



# Historical Mercury Releases from Commercial Products: Global Environmental Implications

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1 **Historical mercury releases from commercial products: global environmental implications**

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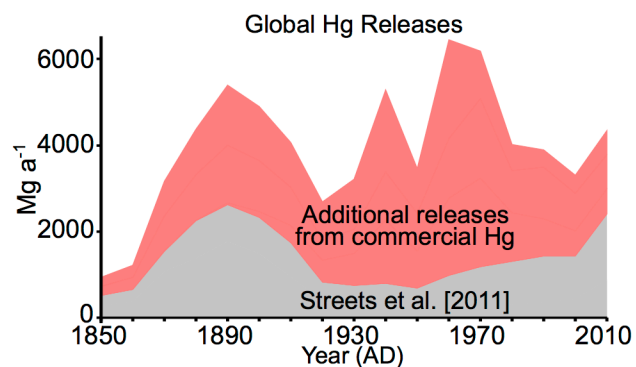
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## 17 ABSTRACT

18 Use and disposal of commercial products has contributed a large and previously unquantified  
 19 anthropogenic source of mercury (Hg) to the global environment over the industrial era, with  
 20 major implications for Hg accumulation in environmental reservoirs. We present a global  
 21 inventory of commercial Hg uses and releases to the atmosphere, water, soil, and landfills from  
 22 1850 to 2010. Previous inventories of anthropogenic Hg releases have focused almost  
 23 exclusively on atmospheric emissions. Cumulative anthropogenic Hg emissions since 1850 have  
 24 recently been estimated at 220 Gg. We find that commercial use of Hg released an additional 540  
 25 Gg to the global environment since 1850 (20% to air, 30% to water, 30% to soil, 20% to  
 26 landfills). Some of this release has been sequestered in landfills and benthic sediments, but 310  
 27 Gg actively cycles among geochemical reservoirs and contributes to elevated present-day  
 28 environmental Hg concentrations. Commercial Hg use peaked in 1970 and has declined sharply  
 29 since. Using our inventory to force a global biogeochemical model improves model consistency  
 30 with observed (1) changes in atmospheric deposition recorded in many remote lake sediments  
 31 and ombrotrophic peat bogs over the industrial era, and (2) declines in atmospheric Hg  
 32 concentrations since the 1990s.

## 33 INTRODUCTION

34 Methylmercury, a toxin formed from inorganic Hg, bioaccumulates in aquatic food  
35 chains and adversely affects human health on a global scale through fish consumption.<sup>1,2</sup> Hg has  
36 been mined since antiquity and extensively used in many commercial products,<sup>3</sup> leading to its  
37 eventual release to the environment upon disposal. Hg cycles rapidly between air, water, and soil  
38 reservoirs so that releases to any of these reservoirs can contribute to oceanic Hg levels.<sup>4</sup>  
39 Background levels of Hg in the environment have increased ~3 fold since pre-industrial times<sup>5</sup>  
40 and may be 5 – 10 times above natural levels.<sup>4,6</sup> This has been conventionally attributed to  
41 atmospheric emissions from mining and combustion.<sup>7,8</sup> Here we show that releases of  
42 commercial Hg to air, water, and soil have contributed a large, previously unquantified source of  
43 Hg to the global environment over the industrial era, with major implications for historical and  
44 present-day conditions.

45 Hg is transferred from its stable lithospheric reservoir to surface environmental reservoirs  
46 by natural geological processes, fossil fuel combustion, and mining (including for commercial  
47 products). It then is exchanged between the atmosphere, surface ocean, and terrestrial  
48 ecosystems on time scales of years to decades.<sup>4</sup> The atmospheric lifetime of gaseous elemental  
49 Hg is of the order of a year, allowing transport and deposition on a global scale.<sup>9</sup> Hg is  
50 eventually transferred to long-lived soil and deep-ocean pools over hundreds of years, and  
51 returns to the lithosphere to close the cycle on a timescale of thousands of years.<sup>10</sup> The lasting  
52 environmental legacy of Hg released to surface reservoirs mandates a historical perspective for  
53 understanding present-day environmental burdens and for evaluating the effectiveness of  
54 regulatory actions such as the Minamata Convention.<sup>11</sup>

55 While an estimated 215 Gg of Hg have been emitted directly to the atmosphere since  
56 1850 from fuel combustion, metals smelting, mining, and chlor-alkali plants, an additional 616

57 Gg have been mined during the same period for numerous commercial uses.<sup>12</sup> These include  
58 Hg-containing products (e.g. batteries) and manufacturing processes that involve Hg (e.g. vinyl  
59 chloride monomer production). Commercial Hg enters the environment either during use or  
60 following product disposal. Previous studies have estimated present-day environmental releases  
61 of Hg from commercial products and processes only for a subset of uses and fate pathways, with  
62 a predominant focus on atmospheric emissions.<sup>13-23</sup> The environmental fate of most of the mined  
63 Hg has not yet been quantified.

