Projecting Global Mercury Emissions and Deposition Under the Shared Socioeconomic Pathways

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November 8, 2023

Abstract

Mercury (Hg) is a naturally occurring element that has been greatly enriched in the environment by activities like mining and fossil fuel combustion. Despite commonalities in some CO2 and Hg emission sources, the implications of long-range climate scenarios for anthropogenic Hg emissions have yet to be explored. Here, we present comprehensive projections of anthropogenic Hg emissions (2020-2300) and evaluate impacts on global atmospheric Hg deposition. Projections are based on four shared socioeconomic pathway (SSP) narratives ranging from sustainable reductions in resource and energy intensity to rapid economic growth driven by abundant fossil fuel exploitation. There is a greater than two-fold difference in cumulative anthropogenic Hg emissions between the lower-bound (110 Gg) and upper-bound (230 Gg) scenarios. Hg releases to land and water are approximately six times those of direct emissions to air (600-1470 Gg). At their peak, anthropogenic Hg emissions reach 2200-2600 Mg a-1 sometime between 2010 (baseline) and 2030, depending on the SSP scenario. Coal combustion is the largest determinant of differences in Hg emissions among scenarios. Decoupling of Hg and CO2 emissions sources occurs under low- to mid-range scenarios, though contributions from artisanal and small-scale gold mining remain uncertain. A projected future shift in speciation of Hg emissions toward lower gaseous elemental Hg (Hg0) and higher divalent Hg (HgII) will result in a higher fraction of locally-sourced Hg deposition. Projected re-emissions of previously deposited anthropogenic Hg follow a similar temporal trajectory to primary emissions, amplifying benefits of primary Hg emissions reductions under the most stringent mitigation scenarios.

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11 Key Points:

- Future (2010-2300) anthropogenic Hg releases to air, land and water (0.7-1.7 Tg) are similar to historical (1510-2010) releases (1.1-2.8 Tg)
- Cumulative anthropogenic Hg emissions to air (2010-2300) vary by a factor of two across scenarios (110-230 Gg)
- Deposition declines from near-term reductions in anthropogenic Hg emissions are
 amplified by reductions in re-emissions from land and ocean

18 Abstract

Mercury (Hg) is a naturally occurring element that has been greatly enriched in the environment 19 20 by activities like mining and fossil fuel combustion. Despite commonalities in some CO₂ and Hg emission sources, the implications of long-range climate scenarios for anthropogenic Hg 21 emissions have yet to be explored. Here, we present comprehensive projections of anthropogenic 22 Hg emissions (2020-2300) and evaluate impacts on global atmospheric Hg deposition. 23 Projections are based on four shared socioeconomic pathway (SSP) narratives ranging from 24 sustainable reductions in resource and energy intensity to rapid economic growth driven by 25 26 abundant fossil fuel exploitation. There is a greater than two-fold difference in cumulative anthropogenic Hg emissions between the lower-bound (110 Gg) and upper-bound (230 Gg) 27 28 scenarios. Hg releases to land and water are approximately six times those of direct emissions to air (600-1470 Gg). At their peak, anthropogenic Hg emissions reach 2200-2600 Mg a⁻¹ sometime 29 between 2010 (baseline) and 2030, depending on the SSP scenario. Coal combustion is the 30 largest determinant of differences in Hg emissions among scenarios. Decoupling of Hg and CO₂ 31 emissions sources occurs under low- to mid-range scenarios, though contributions from artisanal 32 and small-scale gold mining remain uncertain. A projected future shift in speciation of Hg 33 emissions toward lower gaseous elemental Hg (Hg^0) and higher divalent Hg (Hg^{II}) will result in 34 a higher fraction of locally-sourced Hg deposition. Projected re-emissions of previously 35 deposited anthropogenic Hg follow a similar temporal trajectory to primary emissions, 36

- amplifying benefits of primary Hg emissions reductions under the most stringent mitigation
- 38 scenarios.

39 Plain Language Summary

Mercury is a global pollutant that is emitted alongside greenhouse gases like carbon dioxide 40 (CO_2) when fossil fuels such as coal are burned. Researchers have projected how emissions of 41 greenhouse gases and climate are likely to change in the future, but relatively little is known 42 about future Hg releases. Here, we project future Hg emissions between 2020 and 2300 using 43 growth scenarios developed by climate change researchers. Under low emission scenarios, Hg 44 emissions are projected to peak between 2010 and 2030. Under the high emission scenario, Hg 45 releases continue near present-day levels until after 2060, and decline more slowly than other 46 scenarios thereafter. Large variability in projected releases (cumulatively a two-fold difference) 47 is apparent across the low and high scenarios. Globally, the intensity of coal combustion and 48 how quickly it is phased out is the largest driver of future Hg releases. We then use global 49 models to simulate future atmospheric Hg deposition, identifying multiple factors responsible for 50 changing deposition patterns and amounts. We find there is a penalty for delaying reductions in 51 Hg emissions because of increased reemissions from the land and ocean in the future. This work 52 emphasizes the benefits of near-term stringent global reductions in anthropogenic Hg releases. 53

54 **1 Introduction**

55 Mercury (Hg) is a naturally occurring element, but human activities such as mining and fossil-

⁵⁶ fuel combustion have released approximately 1.5 Tg of Hg from stable geologic reservoirs to the

atmosphere, land, and water over the past 500 years (1510-2010) (Streets et al., 2019a). This has

significantly altered the natural biogeochemical Hg cycle (Geyman et al., 2023; Streets et al.,

59 2019a) and adversely affected the health of exposed humans and wildlife (Basu et al., 2023).

60 Reemission of anthropogenic Hg from terrestrial and aquatic ecosystems extends its lifetime in

61 the biosphere (referred to as "legacy Hg"). Past work has quantified the impacts of historical

emissions and legacy Hg cycling on the global Hg cycle (Amos et al., 2013; Angot et al., 2018;

63 Guerrero & Schneider, 2023; Nriagu, 1994). However, estimates of future anthropogenic Hg

emissions based on the most recent future growth scenarios adopted by the Intergovernmental
 Panel on Climate Change (IPCC) and their drivers are not presently available (O'Neill et al.,

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 2016), limiting our ability to project future scenarios of Hg pollution.

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67 Shared Socioeconomic Pathways (SSPs) and associated emission scenarios offer unified

narratives ranging from sustainable development to fossil fuel-driven growth (O'Neill et al.,

69 2016, 2017). They provide a framework for bounding development and greenhouse gas

trajectories extending to the year 2300 (Meinshausen et al., 2020). However, these narratives do

not explicitly address Hg emissions or the technological transformations underpinning future

changes in Hg emission intensity. We fill this gap by developing a methodology for projecting

73 time-dependent change in activity-specific Hg emission factors. This approach is built upon

74 detailed parameterizations developed for historical emission inventories (Streets et al., 2011, 2010) which facilitates interpretering a factor and factors emissions

75 2019a), which facilitates intercomparison of past and future emissions.

- Anthropogenic Hg is released to the atmosphere in both the elemental (Hg^0) and oxidized (Hg^{II})
- forms. Hg^{II} has an atmospheric lifetime against deposition of a few days (Corbitt et al., 2011),

while the lifetime of Hg^0 is greater than one year (Horowitz et al., 2017; Shah et al., 2021).

79 Previous studies projected Hg deposition in 2035 and 2050 based on anthropogenic Hg

80 emissions estimated from older IPCC Special Report on Emission Scenarios (SRES) and

- ⁸¹ independent estimates (Corbitt et al., 2011; Pacyna et al., 2016; Streets et al., 2009). Results
- suggested future increases in Hg^{II} emissions relative to Hg^{0} are likely to increase the proportion
- of regional Hg deposition from emitting countries. The SRES emission trajectories ranged from a
- best-case scenario that showed relatively flat anthropogenic Hg emissions between 2006 and 2050, to a scenario characterized by higher energy and economic growth (A1P) that
- 85 2050, to a scenario characterized by higher energy and economic growth (A1B) that 86 approximately doubled 2006 emissions levels (Streets et al., 2009). Results from Pacya
- approximately doubled 2006 emissions levels (Streets et al., 2009). Results from Pacyna et al.
 (2016) ranged from a slight increase under a current policy scenario to -85% under a maximum
- feasible reduction case. However, these past studies did not consider future changes in marine
- and terrestrial Hg reservoirs that affect reemission of Hg^0 (Amos et al., 2013, 2014). This is
- important because terrestrial and marine Hg^0 evasion are thought to account for large fractions of
- the atmospheric Hg undergoing contemporary and future deposition (e.g., ~60% of 2050
- 92 deposition to the contiguous United States) (Corbitt et al., 2011).
- 93 The main objective of this work was to better understand how future anthropogenic Hg
- emissions and deposition vary among the most recent socioeconomic development pathways
- used by the IPCC. To do this, we developed new decadal projections of primary anthropogenic
- Hg emissions for the years 2020-2300 based on four distinct SSPs spanning a wide range of
- ⁹⁷ radiative forcings. Using a suite of global modeling tools, we quantified how changes in primary
- 98 Hg emissions and re-emissions from terrestrial and marine ecosystems affect regional
- magnitudes and global patterns in atmospheric deposition. This work provides insights into how
- 100 different fossil fuel use scenarios are likely to affect global Hg burdens and the potential effects
- 101 of different pollution control efforts.

