Global Biogeochemical Cycling of Mercury: A Review

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Abstract

Mercury pollution poses global human health and environmental risks. Although mercury is naturally present in the environment, human activities, such as coal burning, have increased the amount of mercury cycling among the land, atmosphere, and ocean by a factor of three to five. Emitted to the atmosphere in its elemental form, mercury travels worldwide before oxidizing to a form that deposits to ecosystems. In aquatic systems, mercury can convert into methylmercury, a potent neurotoxin. People and wildlife are exposed to methylmercury as it bioaccumulates up the food chain. Mercury continues to circulate in the atmosphere, oceans, and terrestrial system for centuries to millennia before it returns to deep-ocean sediments. Areas of uncertainty in the global biogeochemical cycle of mercury include oxidation processes in the atmosphere, land-atmosphere and ocean-atmosphere cycling, and methylation processes in the ocean. National and international policies have addressed direct mercury emissions, but further efforts to reduce risks face numerous political and technical challenges.

Contents

1. INTRODUCTION

Mercury occurs naturally in Earth's biogeochemical system, but centuries of human activities, such as mining and fossil fuel burning, have been mobilizing increasing amounts of the element in the atmosphere, ocean, and terrestrial systems (1). Mercury is a global environmental concern: In the form of methylmercury, it is a potent neurotoxin that affects human and wildlife development and health (2). This review surveys the state of knowledge of the global biogeochemical cycling of mercury, focusing on human influences on the element's biogeochemical cycle and the processes by which mercury changes form and cycles between environmental reservoirs.

Despite longstanding and increasing national and international attention to mercury as an environmental concern, few policies have addressed the full complexity of mercury

Methylmercury: the toxic form of mercury, CH_3Hg^+

behavior in the environment or the degree to which humans have altered its biogeochemical cycle. Mercury emissions controls will have varying benefits at different spatial scales depending on the source and species of mercury controlled. Because of lag times in natural systems, reductions in emissions will not have an immediate, proportional impact on exposure (3–7). The large influence of previously emitted anthropogenic mercury on the current global cycle (representing about one-third of present-day emissions to the atmosphere) means that understanding and managing mercury already circulating in the global land surface and oceans will be an ongoing, long-term challenge. Scientific knowledge of complex biogeochemical interactions in the global mercury cycle remains uncertain.

This review presents current understanding of the global biogeochemical cycle of mercury and the processes by which it travels through the atmosphere, ocean, and land, with special attention to processes important to regional- and global-scale management approaches and policies. After a brief summary of health concerns and related domestic and international mercury reduction policies (Section 2), Section 3 introduces the global biogeochemical cycle and its associated timescales. Sections 4–7 then detail the uncertainties in and constraints on the global budgets of mercury in different portions of the global mercury cycle. Section 4 addresses emissions of mercury to the atmosphere, and Section 5 surveys atmospheric chemistry, transport, and deposition of inorganic mercury. Section 6 addresses one of the less-well understood set of processes involved in land-atmosphere interactions, followed in Section 7 by a discussion of mercury in aquatic systems. The review concludes in Section 8 with a summary and analysis of policy-relevant uncertainties and research needs.

2. HEALTH CONCERNS AND ASSOCIATED POLICY EFFORTS

Methylmercury accumulates through multiple levels in the aquatic food chain. Humans are

exposed mainly through consumption of fish contaminated by methylmercury, particularly those who have high levels of fish consumption (2). Scientific knowledge on methylmercury and its toxic effects have been reviewed in detail by the National Research Council (8) and by Mergler et al. (2). Here, I briefly summarize the health concerns posed by methylmercury and associated policy efforts to address exposure.

High-profile mercury poisoning incidents drew scientific and policy attention to the risks of methylmercury. In the 1950s, in Minamata, Japan, people who ate fish contaminated by high levels of methylmercury experienced neurological damages such as visual, auditory, and sensory disturbances, numbness, and difficulty walking (9). Those exposed in utero experienced more serious effects, including mental retardation, cerebral palsy, deafness, and blindness (8, 9). In Iraq in the 1970s, people were exposed to high levels of methylmercury by eating bread made with grain treated with a mercury-containing fungicide. Health effects included numbness; problems with vision, speech, and hearing; as well as deaths in adults and more serious neurological effects in the offspring of exposed pregnant women (8, 10).

More recently, epidemiological studies have linked exposure to methylmercury in pregnant women to neurological and developmental effects in their offspring. These effects occur at much lower doses than those that caused effects in Japan and Iraq. Three major epidemiological studies have investigated linkages between mercury consumption and health outcomes in the offspring of exposed pregnant women. Studies in the Faroe Islands (11) and New Zealand (12) showed correlations between maternal methylmercury exposure and developmental deficits in offspring, whereas a study in the Seychelles found no association (13). Cardiovascular effects, especially in adult males, have also been linked to methylmercury exposure (14, 15). The U.S. National Research Council reviewed the toxicological effects of methylmercury in 2000 and recommended that the U.S. Environmental Protection Agency

(EPA) reference dose (RfD) for methylmercury be set based on the Faroe Islands study (8). The EPA set the RfD for mercury at 0.1 μg kg body weight^{-1} day^{-1} (16), and the United States and several other countries have advised pregnant women and children to avoid eating fish high in methylmercury, such as tuna, shark and swordfish. An integrative analysis by Axelrad et al. (17) of the Faroe Islands, New Zealand and Seychelles studies estimated that offspring lose 0.18 IQ points for each part per million increase in maternal hair mercury.

Methylmercury also affects the health of wildlife exposed to elevated levels through their diet. Behavioral, neurochemical, hormonal, and reproductive effects have been observed in wildlife at concentrations present in the environment (18, 19). Methylmercury exposure and reproductive impairments are correlated in birds (20), and methylmercury at environmental concentrations could affect fish reproduction (21). Effects of environmental mercury on wildlife are reviewed in more detail by Scheuhammer et al. (21).

Methylmercury is present in both marine and freshwater fish. Freshwater fish are locally caught by recreational anglers and subsistence fishers. These populations, though small, may consume large amounts of fish and thus may be particularly vulnerable to the effects of methylmercury exposure (22). In the United States, however, most seafood consumed by the population are commercial estuarine and marine fish and shellfish, and these species account for >90% of exposure to mercury on a population-wide level (23, 24). Regulations to mitigate methylmercury risks for these different populations require actions at a variety of political scales.

Mercury has been regulated domestically in industrialized countries since at least the 1950s and internationally since the 1970s (25, 26). The United Nations Conference on the Human Environment in Stockholm in 1972 initiated action on high-priority pollutants in the marine environment, including mercury (26a). Numerous early international policies on mercury were developed in the context of regional

Lifetime (e-folding):
average time that a molecule remains in a reservoir; calculated by dividing reservoir mass by the removal rate

water quality cooperation on hazardous substances. Although emissions of mercury have decreased in the past few decades in industrialized countries (27), new knowledge about the effects of methylmercury at lower levels and concerns about long-range transport of increasing emissions from industrializing countries have led to renewed international concern about mercury. Mercury is of particular concern in the Arctic ecosystem, where high levels of mercury have been measured in biota (28). Because indigenous populations in the Arctic eat traditional foods, they are particularly vulnerable to mercury exposure.

