Global biogeochemical implications of mercury discharges from rivers and sediment burial

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Submitted to
Environmental Science & Technology

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Mercury (Hg) is a neurotoxin that cycles globally in the environment and bioaccumulates in aquatic food webs. Rivers link anthropogenic Hg releases on land to marine ecosystems. We estimate global present-day Hg discharges from rivers to ocean margins are $27 \pm 13 \text{ Mmol a}^{-1}$, of which 28% reaches the open ocean and the rest is deposited to ocean margin sediments. This is larger than previously estimated due to accounting for elevated concentrations in Asian rivers and variability in offshore transport across different types of estuaries. Inputs from rivers to the North Atlantic have decreased several-fold since the 1970s but increased to the North Pacific. Our analysis shows these trends in riverine input have large effects at ocean margins but are small offshore and thus do not explain observed declines in vertical seawater profiles in the North Atlantic or increases in the North Pacific. Burial in nearshore marine sediments represents a major sink in the global Hg biogeochemical cycle that has not previously been considered. We find that including this sink in a fully coupled global biogeochemical box model helps to accommodate the large anthropogenic release of Hg from commercial products recently added to global inventories. It also implies that natural environmental Hg levels are lower than previously estimated, suggesting a relatively larger impact from human activity and a potentially faster timescale (centuries instead of millennia) for resequestration of anthropogenic Hg from active cycling.
INTRODUCTION

Human exposure to methylmercury, a potent neurotoxin, is primarily through consumption of marine fish. Anthropogenic mercury (Hg) is transported globally by the atmosphere and the oceans, resulting in worldwide contamination. Most regulatory assessments for Hg have focused on atmospheric emissions and their subsequent fate in the global environment through deposition. On a global scale, anthropogenic releases of Hg to aquatic systems and the impact of rivers as a source of Hg to the marine environment have been comparatively understudied. Previous studies estimated that 15 Mmol a\(^{-1}\) of Hg is presently discharged from rivers to ocean margins, an amount comparable to atmospheric Hg deposition to the oceans (18 Mmol a\(^{-1}\)), and that 90% of this Hg is buried in sediments at ocean margins (estuaries and the continental shelf). Rivers have recently been hypothesized as important contributors to the Hg budgets of the North Atlantic and Arctic Ocean. Here we construct a global, spatially resolved inventory of Hg discharges from major rivers for present-day, including updated information on the fraction reaching the open ocean, and estimate trends between the 1970s and present. We use global Hg models to examine the impacts of rivers on the marine environment and biogeochemical cycling.

Cossa et al. first estimated the global discharge of from rivers was ~5 Mmol a\(^{-1}\). A subsequent study from Sunderland and Mason constructed a global inventory of present-day Hg discharges from rivers to major ocean basins (15 Mmol a\(^{-1}\)) using available data from a few major rivers. More recently, AMAP/UNEP produced a gridded version of this inventory. More than 80% of Hg in rivers is bound to particles (e.g., ref), which can be sequestered during burial of benthic sediments in estuaries and on the continental shelf before reaching the open ocean. Benthic sediments at ocean margins thus serve as a large sink for Hg. However,
the fraction of the suspended particle load in rivers that is buried is highly variable depending on freshwater discharge rates and the physical characteristics of different estuaries \(^{19}\). Here we use a classification scheme for export of particles from major estuarine types \(^{20}\) to better estimate the fraction of particle-bound Hg reaching the open ocean and the global biogeochemical implications of Hg sequestration in ocean margin sediments.

Vertical seawater profiles from the upper ocean (1000 m) near Bermuda indicate a large (>5 pM) decrease in Hg concentrations between 1983 and 2008 \(^{21-23}\), which Soerensen et al. \(^{10}\) postulated could be explained by a declining source of Hg from rivers. Sediment core data from Europe and North America support a decline in Hg discharges from many rivers since the 1970s due to decreases in commercial Hg use, environmental regulations, and wastewater treatment \(^{24-29}\). Conversely, sediment cores and inventories in China and India suggest that Hg discharges from rivers are increasing \(^{30-32}\). Seawater Hg concentrations in the North Pacific Ocean also appear to have increased between the late 1980s and 2006 \(^5\). Here we combine improved estimates of Hg discharges from rivers and their temporal trends with an ocean general circulation model to estimate the plausible role of rivers in driving Hg concentrations in different ocean basins between the 1970s and present.