102 **2 Methods**

- 103 2.1 Description of Development Narratives (SSP Scenarios)
- 104 Forecasts of future anthropogenic Hg emissions were developed in accordance with the scenarios
- used in Phase 6 of the Coupled Model Intercomparison Project (CMIP6), organized under the
 auspices of the IPCC (O'Neill et al., 2016). The scenarios were based on a framework combining
- auspices of the IPCC (O'Neill et al., 2016). The scenarios were based on a framework combining
 narratives of global development with emissions and climate projections from integrated
- assessment and climate models (O'Neill et al., 2016). The first component, the SSPs, are
- comprised of five distinct narratives describing alternative courses of societal development, as
- 10 well as quantitative descriptions of population, economic growth, and urbanization (Dellink et
- al., 2017; Jiang & O'Neill, 2017; Kc & Lutz, 2017). The SSPs were elaborated using integrated
- assessment models (IAMs) to provide quantitative descriptions of energy use, greenhouse gas
- emissions, and land-use change (Riahi et al., 2017).
- 114 For each SSP narrative, multiple IAM trajectories were defined to describe both a baseline
- scenario, which assumes no additional climate policies or climate change impacts, and mitigation
- scenarios, in which further policies are adopted. SSP narratives can be combined with different
- climate forcing pathways to describe the physical response of the climate system. The climate
- forcing is defined according to the long-term global average radiative forcing (W m⁻²; Myhre et
- al., 2013). Following convention, the scenarios used in this work are written as: SSP_{x-y} , where x
- is the SSP and y is the radiative forcing pathway (W m^{-2}) (O'Neill et al., 2016). Throughout the
- rest of this work, scenarios will be referred by their specific name (e.g., SSP1-2.6) or collectively
- 122 as "SSPs."

- For this work, we adopted the three major scenarios that were initially chosen for long-term 123
- 124 extensions (LTE) to 2300 under CMIP6 (O'Neill et al., 2016), namely, SSPs 1-2.6, 5-3.4, and 5-
- 8.5. Scenario SSP1-2.6 is a lower-bound on future emissions, reflecting strong emphasis on 125
- sustainability and intensive control of climate-forcing agents; SSP5-3.4 is known as the 126
- "overshoot" scenario and reflects short-term growth in fossil-fuel use and minimal consideration 127
- of climate control measures until 2040 followed by aggressive mitigation thereafter; SSP5-8.5 is 128
- an upper bound on emissions, in which fossil-fuel use continues with little consideration of 129
- climate mitigation or transition to clean technologies. Subsequently, Meinshausen et al. (2020) 130 extended additional scenarios to beyond 2300, so we added a fourth scenario to our projections, 131
- the so-called "middle-of-the-road" scenario, SSP2-4.5 (Fricko et al., 2017). 132
- The detailed raw activity data that are used in this work to characterize Hg emissions under these 133
- four scenarios out to 2100 follow the work of Rao et al. (2017), with LTE from 2100 to 2300 134
- following the work of Meinshausen et al. (2020). The Hg emissions forecasts reported in this 135
- work map the trajectories of CO₂ forecasts contained in the previously mentioned publications 136
- and the emissions of other species as described in subsequent studies (Gidden et al., 2019; Lund 137
- et al., 2019; Turnock et al., 2020). However, this work contains the first reported projections of 138
- Hg emissions under CMIP6 scenarios. 139
- The SSP scenarios were used primarily to project fossil-fuel use (coal and oil), the manufacture 140
- and use of refined industrial products, and economic parameters. However, there are no specific 141
- variables from which to calculate the extraction and production of raw materials and basic 142
- 143 products because these are not directly relevant to emissions of greenhouse gases. However, they are very important as sources of mercury. Thus, for 21st century production of these materials,
- 144
- we used forecasts specifically generated by industry models, as follows: copper, zinc, and lead 145 (Sverdrup et al., 2019); iron (Morfeldt et al., 2015); mercury (Sverdrup & Olafsdottir, 2020); and 146
- gold (Sverdrup et al., 2012); as well as the basic industrial products steel (Morfeldt et al., 2015) 147
- and cement (Zhang et al., 2018). The work of Watari et al. (2020, 2021) was valuable in guiding 148
- the pathways of metals extraction and use to the end of the century. 149
- 2.2 Mercury Emission Calculation 150
- 151 Mercury emissions under a future climate scenario (f) were calculated using Equation (1):

$$E_{t,r,s,f} = E_{2010,r,s} \times \left(\frac{A_{t,r,s,f}}{A_{2010,r,s,f}}\right) \times \left(\frac{EF_{t,r,s,f}}{EF_{2010,r,s,f}}\right), \#(1)$$

- where E = emissions (Mg a⁻¹); t = decadal future year, r = world region, s = source type; A =152
- activity level (in various units); and EF = emission factor (g per unit of activity). 153
- Future Hg emissions were calculated by extrapolating 2010 base-year emissions, as reported in 154
- 155 (Streets et al., 2017, 2019a), in accordance with the energy and activity forecasts for each of the
- four future scenarios (f) described in the previous section. Emissions were calculated for each 156
- decade between 2020 and 2300, though they reach zero between 2190 and 2250 across scenarios 157
- 158 in accordance with SSP prescriptions. Emissions are zero beyond 2250 across all scenarios.

Emissions were calculated at world region level (r). The SSP forecasts were developed for five 159 world regions: OECD, REF (Russia and Eastern Europe), ASIA, MAF (Middle East and Africa), 160 and LAM (Latin America). This is a very coarse division of the world, which may be adequate 161 for CO_2 studies, but is too aggregated for studies of Hg emissions and transport. In particular, the 162 OECD region is spread across the globe, with contributions from Western Europe, North 163 America, Australasia, and Japan. The prior Hg emission estimates for 2010 (Streets et al., 2019a, 164 2019b), upon which this work is based, were calculated for 17 world regions and subsequently 165 aggregated to seven: North America (NAM), South America (SAM), Western Europe (EUR), the 166 Former Soviet Union (FSU), Africa/Middle East (AFM), Asia (ASA), and Oceania (OCA). By 167 examining 2010 Hg emissions in the world regions used in each of these two studies, a simple 168 equivalence was determined and applied in this work, as follows: NAM = 0.5 OECD + 0.1169 LAM; SAM = 0.9 LAM; EUR = 0.3 OECD; FSU = REF; AFM = MAF; ASA = ASIA + 0.05 170 OECD; and OCA = 0.15 OECD. Emissions at the global scale (GLO) are thus identical. Though 171 this is less than an ideal solution-because of potential future differences in the rates of 172

development among the countries comprising the coarse SSP regions—it is certainly an

improvement over the five SSP regions from the perspective of estimating Hg transport and

175 deposition.

176 The SSP scenarios contain more than 600 activity components (A), covering all aspects of future

energy, industrial, and economic development. The prior 2010 Hg emission estimates were

developed for 18 source types (*s*) (Streets et al., 2017). Because none of the SSP scenarios

provide activity components that can be unequivocally associated with several of the Hg source

types, emissions from 11 source types were projected individually in this work: mining (copper,

zinc, lead, iron, mercury, gold, and artisanal gold), steel production, cement production, coal
 combustion, and oil combustion. Six other source types (municipal waste incineration, other

combustion, and oil combustion. Six other source types (municipal waste incineration, other waste combustion, electrical and measuring equipment, chemicals manufacturing, caustic soda

production, and dental) were projected in aggregate. Silver mining was not included in this work

because emissions from 2010 onwards are expected to be zero.

186 The change in activity levels in the future, A_t/A_{2010} , reflects the growth or shrinkage in activity

187 for a particular source type, in a particular region, under a particular scenario. Some may grow,

some may decline, depending on the influence of the world economy in general and the pressure

of the climate change scenario. For example, coal combustion may increase under a lax climate

scenario or decline under a stringent one. But A_t/A_{2010} only characterizes the change in the size of

the source type, it says nothing about the transformation of it over the time period in question.
New, high-performing technologies will undoubtedly replace older, low-performing ones; and

the imposition of new emission control regulations may or may not force the use of add-on

emission control technologies or even a complete change in production technology. All of these

195 will influence Hg emissions. Except for Carbon Capture and Storage (CCS) and inferences from

 SO_2 emissions, the SSP scenarios say nothing about technology transformation that will

influence Hg emissions. This, therefore, is the most challenging aspect of forecasting future Hg

emissions. Our approach was to develop the ratio $EF_{t,r,s,f}/EF_{2010,r,s,f}$ as an indicator of how future

emission rates will decline from their 2010 values. Note that this ratio is never greater than one

200 (i.e., future emission rates are never higher in the future than in 2010 per unit of activity).

