

Multi-decadal decline of mercury in the North Atlantic atmosphere explained by changing subsurface seawater concentrations

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[1] We analyze 1977–2010 trends in atmospheric mercury (Hg) from 21 ship cruises over the North Atlantic (NA) and 15 over the South Atlantic (SA). We find a steep 1990–2009 decline of $-0.046 \pm 0.010 \text{ ng m}^{-3} \text{ a}^{-1}$ ($-2.5\% \text{ a}^{-1}$) over the NA (steeper than at Northern Hemispheric land sites) but no significant decline over the SA. Surface water Hg^0 measurements in the NA show a decline of $-5.7\% \text{ a}^{-1}$ since 1999, and limited subsurface ocean data show an $\sim 80\%$ decline from 1980 to present. We use a coupled global atmosphere-ocean model to show that the decline in NA atmospheric concentrations can be explained by decreasing oceanic evasion from the NA driven by declining subsurface water Hg concentrations. We speculate that this large historical decline of Hg in the NA Ocean could have been caused by decreasing Hg inputs from rivers and wastewater and by changes in the oxidant chemistry of the atmospheric marine boundary layer. **Citation:** Soerensen, A. L., D. J. Jacob, D. G. Streets, M. L. I. Witt, R. Ebinghaus, R. P. Mason, M. Andersson, and E. M. Sunderland (2012), Multi-decadal decline of mercury in the North Atlantic atmosphere explained by changing subsurface seawater concentrations, *Geophys. Res. Lett.*, 39, L21810, doi:10.1029/2012GL053736.

1. Introduction

[2] Mercury (Hg) is emitted to the atmosphere by natural processes (crustal degassing, volcanoes) and by human activities (fuel combustion, industry, mining). It deposits to the surface, cycles through ecosystems, may be re-emitted to the atmosphere, and is eventually incorporated into stable soil and deep ocean reservoirs [Corbitt *et al.*, 2011; Streets *et al.*, 2011]. Accumulation of mercury in ecosystems and subsequent human exposure to methylmercury through fish con-

sumption is a major environmental concern [Mergler *et al.*, 2007; Mahaffey *et al.*, 2011].

[3] Total gaseous mercury (TGM) or elemental gaseous mercury (Hg^0) has been measured on Atlantic ship cruises since the 1970s [Temme *et al.*, 2003] and at long-term land monitoring sites since the 1990s [Temme *et al.*, 2007; Slemr *et al.*, 2011; Toerseth *et al.*, 2012]. Slemr *et al.* [2011] found that atmospheric Hg decreased worldwide by 20–38% over the 1996–2009 period on the basis of long-term data from Mace Head (Ireland) and Cape Point (South Africa), as well as five Atlantic cruises. Statistically significant declines for the 1995–2009 period have also been reported for Arctic and eastern Canadian sites [Cole *et al.*, 2012]. These decreases are inconsistent with steady or increasing emissions from 1990 to present as indicated by anthropogenic Hg emission inventories [Wilson *et al.*, 2010; Streets *et al.*, 2011]. The long atmospheric lifetime of Hg (~ 0.5 years) suggests that atmospheric observations should reflect hemispheric emission trends [Corbitt *et al.*, 2011].

[4] Observed declines in atmospheric Hg could reflect a decrease in Hg re-emission from geochemical reservoirs to the atmosphere [Slemr *et al.*, 2011]. Oceanic Hg^0 evasion is comparable globally to anthropogenic emissions [Mason *et al.*, 2012]. Elevated Hg in subsurface seawater (below the mixed layer) of the North Atlantic (NA) was previously reported by Laurier *et al.* [2004] and attributed to the legacy of 20th-century anthropogenic sources in North America and Europe [Sunderland and Mason, 2007]. New subsurface data for the NA show that Hg concentrations have declined in recent years [Mason *et al.*, 2012]. Soerensen *et al.* [2010a] suggested that high surface air concentrations observed during NA cruises could be attributed to oceanic evasion of this legacy Hg^0 . Here we extend this idea to an examination of 1977–2010 Hg trends in the atmosphere and the surface ocean using the full ensemble of cruise data available for the NA and South Atlantic (SA).