Recent efforts by the United Nations Environment Programme (UNEP) to address mercury have been the most comprehensive international activities to date addressing the mercury problem. In February 2009, the UNEP Governing Council established a mandate to begin negotiations on a global, legally binding mercury treaty. This decision follows several years of voluntary activities under a mercury program, in which interested countries and other parties established partnerships on topics such as artisanal gold mining, transport and fate research, coal combustion, and waste management. The global treaty negotiations, beginning in 2010, will address atmospheric emissions of mercury as well as mercury in products, processes, wastes, and international trade. A critical issue in these negotiations will be the extent to which technical and financial assistance is available for developing countries to assist in implementation.

In the United States, there has been ongoing controversy over regulation of mercury emissions from coal-fired power plants. In late 2000, after a lengthy legal process, the EPA indicated its intent to regulate mercury emissions from power plants by requiring a so-called MACT (Maximum Achievable Control Technology) standard, which would require plants to reduce as much mercury as available and feasible based on existing technologies. President Bush's administration in 2003 reversed this decision, choosing instead to issue a regulation that

controlled mercury through a cap-and-trade system, which would set a national limit on mercury emissions and allow trading between individual facilities to meet that cap (28a). Although cap-and-trade systems have been successfully used to control acidifying pollutants, environmental organizations, such as the Sierra Club, criticized their application to mercury. They argued that a cap-and-trade approach would allow mercury "hot spots" near sources to persist. The Clean Air Mercury Rule, applying a cap-and-trade approach was issued in 2005, but in 2008, the District of Columbia Circuit Court vacated the rule on procedural grounds, and an appeal by industry organizations to the Supreme Court in 2009 was unsuccessful.

3. THE GLOBAL MERCURY BUDGET

Mercury is naturally mobilized from deep reservoirs in the earth to the atmosphere through volcanic and geological activity (29). The natural biogeochemical cycle of mercury involves atmospheric transport, deposition to land and ocean, and revolatilization (**Figure 1**, in black). The ultimate sink of mercury is burial to the deep-ocean sediments, which occurs very slowly. Calculated from **Figure 1**, the overall (e-folding) lifetime of mercury in the combined atmosphere-ocean-terrestrial system against transfer to the sediments is 3000 years (30). This number is a ratio of two uncertain quantities: the amount of mercury present in the atmosphere-ocean-terrestrial system divided by the preindustrial geogenic source, which is assumed to be at steady state in the preindustrial cycle. Using the same method, Mason & Sheu (1) calculated an overall lifetime of 10,000 years for mercury on the basis of a smaller geogenic source.

Humans have altered the biogeochemical cycle of mercury by coal burning, mining, and industrial activities, which bring mercury from long-term sedimentary storage into the atmosphere (**Figure 1**, in red). Because of these past and present human activities, an increased

amount of mercury is circulating and will continue to cycle for centuries to millennia among the atmosphere, ocean, and land, where it can affect the environment and humans (31). This historical mercury has enhanced the natural fluxes between the atmosphere and the ocean or land (**Figure 1**, red and black dotted lines). The quantities of mercury in environmental reservoirs in both the preindustrial and present-day cycles is uncertain, and estimates of preindustrial and present-day fluxes in the literature from specific reservoirs can vary by a factor of three or more (see **Table 1**).

The critical piece of information needed to construct the preindustrial and present-day mercury cycles is the ratio between present-day and preindustrial mercury deposition. This ratio tells us how much mercury was present in the atmosphere during preindustrial times and therefore provides a constraint on the overall cycle. On the basis of measurements of this ratio from remote lake sediment cores, it is estimated that present-day mercury deposition is three to five times larger than preindustrial deposition. In a review of sediment enrichment, Fitzgerald et al. (32) reported a ratio between present-day and preindustrial deposition in areas not affected by nearby anthropogenic emissions of 2.7 ± 0.9 for 40 U.S. and Canadian lakes, and a ratio of 2.0–2.6 for Scandinavia. Although a growing number of sediment-core records report similar increases since industrialization, some glacial core and peat records show greater enhancement. Using glacial ice core data, Schuster et al. (33) reported a 20-fold increase in mercury deposition since the 1840s, and Roos-Barraclough et al. (34) reported a 15-fold difference between current and preindustrial accumulation in peat records in Switzerland. Biester et al. (35) suggest in a recent review that the differences between peat and lake sediment records may be due to errors in both the lead-210 dating of recent peat ages and the diagenetic remobilization of lead-210 and mercury in peat cores. They thus suggest that lake sediments are a more reliable archive for estimating historical mercury accumulation.

4. EMISSIONS

Mercury is emitted to the atmosphere by both natural processes and human activities. The natural flux of mercury into the atmosphere from primary geological sources is small compared to direct present-day anthropogenic emissions of mercury.

4.1. Preindustrial Emissions

In the natural mercury cycle, primary emissions come from geological sources, which consist of volcanoes and related geological activities as well as land emissions from areas naturally enriched in mercury. The total budget of these sources is estimated (see **Figure 1**) as 500 megagrams (Mg) year^{-1}, although this number is highly uncertain. The distinction between primary emissions (with sources derived from the lithosphere) and the preindustrial secondary flux of mercury from the land, which is derived from soil mercury that originated from the atmosphere (discussed further in Section 6), is critical to understanding the magnitude of human disturbances to the natural cycle.

Primary emissions come from the so-called global mercuriferous belts. These are areas of the earth along plate tectonic boundaries that are geologically enriched in mercury, including western North America, central Europe, and southern China (36). In these areas, mercury is present as cinnabar ore (HgS), which has a characteristic red color. Mercury has been known to human societies for millennia, and early mining activities occurred in these naturally enriched regions. For example, the mercury mine at Almadén, Spain, has produced mercury since at least Roman times (37). Mercury can be released to the atmosphere in these areas through geothermal activity or by natural releases from mercury-enriched rock and soil (38). Varekamp & Buseck (36) estimated, based on measurements from hot springs and geothermal heat transport, that global geothermal emissions total 60 Mg year^{-1}. Gustin et al. (38) estimated the emissions from naturally enriched rock and soil in the United States alone

at 10–20 Mg year^{-1} on the basis of measurements in the western United States. Global estimates for primary land emissions are up to 500 Mg year^{-1} (39). Global emissions from volcanoes have previously been estimated by multiplying global SO_2 emissions estimates by the ratios of mercury to SO_2 measured in volcanic plumes (38). Extrapolation of these measured ratios, which vary over an order of magnitude, result in global estimates of 75–700 Mg year^{-1} for volcanic mercury emissions (29, 40, 41).

4.2. Anthropogenic Emissions

Estimates of direct, present-day anthropogenic emissions of mercury to the atmosphere range from 2200–4000 Mg year^{-1} (see **Table 1**). The most recent global inventory of sources is that of Pacyna et al. (42) for the year 2000 at 2200 Mg year^{-1}. The major anthropogenic source is combustion of fossil fuels, especially coal; stationary combustion represents ~60% of year 2000 emissions, according to the Pacyna et al. (42) source inventory. Other industrial processes that can release mercury to the atmosphere are cement production, nonferrous metal production, pig iron and steel production, caustic soda production, gold production, and waste disposal, as well as direct mercury production. Some have suggested that the Pacyna et al. (42) inventory may underestimate the emissions from Asia (43), and other work has estimated emissions from other sources, such as mercury release from artisanal gold mining (44).