In previous work, we developed a methodology for estimating the time-development of emission factors for historical periods (pre-2010) using transformed normal distribution functions (Streets et al., 2011). These region-specific functions were parameterized based on a detailed review of

experimental measurements around the world, as illustrated in that paper for copper smelting. It

was shown, for example, that the Hg emission rate for copper smelters pre-1900 was an

uncontrolled value of 27.5 gHg/MgCu worldwide. After 1900, emission rates declined, along
 different pathways in different parts of the world, reaching values in 2010 ranging from a low of

207 different pathways in different parts of the world, reaching values in 2010 ranging from a low 208 0.60 gHg/MgCu in western Europe to a high of 11.6 gHg/MgCu in Africa.

209 The challenge in this work was to reasonably represent the continuation of historical *EF* trends

out beyond 2010 in the absence of guidance from the SSP forecasts. In this work, we focused on

the period 2010–2100, where technology transformation will have the biggest effect. For each of the 11 key source types mentioned above, we assumed that by 2100 the emission rates in every

world region will have declined from their 2010 levels to the present-day emission rate of the

214 lowest-emitting region. Thus, in the case of copper smelting, all regions emit at 0.60 gHg/MgCu

by 2100, though the trajectories to reach it vary by region. This 2100 rate then continues for all

216 years beyond 2100. Intermediate decadal years in *EF* were determined as linear trends.

217 For the purposes of atmospheric modeling, speciation of these future atmospheric Hg emissions

into elemental Hg (Hg⁰) and divalent Hg compounds (Hg^{II}) was determined in a similar way, by

extrapolation of the speciation splits in each region in the year 2010. These range rather widely,

from a high value of the Hg^0/Hg ratio of 0.88 in South America to a low of 0.25 in western

Europe. This reflects the different emission characteristics of, for example, mining in South

America and industrial manufacturing/well-controlled coal combustion in western Europe. As

time goes on it can be expected that the Hg^0/Hg ratio will decline everywhere, as artisanal and

small-scale mining techniques (with high Hg^0 emission rates) are retired, and industrial processes

and combustion become increasingly well controlled, leading to conversion of elemental Hg to collectable, oxidized Hg compounds. In this work, we assumed that the 2010 ratios in high-

emitting regions approach the western European level or the North American level (0.33) by

228 2100, eventually leveling out at a technology-limiting value of 0.2 everywhere. Slight variations

were applied among the four scenarios to reflect anticipated patterns of future technology

230 transformations.

Finally, future releases of Hg to land and water were estimated following the method of Streets

et al. (2017, 2019a). In essence, this involves subtracting the air emissions from the total Hg

contained in the raw material that is processed. It was not possible to determine *a priori* the fate

of these releases by source type, technology level, or world region. Uncertainties in the few

quantitative estimates that have been made and the vast quantity of unknown factors for sources

in remote parts of the world essentially rule this out.

237 2.3 Global Atmospheric Hg Deposition

238 We used the GEOS-Chem global chemical transport model to simulate the atmospheric fate and

deposition of mercury emissions under each emissions scenario. The model version (12.8) used

240 here included detailed multi-phase oxidation of elemental Hg and gas-phase photolysis of

oxidized Hg species from Shah et al. (2021). Simulations were run using 2014-2019 MERRA-2

meteorology (Gelaro et al., 2017) on a 2×2.5 -degree horizontal grid with a 72-layer vertical

domain extending through the top of the stratosphere. The first two years of each simulation

- were used for initialization and the final three years were averaged for analysis to reduce the
- effects of meteorological variability on simulated deposition patterns.
- We conducted 5-year simulations for each decade from 2020-2100 for each scenario in addition
- to a common baseline scenario for the 2010 emission year. We hold meteorology constant across
- decadal snapshots to isolate the effects of variation in future emissions. Emissions from the
- seven world regions were distributed onto a 0.25×0.25 -degree grid based on the spatial
- distributions established in prior work (Steenhuisen & Wilson, 2019). Relative fractions of North
 American emissions from Canada, the United States, and Central America were scaled to reflect
- 251 2015 values reported in Streets et al. (2019b) for consistency with past work.
- 253 We constructed source-receptor functions to quantify changes in atmospheric Hg deposition
- resulting from shifts in terrestrial and oceanic Hg^0 evasion and for each anthropogenic emission
- region. Following Corbitt et al. (2011), we define each source-receptor function F_{ij} as:

$$F_{ij} = \frac{D_{ij}}{E_i} \#(2)$$

- where D_{ij} is the total mercury deposition flux to receptor grid cell *j* from emissions in region *i*, and E_i is the magnitude of emissions from region *i*.
- 258 2.4 Global Biogeochemical Box Model (GBBM)
- 259 An updated version of the multi-compartment Global Biogeochemical Box Model (GBBM)
- developed by Amos et al. (2013; 2014) was used to simulate future shifts in atmospheric
- deposition from legacy mercury emissions (Text S1; Table S1-S2). The GBBM represents Hg
- cycling between the atmosphere, terrestrial biosphere (fast, slow, protected), ocean (surface,
- 263 intermediate, deep), and removal by burial in marine sediment.
- 264 We added four new compartments to the GBBM that represent waste reservoirs (fast, slow,
- protected, and immobilized) to explicitly track the fate of the estimated 1.13 Tg Hg released by
- humans to land and water from 1510-2010 (Streets et al., 2019a). These releases included
 tailings and waste from mining and metals production, chlor-alkali plants, and substantial
- tailings and waste from mining and metals production, chlor-alkali plants, and substantial
 contributions from Hg use in commercial products (Horowitz et al., 2014; Streets et al., 2011). In
- prior work, anthropogenic releases to land and water were added to soil compartments of the
- GBBM (Streets et al., 2017). However, it is likely that the majority of such releases are
- sequestered at contaminated sites and do not have the same diffuse impacts on deposition as
- atmospheric sources (Guerrero & Schneider, 2023; Kocman et al., 2017). Therefore, we
- independently tracked land and water Hg releases. We parameterized re-emissions of Hg^0 and
- discharges of Hg^{II} to rivers from waste compartments using the same rate coefficients as for the
- fast, slow, and protected terrestrial Hg in the GBBM. Land and water Hg releases were
- 276 partitioned into waste pools following the methods described in Streets et al. (2017) (Text S1).
- 277 We simulated the time-dependent fate of future anthropogenic Hg releases separately for each
- scenario. In each simulation, the model was initialized by calculating the steady state distribution
- of mercury among reservoirs under a constant geogenic Hg flux of 230 Mg a^{-1} from subaerial
- volcanism and 50 Mg a⁻¹ from hydrothermal vents (Geyman et al., 2023). The model was then

forced with all-time (2000 BCE to 2010 CE) anthropogenic mercury releases from Streets et al.

(2019) followed by emissions specified in each scenario from 2010-2300. Evaluation of model

results for the year 2010 agree with the observational ranges for the atmospheric Hg reservoir,

seawater concentrations, and atmospheric deposition enrichment factors (Table S3, Amos et al.,

285 2015).

286 **3 Results and Discussion**

- 287 3.1 Future Emission Trends
- 288 3.1.1 Long-term perspective on anthropogenic Hg releases

Atmospheric Hg emissions are shown by region (Fig. 1; Table S4) and source sector (Table S5)

for each of the four scenarios considered in this work. All primary Hg emissions are projected to

be zero beyond 2250, consistent with SSP assumptions. Projected cumulative Hg releases to air,

land, and water between 2010 and 2300 range from 709 Gg under SSP1-2.6, to 1710 Gg under

293 SSP5-8.5. Projected anthropogenic emissions between 2010 and 2300 are comparable to

cumulative historical emissions from 1510-2010 (1470 Gg) estimated in past work (Streets et al.,

2019a), suggesting human impacts on the global Hg cycle will be sustained for millennia.

296 Global Hg emissions projected in this work for the year 2050 are substantially lower than similar

forecasts made over a decade prior $(2300-4900 \text{ Mg a}^{-1})$ using the IPCC SRES scenarios (Streets

et al., 2009). Projected anthropogenic emission declines for the year 2050 under SSP1-2.6 (-1080

Mg a^{-1}) are similar to those projected for 2035 by Pacyna et al. (2016) under a scenario involving

full implementation of policy commitments and plans made before 2016. Relative declines

301 projected in this work are partially attributable to lower Hg emission factors from recently

implemented Hg pollution controls (e.g., Zhang et al., 2023). Thus, lower future Hg emissions

303 estimated in this work reflect progress made through global policy efforts in recent decades.

Across all scenarios, future land and water Hg releases follow qualitatively similar trajectories to atmospheric emissions. Land and water Hg releases grow as a fraction of total anthropogenic Hg releases in the future. By 2100, anthropogenic Hg releases to land and water are estimated to be 12 to 25 times greater than anthropogenic Hg emissions to air, compared to only 3.3-fold greater (7330 Mg a⁻¹) in 2010.



Figure 1. Global anthropogenic mercury (Hg) emissions to air by world region and source sector.

