



## Historical and future trends in global source-receptor relationships of mercury



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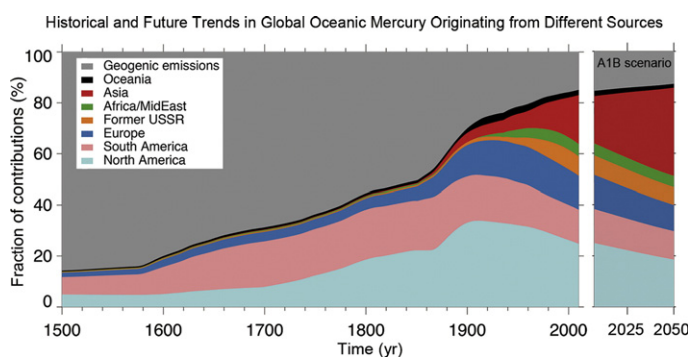
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### HIGHLIGHTS

- Legacy impacts of historical releases are notable on source-receptor relationships.
- North America, Asia and Europe contribute 26%, 16% and 14% of global oceanic Hg.
- Asia will exceed North America as the largest contributor to global ocean in 2019.
- Legacy impacts would lead to the future potential of rising Hg in the Arctic Ocean.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Growing concern about the risk associated with increasing environmental mercury (Hg) concentrations has resulted in a focus on the relationships between intercontinental emitted and accumulated Hg. We use a global biogeochemical Hg model with 8 continental regions and a global ocean to evaluate the legacy impacts of historical anthropogenic releases (2000 BCE to 2008 AD) on global source-receptor relationships of Hg. Legacy impacts of historical anthropogenic releases are confirmed to be significant on the source-receptor relationships of Hg according to our results. Historical anthropogenic releases from Asia account for 8% of total soil Hg in North America, which is smaller than the proportion (~17%) from previous studies. The largest contributors to the global oceanic Hg are historical anthropogenic releases from North America (26%), Asia (16%), Europe (14%) and South America (14%). Although anthropogenic releases from Asia have exceeded North America since the 1970s, source contributions to global Hg receptors from Asia have not exceeded North America so far. Future projections indicate that if Hg emissions are not effectively controlled, Asia will exceed North America as the largest contributor to the global ocean in 2019 and this has a long-term adverse impact on the future environment. For the Arctic Ocean, historical anthropogenic release from North America contributes most to the oceanic Hg reservoir and future projections reveal that the legacy impacts of historical releases from mid-latitudes would lead to the potential of rising Hg in the Arctic Ocean in the future decades, which calls for more effective Hg controls on mid-latitude releases.

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## 1. Introduction

Human activities, such as coal combustion, mining and discarding of commercial products, have been releasing mercury (Hg) to the environment since antiquity, resulting in the increase of Hg in the atmosphere, ocean, and soil by several times over natural levels (Beal et al., 2014; Engstrom et al., 2014; Lamborg et al., 2014). Long atmospheric lifetime of elemental Hg ( $\text{Hg}^0$ ) results in long-range atmospheric transport of emitted Hg. Eventually  $\text{Hg}^0$  is oxidized to divalent Hg ( $\text{Hg}^{\text{II}}$ ) that easily deposits to ecosystems (Lindberg et al., 2007; Corbitt et al., 2011). Methylation and bioaccumulation of Hg in food webs after deposition adversely affects exposed humans and wildlife (Mergler et al., 2007; Mahaffey et al., 2011). Growing concerns about the risk of increasing environmental Hg, have led to the launch of the Minamata Convention on Mercury (<http://www.mercuryconvention.org>) and resulted in a focus on the relationships between emitted and accumulated Hg on an intercontinental scale (Corbitt et al., 2011; Pirrone and Keating, 2011). Here we use a global biogeochemical Hg model to evaluate global source-receptor relationships of Hg on all-time scales and examine the legacy impacts of historical anthropogenic releases.

Source-receptor relationships of Hg on regional and intercontinental scales have been reported in previous studies. Jaffe and Strode (2008) found that 7–20% (~16%) of Hg deposition in North America was attributed to Asian anthropogenic emissions. Lin et al. (2010) estimated that global anthropogenic sources contributed to 75% of Hg deposition over East Asia with the remainder from natural sources. Corbitt et al. (2011) suggested that Asian anthropogenic emissions to air were the largest deposition source of all ocean basins. Durnford et al. (2010) found that Asia was the largest source (29–37%) of atmospheric Hg at Arctic observational sites, and Pirrone and Keating (2011) estimated East Asia to be the largest deposition source of the Arctic Ocean except for natural and reemission sources. Chen et al. (2014) established source-receptor relationships for Hg emissions, transport and deposition among major continents. The study suggested that East Asia was the region with the largest influence on all other continents. These studies, however, all focused on present-day source-receptor relationships of Hg, while historical source-receptor relationships on all-time scales were not included. Meanwhile, legacy impacts of historical anthropogenic releases have been confirmed to influence the global Hg cycle significantly (Amos et al., 2013, 2015). On all-time scales, we could examine the legacy impacts on global source-receptor relationships of Hg.