The global total atmospheric emission of mercury has remained relatively constant since 1990 (42). Mercury emissions in the United States and Europe have declined in the past few decades, mostly owing to the cobenefits of sulfur controls in the 1980s. End-of-pipe technologies used to control sulfur or particulates, such as flue gas desulfurization or electrostatic precipitators, can also reduce mercury emissions to the atmosphere (45). In the United States, mercury-specific emissions regulations on medical waste incineration and municipal waste combustion also led to emissions decreases in the 1990s. U.S. anthropogenic emissions declined from an estimated 220 tons in 1990 to 115 tons in 1999 (46). However, emissions from Asia (about half of present-day anthropogenic emissions) continue to increase, as China and other rapidly developing countries rely more heavily on coal-based electricity. Emissions from Asia represent about 50% of global anthropogenic emissions (see **Figure 2**).

Mercury emissions sources in developed countries such as the United States and those in Europe are better known than those in developing countries. In addition, even though uncertainty in emissions from fuel combustion and various industrial processes are estimated at ±25% and 30%, respectively, uncertainty in waste disposal is a factor of 2–5. As noted above, the use of mercury in small-scale artisanal gold mining operations is potentially a large source that is neglected in most existing inventories; emissions to the atmosphere from this source have been estimated at 450 Mg year^{-1}, although this estimate is very uncertain (30). Historical mining emissions were substantially higher, exceeding 800 Mg year^{-1} (44). Pacyna et al. (42) estimate that mercury emissions in 2020 will be within ±20% of the present-day value but caution that this projection is very uncertain. Streets et al. (44a) estimate that mercury emissions in 2050 will be between 2390 and 4860 Mg year^{-1}, depending on socioeconomic and technology developments. In addition, the speciation of mercury is also an area of uncertainty that is important for its atmospheric fate, as discussed below.

5. ATMOSPHERIC PROCESSES

Mercury is emitted from geological sources and from land and ocean surfaces in its elemental form [Hg(0)]. Hg(0) is also emitted by anthropogenic sources such as coal-fired power plants. However, anthropogenic sources can also emit mercury in two different forms: divalent mercury [Hg(II)] and mercury associated with particulate matter [Hg(P)].

Hg(0) is the most abundant form of mercury in the atmosphere, with a global mean

concentration of about 1.6 ng m^{-3} in surface air. With an atmospheric lifetime of about 0.5–1 year, Hg(0) is well mixed in the global atmosphere (47). Because Hg(II) and Hg(P) are more soluble in water than Hg(0), they are the predominant forms of mercury deposited to ecosystems through wet and dry deposition. Hg(II) and Hg(P) have a much shorter lifetime than Hg(0) in the atmosphere (days to weeks), and their surface concentrations range from 1–100 pg m^{-3}. Understanding the conversions between Hg(0) and Hg(II)/(P) in the atmosphere and their deposition processes are important for assessing the sources and pathways of mercury contamination (1, 48, 49). The sections below discuss the scientific knowledge and uncertainties in two areas: the distribution and atmospheric chemistry of conversions between Hg(0), Hg(II) and Hg(P) in the atmosphere and the understanding and quantification of deposition processes (50–52).

5.1. Distribution and Atmospheric Chemistry

Measurements and models of mercury in the atmosphere can provide insights into the chemistry and distribution of atmospheric mercury (53). Measurements of total gaseous mercury [(TGM) consisting of Hg(0) plus a small contribution of Hg(II) in the gaseous phase] are available predominantly in the northern midlatitudes (54–58), although some data are available over the oceans from cruise measurements (59–61). These measurements show that TGM is relatively well mixed in the global atmosphere. Concentrations are generally higher in the Northern Hemisphere than in the Southern Hemisphere, as most emission sources are in the Northern Hemisphere (62, 63).

The main sink of Hg(0) in the atmosphere is oxidation to Hg(II). The chemical species that acts as the primary oxidant of Hg(0) in the global atmosphere remains unknown. Previously, the most important global oxidant of mercury was thought to be ozone or OH-radicals (64, 65), and these reactions are included in most atmospheric mercury models.

However, thermodynamic calculations and analyses have shown that these reactions may not occur under atmospheric conditions (66). More recent measurements and analyses have measured rate constants for mercury oxidation by bromine (Br) (67). Mercury reduction has been observed in rainwater samples, and thus it has been hypothesized that this reaction may occur in the atmosphere as well (48).

Measurements of Hg(II) in the atmosphere can help constrain uncertainties in redox reactions. These measurements are made by collecting Hg(II) on potassium chloride (KCl)-coated denuders and reducing it to Hg(0) (68). The species measured is referred to as reactive gaseous mercury (RGM), which is an operationally defined quantity. The exact speciation of Hg(II) in the atmosphere remains unknown, but it is thought to be predominantly HgCl$_2$. Oxidation of Hg(0) to Hg(II) is thought to be controlled by photochemistry, and thus production of Hg(II) is expected to peak at midday. Both Br and OH are photochemically produced. RGM has a diurnal cycle that peaks at midday and is at its minimum at night (43). Comparisons of data from Okinawa Island, Japan, (43) with model simulations have shown that while simulated Hg(II) using OH chemistry peaks in the early afternoon, measured RGM peaks earlier—around noon. Because Br rises earlier in the day than OH, this has been interpreted as evidence that Br may be responsible for Hg(0) oxidation (69). RGM concentrations and UV radiation are also correlated in the Atlantic marine boundary layer, suggesting in situ production consistent with Br (70). Even though initial model evaluations have shown that Br could be the dominant global oxidant of mercury (71, 72), further analyses are necessary to confirm this hypothesis.

Br also is thought to be responsible for Arctic mercury depletion events (AMDEs) (73). During AMDEs, which occur throughout polar regions in springtime, Hg(0) decreases rapidly while RGM spikes. AMDEs correlate strongly with tropospheric ozone depletion events, which are the result of rapid oxidation by halogens such as Br and Cl (74). Thus, it is

Dry deposition: settling or uptake to the surface without precipitation

Reactive gaseous mercury (RGM): an operationally defined measurement equivalent to Hg(II)(g)

thought that similar chemistry is responsible for AMDEs. As mercury is of particular concern in the Arctic ecosystem, the fate of the deposited mercury during AMDEs is of substantial interest. Snowpack measurements have shown that a portion of the mercury dry deposits during an AMDE is rapidly reduced and revolatilized (75). However, some deposited mercury may remain in the ecosystem, where it could influence ecosystems at a time of increased productivity (28). The mass balance of mercury deposition and emission to the Arctic ecosystem during AMDEs remains unknown (76).

Recent measurements have shown that RGM concentrations increase with altitude and that Hg(0) decreases. This reflects conversion of Hg(0) to Hg(II) at altitude, as the total mercury remains constant. Measurements at Mt. Bachelor in Oregon (2.7 km above sea level) show mean levels of RGM elevated over surface measurements and very high concentrations (up to 600 pg m^{-3}) in subsiding air (77). Talbot et al. (78) noted the near depletion of Hg(0) in stratospheric aircraft measurements, and Murphy et al. (79) measured mercury associated with particles near the tropopause. Because RGM concentrations in the atmosphere are so low, current measurement techniques cannot reliably measure it from aircraft at high frequency. High-altitude RGM may be an important global reservoir of mercury, and its potential to affect the surface is discussed below.