Trajectories under each scenario are shown by world region in Panels $(\mathbf{a} - \mathbf{d})$. Inset pie charts show fractional emissions occurring during four "snapshot" time periods, arranged clockwise from the bottom: 2010-2049,

2050-2099, 2100-2200, 2200-2300. Cumulative emissions to air (Gg; 2010-2300) are shown in the center of

- each ring. Panels ($\mathbf{e} \mathbf{f}$) provide a sector-specific breakdown of the same emission. ASGM = artisanal and
- 317 small-scale gold mining.
- 318 3.1.2 Scenario-specific patterns in primary anthropogenic Hg releases

Among scenarios considered in this work, SSP1-2.6 is a lower-bound greenhouse gas emissions

case, and it is known as the "2°C scenario." SSP1-2.6 is characterized by aggressive reductions

in greenhouse gas emissions and a nameplate end-century radiative forcing of 2.6 W m^{-2}

322 (Meinshausen et al., 2020). Projected future atmospheric Hg emissions decline continuously after

- 2010 (Fig. 1). In 2010, Asia was the largest regional contributor (1260 Mg a^{-1} ; 57% of total) to
- global atmospheric emissions (2190 Mg a^{-1}). Under SSP1-2.6, a 93% reduction in global Hg emissions is projected (154 Mg a^{-1}) by the year 2100, and Asia remains the largest remaining
- regional emitter (91 Mg a^{-1} , 59%). Beyond 2100, emissions continue to decline at a slower pace
- and reach zero by 2190, in accordance with SSP assumptions.

SSP2-4.5 is a middle-of-the-road scenario that reflects moderate socioeconomic, energy, and

- climate mitigation changes and a continuation of growth and development trajectories following
- the status quo (Meinshausen et al., 2020). Relative to 2010, Hg emissions grow slightly until

- 2030 and then decline at a rate similar to SSP1-2.6 (Fig. 1). Hg emissions are 193 Mg a^{-1} by
- 2100, which is only 25% greater than SSP1-2.6 (Fig. 1). SSP2-4.5 is not nearly as close to the
- middle of the road for Hg emissions as it is for carbon emissions because it relies heavily on a
- switch from coal to natural gas as the primary energy supply under moderate climate mitigation
 efforts. This strategy results in continued greenhouse gas emissions and therefore results in
- efforts. This strategy results in continued greenhouse gas emissions and therefore results in middle of the road climate effects. However, natural gas combustion generates insignificant
- quantities of Hg, and so the trajectory is much lower. Hg emissions under SSP2-4.5 continue at a
- low level into the distant future, eventually reaching zero in 2250 following SSP assumptions
- (Fig. 1). Between 2010 and 2300, cumulative emissions to air are 131 Gg and cumulative
- emissions to land and water are 737 Gg under SSP2-4.5.
- 341 SSP5-3.4 is the overshoot case characterized by a delay in climate-change action until after 2030,
- leading to a major rise in Hg emissions between 2010 and 2030. Hg emissions are projected to

rise to 2580 Mg a^{-1} by 2030, an increase of 17% over 2010 (Fig. 1). Much of that growth occurs

in Asia. After 2030, emissions decline more precipitously than under the other three scenarios,

- reaching levels similar to SSP1-2.6 by 2060. Hg emissions fall to 107 Mg a^{-1} by 2100 and reach zero in the year 2170, which is earlier than SSP1-2.6. Under SSP5-3.4, cumulative emissions to
- zero in the year 2170, which is earlier than SSP1-2.6. Under SSP5-3.4, cumulative emissions to
 air are projected to be 118 Gg. Compared to SSP1-2.6, cumulative emissions are heavily
- weighted toward the first few decades of the projection period. Cumulative emissions to land and
- 349 water are 618 Gg.
- 350 SSP5-8.5 is the upper-bound case, characterized by continued fossil-fuel use and minimal
- 351 consideration of environmental sustainability. Under this scenario, CO₂ concentrations are
- projected to reach levels greater than 2000 ppm by 2200 (Meinshausen et al., 2020). Such levels
- have not occurred on Earth since before the onset of the modern Antarctic glaciation over 40
- Mya (Rae et al., 2021), and are associated with temperatures 3.3°C to 5.7°C higher in 2100 than
- during the early-industrial period (1850-1900) (IPCC, 2023). Emissions of Hg to air remain near
- present levels until after 2060 and then decline to 914 Mg a^{-1} (5-9 times the other scenarios) by
- the end of the 21^{st} century. The dominant Hg emission regions are Asia, where emissions exceed 1000 Mg a⁻¹ through 2070, and Africa and the Middle East, where emissions grow from 383 Mg
- a^{-1} in 2010 to 744 Mg a^{-1} in 2080. Emissions decline slowly beyond 2100, remaining at high
- levels into the 22^{nd} century: 512 Mg a⁻¹ in 2150 and 256 Mg a⁻¹ in 2200 (Fig. 1). Emissions do
- not reach zero until 2250. Cumulative Hg releases (2010-2300) are 235 Gg to air and 1.47 Tg to
- land and water under SSP5-8.5.

363 Several indicators suggest that global anthropogenic Hg emissions are tracking below levels

described in the overshoot scenario (SSP5-3.4) since 2010. While existing emission inventories

report growth in global anthropogenic Hg emissions between 2010 and 2015 (Streets et al.,

2019b), domestic policies have prompted widespread installation of air pollution control devices

367 over the past decade in China (Zhang et al., 2023) and the United States (Dai et al., 2023).

- Trends in Hg^0 concentration and isotopic composition from long-term monitoring sites in China
- also show declines consistent with suggested reductions in regional anthropogenic emissions
- 370 (Wu et al., 2023).

371 3.1.3 Decoupling of Hg emissions and radiative forcing under low coal use scenarios

- Among source sectors in 2010, artisanal and small-scale gold mining (ASGM) was the largest 372
- global source of atmospheric emissions (727 Mg a⁻¹), followed by coal combustion (538 Mg a⁻¹) 373
- and mining and industry (491 Mg a⁻¹). Large-scale (rather than ASGM) gold production was the 374
- 375 largest source of Hg released to land and water (1990 Mg a⁻¹), followed by ASGM (1090 Mg a⁻¹) ¹), electrical and measurement equipment (1010 Mg a⁻¹), chemicals manufacturing (860 Mg a⁻¹),
- 376 and zinc smelting (670 Mg a⁻¹). Coal combustion accounted for a greater fraction of global 377
- atmospheric emissions (25%) in 2010 compared to land and water releases (5%, 370 Mg a^{-1}). 378
- 379 Differences in anthropogenic Hg emissions to air principally reflect differences in coal
- combustion across SSP scenarios. Hg emissions from coal combustion are projected to grow by 380
- 2.3 to 12.5% and peak in 2020 under SSPs 1-2.6 and 2-4.5 (Fig. 1). Near-term increases in Hg 381
- emissions from coal combustion reach levels 65% higher than 2010 under SSP5-3.4. Hg 382
- emissions decline thereafter for lower coal-use scenarios (SSPs 1-2.6, 2-4.5 and 5-3.4). By 2050, 383
- they fall to a fraction of 2010 levels (15% under SSP1-2.6 and 69% under SSP2-4.5) and reach 384
- zero before the end of the century. In contrast, coal combustion persists until 2250 under SSP5-385
- 8.5. Coal-related Hg emissions under SSP5-8.5 reach 990 Mg a⁻¹ (96% higher than 2010) by 386
- 2070 and remain 44% higher than 2010 levels by the end of the century (Fig. 1). 387
- Among lower coal-use scenarios, coal combustion is responsible for 16 Gg (SSP1-2.6) to 28 Gg 388

(SSP2-45) of cumulative (2010-2300) Hg emissions to air. Cumulative Hg emissions to air from 389

coal combustion are ~5 to 9-fold greater under SSP5-8.5 (139 Gg) than the other scenarios, and 390

- coal combustion comprises 59% of all emissions to air between 2010 and 2300 under SSP5-8.5. 391
- Despite relative similarities in the phase-out of coal, SSPs 1-2.6, 2-4.5, and 5-3.4 show very 392
- 393 different greenhouse gas emission trajectories. Such differences arise from rates of natural gas
- and oil combustion, as well as adoption of carbon capture and sequestration. These factors exert 394
- minimal direct influence on primary anthropogenic Hg emissions, though they produce disparate 395
- 396 climate effects. For example, end-century surface temperatures simulated with the NASA GISS-
- E2.1 climate model are $1.8^{\circ}C 2.3^{\circ}C$ higher than the preindustrial (1850-1880) mean under 397 SSP1-2.6 compared to 2.7°C – 3.3°C under SSP2-4.5 (Nazarenko et al., 2022). The degree of
- 398
- warming will modulate future changes in Hg emissions from the natural biosphere (e.g., 399 Krabbenhoft & Sunderland, 2013; Schaefer et al., 2020). Therefore, it is important to consider 400
- the consequences of human activity for both direct anthropogenic Hg releases and warming-401
- driven changes in Hg cycling in the biosphere and ocean (e.g., Schaefer et al., 2020; Schartup et 402
- al., 2019). 403
- 3.1.4 Changes in Hg emission speciation favor local and regional deposition 404
- The fraction of primary anthropogenic Hg emissions released to air as Hg⁰ is projected to decline 405
- in the future (Fig. 2), with implications for transboundary pollution. For SSP1-2.6, the fraction of 406
- Hg⁰ emitted by primary sources is projected to decline by 28% between 2010 and 2100. It 407
- stabilizes from 2100-2140, and then declines continuously thereafter. By 2180, the last decade 408
- with non-zero emissions for SSP1-2.6, Hg⁰ is projected to make up just 20% of total 409
- anthropogenic Hg emissions to air (Fig. 2). Emission speciation under SSP5-3.4 broadly follows that of SSP1-2.6, with slightly higher Hg^0 fractions through 2100, and an accelerated decline 410
- 411 between 2140 and 2160 from 31% Hg⁰ to 20% Hg⁰ (Fig. 2). Under SSP2-4.5, Hg⁰ comprises a
- 412 larger fraction of total Hg emissions to air than under SSPs 1-2.6 and 5-3.4, remaining around 413

