

## Mercury as a Global Pollutant: Sources, Pathways, and Effects

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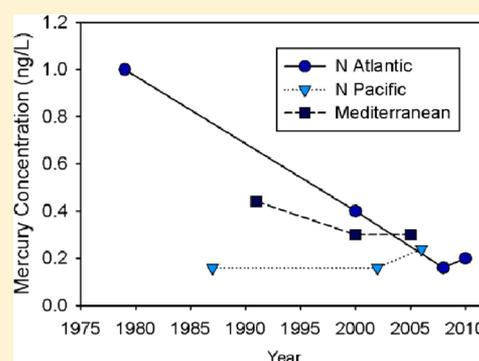
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**ABSTRACT:** Mercury (Hg) is a global pollutant that affects human and ecosystem health. We synthesize understanding of sources, atmosphere-land-ocean Hg dynamics and health effects, and consider the implications of Hg-control policies. Primary anthropogenic Hg emissions greatly exceed natural geogenic sources, resulting in increases in Hg reservoirs and subsequent secondary Hg emissions that facilitate its global distribution. The ultimate fate of emitted Hg is primarily recalcitrant soil pools and deep ocean waters and sediments. Transfers of Hg emissions to largely unavailable reservoirs occur over the time scale of centuries, and are primarily mediated through atmospheric exchanges of wet/dry deposition and evasion from vegetation, soil organic matter and ocean surfaces. A key link between inorganic Hg inputs and exposure of humans and wildlife is the net production of methylmercury, which occurs mainly in reducing zones in freshwater, terrestrial, and coastal environments, and the subsurface ocean. Elevated human exposure to methylmercury primarily results from consumption of estuarine and marine fish. Developing fetuses are most at risk from this neurotoxin but health effects of highly exposed populations and wildlife are also a concern. Integration of Hg science with national and international policy efforts is needed to target efforts and evaluate efficacy.



### INTRODUCTION

The socio-economic consequences of mercury (Hg) contamination and exposure of humans and wildlife to methyl Hg ( $\text{CH}_3\text{Hg}$ ) have prompted policies and regulations to limit Hg releases from human activities and to control its transport within and among the atmosphere, freshwaters, the lithosphere, and the ocean at local, regional, and global scales.<sup>1–4</sup> This synthesis paper is an integration of plenary presentations given at the 2011 meeting of the *International Conference of Mercury as a Global Pollutant* in Halifax, Canada. The overall theme of the conference was “Mercury in Air, Land, Sea and Me”. As such we organize this synthesis around Hg health effects; Hg dynamics in the atmosphere, land and the oceans; and the implications for Hg policy; the topics of the conference plenary sessions. We synthesize the current understanding of Hg and  $\text{CH}_3\text{Hg}$  sources, transport and fate globally, and discuss the potential variation of  $\text{CH}_3\text{Hg}$  exposure to changes in Hg emissions and other environmental perturbations, and impacts on human health and environmental quality. We discuss the most current knowledge on (1) the scale of Hg emission impacts (local-regional-global); (2) the relative importance of geogenic vs anthropogenic emission sources; (3) the recovery response time for ecosystems after emission controls (years-decades-centuries); and (4) the chronic low dose health effects of dietary exposure of  $\text{CH}_3\text{Hg}$  on human populations. We

consider the implications of this knowledge and remaining research gaps for Hg control policies (legally binding or voluntary actions) at regional and global scales.

Mercury is a naturally occurring element, but has been directly mobilized by humans for thousands of years into aquatic and terrestrial ecosystems through mining, the use of Hg in precious metal extraction, its presence as a trace contaminant in many materials (e.g., coal, metal ores), and its use in products (e.g., paint, electronic devices) and by industry (chlor-alkali plants; as a catalyst).<sup>5–8</sup>