2. North and South Atlantic Trends

[5] We analyzed atmospheric data from 21 ship cruises in the NA (5°N – 65°N), and 15 in the SA (70°S – 11°N) (Figure 1 and Table S1 in the auxiliary material) between 1977 and 2010.¹ There is a 1981–1989 gap for which we know of no cruise data. We separate the NA from the SA by the location of the Intertropical Convergence Zone (ITCZ) at the time of the cruise. Atmospheric Hg is measured as either $\text{TGM} \equiv \text{Hg}^0 + \text{Hg}^{\text{II}}$ or as Hg^0 . Hg^{II} accounts for only a few

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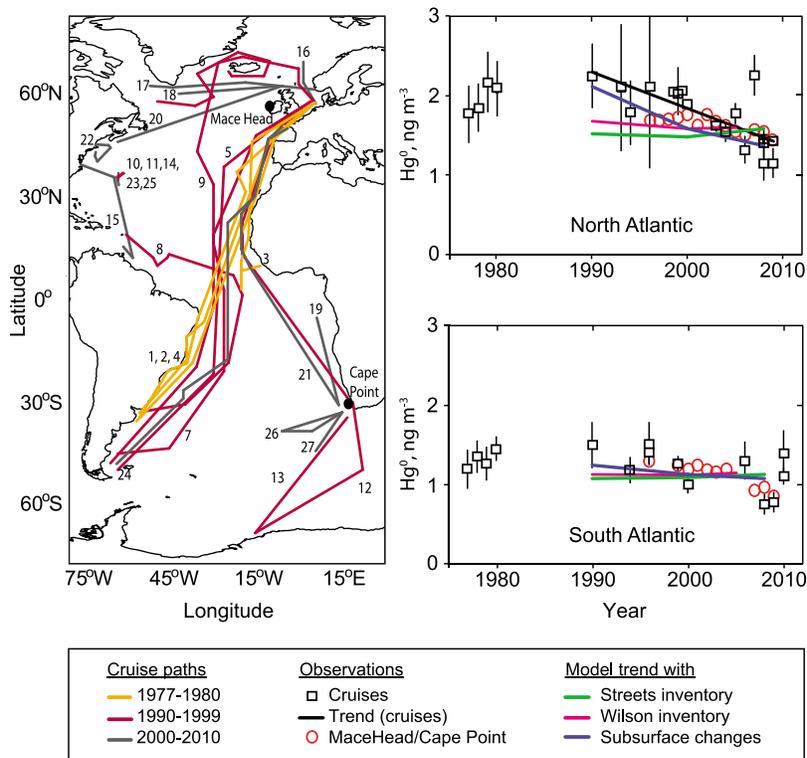


Figure 1. (left) Ship cruises used in the surface air Hg trend analysis. Cruise data are from *Temme et al.* [2003] (numbered on the map as 1, 2, 3, 4, 5, 7, 9, 12, 13); *Mason et al.* [1998] (6); *Lamborg et al.* [1999] (8); *Mason et al.* [2001] (10,11,14); *Laurier and Mason* [2007] (15); *Aspmo et al.* [2006] (16); *Sommar et al.* [2010] (17); *Soerensen et al.* [2010b] (18, 19, 20); *Kuss et al.* [2011] (21, 24); *Andersson et al.* [2008] (22); *Mason et al.* [2009] (23, 25); M. Witt (manuscript in preparation, 2012) (26); and R. Ebinghaus (unpublished data, 2010) (27). Hg concentration trends in surface air of the (top right) NA and (bottom right) SA. Squares represent mean concentrations measured on the cruises, with vertical bars indicating standard deviations. The regression line for the observed NA 1990–2009 trend is shown ($0.046 \pm 0.010 \text{ ng m}^{-3} \text{ a}^{-1}$, $p < 0.001$, $n = 18$). Also shown are GEOS-Chem model trends driven by changes in anthropogenic emissions or in the subsurface NA. Red circles are yearly medians for Mace Head (Ireland) and Cape Point (South Africa) taken from *Slemr et al.* [2011]. Further information on the cruises is in Table S1.

percent of TGM in remote surface air [*Gustin and Jaffe*, 2010], thus we do not distinguish between TGM and Hg^0 in our discussion.

