  | 190.000  | 9.700  | 8.800  | 15.000 | Lake in Michigan's UP with Adirondack's deposition and watershed features |
| Reduction in deposition, 50 % | -0.633 | -1.020 | 4.200    | 0.200  | 1.000  | 2.000  | Eagle Butte Lake, SD  |
| Reduction in deposition, 50 % | -1.041 | -1.020 | 0.810    | 0.120  | -      | 4.800  | Lake Barco, FL  |
| Reduction in deposition, 50 % | -1.163 | -1.020 | 50.000   | 3.600  | 4.000  | 5.000  | Pawtuckaway Lake, NH  |
| Reduction in deposition, 50 % | -1.571 | -1.020 | 220.000  | 35.000 | 25.000 | 2.300  | Lake Waccamaw, NC   |
| Reduction in deposition, 50 % | -1.510 | -1.020 | 2190.000 | 9.900  | 8.000  | 0.250  | Brier Creek, GA   |

Note: For NPS and MFR, "Lakes in Maine" reflect the averaged characteristics of 20 modeled lakes.

**Table S5.** MLRM results testing whether lake characteristics alone predict changes in fish MeHg by 2050 in the model world.

|              | <b>Coef</b> | <b>std err</b> | <b>t</b> | <b>P&gt; t </b> | <b>[0.025</b> | <b>0.975]</b> |
|--------------|-------------|----------------|----------|-----------------|---------------|---------------|
| const        | -0.6521     | 0.432          | -1.509   | 0.175           | -1.674        | 0.370         |
| Watershed    | -0.0003     | 0.000          | -0.924   | 0.386           | -0.001        | 0.000         |
| LakeArea     | -0.0687     | 0.045          | -1.526   | 0.171           | -0.175        | 0.038         |
| Wetland      | 0.0582      | 0.061          | 0.947    | 0.375           | -0.087        | 0.203         |
| AverageDepth | 0.0072      | 0.039          | 0.187    | 0.857           | -0.084        | 0.099         |

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**Table S6.** MLRM results testing lake characteristics together with scenario-specific Hg deposition as predictors of changes in fish MeHg by 2050 in the model world.

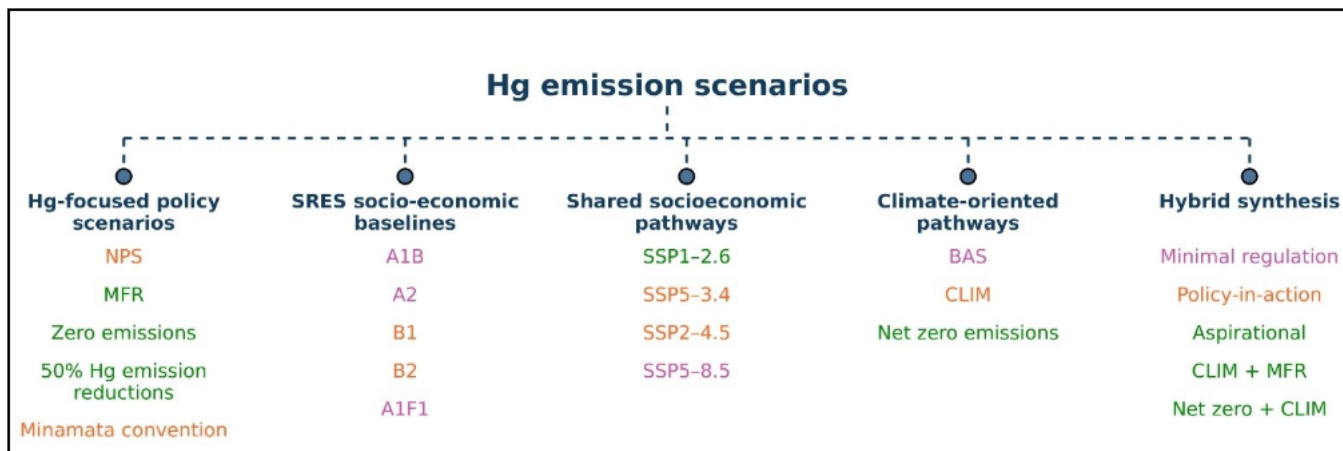
|              | <b>Coef</b> | <b>std err</b> | <b>t</b> | <b>P&gt; t </b> | <b>[0.025</b> | <b>0.975]</b> |
|--------------|-------------|----------------|----------|-----------------|---------------|---------------|
| const        | -0.1249     | 0.275          | -0.454   | 0.666           | -0.799        | 0.549         |
| Watershed    | -0.0002     | 0.000          | -1.219   | 0.269           | -0.001        | 0.000         |
| LakeArea     | -0.0656     | 0.025          | -2.599   | 0.041           | -0.127        | -0.004        |
| Wetland      | 0.0685      | 0.035          | 1.982    | 0.095           | -0.016        | 0.153         |
| AverageDepth | -0.0264     | 0.023          | -1.136   | 0.299           | -0.083        | 0.030         |
| Deposition   | 0.7864      | 0.195          | 4.032    | 0.007           | 0.309         | 1.264         |