5.2. Constraints on Deposition

As noted above, because Hg(II) and Hg(P) are soluble species and have a shorter lifetime in the atmosphere than Hg(0), direct emissions of these species from anthropogenic sources are likely to deposit on a regional scale. Emissions of Hg(0) are longer lived, and large-scale patterns of oxidation and reduction will control conversion to Hg(II) and subsequent deposition. Effective policies to reduce deposition to ecosystems require knowledge of both oxidation and deposition patterns.

Quantifying deposition to ecosystems is limited because measurement networks exist only in a few regions and only for wet deposition. Wet deposition measurements are available from the U.S. Mercury Deposition Network (**Figure 3**) as well as from some stations in Europe (58, 80). As shown in **Figure 3**, the highest wet deposition in the United States is in the Southeast, and another area of elevated deposition covers the Midwest and Northeast United States. The magnitude of mercury dry deposition is uncertain, especially dry deposition of Hg(0), and few measurements are available to constrain model estimates (50, 81). Further measurements of dry deposition, especially in locations where wet deposition measurements are available, would dramatically improve scientific understanding of the mercury cycle (51).

Atmospheric models have been used to analyze total (wet plus dry) deposition patterns in the United States where wet deposition measurements are available for comparison (81a). Model analyses have shown that North American anthropogenic emissions (including the United States, southern Canada, and northern Mexico) contribute between 20% and 30% on average to deposition in the United States (82, 83). The percentage of deposition from North American sources reaches as high as 60% to 81% (82, 83) in the Midwest and Northeast United States, which are downwind of sources emitting Hg(II) and Hg(P) to the atmosphere (**Figure 4**). Although total deposition is highest in the southeastern United States, model results suggest that U.S. domestic emissions controls will have the greatest impact in reducing deposition in the Midwest and Northeast. It has been hypothesized that high deposition in the southeastern United States results from scavenging from the global pool of elevated RGM at altitude through convective activity in the summertime (83, 84), though some have argued that local sources are responsible for part of the elevated deposition there (85–87). Global and regional atmospheric models that are widely used for source attribution do not capture deposition on a very local scale, which is elevated in the vicinity of power plants (88, 89). It is these very local effects that have

Wet deposition: removal from the atmosphere through precipitation

led to criticism of proposed cap-and-trade regulations for mercury, discussed in Section 2.

Chemical reactions that reduce Hg(II) to Hg(0) could lessen the impact of Hg(II) emissions on a regional scale. However, as noted above, the importance of this process is uncertain (90). Some measurements and modeling efforts have shown that reduction of mercury by SO_2 in power plant plumes could decrease wet deposition of Hg(II) up to 10% in areas downwind of coal-fired power plants (91). These and other uncertainties are the subject of ongoing research.

6. TERRESTRIAL CYCLING

Wet and dry deposition brings mercury to the terrestrial surface. Outside of the geologically enriched areas discussed in Section 3, most mercury in the global land surface is deposited as Hg(II) from the atmosphere. Upon deposition, a portion of this mercury will rapidly revolatilize to the atmosphere. The remainder will be incorporated into a long-lived soil pool, where it can slowly evade to the atmosphere on a timescale of centuries to millennia. This section traces the pathway of mercury deposited to the terrestrial system.

6.1. Prompt Recycling

Newly deposited mercury has been shown to preferentially revolatilize, in a phenomenon that has been termed prompt recycling (30). Evidence for this phenomenon comes in part from the METAALICUS experiment (92), in which radiolabeled mercury was added to a watershed in eastern Canada and subsequently monitored over several years. Results from this watershed have shown that recently deposited mercury (diagnosed from measuring the radiolabeled form) is more available for reduction and subsequent emission as Hg(0) than mercury already present in the system. On the basis of this experiment and other studies, it is estimated that 5% to 60% of deposited mercury is promptly recycled to the atmosphere, depending on the surface, with the higher values for

water and surface snow (92–95). This mercury is also preferentially available for conversion into methylmercury (92). The mechanism for this prompt recycling remains a topic of research.

6.2. Incorporation into Vegetation and Soil Pools

Isotope measurements have shown that newly deposited mercury that does not immediately reduce and revolatilize is initially preferentially associated with vegetation (92). In terrestrial vegetation, mercury in aboveground biomass originates primarily from the atmosphere, whereas mercury in the roots comes from the soil (96). Hg(II) deposits on leaves through precipitation (wet deposition) and dry deposition. Uptake of Hg(0) is thought to occur at the leaf interior through gas exchange at the stomata (81). On seasonal scales, mercury deposited to vegetation is incorporated into the soil pool via throughfall and literfall (97).

The majority of mercury in the terrestrial system (>90%) resides in soil and is associated with organic matter, where it binds strongly to reduced sulfur groups (98). Available soil concentration measurements range from 20–70 ng g^{-1} (99–102). On the basis of these measurements, it has been estimated that the total global soil mercury burden is on the order of 10^6 Mg for the top 15 cm of soil (30) and that human activities have enhanced this burden by approximately 15% (1). This calculation underlies the estimate shown in **Figure 1**. Adsorbed mercury remains in the soil, whereas mercury in solution can be methylated or run off into a watershed (103). Factors controlling the adsorption and desorption of mercury in soil systems include the type of soil and dissolved species, such as S-, Cl-, and dissolved organic carbon (103).

6.3. Terrestrial Emissions

Mercury returns to the atmosphere from soils by reduction to Hg(0) and subsequent diffusion or mass transport through soil and into the atmosphere. Total present-day terrestrial emissions are estimated in **Figure 1** at

1700 Mg year^{-1}, with an additional contribution of 600 Mg year^{-1} from biomass burning. Other estimates are shown in **Table 1**. Lindberg et al. (104) estimated the range of potential values of total global vegetation emissions between 1400 and 3200 Mg year^{-1} by extrapolating measurements from a Tennessee forest. Terrestrial emissions are a substantial, yet poorly understood, component of the global total mercury emission. Obrist (96) suggests that terrestrial uptake can in part explain the seasonal variation of Hg(0) atmospheric concentrations observed in the northern midlatitudes, which have a seasonal variability similar to that of CO_2. The seasonal variation of natural emissions is closely linked to the oxidation processes of mercury in the atmosphere.

Mercury reduction and subsequent emission from soils is considered predominantly a physical (abiotic) process, although some recent research has shown that mercury speciation in and emission from soil can be partially controlled by biotic processes (105). The reductants of mercury in soil can be species such as Fe^{2+} and humic and fulvic compounds (103). Reduction and volatilization processes can be enhanced by temperature (106, 107) and solar radiation (108, 109). In some dry ecosystems, increased soil moisture (as a result of precipitation events) promotes volatilization of mercury (110).