METHODS

RIVERINE DISCHARGE OF Hg TO THE OCEANS

We construct a global, spatially distributed estimate of present-day riverine inputs to coastal margins using published measurements collected at or near river mouths (Table S1). We restrict observations to the year 2000 and later, with the exception of the Ob, Lena, and Yenisey rivers in the Russian Arctic \(^{33}\) and Po, Rhone, and Nile rivers in the Mediterranean \(^{34}\) where data
are only available from the early 1990s. Since measurements of Hg in rivers are limited, we aggregate data by continent and ocean basin to calculate flow-weighted mean dissolved concentrations \([\text{Hg(D)}]\) for rivers draining into each ocean basin (Table 1). Fewer measurements of suspended particle Hg concentrations are available, so mean concentrations \([\text{Hg(P)}]\) are estimated from a partition coefficient \(K_D = \frac{[\text{Hg(P)}]}{[\text{Hg(D)}]}\), where \([\text{Hg(P)}]\) is in units of pmol per kg of suspended particulate matter and \([\text{Hg(D)}]\) is in units of pmol per L of river water. From the compilation of data in Table S1 we calculate \(\log_{10} K_D = 4.7 \pm 0.3\). \([\text{Hg(P)}]\) values estimated using this \(K_D\) fall within observed ranges (Table 1).

We multiply the resulting mean riverine \(\text{Hg(D)}\) and \(\text{Hg(P)}\) concentrations by gridded freshwater discharge and suspended sediment data to estimate annual Hg loads entering coastal marine systems (Table 2). Data on total suspended solids (TSS) loads have a 2°x2.5° horizontal resolution \(^{35-37}\) and freshwater discharge data \((Q)\) have a 1°x1° horizontal resolution \(^{38}\).

The fraction of \(\text{Hg(P)}\) transported beyond the continental shelf to open marine waters varies among ocean basins and is estimated from a classification system for sediment dispersal developed by Walsh and Nittrouer \(^{20}\) and adapted by Zhang et al. \(^{39}\). Walsh and Nittrouer \(^{20}\) categorized estuaries into five types of dispersal systems based on river, wave, tide, and margin characteristics. Zhang et al. \(^{39}\) used data on suspended sediment export for representative rivers in each type to estimate the fraction of particle-bound Hg which reaches the open ocean.

We also considered point sources located on coastlines and discharging Hg directly to coastal waters. We isolated coastal grid cells in the AMAP/UNEP \(^6\) global 2010 inventory of anthropogenic releases to water (0.5° x 0.5° horizontal resolution) and discarded any cells overlapping with river mouths to avoid double counting with our riverine Hg source. Although point source discharges can be important locally (e.g., Minamata Bay \(^{40}\)), we find that coastal
point source discharges are negligible globally (<0.01 Mmol $a^{-1}$) relative to rivers and do not discuss them further.

Temporal changes in Hg inputs from rivers to estuaries between the 1970s and present are estimated using dated sediment core data from estuarine river mouths. Time series of Hg concentrations in estuarine river mouths are used as available. In India, sediment core and time series data are unavailable, so we estimate the change in recent decades based on country-specific inventory data. All available observations and associated temporal patterns are provided in the SI. Table 3 summarizes regional enrichment factors (EFs) and their upper and lower bounds used to scale present-day discharges. We assume Hg discharges have been constant at present-day levels since the 1970s in regions with no temporal information.