64 Commercial Hg can enter the environment by various pathways. For example, Hg in  
65 paint rapidly volatilizes to the atmosphere.<sup>24</sup> The chemical manufacturing industry historically  
66 discharged large amounts of Hg directly into waterways, reflected by legacy contamination in  
67 many estuaries today.<sup>25-28</sup> Hg-containing agricultural fungicides and pesticides were applied to  
68 soil in large quantities for much of the 20<sup>th</sup> century.<sup>29</sup> Discarded Hg-containing batteries have  
69 generally been incinerated or landfilled.<sup>30, 31</sup> More recently, many industries have made progress  
70 toward recycling Hg in commercial products or phasing out its use.<sup>32</sup>

71 Here we quantify the global time-dependent historical releases of commercial Hg to  
72 different environmental reservoirs (air, water, soil, landfills), and include these in a global  
73 biogeochemical model<sup>4</sup> to investigate their impact on historical and present-day global Hg  
74 accumulation. Water releases include effluent discharges to both estuaries and inland freshwater  
75 systems, and soil releases are deposited to land or vegetation. Previous work has had difficulty  
76 reconciling the temporal trend of anthropogenic emissions with archival records of historical  
77 atmospheric Hg deposition, many of which show a peak in the 1970s.<sup>33-36</sup> Atmospheric  
78 observations indicate a ~20% to 50% decrease in total gaseous mercury in the Northern  
79 Hemisphere since 1995,<sup>37, 38</sup> at odds with atmospheric emission inventories that estimate

80 increased or flat emissions over this period.<sup>12, 20</sup> Soerensen et al.<sup>38</sup> suggested that the observed  
81 atmospheric trend could be explained by declining oceanic evasion to the atmosphere due to  
82 decreased seawater concentrations. Commercial use of Hg peaked in the 1970s and has declined  
83 dramatically since,<sup>12, 39</sup> and we investigate how the related changes in historical commercial Hg  
84 releases may play a role in reconciling these trends.

## 85 **METHODS**

86 We present a global, historical analysis tracking Hg from when it is mined, through all  
87 known intentional uses and fate pathways, to its final time-dependent releases to different  
88 environmental reservoirs. Commercial Hg uses are grouped by similar environmental release  
89 patterns for a total of 14 use categories (Table 1). We estimate global environmental releases of  
90 Hg and track recycled and landfilled quantities from each use category on a decadal scale  
91 between 1850 and 2010. We first estimate the quantity of Hg consumed in each use category  
92 constrained by the total amount of Hg mined globally. We then apply a series of distribution  
93 factors that vary temporally and by economic development status of different countries to  
94 distribute commercial Hg to its final environmental releases following a simplified substance  
95 flow analysis (e.g. Barr Engineering<sup>14</sup>). Finally, we investigate the impacts of these global, time-  
96 dependent releases on accumulation and cycling among environmental reservoirs using a seven-  
97 box biogeochemical Hg model.

### 98 *Global Commercial Hg Consumption Patterns, 1850 – 2010*

99 We assume that significant Hg release from non-mining commercial Hg uses began in  
100 1850. Anthropogenic releases prior to 1850 were mainly from mining.<sup>12</sup> Overall decadal  
101 estimates of the magnitude of Hg used throughout the industrial era are constrained by global Hg

102 production from mining between 1850 and 1980<sup>12, 39</sup> combined with changes in the Hg supply  
103 available for consumption due to government stockpiling<sup>40</sup> (Figure 1). We assume that mined Hg  
104 that is not stockpiled is incorporated into products within a decade. Secondary Hg sources  
105 (releases from strategic stockpiles, recovery from mining and smelting, and recycling of Hg-  
106 containing products) became significant after 1980.<sup>13, 32, 41</sup> Therefore, for 1990 – 2010, we use  
107 total Hg consumption from Wilson et al.<sup>20</sup> which includes both primary mining and secondary  
108 sources.

109 We then partition the global total Hg supply into individual uses. Methods used to  
110 estimate Hg consumption for each category vary depending on data availability. We divide  
111 countries into two groups, developed countries (North America, OECD/EU Europe, Oceania)  
112 and developing countries including economies in transition (rest of world), based on similarities  
113 in environmental regulations and control technologies within each group that lead to differences  
114 in Hg use and release patterns between them. Data from Streets et al.<sup>12</sup> are used to constrain total  
115 Hg consumed globally for the chlor-alkali industry between 1850 and 1930 and silver and gold  
116 mining between 1850 and 1990, including artisanal and small-scale gold mining (ASGM).  
117 Decadal use patterns available for the United States<sup>42, 43</sup> are used to estimate global consumption  
118 of Hg in the remaining uses for 1850 – 1960. During this period, the US consumed ~25% of total  
119 Hg mined globally and its Hg use data is the most complete.

120 The ca. 1970 onset of environmental regulations targeting Hg releases in developed  
121 countries resulted in major changes in Hg consumption patterns.<sup>25</sup> We therefore estimate Hg  
122 consumption separately for developing countries from 1970 on. For 1970 and 1980, we divide  
123 the global total Hg supply between the developed and developing world by the fraction of global  
124 GDP held by each group of countries.<sup>44</sup> Consumption patterns for the developed world are

125 assumed to follow those of the US in 1970 and 1980.<sup>45</sup> Consumption patterns for the developing  
126 world in 1970 and 1980 are assumed to be similar to those of the US in 1960, when Hg use was  
127 unregulated. For 1990 – 2000, we use regionally resolved Hg consumption data from Wilson et  
128 al.<sup>20</sup> For 2010, we use annual consumption data from AMAP/UNEP<sup>22</sup> for the uses available, and  
129 extrapolate the 2000 – 2005 trend from Wilson et al.<sup>20</sup> for the remaining uses.