25 **Table S7.** Characteristics of atmospheric Hg models, including resolution, primary oxidation pathways, and meteorological drivers.

| <b>Model</b> | <b>Spatial resolution</b> | <b>Vertical resolution</b> | <b>Primary Hg<sup>0</sup> oxidation pathways</b>  | <b>Meteorological data sources (type)</b>                               | <b>References</b>  |
|--------------|---------------------------|----------------------------|---|---|--|
| 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 (1 <sup>st</sup> - stage)<br><br>NO <sub>2</sub> and HO <sub>2</sub> (2 <sup>nd</sup> - stage) | MERRA-2 by NASA's GMAO (Offline);<br><br>NASA GISS GCM ModelE2 (Online) | (Angot et al. 2018; Y. Zhang et al. 2021; H. Zhang et al. 2021)    |
| GLEMOS       | 1°×1°                     | 20 levels                  | O <sub>3</sub> and OH<br><br>Br (Polar regions)   | WRF modelling system, based on ECMWF (Offline)                          | (Pacyna et al. 2016; De Simone et al. 2017; Travnikov et al. 2017) |
| ECHMERIT     | 2.8°×2.8°                 | 19 levels                  | O <sub>3</sub> and OH   | ECHAM5 based on ECMWF (Online)  |  |
| HYSPLIT-Hg   | 2.5°×2.5°                 | 17 levels                  | O <sub>3</sub> and OH   | NCEP/NCAR reanalysis model (Offline)                                    | (Cohen et al. 2016)  |

|                 |           |           |                                 |   |   |
|-----------------|-----------|-----------|---------------------------------|---|---|
| GEM-<br>MACH-Hg | 1°×1°     | 58 levels | OH<br><br>Br (Polar<br>regions) | GEM model, based on<br>ECCC weather<br>prediction model<br>(Online) | (Travnikov et al. 2017)                                     |
| CAM-<br>Chem/Hg | 1.9°×2.5° | 26 levels | O <sub>3</sub> and OH           | CCSM, V3 (Offline)  | (Lei et al. 2014)   |
| CTM-Hg          | 8°×10°    | 9 levels  | O <sub>3</sub> and OH           | GCM (Offline)   | (Bullock Jr. et al. 2009;<br>Vijayaraghavan et al.<br>2014) |
| TEAM            | 10–20 km  | 6 layers  | O <sub>3</sub> and OH           | NCAR-MM5 (Offline)  |   |
| AEROMOD         | 1 km      | -         | O <sub>3</sub> and OH           | NOAA and NCDC<br>(Offline)  |   |

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



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**Figure S1: Classification of future Hg emission scenarios by policy focus and emission level. Emission levels are categorized as high (pink), medium (orange), and low (green).**

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