**Three-dimensional ocean model**

We use the MIT global three-dimensional ocean general circulation model (MITgcm) to examine the impact of 1970s-present changes in riverine discharges on seawater Hg concentrations. The Hg simulation in the MITgcm was developed by Zhang et al. It includes air-sea exchange, redox reactions, and sorption to particles following Soerensen et al. in the surface mixed layer and Zhang et al. in subsurface and deep waters. Particle dynamics driving settling of organic carbon (and Hg) are from the biogeochemical/ecosystem model embedded within the MITgcm. The horizontal resolution is $1^\circ \times 1^\circ$ with 23 vertical levels between the ocean surface and the seafloor. The physical circulation model has been constrained to be consistent with altimetric and hydrographic observations (the Estimating Circulation and Climate of the Ocean (ECCO) state estimates). To isolate the impact of rivers, we initialize seawater Hg concentrations at zero and shut off atmospheric deposition. We then perform two separate
10-year simulations, one forced by present-day Hg discharges from rivers and the other by 1970s inputs. Conclusions drawn from the MITgcm remain the same with 20-year simulations.

**Global biogeochemical box model**

We examine the longer-term impact of rivers on the full global biogeochemical cycle of Hg using an updated version of the 7-box model developed by Amos et al. Mercury is cycled between reservoirs representing the ocean (surface, subsurface, deep), atmosphere, and terrestrial environment (fast, slow, armored pools), and is ultimately removed by burial of marine sediments. Sediments are compacted and subducted to the lithosphere, eventually returning Hg to surface reservoirs by erosion and volcanism on geologic time scales. Exchange of mass between reservoirs is described by first-order rate coefficients. The model is initialized from a natural steady-state simulation without anthropogenic Hg releases, then forced with all-time (2000 BC to 2008 AD) anthropogenic atmospheric emissions from Streets et al. and additional 1850-2008 atmospheric emissions from commercial Hg use. We decrease the terrestrial rate coefficients for loss to the atmosphere via photoreduction and microbial respiration of organic matter by a factor of 10, based on recent field data indicating greater Hg retention by soils.

Amos et al. treated riverine discharges as a first-order process transferring Hg from the terrestrial reservoirs to the surface ocean. The rate coefficient was based on the estimated mass of Hg from rivers that reaches the open ocean (1.9 Mmol a\(^{-1}\)) from Sunderland and Mason. A term representing sequestration in ocean margin sediments was not included, so this Hg was stored in biogeochemically active reservoirs. We account for burial in ocean margin sediments here, which represents an important sink for anthropogenic Hg.
We also further characterize river discharges of Hg to ocean margins by distinguishing between Hg directly released to a river or its watershed by human activity ("primary anthropogenic Hg") versus terrestrial runoff of atmospherically deposited or naturally occurring Hg ("background"). A present-day background is estimated based on Hg concentrations measured in pristine Arctic rivers (Table S1; 15,52,53). Extrapolated globally this amounts to 3.7 Mmol a\(^{-1}\), which is comparable to Kocman et al. 54 (range, 0.85-2.5 Mmol a\(^{-1}\)). From this we infer a first-order rate coefficient for loss of Hg from terrestrial ecosystems to ocean margins by rivers. Between the 1970s and present total Hg discharges to ocean margins from rivers are constrained by observations (Tables 2-3). Prior to 1970, we scale total Hg discharges from rivers using the historical inventory of global releases to water from commercial Hg use 48. The primary anthropogenic contribution is calculated by difference between total and background discharges and treated as an external forcing. Of the total Hg discharged to ocean margins by rivers, 28% is transferred to the surface ocean and 72% removed to ocean margin sediments based on Table 2. All rate coefficients for the updated box model are provided in Table S2.

RESULTS & DISCUSSION

Global riverine inputs and trends

Global present-day Hg inputs from rivers to coastal margins are estimated to be 27 ± 13 Mmol a\(^{-1}\) (Table 2). This is larger than the previous estimates of 15 Mmol a\(^{-1}\) from Sunderland and Mason 8 and 5 Mmol a\(^{-1}\) from Cossa et al. 14 and is driven by recently published data suggesting greater contamination in Asian rivers 30,31,55-59. Riverine Hg inputs to the margins of the Pacific and Indian Oceans account for 80% of our global total (Table 2). Our estimates of
discharges to ocean margins of the Atlantic, Arctic, and Mediterranean are within the ranges of previously published values \(^8, 11, 13, 34, 60, 61\).