### 130 *Environmental Releases of Commercial Hg*

131 Figure 2 shows a generic substance flow analysis diagram that tracks the fate of  
132 commercial Hg for each decade and use category (Table 1) to its eventual release in different  
133 environmental reservoirs. First, the Hg consumed in each use category is divided among direct  
134 releases to air, water, and soil, recycling, and disposal to solid waste or wastewater treatment  
135 ('Tier 1' distribution factors). Certain use categories have additional disposal pathways with  
136 environmental fates that are distinct from general solid waste or wastewater disposal. Hg  
137 entering the three waste disposal pathways is divided further ('Tier 2' distribution factors). Solid  
138 waste is split between disposal in landfills, incineration, and direct disposal to soil, wastewater is  
139 partially captured in sewage sludge and the rest is released to water, and 'Other Disposal' is  
140 distributed between air, land, and landfills. A third tier of distribution factors is needed to  
141 characterize environmental releases for solid waste incineration (emitted to air, or captured in ash  
142 and then deposited to soil or in landfills) and sewage sludge (incinerated and emitted to air,  
143 applied to soil, or disposed of in landfills). Dental amalgam has unique fate pathways not  
144 represented in Figure 2. Some Hg in teeth is permanently stored during burial<sup>46</sup> or partially  
145 released to air, water, and soil upon cremation.



146 Distribution factors for each use category were estimated from a variety of sources  
147 (Tables S1 and S2). Distribution factors are applied globally for 1850 – 1960, and separately for  
148 the developed and developing world between 1970 and 2010. We use previous decadal estimates  
149 from Streets et al.<sup>12</sup> for global atmospheric emissions from chlor-alkali plants, silver and large-  
150 scale gold mining, and ASGM, except for 2010 ASGM emissions for which we use  
151 AMAP/UNEP<sup>22</sup> estimates.

152 Temporal variability in distribution factors is governed by the availability of evidence  
153 that releases have changed from one decade to another due to regulatory controls and other  
154 factors. Global distribution factors are held constant in time through 1950, with variation in 1960  
155 for three use categories (VCM and Other Chemical, Paint, and Pesticides and Fertilizer). This  
156 reflects significant changes in consumption patterns (a change from chemicals to laboratory uses,  
157 marine anti-fouling paint to home interior/exterior latex paint, and agricultural to pulp and paper  
158 fungicides, respectively). Different distribution factors in the developed world are applied for  
159 each decade between 1970 and 2010. Developing world distribution factors are assumed constant  
160 for 1970 – 2010. The “Other” category in Table 1 (9% of 1850 – 2010 global consumption)  
161 encompasses a diverse set of uses with an unknown distribution, and we assume for this category  
162 the average fate of all other categories.

163 Releases to different environmental reservoirs are calculated on a global scale by  
164 multiplying the Hg consumed in each use category and decade by the corresponding distribution  
165 factors. Most products are disposed of and enter the environment within the decade when they  
166 were produced, and manufacturing processes generally consume and release Hg within a year.<sup>13,</sup>  
167 <sup>15, 16, 30, 47-50</sup> Products in the Wiring Devices and Industrial Measuring Devices category are often  
168 in use for 20-50 years before disposal.<sup>13-15, 30, 49</sup> For these products we follow the methods of

169 Jasinski et al.<sup>13</sup> and Cain et al.<sup>16</sup> and assume that 10% are discarded after 10 years, 40% after 30  
170 years, and the remaining 50% after 50 years. Dental amalgams generally remain in living teeth  
171 for 10 to 30 years or more. We estimate excretion and exhalation releases from dental amalgam  
172 to air and water during this time and track the remaining Hg that will be released through  
173 cremation pathways or permanently stored through burial after 30 to 50 years, depending on  
174 average life expectancy<sup>51</sup> and ages of individuals receiving fillings.<sup>16</sup>

175 Recycled Hg is estimated separately for internal reuse of Hg in chemicals manufacturing  
176 and large-scale mining<sup>30, 52, 53</sup> and external recycling of Hg in products like batteries that returns  
177 Hg to the global Hg supply for future use.<sup>53</sup> External recycling did not become widespread until  
178 1990.<sup>13</sup> We do not tie our estimates of recycled Hg to global Hg supply, and instead use  
179 estimated total Hg supply from Wilson et al.<sup>20</sup> which includes all secondary sources for 1990 –  
180 present. We assume that no Hg is released to the environment during the recycling process,  
181 based on available data suggesting releases are negligible.<sup>53</sup>

## 182 *Implications for the Biogeochemical Hg Cycle and Atmospheric Trends*

183 We use the fully-coupled global biogeochemical Hg box model from Amos et al.<sup>4</sup> to  
184 track the fate of commercial Hg after it has been released to the environment. The model  
185 includes seven reservoirs that represent the atmosphere, ocean (surface, subsurface, and deep),  
186 and terrestrial pools (fast, slow, and armored). Fluxes between reservoirs are determined by first-  
187 order rate coefficients applied to the inventory of the exporting reservoir. Atmospheric rate  
188 coefficients are from Holmes et al.<sup>54</sup> and oceanic rate coefficients are from Soerensen et al.<sup>55</sup> and  
189 Sunderland and Mason<sup>56</sup> (for a full list, see Table S3). Riverine rate coefficients have been  
190 updated following Amos et al.<sup>57</sup> to include the settling of particle-bound Hg to benthic estuarine

191 and shelf sediments. This effectively adds a permanent sink from the terrestrial pools. Primary  
192 anthropogenic and geogenic emissions are treated as external forcings.