[6] Figure 1 shows the 1977–2010 surface air concentrations for the ensemble of cruises, separately for the NA and the SA. Also shown are the 1996–2009 data for Mace Head and Cape Point. There are no significant trends between the late 1970s and the early 1990s for either the NA or SA. For 1990–2009, we find a statistically significant decrease in the NA of $-0.046 \pm 0.010 \text{ ng m}^{-3} \text{ a}^{-1}$ (\pm standard error) or $-2.5\% \text{ a}^{-1}$ ($p < 0.001$, $n = 18$), steeper than the 1996–2009 decrease reported by *Slemr et al.* [2011] for Mace Head ($-0.024 \pm 0.005 \text{ ng m}^{-3} \text{ a}^{-1}$). This decrease is apparent over both the West and East NA. Accounting for seasonality [*Soerensen et al.*, 2010b] does not change this result (see Text S1 and Figure S1).

[7] The 1990–2010 SA data show a weaker decrease with low statistical significance (-0.021 ± 0.010 , $p < 0.1$, $n = 11$). *Slemr et al.* [2011] previously reported a significant 1996–2009 decreasing trend of $-0.034 \pm 0.005 \text{ ng m}^{-3} \text{ a}^{-1}$ at Cape Point, corroborated by data from five SA ship cruises. However, these five cruises may not be representative of the ensemble of cruise data in the SA. In particular, the $< 1.0 \text{ ng m}^{-3}$ 2008–2009 cruise concentrations from *Kuss et al.*

[2011] (Figure 1, South Atlantic) that anchor the *Slemr et al.* [2011] trend are not consistent with three other 2006–2010 cruises that show higher values.

[8] *Slemr et al.* [2011] argued that the 1996–2009 decrease in Hg was equally strong in the Northern Hemisphere and Southern Hemisphere, mainly on the basis of the similarity between Mace Head, Cape Point, and their selected cruises. We find otherwise from the ensemble of cruise data, with a sharp contrast between the strong decreasing trend in the NA and the marginally significant trend in the SA. The NA decrease is also stronger than observed elsewhere in the Northern hemisphere over the past 20 years, including nearby continental sites in Ireland and eastern Canada (-0.023 to $-0.035 \text{ ng m}^{-3} \text{ a}^{-1}$ [*Ebinghaus et al.*, 2011; *Slemr et al.*, 2011; *Cole et al.*, 2012]) and at Arctic sites ($-0.009 \text{ ng m}^{-3} \text{ a}^{-1}$ at Alert, no trend at Svalbard [*Cole et al.*, 2012]).

[9] Surface seawater Hg^0 data were collected during five cruises in the Northwest Atlantic in 1998–2000 [*Mason et al.*, 2001] and five cruises in the same area in 2008–2010 [*Andersson et al.*, 2008; *Mason et al.*, 2009; R. P. Mason, unpublished data, 2010]. The data are given in Table S2. Observed mean concentrations were $244 \pm 100 \text{ fM}$ in 1998–2000 and $136 \pm 33 \text{ fM}$ in 2008–2010. This represents a significant decrease of $-5.7\% \text{ a}^{-1}$ during the period