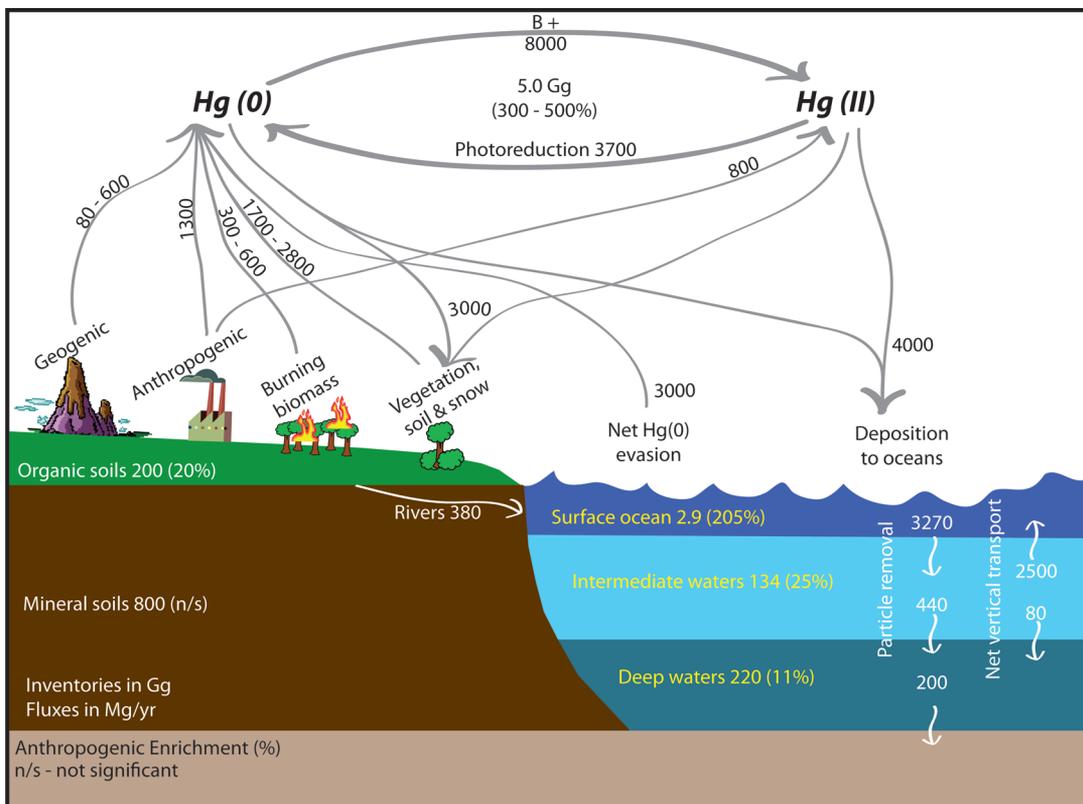
The atmosphere is the foremost transport pathway of Hg emissions, whereas land and ocean processes play an important role in the redistribution of Hg in terrestrial, freshwater, and marine ecosystems and the production of  $\text{CH}_3\text{Hg}$  that drives the major human exposure route, consumption of fish, particularly marine fish. The temporal and spatial scales of Hg transport in the atmosphere and its transfer to aquatic and terrestrial ecosystems depend primarily on its chemical and physical forms. Following emission, elemental Hg ( $\text{Hg}(0)$ ) can be transported long distances before oxidation and removal by

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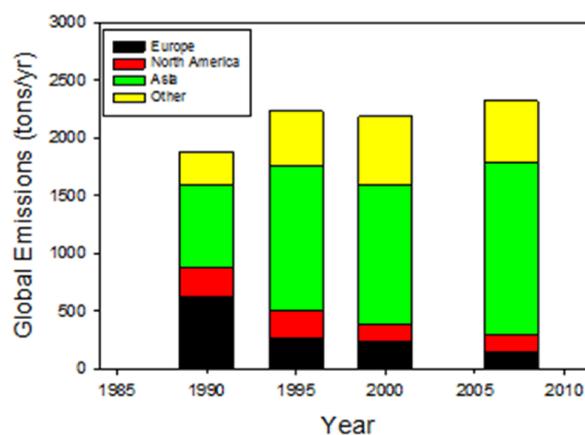
**Figure 1.** Current estimates of the fluxes and pools of mercury at the Earth’s surface derived on this work, and building on previous studies.<sup>99,140,148,181,183,193</sup>  $Hg(II)$  includes both gaseous and particulate forms, plus a negligible contribution (1Mg) from inert particulate mercury. The percentages in brackets are estimated increases in pools and fluxes due to anthropogenic activities over the past 150 years. Fluxes are in  $Mg\ yr^{-1}$  and reservoirs are given in Gg.

particle and gas-phase dry deposition or scavenging by precipitation. The atmospheric residence time of  $Hg(0)$  is several months to a year.<sup>3,6,9,10</sup> Mercury can therefore be transported and deposited to remote locations such as the Arctic and Antarctic.<sup>11-19</sup> Reactive gaseous  $Hg$  (RGM) and particulate bound ionic  $Hg$  ( $Hg(II)$ ) have a shorter atmospheric residence time than  $Hg(0)$  (atmospheric residence time hours to days), and as a result are generally deposited locally or regionally. Inputs to ecosystems largely occur as  $Hg(II)$ , while most  $CH_3Hg$  is produced within ecosystems.

It is important to distinguish between *primary* and *secondary* emission sources of  $Hg$ . Primary sources, both natural and anthropogenic, transfer  $Hg$  from long-lived lithospheric reservoirs to the atmosphere. This  $Hg$  deposits to land and oceans. Deposited  $Hg$  can be reduced to  $Hg(0)$  and then re-emitted. Re-emission processes represent *secondary sources* that exchange  $Hg$  among surface reservoirs using the atmosphere as a vehicle. Primary sources increase the global pool of  $Hg$  in surface reservoirs, while secondary sources redistribute it among and within ecosystems.