- 55% until 2040 (Fig. 2). The Hg⁰ fraction then exhibits a slow decline with overall emissions and 414
- reaches 22% by 2240 (Fig. 2). SSP5-8.5 consistently represents an upper bound for the Hg^0 415
- fraction, increasing to 57% in 2060, followed by near-continuous declines to reach 22% by 2240. 416
- While trajectories vary across scenarios, greater fractions of Hg^{II} relative to Hg⁰ in primary 417 anthropogenic emissions are expected to produce future deposition patterns from primary
- 418
- emissions that increasingly reflect local and regional rather than global sources (Fig. S1). 419







The percentage of total Hg emissions to air released as elemental Hg (Hg⁰) is shown for each scenario and the 422

- remaining fraction consists of divalent mercury (Hg^{II}). 423
- 3.2. Future Deposition Patterns from Primary Anthropogenic Emissions 424
- Modeled global Hg deposition to land increases by 22 Mg a^{-1} (0.9%) between 2010 and 2020 425
- under SSP1-2.6 emissions. This reflects increases from both primary anthropogenic emissions 426 $(+8 \text{ Mg a}^{-1})$ and higher re-emissions from terrestrial and ocean surfaces due to increasing Hg 427 reservoirs (+14 Mg a⁻¹) (Fig. S2). Modeled increases in regional Hg deposition occur during this 428
- period over all global regions except for Europe and North America (Fig. 3a; 4a). There, declines 429
- in deposition from primary anthropogenic sources exceed increases in deposition from growing 430
- global terrestrial and oceanic Hg reservoirs and subsequent re-emissions. From 2040 through the 431
- end of the century, declines in atmospheric Hg deposition are projected for all regions. By 2100, 432
- total atmospheric Hg deposition is projected to be less than half of 2010 levels (46%, 1120 Mg a⁻ 433
- ¹) (Fig. S2). The largest regional deposition declines are projected over Asia (-510 Mg a^{-1} ; -434
- 73%) because it was the largest source region in 2010 (Fig. 3a). 435
- Temporal patterns in atmospheric Hg deposition under SSP2-4.5 are qualitatively similar to 436 those of SSP1-2.6, characterized by slight but regionally heterogeneous near-term increases that 437 give way to continuous decreases. Relative to 2010, global deposition increases by 1.7% in 2020 438 and 1.6% in 2030. These increases occur over all regions except Europe, where declines are 439 smaller than for SSP1-2.6 (-17% in 2020 and 2030) (Fig. 3b). By the end of the century, 440 deposition to land is 1270 Mg a⁻¹ lower than in 2010 (Fig. S2), with regional declines reaching 441 71% (-25 µg m⁻² a⁻¹) in Asia (Fig. 3b). 442

- 443 Under SSP5-3.4, near-term deposition increases considerably, growing to 2690 Mg a^{-1} (+9%) in
- 444 2030 over land (Fig. S2). The greatest increases occur in Asia, where total Hg deposition reaches
- 445 41 μ g m⁻² a⁻¹ (+18%) in 2030. Deposition grows by greater than 8% over all regions except 446 Europe. After 2030, accelerated emission reductions relative to SSP1-2.6 result in comparable
- end-century deposition, which is 1340 Mg a^{-1} lower than in 2010 (Fig. S2).

448 Atmospheric deposition under SSP5-8.5 is much higher than the other scenarios through most of

- the century. However, 2020 deposition is the lowest of all scenarios, and 2030 deposition is
- lower than all scenarios other than SSP1-2.6 (Fig. S2). Growth in emissions from Africa and the
- 451 Middle East, combined with sustained emissions elsewhere, produce increasing deposition over
- 452 most regions from 2020 until 2070. Most notably, deposition to Africa and the Middle East reach
- 453 levels 18% higher (+2.5 μ g m⁻² a⁻¹) by 2070 and remain above baseline levels through 2100 (Fig. 454 3d; Fig. S2). End-century total deposition to land is 650 Mg a⁻¹ lower than in 2010.



in atmospheric Hg deposition are shown by world region (represented by colored bars) for each decade from

459 2020 to 2100 (x-axis). Each subplot represents temporal trends under a different Shared Socioeconomic

⁵⁷ Figure 3. Relative changes in regional atmospheric mercury (Hg) deposition compared to 2010. Changes

⁴⁶⁰ Pathway (SSP) scenario (O'Neill et al., 2016).



Figure 4. Trajectories of global atmospheric mercury (Hg) deposition. Panels represent fractional change
 in total deposition relative to the 2010 baseline by scenario (rows) for three time periods: 2030 (left), 2050
 (center) and 2100 (right). Deposition is calculated as the sum of deposition from primary, legacy, and natural

465 Hg emissions using the GEOS-Chem atmospheric mercury model (Shah et al., 2021) and the Global

466 Biogeochemical Box Model (GBBM; Amos et al., 2013, 2014).

467 3.3 Implications for Legacy Hg Deposition from Terrestrial and Aquatic Emissions

468 Scenarios for deposition of legacy Hg vary among the four SSPs. Legacy deposition peaks

469 before 2035 at levels 20 - 62 Mg a⁻¹ higher than 2010 under SSPs 1-2.6, 2-4.5, and 5-3.4 (Fig.

5). Legacy deposition under SSP5-8.5 exhibits a larger and later peak, exceeding 2010 deposition

by 66 Mg a^{-1} in 2071 (Fig. 5). By 2100, relative trends in legacy deposition are notably different,

with declines of 330 - 380 Mg a⁻¹ relative to 2010 for SSPs 1-2.6, 2-4.5 and 5-3.4, compared

- 473 with a decline of 47 Mg a^{-1} for SSP5-8.5 (Fig. 5). These end-century declines in legacy
- emissions are responsible for 26 28% of total declines in atmospheric Hg deposition to land
- 475 under SSPs 1-2.6, 2-4.5, and 5-3.4, compared to 7% for SSP5-8.5.





Figure 5. Mercury (Hg) deposition to land from legacy and natural emissions. Trajectories under future
emissions (2010-2300) are shown for SSPs 1-2.6 (blue line), 2-4.5 (orange line), 5-3.4 (yellow line), and 5-8.5
(red line), in addition to a hypothetical scenario where primary anthropogenic emissions are zero after 2010
(dotted black line). Implications of future trajectories for upper ocean reservoirs are shown in Figure S4.

482 Comparing SSPs 1-2.6 and 5-3.4 provides insights into the consequences of delaying emission

reductions. Cumulative emissions through 2100 are comparable for SSP1-2.6 (101 Gg) and
SSP5-3.4 (112 Gg). Additionally, anthropogenic Hg emissions are lower in 2100 under SSP5-3.4
than SSP1-2.6 (107 Mg a⁻¹ and 154 Mg a⁻¹). Higher deposition to anthropogenic receptor regions

in 2100 under SSP5-3.4 reflects a legacy deposition penalty for the unrestrained growth of the
 first three decades of the century. Deposition from legacy emissions is 20 Mg higher in 2100 for

first three decades of the century. Deposition from legacy emissions is 20 Mg higher in 2100 fo
 SSP5-3.4 compared to SSP1-2.6, whereas deposition from primary anthropogenic emissions is

488 SSP5-3.4 compared to SSP1-2.6, whereas deposition from primary anthropogenic emissions is
 489 16 Mg lower. This example demonstrates the long-term benefits associated with near-term

490 emissions mitigation, as discussed previously in Angot et al. (2018) and Amos et al. (2013).