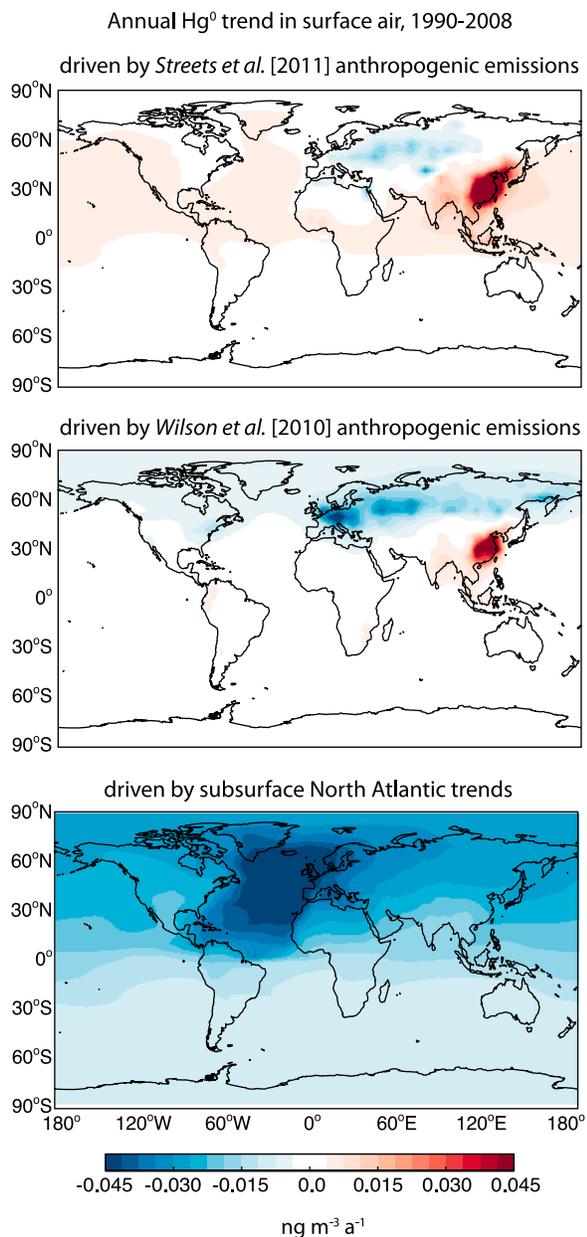


Figure 2. Simulated differences of Hg⁰ surface air concentrations in GEOS-Chem as driven by changes in anthropogenic emissions by *Streets et al.* [2011] (1990–2008) and by *Wilson et al.* [2010] (1990–2005) and subsurface North Atlantic (0°–70°N) seawater concentrations. Results are expressed as an annual trend (ng m⁻³ a⁻¹).

(t-test paired means difference, $p < 0.05$). This decline is even steeper than that in NA surface air over the same 1999–2009 period (surface air: -0.066 ng m⁻³ a⁻¹, -4.0% a⁻¹, $n = 14$). As we will see below, this is consistent with oceanic forcing of the atmospheric trend.

3. Cause of the North Atlantic Surface Air Decline

[10] We use the GEOS-Chem global biogeochemical mercury model (v 9-01-02; <http://www.geos-chem.org>) to investigate possible causes of the 1990–2009 Hg decline in the NA, including changes in emissions and in subsurface

ocean concentrations. The model is described by *Soerensen et al.* [2010a], *Holmes et al.* [2010], *Amos et al.* [2012], and *Zhang et al.* [2012]. It dynamically couples a 3-D atmospheric transport and chemistry model with a 2-D surface ocean (depth specified from observations) and land reservoir on a $4^\circ \times 5^\circ$ horizontal grid and with 1-h time steps. Here ‘surface ocean’ denotes the ocean mixed layer (annual global mean depth ~ 50 m). The surface ocean interacts with the atmosphere by air-sea exchange of Hg⁰ and Hg^{II}, and wet deposition of Hg^{II}. It interacts with subsurface waters through particle settling and vertical seawater transport but does not include lateral transport. Fixed subsurface ocean concentrations in individual basins are specified from observed vertical profiles extending from the bottom of the mixed layer to the top of the permanent thermocline [*Soerensen et al.*, 2010a]. Within the surface ocean, redox cycling between Hg⁰ and dissolved Hg^{II} is driven by photochemical and biological processes. We updated the simulation of aqueous-particle partitioning of Hg^{II} in the surface ocean [*Soerensen et al.*, 2010a] with a specific K_D (affinity of aqueous Hg^{II} for the solid phase) of 1×10^5 L kg⁻¹ for the NA based on observed values from *Mason et al.* [1998].

[11] Model Hg concentrations in the surface ocean and atmosphere respond to changes in emissions and subsurface ocean concentrations on a timescale of less than a year [*Corbett et al.*, 2011]. We can therefore diagnose the long-term trends in the model from changing emissions or subsurface ocean concentrations by conducting 1-year simulations (with four years of initialization) for decadal time slices. All simulations use the same meteorological year (2008) to remove the effect of interannual meteorological variability. We do this since long-term time series at Mace Head and Cape Point show little year-to-year variability (Figure 1), and GEOS-Chem Hg simulations driven by 30-year (1980–2010) assimilated meteorological data show no significant long-term trends driven by meteorology (J. A. Fisher, personal communication, 2012). Furthermore studies of CO₂ air-sea exchange in the North Atlantic show no significant decadal trends associated with climatic modes such as the, North Atlantic Oscillation and the El Niño - Southern Oscillation [*Bates*, 2001, 2007].