193 We added a landfill reservoir to this model. Landfills can emit Hg into the atmosphere  
194 through vent pipes, diffusion from cover soil, and from the “working face” where waste is  
195 exposed and actively dumped.<sup>31</sup> They can also release Hg into groundwater and soils through the  
196 base of the landfill but this appears to be negligible.<sup>58</sup> Data available on Hg emissions for  
197 managed landfills in China,<sup>31</sup> the United States,<sup>59</sup> and Korea<sup>60</sup> indicate low values. We find a  
198 mean lifetime of 20,000 years for Hg in landfills by combining emissions values for each site  
199 with estimated Hg reservoirs inferred from the waste content and disposal magnitudes. Landfills  
200 as defined here can therefore effectively be viewed as a permanent sink.

201 Commercial Hg enters the biogeochemical cycle via the atmosphere, water, soil, and  
202 landfills. Hg releases to soil are distributed among the three terrestrial reservoirs of the model in  
203 the same manner as atmospherically deposited Hg in Amos et al.<sup>4</sup> Hg releases to water are  
204 distributed as 90% to inland freshwater systems and 10% to estuaries based on the distribution of  
205 present-day point sources.<sup>57, 61</sup> We assume that 75% of the Hg input to inland freshwater systems  
206 is sequestered permanently in sediments and 25% evades to the atmosphere, based on models for  
207 a variety of lakes and rivers.<sup>62</sup> Based on previous work, we assume that 50% of the Hg directly  
208 released to estuaries is transported to the surface ocean, 10% evades to the atmosphere, and 40%  
209 is sequestered permanently in coastal sediments.<sup>63, 64</sup> In this manner, we estimate that 70% of Hg  
210 released to water is permanently sequestered, 23% enters the atmosphere, and 7% enters the  
211 surface ocean.

212 We apply the box model to time-dependent simulations from 2000 BC to 2010 following

213 the methods described in Amos et al.<sup>4</sup> We simulate 1850 – 2010 with anthropogenic atmospheric  
214 emissions from Streets et al.<sup>12</sup> plus the releases from commercial Hg quantified in this study. We  
215 evaluate the model with three global observational constraints, following Amos et al.<sup>4</sup>: (1) the  
216 present-day atmospheric inventory (best estimate of 5000 Mg, range 4600 – 5600 Mg), (2) the  
217 present-day mean upper (0 to 1500 m) ocean concentration (best estimate of 1.0 pM, range 0.5-  
218 1.5 pM), and (3) the relative anthropogenic enrichment factor (AEF) in atmospheric deposition  
219 between pre-industrial and present-day (best estimate of 3, range 2 to 5). We define our model  
220 AEF as the ratio of average 1985-2000 deposition to average 1760-1880 deposition.<sup>65, 66</sup> These  
221 two time intervals are empirically determined from the compilation of lake sediments described  
222 in Biester et al.<sup>67</sup> to provide a more consistent comparison between models and observations.<sup>65</sup>

## 223 **RESULTS AND DISCUSSION**

### 224 *Global Commercial Hg Consumption Patterns, 1850 – 2010*

225 Figure 3 shows the global commercial use of Hg since 1850 for all categories described  
226 in Table 1, and partitioned between the developed and developing world after 1970. Prior to  
227 1900, almost all mined Hg was used in silver and gold mining. In the 20<sup>th</sup> century, Hg uses  
228 diversified greatly. Hg use in large-scale mining declined following the end of the gold rush and  
229 as extraction methods that did not require Hg became widespread. The 1940s peak in Figure 3 is  
230 driven by chemicals production for munitions during WWII. The 1970s peak represents the  
231 height of Hg use in consumer products like paint and batteries. Total consumption declined after  
232 the 1970s when many developed countries implemented regulations on Hg uses and  
233 environmental releases. Since 1990, the developing world has dominated global Hg

234 consumption. ASGM in developing countries is presently the largest use of Hg globally, and is  
235 increasing.<sup>22, 68</sup>

### 236 *Environmental Releases of Commercial Hg*

237 Figure 4 compares the historical atmospheric emissions inventory of Streets et al.<sup>12</sup>  
238 (including sources from combustion, metals smelting, mining, and chlor-alkali plants) with our  
239 best estimate of additional releases to air, water, soil, and landfills from commercial Hg. Streets  
240 et al.<sup>12</sup> estimate total emissions to air of 215 Gg since 1850. Additional releases in our inventory  
241 over the same period total 540 Gg. Of these, 230 Gg are permanently sequestered in landfills or  
242 in benthic sediments of freshwater and estuarine systems. The remaining 310 Gg (including 110  
243 Gg emitted to air) cycle between biogeochemical reservoirs and represent a larger anthropogenic  
244 perturbation to the global Hg cycle than the 215 Gg of Streets et al.<sup>12</sup> Commercial Hg thus  
245 represents a major, previously unquantified source of Hg to the global environment. The  
246 temporal pattern of anthropogenic Hg releases is also affected. Whereas Streets et al.<sup>12</sup> indicate  
247 rising emissions since 1950, our estimate shows a decline in total releases from 1970 to 2000.

