Science of the Total Environment 610–611 (2018) 24–31

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

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

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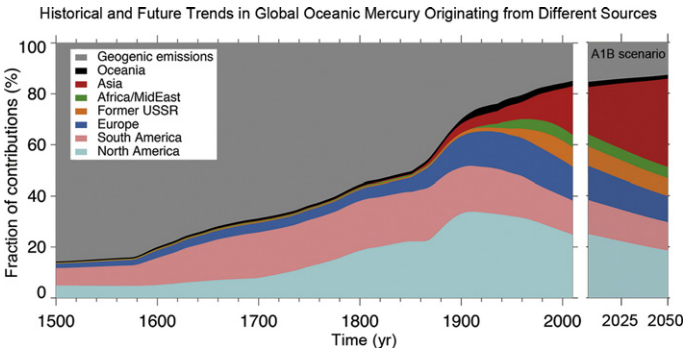
HIGHLIGHTS

- 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



ARTICLE INFO

Article history:
Received 28 April 2017
Received in revised form 19 July 2017
Accepted 20 July 2017
Available online 9 August 2017

Editor: D. Barcelo

Keywords:
GEOS-Chem
Global Terrestrial Mercury Model (GTMM)
Legacy impacts
Global simulations
The Arctic
Future projections

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 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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http://dx.doi.org/10.1016/j.scitotenv.2017.07.182
0048-9697/

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 (Hg^0) results in long-range atmospheric transport of emitted Hg. Eventually Hg^0 is oxidized to divalent Hg (Hg^{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, Hg^0 and Hg^{II}, are tracked in the model with oxidation of Hg^0 by Br atoms to Hg^{II}, photoreduction of Hg^{II} to Hg^0 in droplets and gas-particle partitioning between gaseous and particulate Hg^{II} (Holmes et al., 2010; Amos et al., 2012). Fast oxidation of Hg^0 to Hg^{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