Figure 1 illustrates the spatial distribution of our present-day riverine inputs of total Hg to ocean margins, which is driven by both variability in Hg(P) concentrations (Table 1) and suspended sediment discharges \(^35\). Major rivers and highly contaminated systems are prominent (e.g., Yangtze, Amazon, Ganges). Large Hg discharges from Mexican rivers to the Pacific Ocean reflect high TSS values. Discharges from remote Asian rivers draining to the North Pacific Ocean in Figure 1 are based on observations from major Chinese rivers and may be overestimated. Figure 1 shows low Hg discharges from African rivers, based solely on data for the Nile \(^34\) and might be an underestimate. For example, the Congo River is known to be polluted by unregulated discharges from chemical industries, leaching from solid waste dumps, and artisanal gold mining \(^6, 62\).

We estimate that 28% of particle-bound Hg in rivers is exported to the open ocean globally (Table 2), a substantial increase from the 10% suggested in Sunderland and Mason \(^8\). The export fraction is higher for the North Pacific (29%) than for the North Atlantic (11%) due to a greater prevalence of large rivers that efficiently transport suspended particles containing Hg offshore \(^63, 64\). We find that 7.5 ± 3.7 Mmol a\(^{-1}\) of Hg discharged by rivers is delivered to the open ocean. The uncertainty is estimated by adding the uncertainties in discharge at coastal margins for individual ocean basins in quadrature, scaled by the corresponding export fractions (Table 2). Our best estimate (7.5 Mmol a\(^{-1}\)) amounts to ~40% of present-day atmospheric Hg(II) deposition to the oceans (18 Mmol a\(^{-1}\)) \(^9\).

Discharges of Hg are decreasing in North American and European rivers but increasing in India and China. Mercury discharges from rivers to the margins of the North Atlantic Ocean
peaked around the 1970s, likely due to the large quantities of Hg used and released from commercial products and industrial manufacturing at that time\textsuperscript{48,65-67}. Sediment core data suggest that Hg discharges from rivers bordering the North Atlantic in the 1970s were a factor of 9 (range, 4 to 20) larger than at present (Table 3). In India and China, riverine Hg discharges to the marine environment have increased by 40-400\% since the 1970s based on sediment core data and country-level inventories of Hg releases to water\textsuperscript{30,32,68}. The increase is likely driven by dense development and urbanization along major rivers\textsuperscript{30,69,70}, greater use of Hg in industrial processes (e.g., in vinyl chloride monomer production)\textsuperscript{71,72}, and increasing agricultural application of Hg-containing phosphate fertilizers\textsuperscript{73}.

Figure 2 shows the changes in riverine contributions to total dissolved Hg in the surface ocean (0 to 55 m) as simulated by the MITgcm given 1970s and present discharges. Results for subsurface waters (100 to 500 m) are shown in Figure S2. Concentrations of Hg in seawater have decreased throughout the North Atlantic but increased over much of the North Pacific. The trends driven by rivers are large in coastal areas (>1 pM) but fall to less than 0.1 pM in the open oceans. Soerensen et al.\textsuperscript{10} suggested that decreasing inputs of Hg to the North Atlantic Ocean from rivers might explain the observed 5 pM decline in seawater Hg concentrations over a 1000 m vertical profile near Bermuda between 1979 and 2008\textsuperscript{22}. In the North Pacific Ocean, Sunderland et al.\textsuperscript{5} reported an increase of up to 0.5 pM in vertical profiles from the North Pacific measured between 1987 and 2006. Our simulation shows that rivers alone are insufficient to explain the magnitude of seawater trends in the North Atlantic or North Pacific. The trend in the North Pacific is likely attributable to atmospheric deposition\textsuperscript{5}, but the observed 5 pM decline near Bermuda is challenging to explain.
Broader biogeochemical implications