Mercury can also be released to the atmosphere when the organic matter it is bound to is burned. Evidence for mercury emissions in biomass burning comes in part from atmospheric measurements, where mercury has been measured in atmospheric plumes that originate from burning areas. Biomass-burning emissions have been estimated at 670 $\pm$ 330 Mg year^{-1}, with 168 $\pm$ 75 Mg of that total from boreal forests (111). Turetsky et al. (112) calculated that emissions from boreal forests and peat soils varied over an order of magnitude between low- and high-fire years, and in high-fire years, were nearly comparable in magnitude to North American industrial emissions. Wildfires are expected to increase as a result of future climate warming (113). Changes in burning and other climate-related changes, such as loss of peatlands, could mobilize substantial amounts of mercury from soils into the atmosphere.

7. AQUATIC CYCLING

Human concerns about mercury are largely driven by the dangers of methylmercury contamination. The dominant pathway of human methylmercury exposure is through eating contaminated fish. For this reason, understanding the cycling of mercury in aquatic systems is critical to environmental risk analysis. Both freshwater and marine systems are of concern, though the cycling of mercury differs in these two environments.

7.1. Freshwater Systems

Much of the research that has been conducted on aquatic mercury cycling has occurred in freshwater ecosystems. Mercury contamination of freshwater systems is widespread: In the United States in 2006, 48 out of 50 states advised residents to avoid consuming fish from certain water bodies because of methylmercury risks (114). Ecosystems are also affected by point sources of mercury owing to local contamination issues (115–117).

In areas away from point sources, atmospheric mercury reaches freshwater ecosystems by direct deposition to lake surfaces and through runoff from watersheds. Wet and dry deposition to watersheds and lake surfaces, as in terrestrial systems, is predominantly as Hg(II). Hg(II) can reduce to Hg(0), which may then volatilize to the atmosphere, as depicted in **Figure 5** (118). A small portion of Hg(II) is converted to the more toxic form of methylmercury (MeHg). Methylation of mercury is a biologically mediated process known to be facilitated by some strains of sulfate- and iron-reducing bacteria (119–121). Wetlands (122) and lake sediments (123) are important environments where methylation occurs. The spatial extent of these areas in particular ecosystems influences net methylation (124).

Methylmercury can bioconcentrate in living organisms and then further biomagnify up the food chain, as predators eat prey contaminated with methylmercury and further concentrate it in their tissues. This process of bioaccumulation means that methylmercury concentrations in predatory fish can be elevated relative to water by a factor of $\geq 10^6$ (118).

In aquatic sediments, microbes convert a small fraction of inorganic mercury (Hg(II)) to MeHg over time. Sulfur, organic carbon, sediment structure, and composition affect methylmercury production by changing the amount of bioavailable inorganic mercury and by stimulating microbial activity. In coastal sediments, sulfate-reducing bacteria are thought to be the principal agents responsible for MeHg production (123, 125, 126). These microbes thrive at the geochemical interface between oxic and anoxic conditions (127). A number of environmental factors are known to affect the rate of MeHg formation by influencing the supply of bioavailable Hg(II) and/or the activity of methylating microbes. In addition to Hg(II) concentrations, effective proxy indicators for MeHg production and accumulation identified by previous research include sulfide concentrations, total organic carbon, and redox potential (128–133). Methylmercury production has also been measured in the water column in lakes (134, 135) and in anoxic coastal waters (R. Mason, personal communication).

Ecosystems respond to changes in deposition on varying timescales, depending on ecosystem type and the influence of watersheds (97). Knightes et al. (3) used coupled ecosystem models to illustrate fish methylmercury responses to a 50% decline in deposition in a variety of lake ecosystems. In their model, concentration decreases were characterized by an initial decline on a timescale of one to three decades, followed by a slower response on the order of centuries. Response times were longest in watershed-dominated ecosystems, and shortest in ecosystems that received mercury from direct deposition to the water surface. These findings are similar to those of the METAALICUS experiment, discussed above (6), in which fish methylmercury concentrations responded rapidly to mercury added directly to the lake surface, and they are also consistent with mesocosm studies of ecosystem loading (136).

7.2. Marine Systems

The forms of mercury in the ocean are Hg(0), Hg(II), methylmercury, dimethylmercury, and particulate and colloidal mercury (137, 138). Concentrations of total mercury differ among the global oceans. Average ocean concentrations of total mercury are on the order of 1.5 picomolar (pM) (139), although higher concentrations have been measured in the Mediterranean (2.5 pM) (140) and North Atlantic (2.4 pM) (141) and lower concentrations in the Pacific (1.2 pM) (142, 143). Recent modeling has shown that concentrations in most ocean basins are not at steady state with respect to atmospheric inputs and will likely continue to increase over the next several decades (143).

Exchange of mercury at the surface of the ocean is thought to be rapid and extends the atmospheric lifetime of mercury (144). Similar to processes described in more detail above in freshwater systems, Hg(II) can be deposited to the ocean by dry and wet deposition, and Hg(0) by dry deposition. Hg(II) deposited to the ocean can be reduced to Hg(0), adsorbed onto particles, or methylated. Reduction from Hg(II)$_{(aq)}$ to Hg(0) can be both biologically and photochemically mediated, and oxidation processes can also be significant (1).

A topic of some debate in the literature is the relative importance of different mercury methylation processes and locations in the ocean (4, 145). Historically, mercury species have been difficult to measure in seawater owing to analytical detection limits (146). Reported concentrations of methyl mercury range from 2% to 35% of total mercury concentrations in the ocean (4, 146a). Recent advances in capabilities mean that new data are now being collected for many regions (147–149). Mercury methylation can occur in the sediments

of continental shelf regions (150) and estuaries (151–153), within the water column (154), or at deep-ocean hydrothermal vents (155). Mercury methylated through these different processes is likely to affect fish concentrations on different timescales (7). Better knowledge of these ocean processes could help decision makers to assess the potential benefits of reductions in mercury deposition to methylmercury concentrations in marine fish.

8. POLICY-RELEVANT UNCERTAINTIES AND RESEARCH NEEDS

In order to effectively reduce methylmercury exposures, future management approaches need to consider the dynamics of the mercury problem on various spatial and temporal scales. Reducing deposition in different locations, such as the Southeast and Northeast United States, will require controls on different sources and mercury species. Monitoring the influence of emissions reduction policies, and adapting them, should take into account how ecosystems and oceans might respond to mercury changes over time. Further research can help to illuminate these policy choices.

To date, policies to mitigate methylmercury exposure have focused on controlling direct anthropogenic emissions. On the basis of current knowledge of the global mercury cycle, these fluxes represent only about a third of present-day emissions and deposition. Of similar importance is the historical legacy of previous anthropogenic emissions, which continue to cycle in the environment. Preventing this mercury from emitting, depositing, and converting to methylmercury could become a policy objective if direct emissions reductions are judged insufficient to minimize health and environmental risks. This would require improved knowledge of land-atmosphere cycling and biogeochemical controls on methylation processes, and it might involve coordinating mercury reductions with other emissions reduction policies or land-use choices. For example, a large portion of the anthropogenic mercury released since indus-

trialization currently resides in soils. Increased biomass burning or loss of peatlands could cause large mercury releases, and preventing these changes would avoid releases.