248 Figure 5 presents the historical contributions of individual commercial use categories to  
249 global environmental releases to air, water, soil, and landfills. Releases to air, water, and soil all  
250 show similar enhancements from late 19<sup>th</sup>-century mining. Differences are attributable to  
251 changes in the fate of commercial Hg over time. Emissions to air peak in 1970, mainly due to  
252 paint volatilization and incineration of batteries. Although Hg use in batteries increased from  
253 1970 to 1980, open-air waste burning at landfills was eliminated during this period following  
254 solid waste regulations.<sup>69, 70</sup> Use of explosives and weapons was a major emitter to air during  
255 1900 – 1950 with a peak in WWII.

256 Water releases also exhibit a peak during WWII, due to laboratory uses and chemicals  
257 manufacturing. The overall maximum occurs in 1960, with a steep subsequent decline following  
258 implementation of chlor-alkali liquid effluent regulations in the early 1970s in North America  
259 and Europe.<sup>71, 72</sup> Implementation of wastewater treatment from the 1980s onward led to even  
260 greater declines in water releases,<sup>73</sup> but contributes a small amount to soils due to application of  
261 Hg-containing sludges.<sup>16, 53</sup> Similarly, chlor-alkali plants began capturing Hg in sludges in the  
262 1970s, which were subsequently dumped on land or landfilled on-site.<sup>72</sup> The 1970 peak in soil  
263 releases is driven by Hg used in chlor-alkali plants and Hg-containing pesticides and fertilizer  
264 that were applied directly to land.

#### 265 *Implications for the Biogeochemical Hg Cycle and Atmospheric Trends*

266 Figure 6 shows simulated atmospheric Hg from 1850-present after adding our inventory  
267 of commercial Hg releases to the updated global biogeochemical Hg model, which includes  
268 burial of riverine Hg in benthic sediments of coastal marine systems from Amos et al.<sup>57</sup> and an  
269 additional reservoir representing landfills. Our simulated present-day atmospheric reservoir is  
270 5800 Mg, the mean Hg concentration in the upper ocean is 1.5 pM, and the AEF for atmospheric  
271 deposition is 4.4. The original Amos et al.<sup>4</sup> simulation not including commercial Hg yielded a  
272 present-day atmosphere of 5300 Mg, but did not account for burial of riverine Hg. Accounting  
273 for burial without commercial Hg results in an atmosphere that is too low (2700 Mg).  
274 Conversely, including commercial Hg in the original Amos et al.<sup>4</sup> simulation without burial  
275 would yield a present-day atmospheric reservoir of 10,000 Mg, much higher than observed.

276 Our simulated present-day atmospheric reservoir of 5800 Mg is still slightly higher than  
277 the observational range (4600-5600 Mg). This is due to the increase over the past decade (Figure

278 6) driven by rising anthropogenic atmospheric emissions in the Streets et al.<sup>12</sup> inventory  
279 (primarily from coal burning in Asia) and rising releases from ASGM<sup>22</sup> (Figures 3 and 4).  
280 However, Wilson et al.<sup>20</sup> suggest that global anthropogenic emissions have in fact remained  
281 relatively constant since 2000, and AMAP/UNEP<sup>22</sup> suggest that the ASGM increase since 2000  
282 may be an artifact of improved reporting. We conducted a sensitivity simulation holding constant  
283 anthropogenic atmospheric emissions and ASGM releases for the 2000-2010 period, and results  
284 are shown as the dashed line in Figure 6. We obtain in that simulation an atmospheric reservoir  
285 of 5000 Mg, more consistent with observations, and unchanged upper ocean concentration of 1.5  
286 pM and AEF of 4.4. Alternatively, the atmospheric reservoir could be reduced within the range  
287 of observational constraints if the efficacy of Hg re-emission from soils were decreased, a  
288 change supported by recent observational evidence.<sup>74, 75</sup>

289 Inclusion of commercial Hg releases in our global biogeochemical model improves  
290 model consistency with archival records of atmospheric deposition. Lake sediments and  
291 ombrotrophic peat bogs generally indicate a gradual rise over the industrial era with a peak in the  
292 1970s.<sup>33-36</sup> Figure 6 indicates a 1970s peak in simulated atmospheric Hg and a muted 19<sup>th</sup>  
293 century mining signal. Without the inclusion of commercial Hg, model deposition increases from  
294 1970 to present and there is a prominent 19<sup>th</sup> century peak.<sup>4</sup> Our inventory also helps to explain  
295 the observed 1990-present declines in atmospheric Hg over North America and Europe.<sup>37, 38</sup>

296 Our work shows that environmental releases of commercial Hg to air, water, and soil  
297 over the industrial period have represented a major and previously unquantified perturbation to  
298 Hg in the global environment. The legacy of this source in oceanic and terrestrial reservoirs has  
299 important implications for policy. Future work should examine the geographical distribution of  
300 commercial Hg releases and its impact on regional-scale environmental Hg loadings and

301 atmospheric trends. Better understanding of the role of soils for long-term storage of  
302 anthropogenic Hg is also critically needed.