A range of global and national policy actions have been implemented to reduce mercury contamination in the environment over the past decades, such as the UN Minamata Convention on Mercury and the US Mercury and Air Toxics Standards (MATS) (Selin and Selin, 2006; Agency, 2011). Based on the knowledge of policies, technologies and economic growth, numerous studies have projected future emission scenarios and evaluated their influences on global Hg cycle and exposure risks (Streets et al., 2009; Selin, 2014; Giang and Selin, 2016). Control policies may decrease environmental Hg, and benefit humans and societies (Selin, 2014; Giang and Selin, 2016). For instance, despite that fact quantitative reduction targets are not given, the Minamata Convention gives requirements of best available techniques (BAT) approach in schedules for each emission category and provides for mechanisms of reduction of Hg by category-specific provisions. By combining the legacy impacts of historical anthropogenic releases with future emission scenarios, we investigate future global source-receptor relationships of Hg up to 2050, and evaluate the potential effectiveness of suggested control policies under the legacy impacts of historical anthropogenic releases.

Considering the heavy dependence on marine-based diets from Arctic indigenous peoples, Hg pollution in the Arctic Ocean is of particular concern (AMAP, 2011). A ten-fold increase of Hg levels was observed for marine animals in the Arctic Ocean over the past 150 years, which was attributed to the rapid increase of global anthropogenic sources after the Industrial Revolution (Dietz et al., 2009;

Braune et al., 2015). The source apportionment of accumulated Hg in the ocean is of importance for Arctic Hg controls. Previous studies have investigated the source of atmospheric Hg over the ocean and illustrated the critical role of Asia (Durnford et al., 2010; Pirrone and Keating, 2011). However, source apportionment of oceanic Hg in the Arctic has been understudied. Under the global simulations of source-receptor relationships in this study, we could conduct a nested simulation for the Arctic Ocean and examine the source of accumulated Hg in the ocean on all-time scales.

## 2. Material and methods

### 2.1. Model description

We use the fully coupled, seven-reservoir box model of GEOS-Chem for global Hg cycle developed by Amos et al. (2013, 2014) (<http://bgc.seas.harvard.edu/models.html>) to evaluate global source-receptor relationships of Hg on all-time scales. Model reservoirs include the atmosphere, ocean (surface, subsurface, and deep ocean), and terrestrial ecosystems (fast terrestrial, slow soil, and armored soil). A set of coupled ordinary differential equations based on first-order rate coefficients ( $k$ ) is applied to represent Hg cycling in the model. The rate coefficients ( $k$ ) are derived from the literature estimates of present-day masses and flows (Selin et al., 2008; Holmes et al., 2010; Smith-Downey et al., 2010; Soerensen et al., 2010), as described in details by Amos et al. (2013, 2014).

We divide the Earth surface into eight continental regions and a global ocean to evaluate the relationships between emitted and accumulated regions. The continental regions include North America, South America, Europe, Former USSR, Africa/Middle East, Asia, Oceania and Antarctic (Fig. A.1). Each continental region has individual atmosphere and terrestrial ecosystems (fast terrestrial, slow soil, and armored soil) reservoirs. We add some ordinary differential equations based on first-order rate coefficients ( $k$ ) to represent Hg cycling of atmospheric Hg lateral transport and soil Hg exchange. The rate coefficients ( $k$ ) for atmospheric Hg lateral transport are simulated by the GEOS-Chem Hg model v9-01-03 (<http://geos-chem.org>), which includes a 3-D atmosphere model coupled to a 2-D surface slab ocean and a 2-D soil reservoir (Selin et al., 2008; Holmes et al., 2010; Soerensen et al., 2010). Two Hg species,  $\text{Hg}^0$  and  $\text{Hg}^{\text{II}}$ , are tracked in the model with oxidation of  $\text{Hg}^0$  by Br atoms to  $\text{Hg}^{\text{II}}$ , photoreduction of  $\text{Hg}^{\text{II}}$  to  $\text{Hg}^0$  in droplets and gas-particle partitioning between gaseous and particulate  $\text{Hg}^{\text{II}}$  (Holmes et al., 2010; Amos et al., 2012). Fast oxidation of  $\text{Hg}^0$  to  $\text{Hg}^{\text{II}}$  and subsequent loss by deposition is caused by high Br atom concentrations in polar springtime (Fisher et al., 2012). We obtain simulated reservoir masses and lateral transport fluxes among the specified regions (Table A.1), which are used to calculate the rate coefficients ( $k$ ) for atmospheric Hg lateral transport. We also obtain simulated deposition fluxes and calculate regional deposition rates with the ratio of deposition fluxes to atmospheric reservoir masses. The rate coefficients ( $k$ ) for soil Hg exchange are simulated by the Global Terrestrial Mercury Model (GTMM) (Table A.2). GTMM is a global  $1^\circ \times 1^\circ$  biogeochemical model for soil Hg cycle and used to simulate the continuous evolution from preindustrial to present day with a monthly time step. Further model details are found in Smith-Downey et al. (2010).

Considering the source apportionment of accumulated Hg in the Arctic Ocean, we couple a five-box geochemical model of the Arctic Hg cycle to the global box model to conduct a nested simulation for the Arctic Ocean. The description for the Arctic model is found in Soerensen et al. (2016). The coupling process includes: (1) exchange of atmospheric Hg through lateral atmospheric transport, (2) exchange of oceanic Hg through lateral oceanic currents, and (3) riverine discharge to the Arctic Ocean from adjacent continents. The reservoir masses and fluxes for lateral atmospheric transport and oceanic currents are shown in Table A.3. The updated model is evaluated through comparison with observational constraints and literature values, which is illustrated in Text A.1. We

also apply perturbation analysis in Text A.2 to examine how the model results are influenced by the uncertainties in model inputs, such as anthropogenic releases, process rates and atmospheric Hg chemistry.