A critical challenge for national and international policies to reduce mercury, as shown by the controversy over the U.S. Clean Air Mercury Rule, is determining how to assess the benefits of reductions in emissions of different forms of mercury from different locations. Although policies have to date focused on total mercury, reducing Hg(II) and Hg(P) will largely have local and regional benefits, whereas Hg(0) reductions will have global benefits. The technologies and costs of regulating the various forms of mercury differ. This is a particular challenge for international regulations. China, for example, may want to limit Hg(II) and Hg(P) emissions owing to domestic concerns, but international transport of Hg(0) motivates its inclusion in international negotiations. These distinctions become important especially where international activities provide technical and financial assistance for developing country actions. From an atmospheric perspective, identifying and quantifying the chemical processes of atmospheric oxidation and reduction would provide more knowledge of where and how mercury circulates globally and allow managers to target monitoring and abatement activities to sensitive ecosystems. Correspondingly, improved measurements of dry deposition would provide not only constraints on the global budget of mercury and on the balance of direct anthropogenic, historical, and natural emissions, but also give improved quantitative information on how much mercury is entering sensitive ecosystems.

Reductions in mercury emissions will not necessarily result in immediate reductions in methylmercury exposure. In some cases, such as certain ocean basins, concentrations may continue to rise despite emissions cuts because they are not yet at steady state with respect to deposition. However, mercury emissions reductions could alter these trajectories. In other systems, such as certain lake ecosystems, responses could be rapid. Further knowledge of the mechanisms

and timescales of methylmercury responses to changes in deposition in both freshwater and marine systems could help decision makers monitor and adapt mercury reduction policies at a variety of spatial scales.

Finally, it is well-known that the biogeochemical timescales in the mercury cycle are quite long, on the order of centuries to millennia, compared with those normally considered in policy analysis. Human activities over the past 150 years have already modified the global biogeochemical cycle of mercury in ways that will not recover on timescales of years to decades, and human alterations of these cycles are continuing. This suggests that, similar to addressing human influences on the global carbon cycle, policy interventions to reduce risks will be most effective if they combine aggressive mitigation with potential adaptations.

SUMMARY POINTS

1. Human activities have increased the amount of mercury cycling through the atmosphere-ocean-terrestrial system by about a factor of three.

2. Most anthropogenic emissions of mercury (60%) are from combustion of fossil fuels, especially coal. Emissions have declined in recent years in the United States and Europe but are increasing in Asia, which is responsible for 50% of the global anthropogenic emissions.

3. Mercury emitted to the atmosphere will remain in the atmosphere-ocean-terrestrial system for ~3000 years before returning to the sediments.

4. Outside geologically enriched areas, most mercury in the land system originates from the atmosphere and is present in soils, where it is bound to reduced sulfur groups in organic matter. Through reduction and diffusion to the surface, this mercury can be emitted back to the atmosphere.

5. In the oceans, mercury levels have not yet reached steady state with respect to current levels of deposition. This means that if anthropogenic emissions continue at their present level, ocean concentrations in many ocean basins will increase in the future.

6. Sulfate-reducing bacteria convert mercury into the toxic form of methylmercury. This process is affected by factors such as the sulfur cycle, ecosystem pH, and the presence of organic matter.

7. Mercury has been regulated since the 1950s in industrialized countries and internationally since the 1970s, although global transport of mercury continues to be of concern, especially in the Arctic ecosystem. Bioaccumulation of mercury in the Arctic contaminates wildlife and traditional food sources.

8. The United Nations Environment Programme will begin negotiations in 2010 for a global treaty to regulate mercury.

FUTURE ISSUES

1. The predominant atmospheric oxidant of mercury, which converts elemental mercury to the deposited form of divalent mercury, remains unknown. Ozone, OH radicals, and Br are potential candidates.

2. No existing monitoring network measures dry deposition of mercury, and wet deposition is systematically monitored only in North America and Europe. Improved deposition monitoring would help constrain the global biogeochemical cycle of mercury.

3. The processes by which mercury is methylated in the ocean are a topic of continuing research. The importance of methylation in coastal regions, in the water column, or at deep-ocean hydrothermal vents remains uncertain.

4. Methylmercury concentrations do not respond linearly and instantaneously to changes in anthropogenic emissions. More research is necessary on the timescales of these responses in order to inform policy.

5. Negotiations for a global treaty on mercury will address the international types and extent of mercury regulations as well as potential technical and financial assistance for developing countries for implementation of the regulations.

6. Future efforts to control mercury need to address reducing different forms of mercury emissions (Hg(0) and Hg(II)), which will benefit global and regional scales, respectively.

7. The historical and persistent nature of human modifications to the global mercury cycle suggest that policy interventions to reduce risks will be most effective if they combine mitigation and adaptation.

DISCLOSURE STATEMENT

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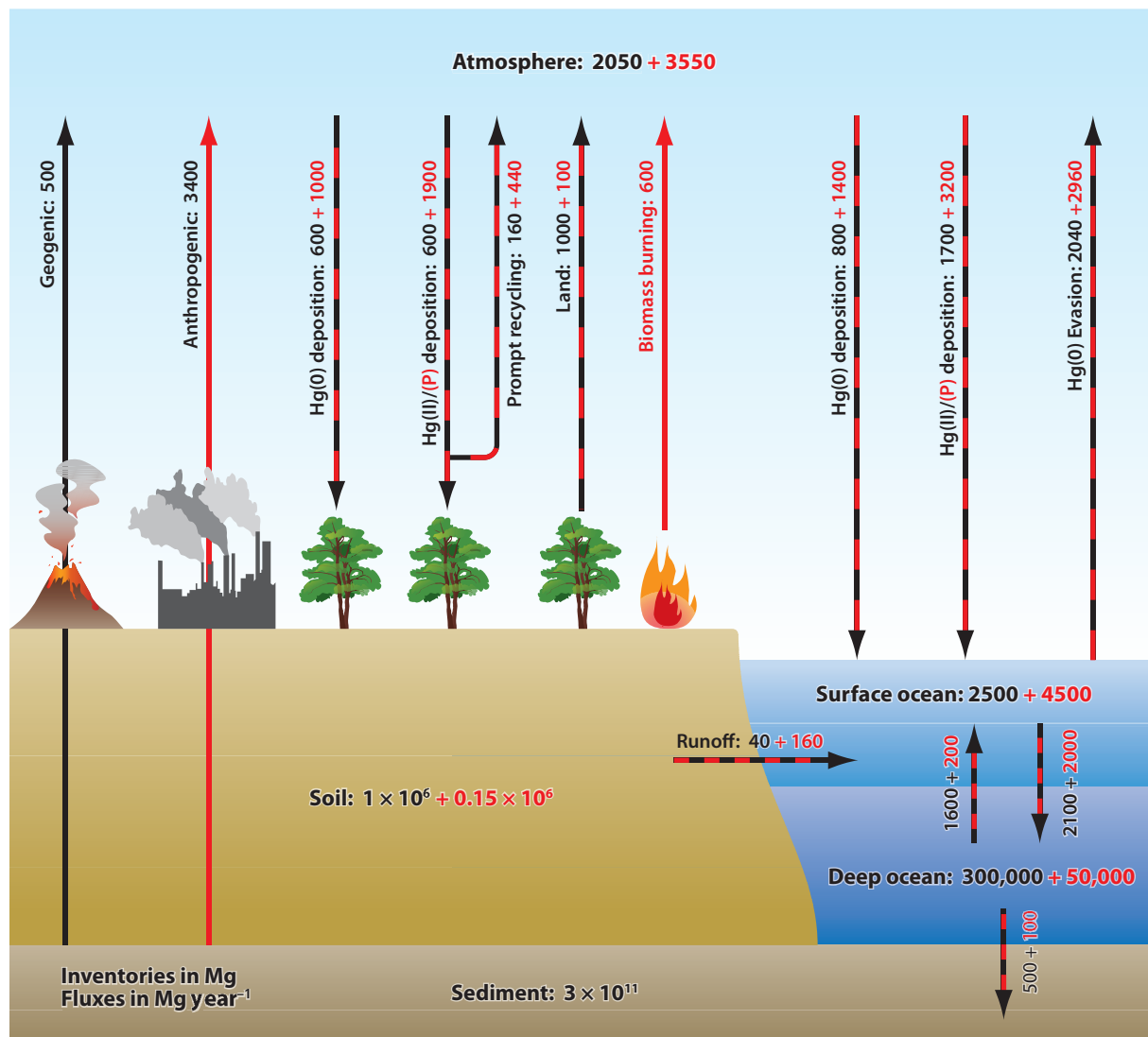
Figure 1