[12] According to the *Streets et al.* [2011] inventory, global anthropogenic Hg emissions increased by 30% between 1990 and 2008 but Hg^{II} emissions in the USA and Europe decreased by 20% and 40%, respectively. Large regional decreases in Hg^{II} emissions are also found in the *Wilson et al.* [2010] inventory where global emissions decrease by 2.5% between 1990 and 2005. Hg^{II} is removed regionally by deposition, in contrast to Hg⁰. To see if regional decreases in Hg^{II} emissions could explain the NA Hg⁰ trend we conducted GEOS-Chem simulations using 1990, 2000, and 2008 anthropogenic emissions from *Streets et al.* [2011] and 1990, 2000, and 2005 anthropogenic emissions from *Wilson et al.* [2010] with all else kept constant. As shown in Figure 2, Hg⁰ concentrations over the NA increase with the *Streets et al.* [2011] emission trend, as the regional decrease in Hg^{II} emissions is insufficient to compensate for the rise of Hg emissions in East Asia. Even with the essentially flat global emission trend of *Wilson et al.* [2010], the decline over the NA is far too weak to explain observations. GEOS-Chem includes fast in-plume reduction of Hg^{II} from power plant emissions [*Zhang et al.*, 2012], but

a sensitivity simulation without this in-plume reduction still does not reproduce the observed 1990–2008 decline over the NA.

[13] Total Hg in NA subsurface waters in the 1980s and 1990s was enriched relative to the North Pacific [Gill and Fitzgerald, 1988; Laurier et al., 2004; Mason et al., 2012] but appears to have declined sharply over the past decades. Vertical profile (50–1000 m) Hg concentrations measurements in the West Atlantic (BATS station, 32°N, 64°W) indicate mean values of 6.10 ± 2.35 pM in July 1979 and September 1983 [Gill and Fitzgerald, 1988], 1.95 ± 1.59 pM across several seasons in 1999–2000 [Mason et al., 2001], and 0.78 ± 0.40 pM in June 2008 [Mason et al., 2012] (Table S3). Similarly, Scotian Shelf inflow water measured by Dalziel [1992] in the mid-1980s had two-fold higher concentrations than those measured in 2002 [Sunderland et al., 2012]. These long-term trends are unlikely to be aliased by seasonal variability (less than 40% [Laurier et al., 2004; Mason et al., 2012]) or by interannual variability (which would then propagate to the atmospheric data) [Laurier et al., 2004; Mason et al., 2012]. Clean measurement techniques were established in the 1980s [Gill and Fitzgerald, 1987] and allow confidence in the trend [Mason et al., 2012].

[14] We simulated the effect of this subsurface ocean concentration trend in the NA by conducting GEOS-Chem simulations for 1990, 2000, and 2008, with all else constant including 2008 anthropogenic emissions from Streets et al. [2011] and subsurface ocean concentrations outside the NA from Soerensen et al. [2010a]. Subsurface ocean concentrations for the NA were specified based on the means of measurements at the BATS station (1.95 pM in 2000 and 0.78 pM in 2008) and a conservative estimate of 5 pM in 1990.

[15] Figure 2 shows that these changes in NA subsurface seawater concentrations cause a -0.043 ng m⁻³ a⁻¹ (-2.4% a⁻¹) mean model decline in surface air concentrations over the NA for 1990–2008, closely reproducing the observed trend of -0.046 ± 0.010 ng m⁻³ a⁻¹ (Figure 1). Simulated surface ocean Hg⁰ concentrations in the NA decline by -6.3% a⁻¹ between 2000 and 2008, consistent with the observed -5.7% a⁻¹ decline. The steeper decline observed in the NA surface ocean relative to the atmosphere is reproduced by the model, providing further support for subsurface ocean forcing of the atmospheric trend.

[16] Changing subsurface seawater Hg has a more pronounced impact on surface air Hg⁰ over the NA than over Europe, eastern North America and the Arctic (Figure 2), consistent with the observed trends mentioned previously. We reproduce the trends at mid-latitude terrestrial sites within an average of 30% [Ebinghaus et al., 2011; Cole et al., 2012]. The simulated 1990–2008 trend in SA surface air is much weaker than in the Northern Hemisphere (0.9% a⁻¹, Figure 1), consistent with cruise observations. A bootstrap analysis indicates that such a weak trend would not be statistically detectable from the cruise data given their variability (Figure 1).