## 2.2. Anthropogenic releases

The model is driven by all-time (2000 BCE to 2008 CE) anthropogenic atmospheric emissions from Streets et al. (2011) and additional 1850–2008 atmospheric emissions from commercial Hg use (Horowitz et al., 2014). They estimated that total global anthropogenic atmospheric Hg emissions were 137 Gg prior to 1850 and 320 Gg during 1850–2008 (105 Gg for commercial Hg use). The model is also driven by 1850–2008 Hg discharges from rivers (Amos et al., 2014) and global geogenic emissions (90 Mg yr<sup>-1</sup>) derived from Pirrone et al. (2010) and Bagnato et al. (2011).

In order to evaluate the global source-receptor relationships, we distribute the global anthropogenic releases regionally for specified continents (Fig. 1). Anthropogenic atmospheric emissions from Streets et al. (2011) during 1850–2008 (215 Gg) were already distributed regionally. The additional 1850–2008 atmospheric emissions from commercial Hg use (105 Gg) (Horowitz et al., 2014) are distributed differently during different periods in this study. As recommended by Horowitz et al. (2014), we distribute the emissions during 1970–1980 based on the fraction of global GDP held by each continent of countries (<http://unstats.un.org/>). We distribute the emissions during 1990–2008 based on the regionally resolved Hg consumption data from Wilson et al. (2010) and AMAP/UNEP (2013). The 1850–1970 emissions are distributed based on the regional emission fractions from Streets et al. (2011) due to limited information of Hg consumption during this period. The atmospheric emissions prior to 1850 were primarily emitted from silver mining in colonial Spanish America during 1500s to 1800s (88%). We specify Hg emissions for North America (Mexico), South America (Bolivia and Peru) and other continental regions during 1500s to 1800s based on the percentage of global silver production referred from Nriagu (1994). Due to lack of information, the remaining small amount of emissions before 1500s are distributed equally to each year and the annual emissions are distributed to each region based on the emission fraction in 1500s. Based on the specified continents (Fig. A.1), and global distribution of riverine Hg discharges referred from Amos et al. (2014), we calculate the riverine Hg discharges from each specified continents. Then we calculate the Hg discharges to the open ocean from different

continents based on the fraction of particulate Hg reaching each open ocean basin (Walsh and Nittrouer, 2009).

For the evaluation of future source-receptor relationships, we involve four projected scenarios from Streets et al. (2009) and Amos et al. (2013) including A1B scenario, B1 scenario, “mercury controls” scenario and “zero emissions” scenario. The A1B scenario is characterized by rapid economic growth and continuous emission growth, and the B1 scenario is the most optimistic scenario which has effectively constant emissions relative to present levels in the study. The “mercury controls” scenario assumes that primary anthropogenic emissions decline to 50% of their present levels by 2050 due to the implementation of Hg-specific controls across multiple emission categories. The “zero emissions” scenario assumes that primary anthropogenic emissions are completely stopped after 2015, which indicates the maximum achievable benefit of emission controls. All scenarios are implemented in 2015, and we assume constant releases for riverine Hg discharges up to 2050 due to limited projection information.

## 3. Results and discussion

### 3.1. Different anthropogenic enrichment among continents

Table 1 illustrates masses and anthropogenic enrichment for Hg reservoirs in each region at present day (post-1990). Hg in the fast terrestrial reservoir of the Former USSR is the largest fast terrestrial reservoir, while Hg reservoirs in the slow and armored soils or those that have the longest turnover time for carbon and the most recalcitrant, of Africa/Middle East exceed that of the Former USSR. This indicates that the transfer rates from fast to slow and armored soils of Africa/Middle East are higher than that of the Former USSR. Low temperature in high latitude (such as the Former USSR) inhibits the decomposition of litter and soil organic matter in top soils, which results in high retention of Hg in the fast terrestrial reservoir (Van der Werf et al., 2003; Smith-Downey et al., 2010).

Two enrichment factors (EFs) are defined to illustrate how the present-day soil and oceanic Hg are influenced by all-time human activities. EF<sub>1760–1880</sub> is defined as the ratio of Hg content at present day to that in pre-industrial era (1760–1880), and EF<sub>2000 BCE–1550 CE</sub> is defined as the ratio of Hg content at present day to that in pre-large-scale mining era (2000 BCE–1550 CE). Large differences between EF<sub>1760–1880</sub> and EF<sub>2000 BCE–1550 CE</sub> are found for fast reservoirs (fast terrestrial reservoir and surface ocean), while small discrepancies are found for slow reservoirs (slow soil, armored soil, and deep ocean). This indicates that the anthropogenic releases after 1850 primarily accumulate in the fast reservoirs, and natural emissions contribute a lot to the slow reservoirs. Differences among regional EFs highlight the different processes of Hg cycling in different terrestrial ecosystems and ocean. In Asia, we find higher EFs for fast, slow and armored soils than other continents, consistent with the recent large anthropogenic releases from Asia. On the other hand, the Former USSR has the smallest EFs and largest residence times among continents due to the remote location and large retention of natural Hg in the slow-overturning soil pools in a cold environment.