491 By the end of the century, Hg re-emissions from the ocean and land will become relatively more important as sources of Hg deposition (Fig. S3), but deposition will not be evenly distributed by 492 world region. Our source-receptor modeling suggests that South America receives the largest 493 share of legacy Hg deposition on a per-area basis, receiving 23% higher deposition per unit of 494 terrestrial emissions and 48% higher deposition per unit of ocean evasion than the area-weighted 495 average of all world regions (Fig. 6). Such high rates of deposition over South America are 496 driven by high rates of foliar uptake and wet deposition. In contrast, Oceania receives the lowest 497 areal share of terrestrial emissions (73% of average), and the Former USSR receives the smallest 498 areal share of Hg sourced from oceanic evasion (74% of average) (Fig. 6). Such low deposition 499 is driven by the relative isolation of Oceania from terrestrial emissions and of the Former USSR 500 from ocean emissions. These source-receptor relationships are subject to change in the future due 501 to shifting patterns of historical anthropogenic Hg loading (e.g., Zolkos et al., 2022) and 502

changing biogeochemical dynamics mediating deposition (e.g., Alexander & Mickley, 2015;

504 Krabbenhoft & Sunderland, 2013; Yang et al., 2019).



506 Figure 6. Regional differences in area-normalized atmospheric Hg deposition from natural and legacy

507 **sources.** Bars represent differences in areal deposition rates between individual receptor regions and the 508 average over land. Positive values mean that receptor regions receive greater deposition per unit area than

average over land. Positive values mean that receptor regions receive greater deposition per unit area than average, and negative values mean that receptor regions receive less deposition than average. Individual panels

show trends in deposition resulting from emissions from the terrestrial biosphere (\mathbf{a}) and from the ocean (\mathbf{b}).

511 Receptor regions are Africa and the Middle East (AFM), Asia (ASA), Europe (EUR), North America (NAM),

512 Oceania (OCA), South America (SAM), and the Former Soviet Union (FSU).

513 Globally averaged seawater Hg concentrations in the upper ocean (0-1500 m) are projected to

514 increase over the coming decades across all scenarios in this study. The Hg reservoir in the upper

ocean peaks at 142 to 151 Gg between 2037 (SSP1-2.6) and 2081 (SSP5-8.5) (Fig. S4). Near-

516 term increases in upper ocean Hg concentrations are driven by future rather than historical

517 emissions. Simulated seawater Hg concentrations begin declining in 2015 under a scenario with

no future primary anthropogenic Hg releases (Fig. S4). By the end of the 21st century, upper

ocean Hg concentrations are projected to be 15% lower than 2010 under SSP1-2.6 and are 11%

520 higher than 2010 under SSP5-8.5.

521 4 Conclusions

505

522 The SSP scenarios evaluated in this work result in a greater than two-fold difference in

523 cumulative anthropogenic Hg emissions between 2010 and 2300. Cumulative anthropogenic

emissions to air and releases to land and water between 2010 and 2300 range from 710 Gg under

the low-bound scenario (SSP1-2.6) to 1710 Gg under the upper-bound scenario (SSP5-8.5).

526 These future releases are comparable to all-time historical anthropogenic emissions of 1540 Gg

527 (80% CI: 1060 – 2800 Gg) (Streets et al., 2019a).

528 Transition of the energy sector away from coal combustion is the largest determinant of

differences among scenarios, with lower bound and mid-range scenarios (SSPs 1-2.6, 2-4.5, 5-

530 3.4) all exhibiting similar cumulative emissions due to declining coal usage. By contrast,

industrial Hg mining and ASGM releases are projected to grow in relative importance in the

future. These results imply that under the most stringent climate policies, the largest sources of

533 Hg and CO_2 are likely to become more distinct from one another.

- 534 Numerous factors may affect regional Hg deposition patterns in the future. For most regions,
- reducing primary anthropogenic Hg emissions remains the most effective way to reduce
- deposition, since 55 71% of Hg^{II} emissions redeposit to the region of origin, compared to 5 12%
- 537 13% for Hg^0 (Fig. S5). However, the scenario-based trajectories described in this work indicate 538 that anthropogenic Hg^{II} emissions to air will decline more slowly than Hg^0 , causing Hg^{II} to grow
- that anthropogenic Hg^{''} emissions to air will decline more slowly than Hg^{''}, causing Hg^{''} to grow as a fraction of total anthropogenic Hg emissions to air. As the speciation of anthropogenic Hg
- emissions shifts towards lower fractions of Hg^0 , a greater proportion of regional emissions will
- redeposit to the region of origin. Across all regions, the fraction of self-sourced atmospheric Hg
- deposition is projected to increase from 33% in 2010 to 35% 45% in 2100 (Fig. S1).
- 543 Regional atmospheric Hg deposition patterns over the next century reflect trends in the
- magnitude and spatial distribution of primary anthropogenic emissions as well as re-emissions
- from the land and ocean. In regions where anthropogenic Hg emissions reductions are projected
- to occur, total Hg deposition is expected to decrease, even during periods where global
- 547 deposition is increasing. This means that domestic policies have significant leverage on domestic
- Hg deposition (e.g., Dai et al., 2023). However, greater reductions in anthropogenic emissions
- are accompanied by larger declines in emissions from the land and ocean. Under SSP1-2.6, 28%
- of the reduction in atmospheric deposition from 2010 to 2100 was the result of declines in legacy
- emissions. As a result, globally-coordinated efforts to reduce near-term anthropogenic Hg emissions will produce amplified benefits in terms of long-term declines in atmospheric
- 553 deposition (e.g., Angot et al., 2018).

554 Acknowledgments

- 555 Financial support for this work was provided by the U.S. National Science Foundation (award
- numbers 2210173 and 2108452). The authors declare no competing financial interest. We thank
- 557Zig Klimont of IIASA for providing the detailed activity and emissions datasets for the SSPs
- 558 used in this work.

559 **Open Research**

- 560 Mercury emission files, code and model output are available in an OSF repository (peer review
- 561 link provided with submission). The version of GEOS-Chem used in this study is available from
- 562 (<u>https://doi.org/10.5281/zenodo.3784796</u>).
- 563

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723	

1 Supporting Information for: **Projecting Global Mercury Emissions and Deposition**

2 Under the Shared Socioeconomic Pathways

3 4

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12 Contents of this file

- 13
 Text S1

 14
 Text S1

 15
 Tables S1 to S5

 16
 5
- 16 Figure S1 to S5

17 Introduction

- 18 The supporting information contains 1 text section, 5 tables, and 5 figures.
- 19 Text S1. Global Biogeochemical Box Model description [p.2]
- Table S1. Modern mercury reservoir magnitudes and fluxes [p.3-4]
- Table S2. Rate coefficients used in Global Biogeochemical Box Model [p.5]
- Table S3. Global Biogeochemical Box Model evaluation [p.6]
- Table S4. Emissions by region for the base year and key future years (Mg a⁻¹) [p.7]
- Table S5. Emissions by source type for the base year and key future years (Mg a⁻¹) [<u>p.8</u>]
- Figure S1. Trends in the fraction of anthropogenic emissions redepositing to region of origin [<u>p.9</u>]
- Figure S2. Trajectories of atmospheric mercury (Hg) deposition from primary anthropogenic and
 legacy + natural emissions [p.10]
- Figure S3. Change in atmospheric mercury (Hg) deposition from legacy emissions, 2010-2100
 [p.11]
- Figure S4. Scenario-specific mass trajectories of mercury (Hg) in the upper ocean (0-1500 m) [p.12]
- Figure S5. Mercury source-receptor matrices, by species [p.13]

32 Text S1. Global Biogeochemical Box Model Description

33

34 We simulate the temporal evolution of mercury in eleven global compartments using a modified version

of the geochemical box model described in Amos et al. (2013). Model compartments include the

36 atmosphere (ATM), three ocean compartments delineated based on depth (surface: OCS, intermediate:

37 OCI, and deep: OCD), and three compartments for terrestrial vegetation and soils delineated based on

38 organic carbon turnover time (fast: TF, slow: TS, and protected: TP).

39

40 We added four waste compartments to the existing model to track the fate of anthropogenic Hg

41 emissions to land and water. Waste compartments are delineated based on turnover time in a manner

42 analogous to terrestrial compartments, and are defined as fast (WF), slow (WS), protected (WP), and

43 immobilized (WI). Model code is implemented in python and will be made available in a public

- 44 repository upon publication.
- 45

46 Following Amos et al. (2013), the model is defined as a system of first-order differential equations:

47

 $\frac{\mathrm{dm}}{\mathrm{dt}} = \mathbf{K}\mathbf{m} + \mathbf{s}$

48 49

50

51

In which *m* is a vector containing the mass in each of the seven reservoirs, *s* is a vector containing inputs to each reservoir and **K** is the 11 x 11 matrix of rate coefficients between each reservoir pair. We simulate the pre-anthropogenic natural steady state by solving dm/dt = 0 under a constant geogenic

52 simulate the pre-anthropogenic natural steady state by solving dm/dt = 0 under a constant geogenic 53 source of 230 Mg Hg a⁻¹ to the atmosphere (Geyman et al., 2023) and 50 Mg a⁻¹ to the deep ocean,

54 reflecting the central estimate from Lamborg et al. (2006).

55

56 We construct **K** using values adapted from Amos et al. (2014). We updated the atmospheric budget for 57 consistency with the most recent GEOS-Chem simulation from Shah et al. (2021). We adjusted setting 58 fluxes associated with marine particles in the ocean based on more recent measurements of Hg 59 partitioning to particles (Cui et al., 2021; Lamborg et al., 2016).