4. Possible Causes of the North Atlantic Subsurface Hg Decline

[17] We find from the model that the 1990–2009 atmospheric decrease of Hg in the NA can be explained by the observed 80% Hg decline in the subsurface NA over the past two decades. However, the cause of this subsurface decline requires further consideration. A parallel can be drawn with

lead, which declined by 85% in the subsurface NA between 1979 and 2008 after leaded gasoline was phased out [Wu and Boyle, 1997; Lee et al., 2011]. Tracer data for NA subsurface waters indicate a mean age of 10–30 years since last contact with the atmosphere [Fine, 2010]. Subsurface Hg trends over the last 30 years are thus sensitive to trends in inputs going back 50 years to the 1960s. We speculate here on possible causes for the Hg subsurface decline.

[18] A first possibility would be a decrease in Hg^{II} deposition to the NA over the last 50 years, decreasing the reservoir of Hg in subsurface waters available for evasion. Large reductions in Hg^{II} emissions occurred in both North America and Europe over this period. However, sensitivity simulations of deposition in 1970 and 2000 based on the historical emission scenario from Streets et al. [2011] indicate less than 2% change in total deposition to the North Atlantic due to simultaneous increases from global sources. Most Hg deposition to the NA is thought to be driven by Hg⁰ oxidation by Br atoms in marine surface air [Hedgecock and Pirrone, 2004; Holmes et al., 2009]. Ozone in marine surface air has doubled over the NA in the past 50 years [Lelieveld et al., 2004; Parrish et al., 2009], which would cause a proportional decrease in Br atom concentrations by shifting the Br/BrO radical equilibrium toward BrO [Parrella et al., 2012]. Atmospheric deposition to the NA would have correspondingly decreased.

[19] Another possible explanation for the declining trend in NA seawater is decreased riverine and wastewater inputs at ocean margins. Peak river inputs in the 1970s reflected in sedimentary archives were 2–6 fold higher than present [e.g., Steinberg et al., 2004; Sunderland et al. 2010; Boutier et al., 2011; Elbaz-Poulichet et al., 2011]. Sunderland and Mason [2007] estimated that present-day riverine inputs to the Atlantic (in their model defined as 35°S–55°N) are approximately 25% of atmospheric deposition, suggesting that NA riverine inputs were comparable to atmospheric deposition in the 1970s. Declines in river inputs since the 1970s can be attributed to regulation of large industrial sources such as the chlor-alkali industry [Euro Chlor, 2005; Chlorine Institute, 2009] and phase-out of Hg in many consumer products that contributed to wastewater inputs [Cain et al., 2007]. Hg discharge from municipal wastewater into NA urban estuaries decreased by over 95% following implementation of secondary wastewater treatment in the 1980s and 1990s [Sunderland et al., 2012; NOAA, 1994; Wu, 2011].

[20] Regardless of the cause of declining oceanic Hg levels observed in the NA subsurface ocean since 1990, our results show that the resulting changes in NA Hg⁰ evasions are sufficient to explain the observed atmospheric trends over NA and adjacent continents. Declining subsurface oceanic Hg concentrations in the NA have major global implications for the atmospheric Hg budget, causing a modeled global decrease of -1300 Mg a⁻¹ in the Hg flux from the ocean to the atmosphere over the 1990–2008 period. This is much larger than the global change in anthropogenic emissions over the same period (-47 Mg a⁻¹ according to Wilson et al. [2010] or $+480$ Mg a⁻¹ according to Streets et al. [2011]). As previously hypothesized by Stenr et al. [2011], long-term trends in re-emission can overwhelm primary anthropogenic emissions in driving the atmospheric Hg trend.

[21] NA atmospheric concentrations are presently similar to the SA (Figure 1), implying that most of the historical

enrichment of subsurface Hg in the NA has been re-distributed globally or transferred to the deep ocean by now. Thus, we expect observed decreases in NA atmospheric Hg to slow down over the next decade and possibly reverse if global emissions rise. Our work highlights the role of the ocean in mediating the temporal response of the atmosphere to changes in anthropogenic emissions. It shows a need to better understand the contributions of effluent Hg releases from rivers and wastewater to the global biogeochemical cycling of Hg.

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