### 3.2. Present-day source-receptor relationships of Hg

Fig. 2 illustrates the fractional contributions of Hg content in the continents and global ocean originating from seven continental sources and global geogenic emissions at present day (post-1990). Historical anthropogenic releases account for a larger fraction of Hg content in the global ocean than continents. The fraction of anthropogenic contribution to the global ocean is 84%, and to the continental receptors is averaged at 59%. Natural emissions (global geogenic emissions) contribute a lot to the continental receptors (~41%) due to the long lifetime of Hg in armored soil which accumulates much Hg from global geogenic emissions in the pre-large-scale mining and pre-industrial era (Amos et al., 2013).

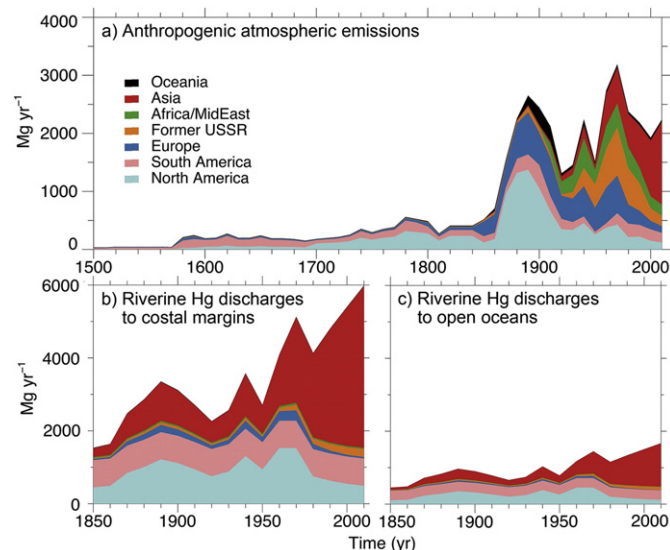


Fig. 1. Historical trends in anthropogenic atmospheric Hg emissions and riverine Hg discharges to the global ocean from seven specified continental regions.

**Table 1**  
Masses and anthropogenic enrichment for Hg reservoirs in each region at present day (post-1990).

		North America	South America	Europe	Former USSR	Africa/Middle East	Asia	Oceania	Global ocean
Fast terrestrial reservoir (Surface ocean) <sup>a</sup>	Reservoirs ( $\times 10^3$ Mg)	3.0	3.0	1.2	8.3	3.4	1.8	2.1	3.8
	EF <sub>1760–1880</sub> <sup>b</sup>	4.1	4.1	4.1	4.1	4.1	4.2	4.1	4.7
	EF <sub>2000 BCE–1550 CE</sub> <sup>c</sup>	13.3	12.9	12.9	9.0	10.6	14.2	10.2	14.6
	Residence time (year) <sup>d</sup>	7.2	5.3	9.7	22.6	5.1	3.1	11.2	
Slow soil (Subsurface ocean)	Reservoirs ( $\times 10^3$ Mg)	13.8	14.7	4.9	22.3	23.1	13.6	10.0	156.8
	EF <sub>1760–1880</sub>	3.7	3.9	3.5	2.9	3.6	4.1	3.3	4.9
	EF <sub>2000 BCE–1550 CE</sub>	8.4	9.3	7.4	4.3	6.8	10.0	5.9	13.5
	Residence time (year)	138.7	98.5	172.2	232.3	157.5	110.3	186.1	
Armored soil (Deep ocean)	Reservoirs ( $\times 10^3$ Mg)	17.1	15.8	4.5	26.5	43.9	21.1	17.7	130.9
	EF <sub>1760–1880</sub>	1.4	1.5	1.4	1.2	1.2	1.4	1.2	3.0
	EF <sub>2000 BCE–1550 CE</sub>	1.7	1.7	1.7	1.2	1.3	1.7	1.3	5.7
	Residence time (year)	4557.1	3301.5	4183.1	8978.9	7192.6	3747.6	9434.0	
The whole terrestrial reservoir (ocean)	Reservoirs ( $\times 10^3$ Mg)	33.9	33.4	10.6	57.0	70.4	36.5	29.9	291.5
	EF <sub>1760–1880</sub>	2.0	2.2	2.2	1.8	1.6	1.9	1.6	3.8
	EF <sub>2000 BCE–1550 CE</sub>	2.8	3.0	3.1	2.0	1.9	2.6	1.9	8.4
	Residence time (year)	162.0	143.0	206.3	375.8	328.0	110.0	546.4	

<sup>a</sup> The reservoirs in the parentheses are only defined for the global ocean.

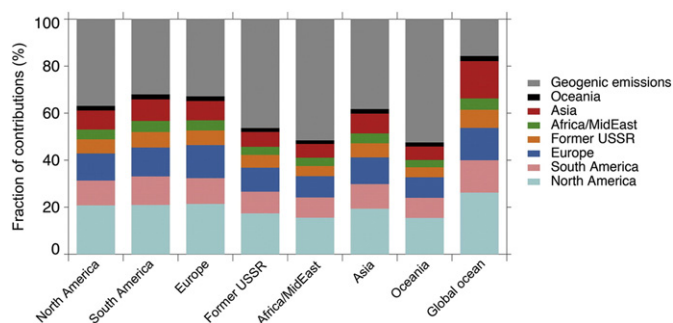
<sup>b</sup> EF<sub>1760–1880</sub> is defined as the ratio of Hg content at present day to that in pre-industrial era (1760–1880).