60

61 We force the model with historical anthropogenic emissions (1510-2010) from Streets et al. (2019) 62 followed by projected future emissions under each SSP scenario. Cumulative historical emissions are 345 63 Gg Hg to the atmosphere, and 1127 Gg Hg to land and water. Land and water releases are categorized as 64 mining (810 Gg) or non-mining (326 Gg). In each simulation year (*t*), the input vector, *s*(*t*), was updated 65 to contain both geogenic sources and anthropogenic emissions.

66

Mining releases to land/water were allocated to terrestrial reservoirs, with 5% to the slow pool and 95%
to the armored pool. All other releases followed the parameterization described in Streets et al. (2017)
with 40% assumed to be sequestered and considered unavailable for active cycling. The remaining 60%
was allocated among the new fast, slow and protected waste pools according to the relative carbon
distribution in the analogous soil pools from the Global Terrestrial Mercury Model (Smith-Downey et al.,

Anthropogenic release magnitudes are linearly interpolated from the original decadal time
 resolution for use in the model.

75 Table S1. Present-day reservoirs and fluxes used to calculate first-order rate coefficients in

76 **11-box model of Hg global biogeochemical cycling.**

Atmosphere (ATM): 4.0 Gg ^a	Flux (Mg a ⁻¹)
Hg ^{II} deposition to ocean	3900 ^a
Hg ^o uptake by ocean, gross	2000 ^a
Hg ^{II} deposition to land	1600 ^a
Hg ⁰ deposition to land	1200ª
Surface Ocean (OCS): 2.9 Gg ^b	
Hg ⁰ gross evasion to the atmosphere	4800 ^a
Hg ⁰ net evasion	2800 ^a
Particle settling to intermediate ocean	3300 ^b
Gross detrainment flux to intermediate ocean	5100 ^b
Intermediate Ocean (OCI): 130 Gg ^c	
Particle settling to deep ocean	600
Upwelling intermediate to surface	7100 ^c
Downwelling intermediate to deep	340 ^c
Deep Ocean (OCD): 220 Gg ^c	
Particle settling; burial to deep sediment	210 ^c
Upwelling deep to intermediate	180 ^c
Fast Terrestrial (TF): 9.6 Gg ^d	
Evasion due to heterotrophic respiration of SOM	40 ^e
Photoreduction	80 ^e
Biomass burning; fast to atmosphere	290 ^f
Riverine export to surface ocean	110 ^g
Riverine export to continental margin sediment	600 ^g
Decomposition; fast to slow	330 ^d
Decomposition and mineral stabilization; fast to protected	10 ^d
Slow Soil (TS): 35 Gg ^d	
Evasion due to heterotrophic respiration of SOM	20 ^e
Biomass burning; slow to atmosphere	8 ^f
Riverine export to surface ocean	3 ^g
Riverine export to continental margin sediment	17 ^g
Decomposition; slow to fast*	210 ^d
Decomposition and mineral stabilization; slow to protected	0.5 ^d
Protected Soil (TP): 190 Gg ^{d,**}	
Evasion due to heterotrophic respiration of SOM	3 ^e
Biomass burning; protected to atmosphere	4 ^f
Riverine export to surface ocean	2 ^g
Riverine export to continental margin sediment	10 ^g
Decomposition; protected to fast*	20 ^d
External Inputs	
Subaerial volcanism emissions to atmosphere	230 ^h
Submarine volcanism emissions to deep ocean	50 ⁱ
Anthropogenic emissions to air, land, water	$f(t)^{j}$

First-order rate coefficients, k, are calculated as $k_{ij} = F_{ij}/m_i$, where F_{ij} and m_i are the fluxes (Mg a⁻¹) and reservoir sizes (Gg) provided in the table above.

79 ^a Shah et al. (2021)

80 ^b Soerensen et al. (2010)

81 ° Sunderland & Mason (2007)

82 ^d Smith-Downey et al. (2010)

83 e Amos et al. (2014). Values reflect downward revision of terrestrial evasion fluxes relative to Smith-Downey et al. (2010) based

84 on observations and empirical models suggesting greater retention of deposited Hg than previous estimates (Hararuk et al.,

85 2013; Obrist, 2012; Obrist et al., 2014).

86 ^f Total biomass burning is 300 Mg a⁻¹ (Holmes et al., 2010) of which 95% is estimated to come from vegetation and 5% from the

soil pools based on their carbon content (Smith-Downey et al., 2010).

88 g Based on present-day "background" global river discharge to terrestrial margin of 740 Mg a⁻¹ from Amos et al. (2014), and

89 assumption that 16% of riverine reaches open ocean based on estimate that all dissolved riverine Hg reaches the open ocean

90 (9% of total) and 6-7% of particulate riverine Hg reaches the open ocean (Zhang et al., 2015). Riverine Hg not reaching the open

- 91 ocean is buried in continental margin sediment. Riverine Hg fluxes are sourced from terrestrial pools in the same manner as
- 92 biomass burning, with 95% is estimated to come from vegetation and 5% from the soil pools based on their carbon content
- 93 (Smith-Downey et al., 2010).
- ^h Geyman et al. (2023)
- 95 Lamborg et al., (2006)
- 96 ^j Function of time; see Methods in main text and Text S1 for more details.
- 97 * Transfer of soil Hg from longer-lived to shorter-lived reservoirs represents processes such as priming and changes in the
- 98 degree of mineral stabilization of soil organic matter.
- 99 ** Soil Hg reservoirs are based on global estimates made for organic soils using the Global Terrestrial Mercury Model (GTMM;
- 100 Smith-Downey et al., 2010), a mechanistic global model facilitating self-consistent internal transfer and external Hg fluxes for
- 101 the terrestrial biosphere. Large-scale geochemical soil survey results suggest greater total Hg mass in global topsoil than in the
- 102 GTMM (Ballabio et al., 2021; Olson et al., 2022), but more work is needed to understand the implications of these findings for
- 103 the terrestrial Hg budget.
- 104

C	Compartment To (<i>i</i>)		Rate (a ⁻¹)*
	Terrestrial Fast	!	5.00×10^{-1}
	Terrestrial Slow	:	1.28×10^{-1}
Te	errestrial Protected	-	7.20 × 10 ⁻²
	Ocean Surface		$1.47 imes 10^{\circ}$
	Atmosphere	4	4.36 × 10 ⁻²
	Terrestrial Slow	:	$3.44 imes 10^{-2}$
Te	errestrial Protected	:	1.04 × 10 ⁻³
	Ocean Surface		1.24 × 10 ⁻²
lar	gin Sediment Burial	(5.16 × 10 ⁻²
	Atmosphere	9	9.43 × 10 ⁻⁴
	Terrestrial Fast	(5.00 × 10 ⁻³
Te	errestrial Protected	:	1.43 × 10 ⁻⁵
	Ocean Surface	5	8.97 × 10 ⁻⁵
lar	gin Sediment Burial	4	4.45 × 10 ⁻⁴
	Atmosphere		3.68 × 10 ⁻⁵
	Terrestrial Fast	-	7.89 × 10 ⁻⁵
	Ocean Surface	(9.01 × 10⁻ ⁶
lar	gin Sediment Burial	4	4.47 × 10 ⁻⁵
	Atmosphere		1.66×10^{0}
C	Ocean Intermediate		2.90×10^{0}
	Ocean Surface	ļ	5.46 × 10 ⁻²
	Ocean Deep	-	7.23 × 10 ⁻³
C	Ocean Intermediate	5	$8.18 imes 10^{-4}$
lari	ine Sediment Burial	9	9.55 × 10 ⁻⁴
	Atmosphere	4	4.36 × 10 ⁻²
	Terrestrial Slow	3	3.44 × 10 ⁻²
Τe	errestrial Protected		1.04 × 10 ⁻³
	Ocean Surface	:	1.24 × 10 ⁻²
lar	gin Sediment Burial	(5.16 × 10 ⁻²
	Atmosphere	(9.43 × 10 ⁻⁴
	Terrestrial Fast	(5.00 × 10 ⁻³
Te	errestrial Protected		1.43 × 10 ⁻⁵
	Ocean Surface		8.97 × 10 ⁻⁵
lar	gin Sediment Burial	4	4.45 × 10 ⁻⁴
	Atmosphere		3.68×10^{-5}
	Terrestrial Fast	-	7.89 × 10⁻⁵
	Ocean Surface	(9.01 × 10⁻ ⁶
lar	gin Sediment Burial	4	4.47 × 10 ⁻⁵

105	Table S2. Rate coefficients used in global biogeochemical box model.
100	

Compartr	nent From (i)	Compartment To (j)	Rate (a ⁻¹) [*]
Wa	ste Immobile	Atmosphere	1.00×10^{-20}

^{*}Rates are calculated from the sum of fluxes in cases where mass transfer between a pair of reservoirs

107 (*i,j*) is composed of multiple fluxes.