While the 3D ocean model provides the spatially resolved response of seawater Hg concentrations to changes in Hg discharges from rivers, we use a box model to examine the impacts over longer timescales and across all geochemical reservoirs. In our updated global biogeochemical box model, cumulative anthropogenic Hg forcings since 1850 include 1.6 Gmol of Hg emitted to the atmosphere (1.1 Gmol from ref 47, and an additional 0.55 Gmol from ref 48), plus 1.9 Gmol of primary anthropogenic Hg in river discharge. Sequestration of Hg in ocean margin sediments in the model provides a sink of 1.3 Gmol for anthropogenic Hg since 1850. Thus we estimate that ocean margin sediments have sequestered up to one third of post-1850 anthropogenic Hg releases. Removal of Hg to ocean margin sediments and decreased soil re-emissions helps balance the increase in anthropogenic sources. Our simulated present-day atmosphere is 27 Mmol (compared to 23-28 Mmol supported by observations 46), the upper ocean (0-1500 m) Hg concentration is 1.7 pM (compared to the range in mean total Hg across ocean basins 0.6-2.9 pM 22), and storage in organic soils is 1.2 Gmol (compared to >1.5 Gmol from Hararuk et al. 49). Our analysis suggests that further increasing modeled Hg retention in terrestrial reservoirs requires a revision of the rates of Hg exchange between soil pools.

Figure 3 shows the time-dependent fate of a pulse of Hg emitted to the atmosphere in our updated model and then cycled through the different model reservoirs. We find a characteristic time scale of centuries for removal to ocean margin sediments, versus thousands of years for removal to deep ocean sediments, so that ocean margin sediments are the dominant long-term sink of Hg. The Hg pulse cycles among surface reservoirs for decades through the legacy of storage in the subsurface ocean. Unlike in Amos et al. 46, the deep ocean does not become a
dominant reservoir over centurial time scales because of the competing sink from ocean margin sediments.

Figure 4 shows the future recovery of the atmosphere and ocean under a hypothetical scenario of zero releases of primary anthropogenic Hg after 2015. The effect of sequestration in ocean margin sediments is illustrated by comparing model results with and without this sink, where the latter is more similar to the original model version in Amos et al.\textsuperscript{46}. Sequestration in ocean margin sediments in our updated model hastens recovery in the atmosphere, surface, and subsurface ocean over the 21\textsuperscript{st} century. The subsurface ocean decreases by 50\% by 2100, as compared to 35\% when we do not account the sediment sink (Figure 4), and 25\% in Amos et al.\textsuperscript{46}. Figure 4 also shows that the difference between simulations widens over time due to legacy Hg being more efficiently removed from active cycling.

Accounting for the loss of Hg to ocean margin sediments lowers the simulated steady-state natural budget of Hg in all reservoirs. This increases the relative perturbation from human activity and suggests an all-time enrichment in surface reservoirs possibly twice as large as previously estimated. Simulated natural deposition is 0.8 µg m\textsuperscript{-2} a\textsuperscript{-1}, which falls within the range of background deposition (0.6 – 1.7 µg m\textsuperscript{-2} a\textsuperscript{-1}) estimated from multi-millennia peat archives\textsuperscript{74}. However, all-time enrichment is sensitive to changes in the efficiency of long-term storage in terrestrial soils and sequestration in ocean margin sediments. Benthic sediments at ocean margins have been highly perturbed by activities such as coastal development, dredging, and trawling\textsuperscript{75}. Such disturbance would prolong the biogeochemical lifetime of anthropogenic Hg. Better information is needed on the long-term fate of Hg in benthic ocean margin sediments.
Acknowledgements

The US National Science Foundation Divisions of Atmospheric Chemistry (ATM0961357) and Chemical Oceanography (OCE1130549) supported this work. HMA acknowledges support from NSF GFRP. JSI thanks the support from the 7FP GMOS and ARRS program P1-0143. We thank Anne Soerensen, David Krabbenhoft, Mark Brigham, and Rob Mason for their thoughtful discussion. HMA acknowledges use of NCL software version 6.1.2 (http://www.ncl.ucar.edu/) to create Figures 1, 3, and 4.
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## Table 1. Mean present-day riverine Hg concentrations