<sup>c</sup> EF<sub>2000 BCE–1550 CE</sub> is defined as the ratio of Hg content at present day to that in pre-large-scale mining era (2000 BCE–1550 CE).

<sup>d</sup> The residence time  $\tau$  is calculated based on the equation  $\tau_i = 1 / \sum_j k_{ij}$ , where the rate coefficients  $k_{ij}$  describing the flow from reservoir  $i$  to reservoir  $j$ .

North America is the largest anthropogenic contributor to the continental receptors (~19%) followed by Europe (~11%) and South America (~10%). Historical anthropogenic releases from Asia account for 8% of soil Hg in North America, which is smaller than the proportion (~17%) from previous studies (Jaffe and Strode, 2008; Chen et al., 2014). Chen et al. (2014) used the AMAP/UNEP atmospheric Hg emission inventory for the year 2005 (Pacyna et al., 2010) to simulate the source-receptor relationships among eleven continents, and they suggested that East Asia was the largest continental source for all other continents in 2000s. However, in the all-time perspective, the developed world (North America and Europe) that released a large quantity of Hg in the late 19th century (Fig. 1), is the largest anthropogenic source for the continental receptors. The difference results from the all-time simulation (2000 BCE–present) in this study and only the present-day simulation (2000s) in previous studies. For present-day simulation, modern anthropogenic emission inventories are used and large contributions from natural sources (weathering, volcanoes) and reemissions (terrestrial and oceanic reemissions) are estimated (Corbitt et al., 2011; Pirrone and Keating, 2011; Chen et al., 2014). All-time simulation separates the legacy contributions of historical anthropogenic releases from the large reemissions.

The global ocean is more influenced by historical anthropogenic releases than the continents. The contributions of historical anthropogenic releases from North America, Asia, Europe, and South America are 26%, 16%, 14% and 14%, respectively. Some previous studies reported that Asia was the largest anthropogenic contributor to the global ocean

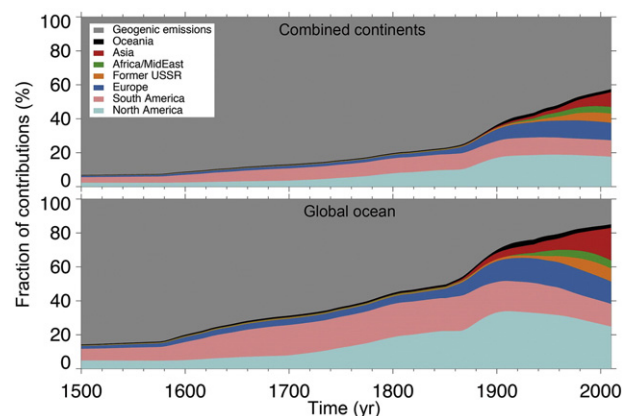


**Fig. 2.** Fractional contributions of Hg content in the continental receptors and global ocean originating from different sources at present day (post-1990).

(Corbitt et al., 2011; Pirrone and Keating, 2011). However, we find that North America is the largest anthropogenic source. Amos et al. (2013) also showed the largest anthropogenic contribution from North America, but the magnitude was smaller than our result. That study only investigated the historical anthropogenic releases during 1850–2008 and did not include riverine Hg discharges. Above all, the legacy impacts of historical anthropogenic releases are significant on the present-day source-receptor relationships of Hg.

### 3.3. Historical trend in source-receptor relationships of Hg

Fig. 3 compares the trends in contributions of Hg content in the combined continents and global ocean originating from seven continental sources and global geogenic emissions since 1500. With the increase of anthropogenic releases (Fig. 1), the total contribution from continental anthropogenic sources has increased from 7% in 1500 to 57% in 2008 for the combined continents, and from 14% in 1500 to 85% in 2008 for the global ocean. Despite the steady global geogenic emissions assumed in this study, the decrease in contribution from the geogenic emissions could be attributed to the growth of anthropogenic releases and relative growth of Hg content in all these reservoirs. The anthropogenic releases from North America, South America, and Europe have dominated the total anthropogenic contribution before 1880s. With the increase of



**Fig. 3.** Trends of Hg in the global ocean and combined continents originating from different sources (1500–2008), given as the fractional contributions.

anthropogenic releases, the anthropogenic contribution from the developing world (Asia, Africa/Middle East and the Former USSR) has increased from 1% in 1880 to 18% in 2008 for the combined continents, and from 3% in 1880 to 32% in 2008 for the global ocean. This indicates that the developing world has been an important source region since the 1880s. The anthropogenic releases from North America peaked in the 1880s, and subsequently a rapid growth in North American contribution was observed during the 1880s to the 1920s. Due to timing delay of impacts from historical releases (Amos et al., 2013), the contribution from North America to the global ocean reached the peak of 34% in the 1910s. The timing delay could be also inferred from the revised chronology for the Upper Fremont Glacier (UFG) ice cores (Chellman et al., 2017). The increases of Hg in UFG were observed in the early-20th century rather than the mid-19th century Gold Rush with large anthropogenic releases, indicating more than half a century for the timing delay of impacts from historical releases. However, the peak was not observed for the combined continents and North American contribution has been constant since the 1920s. This is mostly attributed to larger Hg retention by soils than oceans (Smith-Downey et al., 2010; Obrist et al., 2014), which leads to slower timing delay on soils than oceans. Therefore, the decrease of North American contribution to the

continents on a global scale has not been observed so far. Asia has been an important continental source since the 1920s, and its contribution increases from 1% in 1920 to 9% in 2008 for the combined continents, and from 4% in 1920 to 19% in 2008 for the global ocean.