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306 **SUPPORTING INFORMATION.** Additional information on data sources used to estimate  
307 distribution factors (Table S1), distribution factors used in this study (Table S2), and model rate  
308 coefficients (Table S3). This material is available free of charge via the Internet at  
309 <http://pubs.acs.org>.

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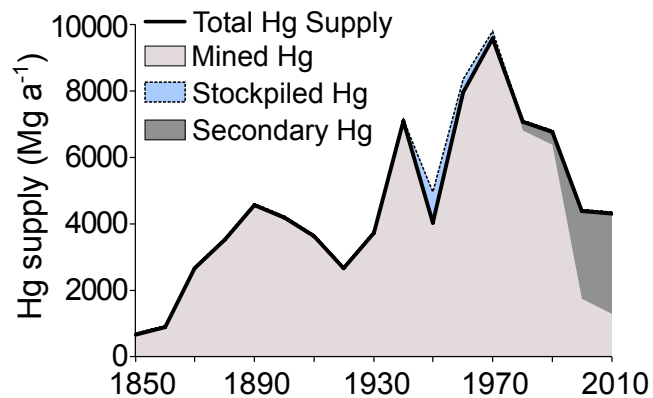
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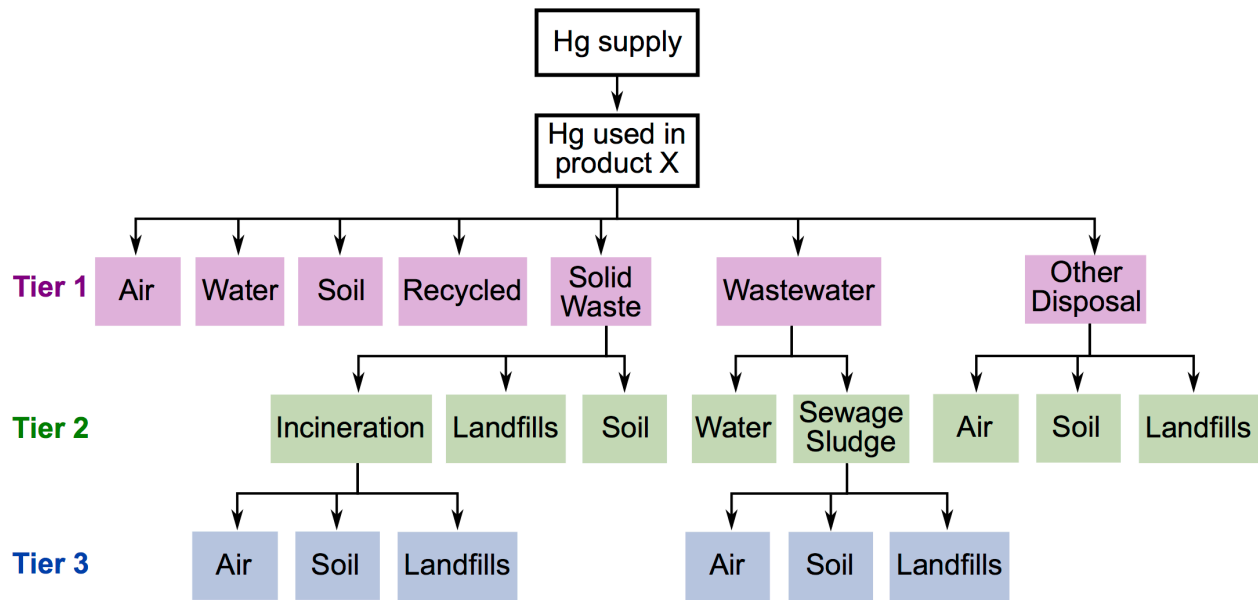
524 Presented at 11th International Conference on Mercury as a Global Pollutant, Edinburgh,  
525 Scotland, July 2013.



526  
 527 **Figure 1. Historical global supply of Hg for commercial uses.** This supply includes primary  
 528 mined Hg (1850 – 2008 data from Streets et al.<sup>10</sup>), minus the amount stockpiled by the United  
 529 States between 1945 and 1970, and augmented by secondary Hg including recycled, recovered as  
 530 a byproduct, and released from stockpiles after 1980.