Constraint Description	Model Value	Reference Value
Mass Hg in modern troposphere (Gg)	3.9	4.0ª
Upper ocean Hg concentration (0-1500 m) (pM)	1.2	$0.8 - 1.8^{b}$
Deep ocean (>1500m) Hg concentration (pM)	1.2	1.1 – 1.7 ^b
Pre-industrial to modern atmospheric Hg deposition enrichment factor (1840-20 th century maximum)	4.1	3 – 5°

109 Table S3. Global Biogeochemical Box Model Evaluation

110 ^a Shah et al. (2021)

111 ^b (Lamborg et al., 2014; Sunderland & Mason, 2007)

112 ^c (Fitzgerald et al., 2005; Li et al., 2020; Sonke et al., 2023)

	1-2.6	2010	2030	2050	2100	2150	2200	2250
	NAM	109.3	79.4	44.8	12.8	8.8		
	SAM	263.6	253.9	153.4	9.8	6.7		
	EUR	85.7	30.7	16.7	7.0	4.8		
	FSU	86.5	107.7	53.9	9.3	6.4		
	AFM	383.2	363.4	251.1	21.1	14.5		
	ASA	1255.5	1115.3	597.8	90.9	62.6		
	OCA	25.6	15.3	8.3	3.5	2.4		
	GLO	2209.4	1965.8	1126.0	154.3	106.3	0	0
	2-4.5	2010	2030	2050	2100	2150	2200	2250
	NAM	109.3	92.2	61.4	15.8	10.5	5.3	
	SAM	263.6	261.0	159.9	13.4	8.9	4.5	
	EUR	85.7	37.9	26.2	8.6	5.7	2.9	
	FSU	86.5	124.2	74.8	11.1	7.4	3.7	
	AFM	383.2	388.1	293.7	34.4	23.0	11.5	
	ASA	1255.5	1325.3	841.0	104.9	69.9	35.0	
	OCA	25.6	18.9	13.1	4.3	2.9	1.4	
	GLO	2209.4	2247.7	1470.2	192.5	128.3	64.2	0
	5-3.4	2010	2030	2050	2100	2150	2200	2250
	NAM	109.3	99.8	44.7	9.5	6.3		
	SAM	263.6	270.7	155.6	7.4	4.9		
	EUR	85.7	41.8	16.5	5.2	3.5		
	FSU	86.5	123.1	52.7	5.4	3.6		
	AFM	383.2	452.0	278.3	17.3	11.5		
	ASA	1255.5	1568.6	678.0	59.0	39.1		
	OCA	25.6	20.9	8.2	2.6	1.7		
	GLO	2209.4	2576.9	1234.0	106.6	70.6	0	0
_	5-8.5	2010	2030	2050	2100	2150	2200	2250
	NAM	109.3	81.4	82.5	38.5	21.5	10.8	
	SAM	263.6	264.7	184.7	31.4	17.6	8.8	
	EUR	85.7	31.2	37.2	21.0	11.8	5.9	
	FSU	86.5	80.9	103.7	33.4	18.7	9.3	
	AFM	383.2	428.7	527.9	422.9	236.9	118.4	
	ASA	1255.5	1198.8	1155.8	356.7	199.8	99.9	
	OCA	25.6	15.6	18.6	10.5	5.9	2.9	
	GLO	2209.4	2101.2	2110.3	914.3	512.1	256.1	0
•								

113 Table S4. Mercury emissions by region for the base year and key future years (Mg a⁻¹)

114 **Region names**: NAM = North America, SAM = South America, EUR = Western Europe, FSU = Former

115 Soviet Union, AFM = Africa and the Middle East, ASA = Asia, OCA = Oceania.

-		-	-			•	-
2250	2200	2150	2100	2050	2030	2010	1-2.6
		83.7	121.5	445.3	556.9	491.1	Mining and Industry
		0	0	457.3	726.8	726.8	Artisanal Gold Mining
		3.5	5.1	79.6	375.2	538.2	Coal Combustion
		2.5	3.7	13.0	17.1	14.5	Oil Combustion
		16.6	24.1	130.8	289.8	438.9	Other
0	0	106.3	154.3	1126.0	1965.8	2209.4	Total
2250	2200	2150	2100	2050	2030	2010	2-4.5
	40.5	81.0	121.5	445.3	556.9	491.1	Mining and Industry
	0	0	0	457.3	726.8	726.8	Artisanal Gold Mining
	3.6	7.3	10.9	372.0	584.7	538.2	Coal Combustion
	5.6	11.2	16.9	23.9	20.9	14.5	Oil Combustion
	14.4	28.9	43.3	171.7	358.4	438.9	Other
0	64.2	128.3	192.5	1470.2	2247.7	2209.4	Total
2250	2200	2150	2100	2050	2030	2010	5-3.4
		56.7	85.6	443.4	565.3	491.1	Mining and Industry
		0	0	457.3	726.8	726.8	Artisanal Gold Mining
		0.5	0.8	187.8	888.4	538.2	Coal Combustion
		4.9	7.4	22.7	18.4	14.5	Oil Combustion
		8.5	12.8	122.8	378.0	438.9	Other
0	0	70.6	106.6	1234.0	2576.9	2209.4	Total
2250	2200	2150	2100	2050	2030	2010	5-8.5
	24.0	48.0	85.6	443.4	565.3	491.1	Mining and Industry
	0	0	0	457.3	726.8	726.8	Artisanal Gold Mining
	217.1	434.2	775.3	985.8	539.6	538.2	Coal Combustion
	4.2	8.5	15.2	32.7	28.5	14.5	Oil Combustion
	10.7	21.5	38.3	191.1	241.0	438.9	Other
0	256.1	512.1	914.3	2110.3	2101.2	2209.4	Total

117 Table S5. Mercury emissions by source category for the base year and key future years (Mg a⁻¹)



120 **Figure S1. Trends in the fraction of anthropogenic emissions redepositing to region of origin.** Lines represent the

range among individual world regions and points represent the fraction of total anthropogenic emissions that redeposit to the region of origin. Values are separated by scenario (SSP1-2.6, SSP5-3.4, SSP2-4.5, SSP5-8.5) and

presented for selected years (2030, 2050, 2070, 2100). Values for the baseline year (2010) are shown at the top of

124 the figure in black, with the central value (vertical grey line) and regional range (grey shaded area) shown for

125 comparison. Trends are driven by changes in the speciation of anthropogenic emissions. As the percentage of total

126 Hg released as elemental Hg (Hg⁰) declines in anthropogenic Hg emissions to air, the fraction of anthropogenic

127 emissions redepositing to the region of origin increases. Note that the speciation of anthropogenic emissions

128 continues to shift towards lower Hg⁰ percentages beyond 2100 (Fig. 2).



129

130 Figure S2. Trajectories of atmospheric mercury (Hg) deposition from primary anthropogenic and legacy + natural

emissions. Changes in the emission drivers of deposition can be visualized over sequential decadal snapshots from 2010 (blue) to 2100 (red). Deposition is calculated over ice-free land surfaces, with total deposition being the sum

133 of deposition from legacy + natural emissions (y-axis) and deposition from primary anthropogenic emissions (x-

134 axis). Panels **a** – **d** represent different Shared Socioeconomic Pathway (SSP) scenarios. The background of each

panel is shaded to show the fraction of total deposition from legacy + natural sources. Inset annotations indicate

136 the change in deposition between 2010 and 2100 for each category. Note that all temporal change in the "legacy +

137 natural" deposition category is due to change in the legacy component because natural deposition is fixed.



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141 Figure S3. Change in atmospheric mercury (Hg) deposition from legacy emissions, 2010-2100. Patterns of

142 deposition for 2010 and 2100 are calculated based on emissions from the ocean and terrestrial biosphere, which

143 are quantified by forcing the global biogeochemical box model (GBBM) with historical (1510-2010) emissions from

144 Streets et al. (2019) and scenario-specific future emissions. Emissions from the ocean and terrestrial biosphere

145 include a "natural" component, though all change in the deposition shown here is attributable to change in legacy

emissions because natural emissions are fixed. Deposition change is calculated as the difference between 2100and 2010 values.



149 Figure S4. Scenario-specific mass trajectories of mercury (Hg) in the upper ocean (0-1500 m). Trajectories under

150 future emissions (2010-2300) are shown for SSPs 1-2.6 (blue), 2-4.5 (orange), 5-3.4 (yellow), and 5-8.5 (red), in

addition to a reference scenario in which primary anthropogenic emissions cease in 2010 (dashed black).





153 Figure S5. Mercury source-receptor matrices, by species. Matrix elements represent the fraction of a unit 154 emission from a given source (column) to a given receptor (row). Source-receptor matrices are presented for

155 emissions of gaseous elemental mercury (panels a and c) and gaseous oxidized mercury (panels b and d). Note that

156 the sum down an entire column in (a, c) and (b, d) is close to, but not quite 1, resulting from a small fraction of

- 157 deposition which occurs to global ice surfaces and a rapid re-emission process in the model. Also note the 158 differences in color scale between species.
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161 **References**

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