For the global ocean, the critical sources have changed during the past 500 years. Before the 1920s, global geogenic emissions were the dominant source, followed by historical anthropogenic releases from North America and South America. During the 1920s to the 1990s, historical anthropogenic releases from North America were the largest source, followed by global geogenic emissions, historical anthropogenic releases from South America and Europe. In the modern era, historical anthropogenic releases from North America and Asia have been the two largest sources. However, for the combined continents, global geogenic emissions have always been the dominant source during the past 500 years. Comparison between Fig. 1 and Fig. 3 suggests that although the anthropogenic releases from Asia have exceeded North America since the 1970s, Asian source contributions to the global ocean and combined continents have not exceeded North America so far. Large anthropogenic releases in the late 19th century from North America and South America result in their largest contributions of Hg in the global ocean and combined continents up to the modern era.

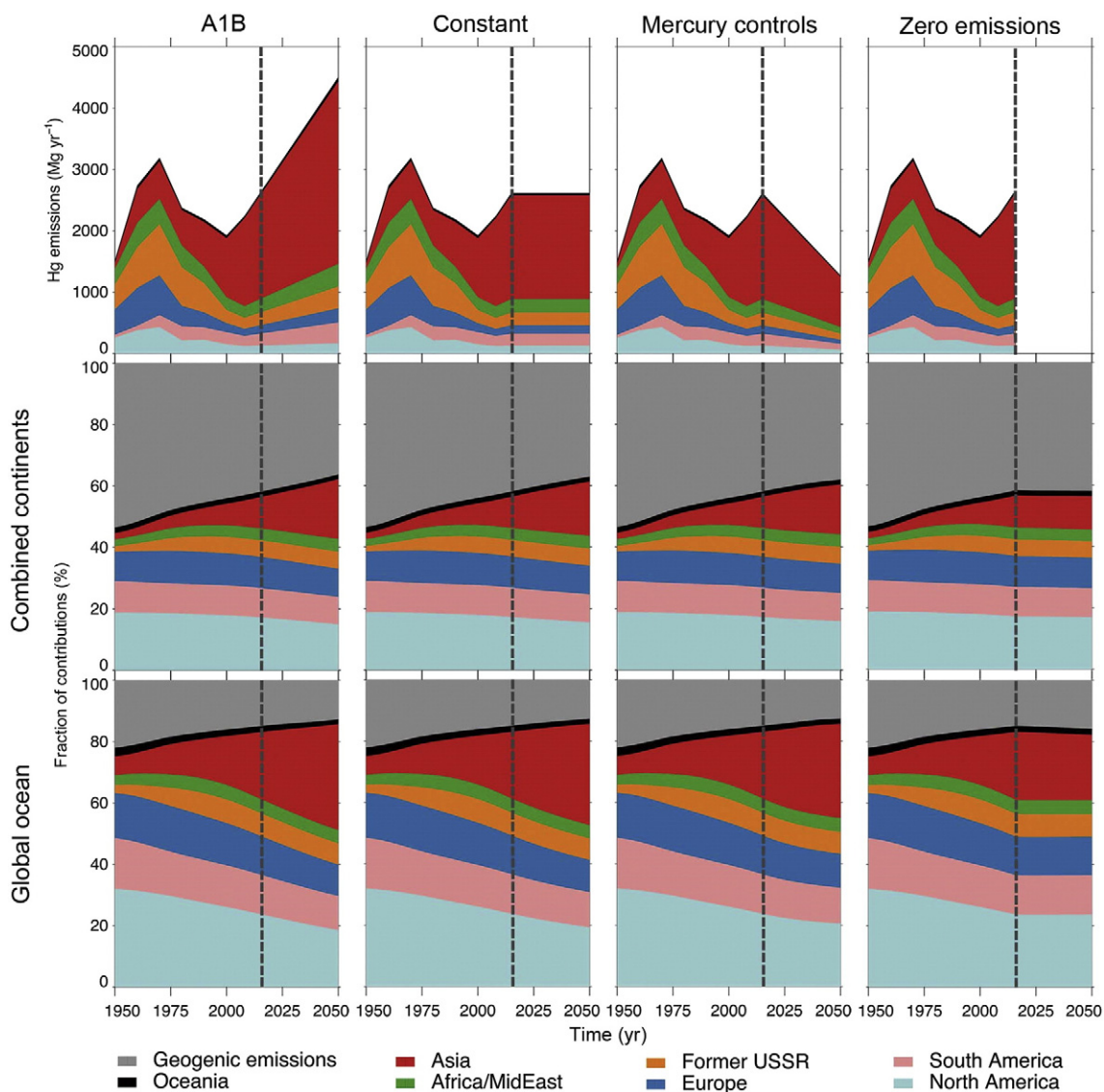


Fig. 4. Trends of Hg in the global ocean and combined continents originating from different sources under different hypothetical future emission scenarios up to 2050 (fractional contributions). The four scenarios (A1B, "constant", "mercury controls" and "zero emissions") are described in Section 2.2. The dash line indicates the year 2015.