532 **Table 1. Intentional uses of Hg in products and processes.**

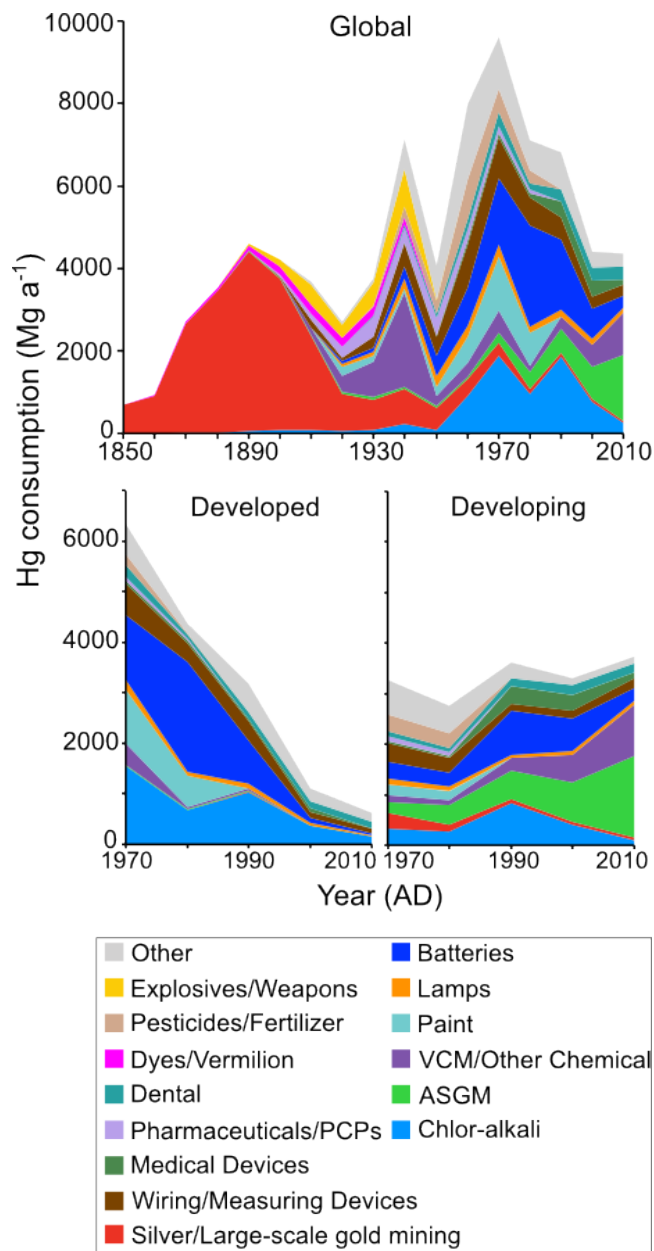
<b>Category name</b>	<b>Description</b>
Chlor-Alkali Plants	Electrochemical production of caustic soda and chlorine with Hg cathode
Silver and Large-scale Gold Mining	Extraction from ore by Hg amalgamation
Artisanal and Small-Scale Gold Mining (ASGM)	Hg amalgamation by individual miners
Vinyl Chloride Monomer (VCM) and Other Chemical	Production of chemicals with Hg catalysts, felt hat manufacturing, and laboratory uses
Paint	Hg fungicide in marine anti-fouling paint, interior and exterior latex paint
Lamps	All types of Hg-containing lightbulbs (fluorescent, high intensity discharge, etc.)
Batteries	Button cells and cylinders using Hg as cathode or to prevent corrosion
Wiring Devices and Industrial Measuring Devices	Switches and relays, thermostats, barometers, manometers
Medical Devices	Thermometers and sphygmomanometers (blood pressure meters)
Pharmaceuticals and Personal Care Products	Vaccines and medicines, soaps, cosmetics
Dental amalgam	Cavity fillings with Hg/silver/tin/copper amalgam
Dyes/Vermilion	Pigments containing Hg compounds
Pesticides and Fertilizer	Fungicides used in agriculture and pulp and paper
Explosives/Weapons	Munitions, blasting caps, fireworks
Other	Ritual, cultural, and miscellaneous uses