<table>
<thead>
<tr>
<th>Ocean Basin</th>
<th>[Hg(D)] (pM) mean ± SE (range)</th>
<th>[Hg(P)] (nmol g⁻¹)</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Arctic</strong></td>
<td></td>
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<tr>
<td>Canada and USA</td>
<td>7.3 ± 2.1 (2.5-17)</td>
<td>0.39 ± 0.35 (0.01-0.63)</td>
<td>7</td>
</tr>
<tr>
<td>Russia</td>
<td>3.1 ± 1.1 (1.5-5.0)</td>
<td>0.17 ± 0.15 (0.03-0.30)</td>
<td>3</td>
</tr>
<tr>
<td><strong>North Atlantic</strong></td>
<td></td>
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<tr>
<td>Europe</td>
<td>9.1 ± 2.1 (0.10-390)</td>
<td>0.49 ± 0.42 (0.40-35)</td>
<td>3</td>
</tr>
<tr>
<td>Canada and USA</td>
<td>8.7 ± 5.7 (1.8-17)</td>
<td>0.47 ± 0.32 (0.40-8.9)</td>
<td>4</td>
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<tr>
<td><strong>South Atlantic</strong></td>
<td></td>
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<tr>
<td>South America</td>
<td>28 ± 18 (0.50-330)</td>
<td>1.5 ± 1.2 (0.20-2.5)</td>
<td>2</td>
</tr>
<tr>
<td>Africa</td>
<td>2.0 ± 1.3 (n/r)</td>
<td>0.11 ± 0.10 (n/r)</td>
<td>1</td>
</tr>
<tr>
<td><strong>North Pacific</strong></td>
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</tr>
<tr>
<td>Canada and USA</td>
<td>6.4 ± 3.2 (2.6-10)</td>
<td>0.34 ± 0.33 (0.55-7.7)</td>
<td>3</td>
</tr>
<tr>
<td>China</td>
<td>110 ± 55 (5-3000)</td>
<td>5.9 ± 5.8 (1.8-25)</td>
<td>3</td>
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<tr>
<td><strong>South Pacific</strong></td>
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<tr>
<td>Southeast Asia</td>
<td>3.6 ± 2.3 (3.2-55)</td>
<td>0.19 ± 0.19 (0.3-0.5)</td>
<td>1</td>
</tr>
<tr>
<td>South America</td>
<td>28 ± 18 (0.50-330)</td>
<td>1.5 ± 1.2 (0.60-2.5)</td>
<td>-</td>
</tr>
<tr>
<td>China</td>
<td>110 ± 55 (5-3000)</td>
<td>5.9 ± 5.8 (1.8-25)</td>
<td>-</td>
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<tr>
<td><strong>Indian</strong></td>
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<tr>
<td>India</td>
<td>50 ± 33 (56-370)</td>
<td>2.7 ± 2.9 (15-17)</td>
<td>1</td>
</tr>
<tr>
<td>Southeast Asia</td>
<td>3.6 ± 2.3 (3.2-55)</td>
<td>0.19 ± 0.19 (0.3-0.5)</td>
<td>-</td>
</tr>
<tr>
<td>Africa</td>
<td>2.0 ± 1.3 (n/r)</td>
<td>0.11 ± 0.10 (n/r)</td>
<td>1</td>
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<tr>
<td>Mediterranean</td>
<td>1.8 ± 0.8 (1.8-120)</td>
<td>0.10 ± 0.09 (0.30-25)</td>
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</tbody>
</table>

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**Notes:**

- Flow-weighted mean dissolved concentrations [Hg(D)] are based on a survey of published measurements collected in rivers flowing into estuaries. The complete compilation of observations is in Table S1. The standard error on the weighted mean (SE) is calculated when n is ≥ 3 and ranges from 30% to 65%. When n < 3 we assume SE = 65%. See text for estimation.
of [Hg(P)]. The SE of [Hg(P)] is calculated by propagating the relative SEs of [Hg(D)] and of the partition coefficient $K_D$ in quadrature. The ranges for both [Hg(D)] and [Hg(P)] are based on reported observations (Table S1).