### 3.4. Projections for the future source-receptor relationships of Hg

Fig. 4 compares the future changes of global source-receptor relationships under the four projected scenarios (A1B, “constant”, “mercury controls” and “zero emissions”). Under the A1B scenario, with the increase of anthropogenic emissions from Asia, the contribution from Asia to Hg reservoirs will increase rapidly after 2015, with a 90% increase for the combined continents and a 58% increase for the global ocean during 2015 to 2050. On the other hand, a sustained decrease will be observed for the anthropogenic contributions from the developed world. Asia will exceed North America as the largest contributor to the global ocean in 2019. Under the “constant” scenario, the contribution from Asia will still increase even if the future emissions stay constant. Under the “constant” scenario, compared with the A1B scenario, the contribution from Asia will increase with 73% for the combined continents and 52% for the global ocean during 2015 to 2050. Under the “mercury controls” scenario, although Asia will also exceed North America as the largest contributor to the global ocean in 2019, but the increase of Asian contributions during 2015 to 2050 is only 41% for the global ocean, which is smaller than the two scenarios above, especially smaller in the period 2030–2050. Under the “zero emissions” scenario, the contributions from continental sources will nearly stay constant, with a slight decrease for Asia (−2%) to the global ocean. This bounds to the maximum benefits from emission controls.

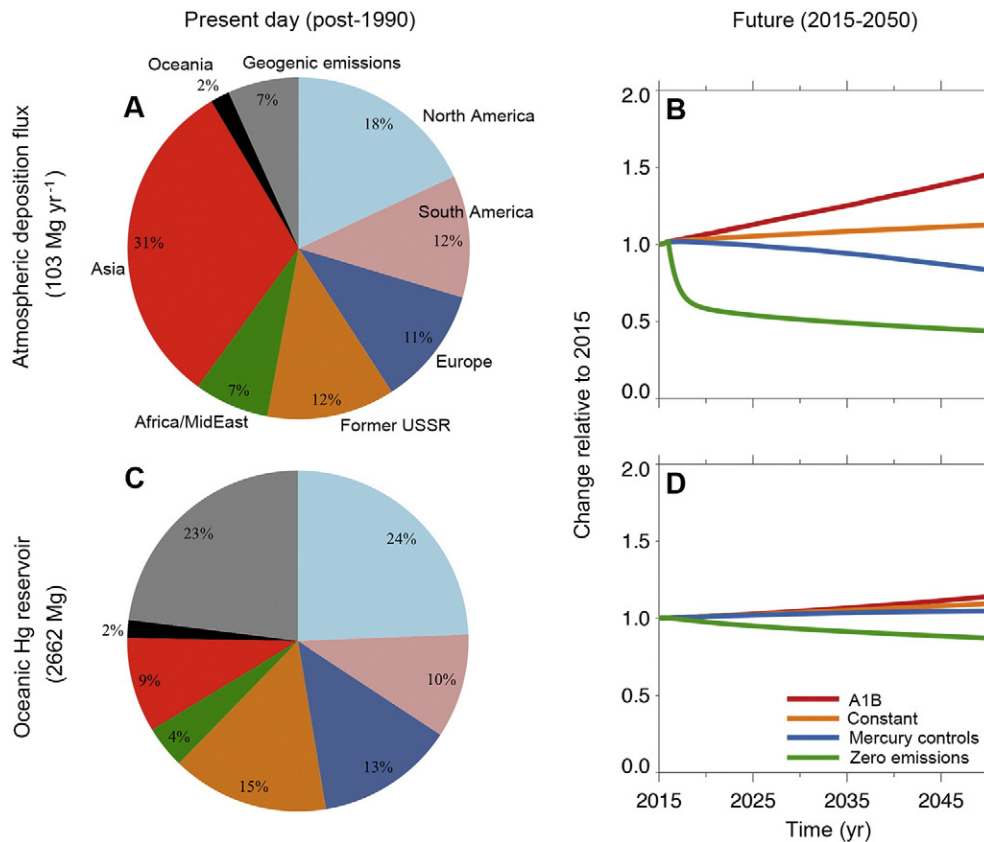
The legacy impacts of historical anthropogenic releases reveal a timing delay of impacts from historical releases on global source-receptor relationships of Hg in the future. With the increase of anthropogenic emissions from Asia since 1970, the contributions from Asia have increased and the contributions from the developed world have decreased. Although the emission projections may differ after 2015, such as the A1B, “constant”, and “mercury controls” scenarios, the changes in the spatial

contributions from the continental sources will be similar during the initial period (2015–2030), and Asia will exceed North America as the largest contributor to the global ocean in 2019. This is attributed to the legacy impacts of large historical anthropogenic releases from Asia since the 1970s. Reductions of Hg emissions under “mercury controls” scenario will moderate the changes of anthropogenic contributions after the year 2030 and reduce the future contributions from Asian anthropogenic emissions relative to the A1B scenario. Comparison between “mercury controls” and “zero emissions” indicates that stricter regulations, especially regulations on Asia, will reduce the large contributions from Asia in the future with up to 30%.