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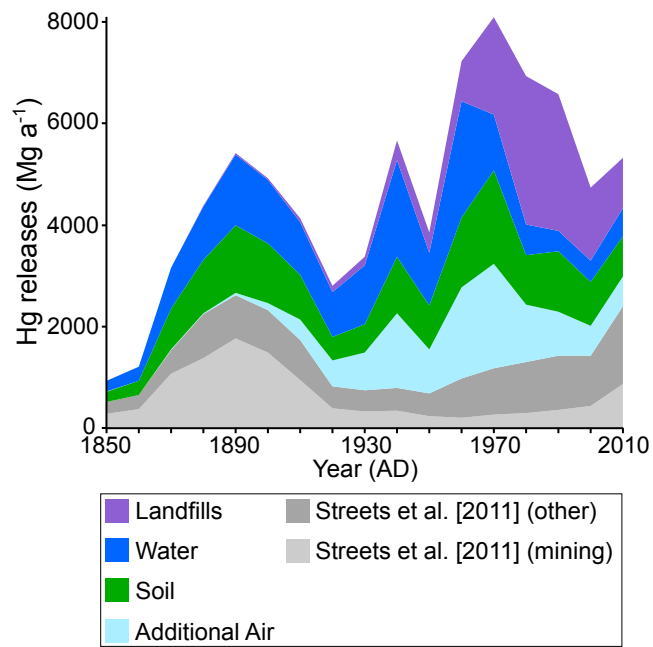
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536 **Figure 2. Generic substance flow analysis diagram for commercial Hg.** Each arrow is a  
 537 distribution factor quantified in this work (Table S3). This diagram applies to all intentional use  
 538 categories with the exception of dental amalgam, which has additional pathways (see Methods).  
 539 “Other disposal” refers to medical waste incineration (for Medical Devices and Dental use  
 540 categories) and iron and steel recycling (for Wiring Devices and Industrial Measuring Devices  
 541 use category).  
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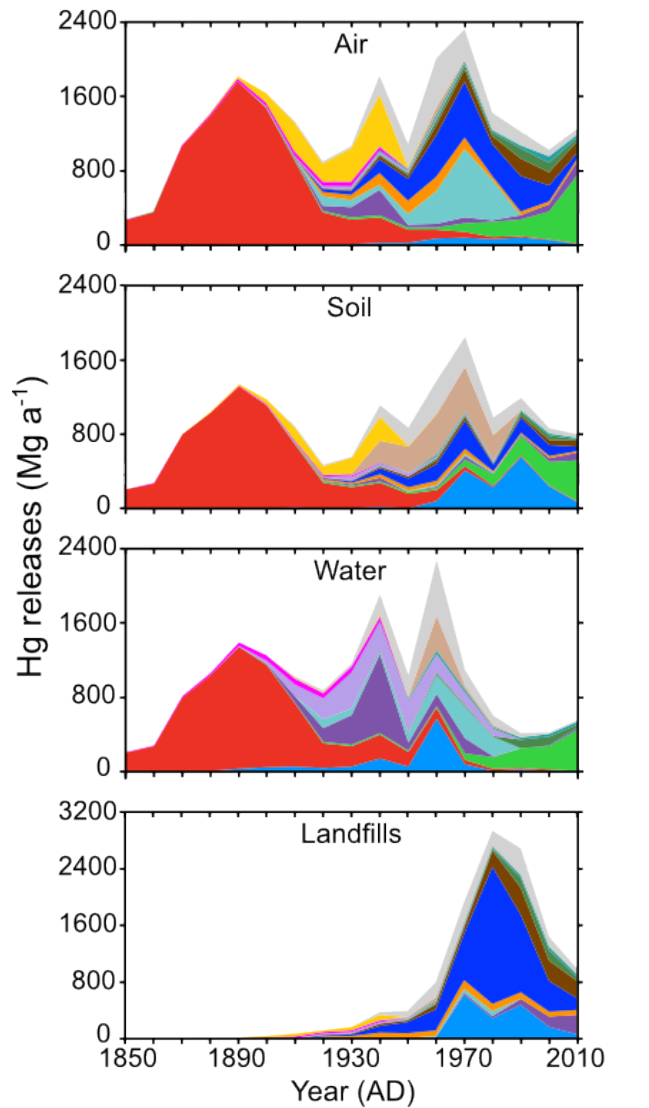
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**Figure 3. Global historical Hg consumption in commercial products.** Consumption is partitioned for each decade between the different use categories of Table 1, and further partitioned between developed countries and developing countries after 1970.



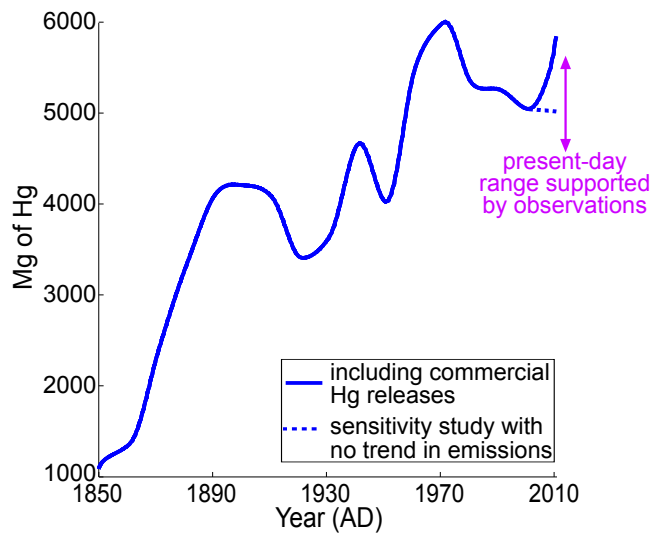
547  
 548 **Figure 4. Historical global releases of Hg to the environment.** The Streets et al.<sup>10</sup> inventory  
 549 includes atmospheric releases from combustion, smelting, mining, and chlor-alkali plants.  
 550 Additional air, soil, water, and landfill releases shown are associated with commercial Hg  
 551 products as quantified in this work.





552  
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554

**Figure 5. Global historical releases of commercial Hg to environmental reservoirs by use category.**



555  
 556 **Figure 6. Trend in simulated atmospheric Hg mass from 1850 to 2010.** Results shown using  
 557 the updated biogeochemical model described in the text, including additional commercial Hg  
 558 releases (blue) and a sensitivity simulation with ASGM releases and Streets et al.<sup>10</sup>  
 559 anthropogenic atmospheric emissions held constant from 2000-2010 (dotted blue line).