Global biogeochemical cycle for mercury. Natural (preindustrial) fluxes [megagrams (Mg) year^{-1}] and inventories, in Mg, are noted in black. Anthropogenic contributions are in red. Natural fluxes augmented by anthropogenic activities are noted by red-and-black dotted lines. Modified from Selin et al. (30). A mean enrichment factor of three between the preindustrial and present-day mercury deposition, based on remote sediment cores, is used as a constraint.

Table 1 Global biogeochemical cycles for mercury in the literature (Mg year^{-1}) (158)

Flux category	Selin et al. (30)	Sunderland & Mason (143)[a,b]	Mason et al. (157)[a]	Lamborg et al. (139)[a]	Mason & Sheu (1)[a]
Atmospheric sources					
Land	2,800	2,100	1,000	1,000	1,600
Preindustrial[c]	1,660	880	1,000	1,000	900
Anthropogenic	1,140[d]	1,220	0	0	700
Ocean	5,000	2,620	2,000	800	2,600
Preindustrial	2,040	1,360	600	400	1,300
Anthropogenic	2,960	1,260	1,400	400	1,300
Anthropogenic primary	3,400	2,260–3,380	4,000	2,600	2,400
Atmospheric sources (total)	**11,200**	**6,980–8,100**	**7,000**	**4,400**	**6,600**
Atmospheric sinks					
Land	4,100	2,640	5,000	2,200	3,520
Preindustrial	1,200	880	1,000	800	850
Anthropogenic	2,900	1,760	4,000	1,400	2,670
Ocean	7,100	2,800–5,800	2,000	2,000	3,080
Preindustrial	2,500	1,360	600	600	1,350
Anthropogenic	4,600	1,440–4,440	1,400	1,400	1,730
Atmospheric sinks (total)	**11,200**	**5,440–8,440**	**7,000**	**4,400**	**6,600**
Other fluxes					
Runoff	200	380	200	–	200
Preindustrial	40	80	60	–	40
Anthropogenic	160	300	140	–	160
Burial	600	220	200	400	200
Preindustrial	500	80	60	400	90
Anthropogenic	100	140	140	0	110
Atmospheric burden					
Preindustrial	2,050	1,600	1,600	1,800	1,600
Anthropogenic	3,550	4,000	3,400	3,400	3,400
Atmospheric burden (total)	**5,600**	**5,600**	**5,000**	**5,200**	**5,000**

[a]Converted from megamole (Mmol) year^{-1}.
[b]Ranges reflect different model simulations.
[c]Including geogenic source.
[d]Including biomass burning.
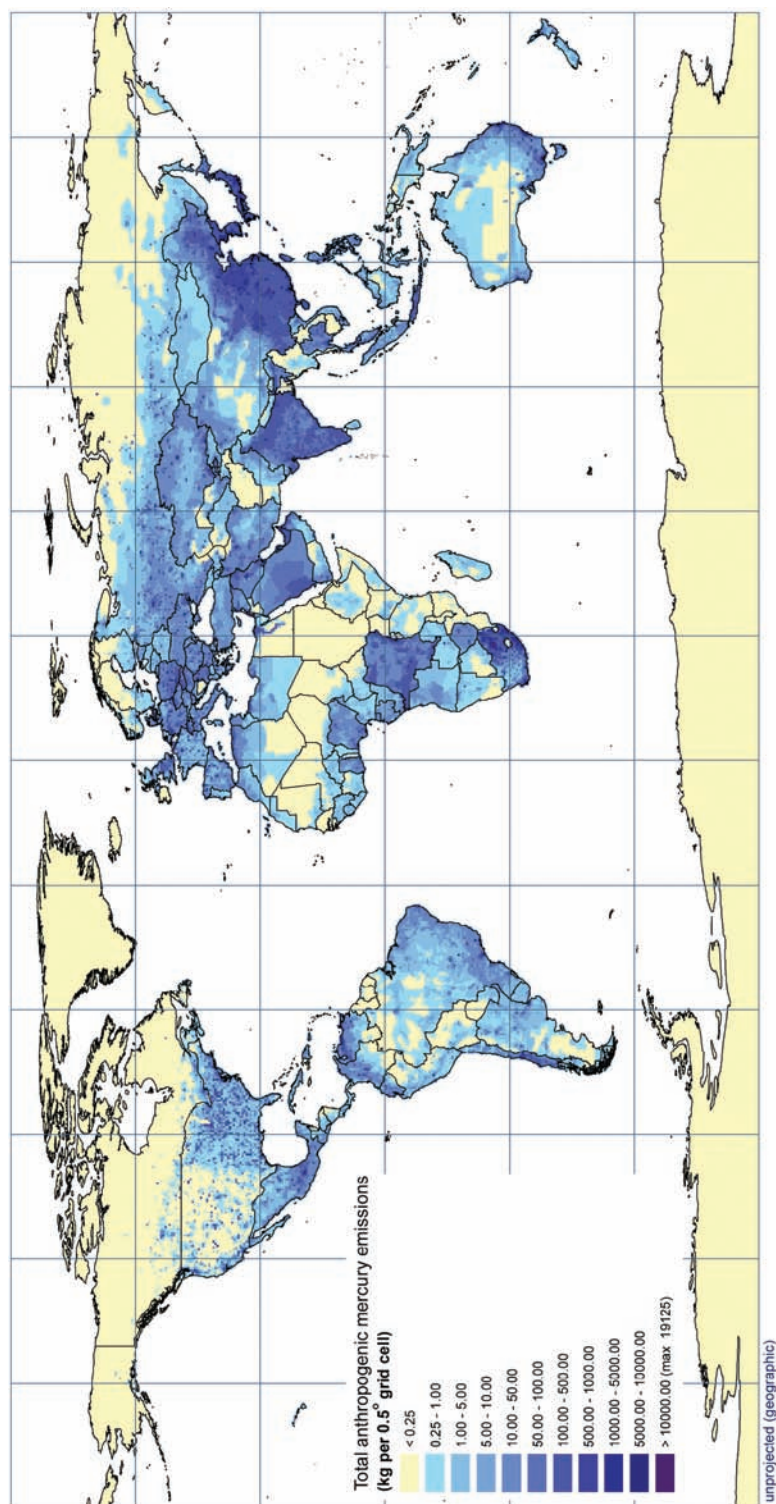
Figure 2

Spatial distribution of anthropogenic mercury emissions to the atmosphere for 2000, Pacyna et al. (156).