Observations are aggregated by continental regions flowing into each ocean basin (indented). Ocean basin boundaries are given in Figure S1.

\(^{c}\) Per gram dry weight suspended sediment.

\(^{d}\) Number (n) of estuarine regions used to calculate the flow-weighted mean.

\(^{e}\) South American mean Hg concentrations are also applied to Central America.

\(^{f}\) Based on data for the Nile River \(^{34}\).

\(^{g}\) The original publication \(^{77}\) only provides a range. We use the mid-range value as estimate of the mean.

\(^{h}\) Based on observations for South American rivers entering the South Atlantic.

\(^{i}\) Based on observations for Chinese rivers entering the North Pacific.

\(^{j}\) Limited observations available suggest that rivers are highly contaminated ([Hg(D)] = 50 - 400 pM [Ram et al., 2003], and [THg] = [Hg(D)] + [Hg(P)] > 6000 pM \(^{59}\)). We use [Hg(D)] = 50 pM as a conservative estimate.

\(^{k}\) Based on observations for Southeast Asian rivers entering the South Pacific.

\(^{l}\) Based on rivers draining from the European continent and the Nile River (Table S1). Ranges not reported (“n/r”).
Table 2. Present-day discharges to ocean margins.

<table>
<thead>
<tr>
<th>Ocean</th>
<th>Hg(D) Reaching Fraction of Pre-satellite Total Discharge</th>
<th>(Pm a(^{-1}))</th>
<th>(Pm a(^{-1}))</th>
<th>Fraction of Hg(P) Reaching Open Ocean</th>
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<tbody>
<tr>
<td>Arctic</td>
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<td>Mediterranean</td>
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<td>Southern Ocean</td>
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<td>Global</td>
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</table>

a Geographical boundaries from Figure S1.

b Geographical boundaries from Figure S1.

c Based on the Walsh and Nittrouer 20 classification scheme for dispersal of suspended sediment and observations from rivers.

d Product of [Hg(D)] and Q, where [Hg(D)] is from Table 1 and has units of nmol a\(^{-1}\).

e Product of [Hg(P)] and TSS, where [Hg(P)] is from Table 1 and has units of pmol a\(^{-1}\).

f Based on the Walsh and Nittrouer 20 classification scheme for dispersal of suspended sediment and observations from rivers.

ghi Errors for individual ocean basins are added in quadrature to estimate the error on global total Hg(D) inputs.
Table 3. Enrichment factors relative to 2008 for riverine Hg inputs to oceans

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a Best estimates and observational ranges from estuarine sediment cores collected at the mouths of major freshwater tributaries and riverine Hg concentration time series. See SI for details.
Figure 1. Present-day annual discharges of total (dissolved + particulate) mercury to ocean margins from rivers.
Figure 2. The simulated change in the riverine contribution to annual mean dissolved Hg concentrations in the surface ocean (0 to 55 m) given 10 years of discharge representative of the 1970s and then present day. Model results are from a 3D ocean general circulation model (MITgcm) as described in the text. Red indicates an increase in oceanic Hg relative to present-day and blue indicates a decrease.
**Figure 3.** Time-dependent fate of a unit pulse of Hg emitted to the atmosphere at time $t = 0$ and then tracked by the model as it cycles between the ocean, atmosphere, and terrestrial reservoirs before eventually being removed to ocean margin sediments. This figure updates the top panel of Figure 6 of Amos et al.\(^{46}\).
Figure 4. Change in global reservoir masses of Hg relative to 2015 under a hypothetical scenario of zero releases of primary anthropogenic Hg after 2015. Results are shown for the updated biogeochemical box model with sequestration in ocean margin sediments (solid) and without (dashed).