Projections for the future source-receptor relationships indicate developing regions, especially Asia, would strengthen their reductions even though the source contributions to the global ocean and combined continents have not exceeded North America so far. If Hg emissions are not effectively controlled, such as the A1B and B1 scenarios, Asia will exceed North America as the largest contributor to the global ocean in 2019 and this has a long-term adverse impact on the future environment due to the legacy impacts of large historical anthropogenic releases from Asia since the 1970s. Therefore, effective control measures are needed for Asian anthropogenic releases, as well as other similar developing regions.

### 3.5. Implications for the Arctic Ocean

Fig. 5 illustrates the source contributions of Hg in the Arctic atmosphere and ocean, and predicts the future trends in the deposited Hg and oceanic Hg under different hypothetical emission scenarios. Panel A shows that present-day atmospheric Hg deposition over the Arctic Ocean is mainly from historical anthropogenic releases from Asia, and the contribution (31%) is consistent with the finding (29–37%) from



**Fig. 5.** Source contributions of Hg in the Arctic atmosphere and ocean, and projected trends under different hypothetical future emission scenarios. Panels A and C show the source contributions of Hg in the atmospheric deposition and oceanic reservoir at present day (post-1990), respectively. Panels B and D show the projected trends of Hg in atmospheric deposition and oceanic reservoir up to 2050, respectively. The projected trends are expressed as the changes relative to the value in 2015.

Durnford et al. (2010). Besides Asia, historical anthropogenic releases from North America (18%), South America (12%), the Former USSR (12%) and Europe (11%) also contribute a lot to atmospheric Hg deposition over the ocean. Geogenic emissions show smaller contributions than these anthropogenic sources. Hg from the continental and geogenic sources flows into the Arctic Ocean via atmospheric deposition, river discharges and oceanic currents. Atmospheric deposition is the most critical intermediate process among the three processes, and Hg flows into the ocean from atmospheric deposition is about two times larger than that from oceanic currents (Table A.3). We simulate Hg flows via the three intermediate processes and classify the source contributions of continental and geogenic sources to the Arctic Ocean Hg. Results show Asia is not the area of the largest anthropogenic source. Releases from North America rank the first with the contribution of 24%, followed by geogenic source (23%), the Former USSR (15%) and Europe (13%). Compared to oceanic Hg, the lifetime of atmospheric Hg is very short, and atmospheric deposition could only reflect emission information in recent years, but limited information for legacy impacts of historical releases. Similar to the global ocean, legacy impacts of the large historical releases from developed regions and geogenic sources are also significant for Hg in the Arctic Ocean.

Fig. 5B and D show the projected trends of Hg in atmospheric deposition and oceanic reservoir up to 2050, respectively, under the four projected scenarios. Trend of atmospheric Hg deposition is more responsive to the trend of emission scenarios than trend of Hg in oceanic reservoir. For instance, under the “zero emissions” scenario, atmospheric Hg deposition declines by  $-50\%$  rapidly and oceanic Hg declines slowly. Under the “constant” scenario, although the anthropogenic emissions keep constant, Hg in atmospheric deposition and oceanic reservoir would continue to increase in the future decades, and the increases would be  $+12\%$  and  $+9\%$  respectively during 2015–2050. We attribute the phenomenon to the legacy impacts of large historical anthropogenic releases from Asia since the 1970s. The legacy impacts of historical anthropogenic releases from mid-latitudes would lead to the potential of rising Hg in the Arctic Ocean in the future decades, which calls for more effective Hg controls on mid-latitude releases. Under the “mercury controls” scenario, atmospheric Hg deposition would decrease by  $-16\%$  and oceanic Hg would increase by  $+5\%$  instead during 2015–2050. We could predict oceanic Hg would transfer from increase to decrease after 2050. The difference indicates that more time is needed for Hg controls on the mitigation of Hg pollution in the ocean than the atmosphere.

#### 4. Conclusions

In summary, this study highlights the significant legacy impacts of historical anthropogenic releases on global source-receptor relationships of Hg. Notable findings include: 1) results indicate the source-receptor relationships are different between the all-time simulation in this study and the present-day simulation in previous studies; 2) North America is the largest contributor to the global oceanic Hg. Although anthropogenic releases from Asia have exceeded North America since the 1970s, source contributions to global Hg receptors from Asia have not exceeded North America so far; 3) future projections indicate that Asia will exceed North America as the largest contributor to the global ocean in 2019 if Hg emissions are not effectively controlled. The scientific community should revisit the contributions from Asia and North America to global Hg receptors on different time scales.

In consideration of the particular concern for Hg pollution in the Arctic Ocean, we conduct a nested simulation for the Arctic Ocean under the global simulations to examine the source of accumulated Hg in the ocean. Historical anthropogenic release from Asia is the largest source for atmospheric Hg deposition, but that from North America contributes most to oceanic Hg reservoir. Future projections reveal that the legacy impacts of historical anthropogenic releases from mid-latitudes would lead to the potential of rising Hg in the Arctic Ocean in the future decades, which calls for more effective Hg controls on mid-latitude releases.

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#### Appendix B. Appendix A. Supplementary data

The supplementary material provides further details of the method and data used in this study. Supplementary data associated with this article can be found in the online version, at doi: <http://dx.doi.org/10.1016/j.scitotenv.2017.07.182>.

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