National Atmospheric Deposition Program/Mercury Deposition Network
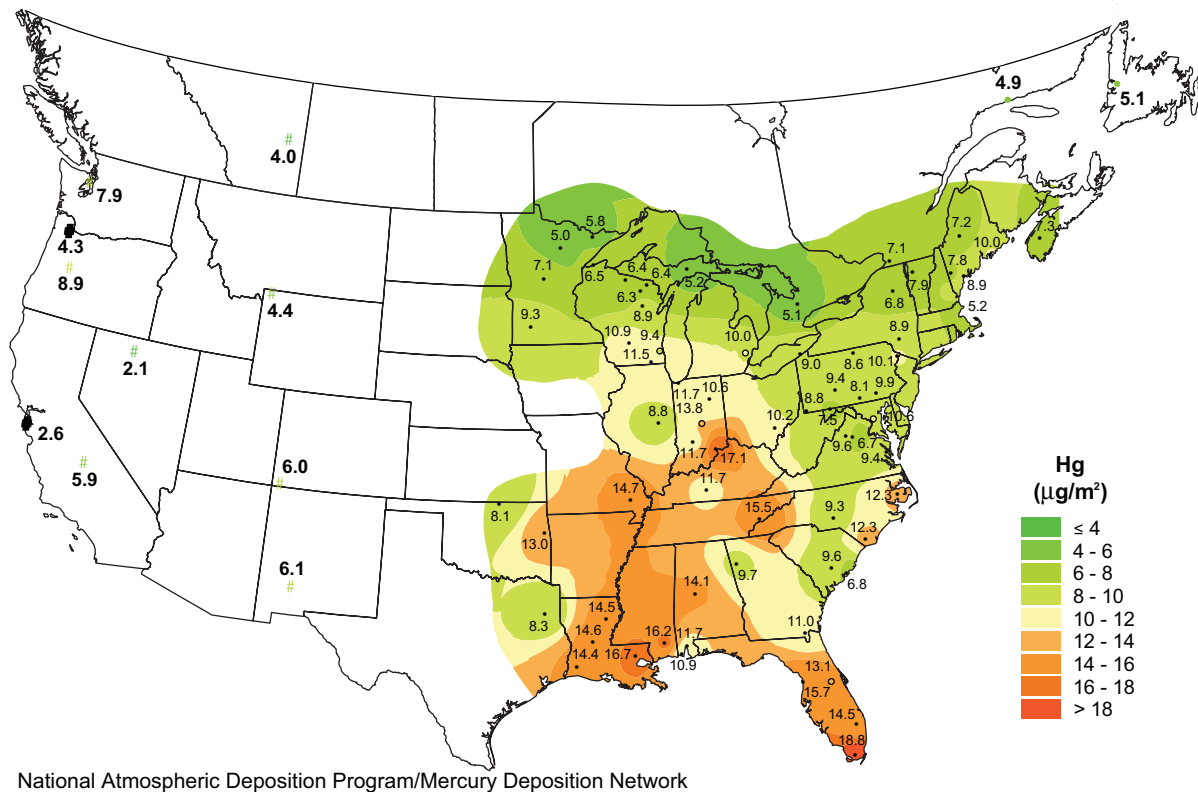
Figure 3

Wet deposition of mercury to the United States for 2006, measured by the Mercury Deposition Network, National Atmospheric Deposition Program (NRSP-3) (80).
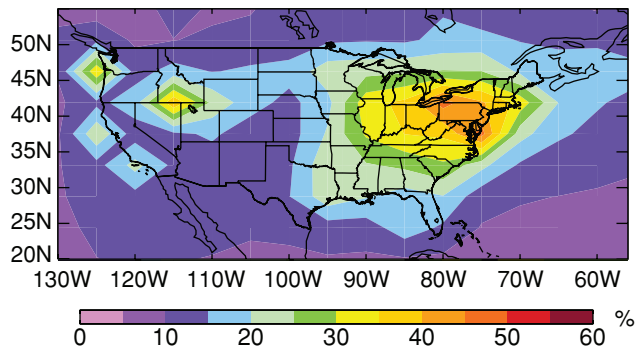
Figure 4

Percentage contribution from North American primary anthropogenic sources to total (wet plus dry) annual mercury deposition simulated by the GEOS-Chem global mercury model for 2004–2005. North America is defined as the geographical domain shown in the figure. Reproduced with permission from Selin & Jacob (83).
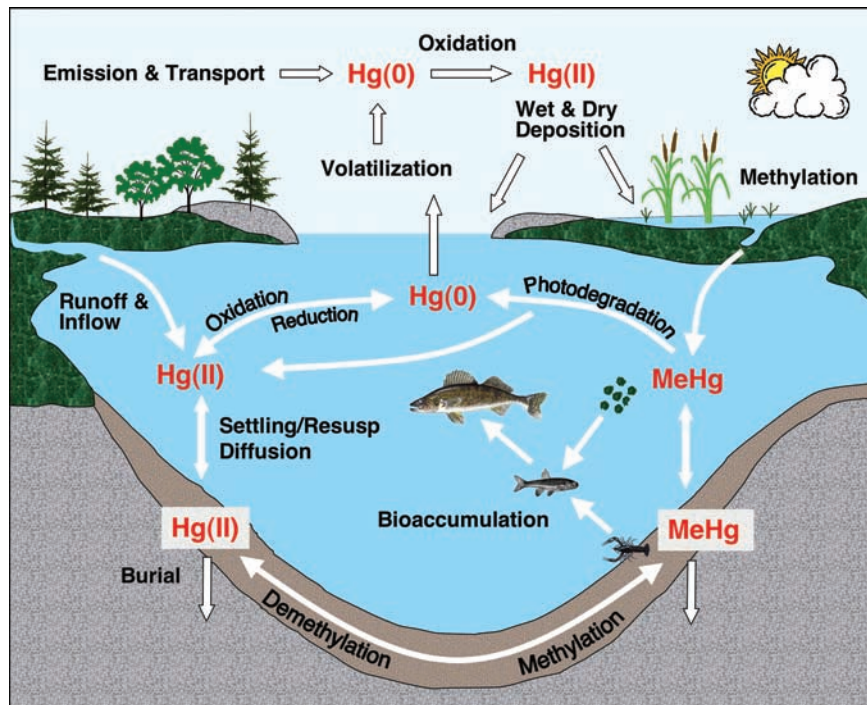
Figure 5

Mercury cycling in a lake and its watershed. Reproduced with permission from Engstrom (118), copyright 2007 National Academy of Sciences, U.S.A. Abbreviations: Hg(0), mercury in its elemental form; Hg(II), divalent mercury; MeHg, methylmercury; Resusp, resuspension.

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