Recent global assessments and associated modeling have improved understanding of  $Hg$  emissions from both primary and secondary sources,<sup>6,8,20-22</sup> as well as their atmospheric transport and deposition.<sup>2,17,23-28</sup> Overall, total global  $Hg$  emissions to the atmosphere range from 6500 to 8200  $Mg\ yr^{-1}$ , of which 4600 to 5300  $Mg\ yr^{-1}$  are from natural processes and sources (primary geogenic plus secondary emissions). Primary anthropogenic sources release 1900–2900  $Mg\ yr^{-1}$  compared with primary natural (geogenic) inputs of 80–600  $Mg\ yr^{-1}$

(Figure 1). East Asia currently accounts for almost 40% of total primary global anthropogenic emissions of  $Hg$  (Figure 2).<sup>6,23</sup>



**Figure 2.** Trends in global emissions of mercury; note emissions in Asia are increasing while Europe and North America are decreasing.<sup>6,21</sup>

Impacts of  $Hg$  have been assessed for three potential future anthropogenic greenhouse gas (GHG) emission scenarios under the Intergovernmental Panel on Climate Change Special Report on Emission Scenarios (IPCC SRES).<sup>7,8</sup> Under most GHG scenarios, primary  $Hg$  emissions will increase unless emission controls are widely implemented. The main driver for increased  $Hg$  emissions is the expansion of coal-fired electricity generation in the developing world, particularly Asia, and the

degree to which control technologies are implemented. An evaluation of these scenarios by Sundseth et al.<sup>29</sup> confirmed the high socio-economic cost of these projected increases in emissions. They estimate that by 2020, the annual cost of the associated decrease in IQ would reach US \$3.7 billion (2005 dollars).

An important distinction between Hg and most other atmospheric pollutants is that environmental and health impacts are only indirectly related to ambient atmospheric concentrations of Hg(II) or Hg(0). Effects and toxicity result from the net conversion of Hg(II) to the more bioaccumulative CH<sub>3</sub>Hg, a process that generally occurs under reducing conditions in wetlands and sediments in watersheds and coastal zones, and in the upper ocean (see below). Exposure to humans and wildlife overwhelmingly occurs by the consumption of CH<sub>3</sub>Hg contaminated ocean, freshwater and terrestrial organisms.<sup>1,30–33</sup> As a result, the impact of Hg on ecosystem health is related not only to the magnitude of regional and global emissions and deposition, but also the potential for watersheds and the oceans to convert Hg(II) to CH<sub>3</sub>Hg; of food webs to biomagnify CH<sub>3</sub>Hg; and the processes and ecosystem disturbances that alter net methylation and trophic transfer.<sup>34,35</sup>

Global concern of widespread CH<sub>3</sub>Hg contamination has motivated international initiatives to assess current knowledge on atmospheric Hg emissions, transport and its effects on terrestrial and aquatic ecosystems. These include activities of the United Nations Environment Programme (UNEP) ([www.unep.org/PDF/PressReleases/GlobalMercuryAssessment2013.pdf](http://www.unep.org/PDF/PressReleases/GlobalMercuryAssessment2013.pdf)) and that of the UNECE-LRTAP Task Force on Hemispheric Transport of Air Pollution (TF HTAP).<sup>36,37</sup> The UNEP initiative is aimed at decreasing human health and environmental risk from the release of Hg, as well as improving the understanding of international Hg emissions and their transport and fate.<sup>2,17</sup> An international treaty (Minamata Convention) was signed in January 2013 ([www.unep.org/hazardoussubstances/mercury/Negotiations/INC5/INC5PressReleases/tabid/106835/Default.aspx](http://www.unep.org/hazardoussubstances/mercury/Negotiations/INC5/INC5PressReleases/tabid/106835/Default.aspx)) to control the global release of Hg to the environment. An observational infrastructure system is also being developed to monitor Hg in the atmosphere, and marine and terrestrial ecosystems through international Hg agreements<sup>38</sup> involving partnerships among regional Hg monitoring programs. Such information will improve understanding of the global impacts of Hg pollution and help evaluate the effectiveness of national and international policies.

## ■ EXPOSURE AND HEALTH OF HUMANS AND WILDLIFE

**Exposure Pathways and Toxicology.** The driver of most environmental Hg and CH<sub>3</sub>Hg studies is the perception that human health risk from CH<sub>3</sub>Hg exposure has increased with industrialization and the increasing cumulative release of Hg to the environment from anthropogenic sources. CH<sub>3</sub>Hg concentrations are elevated in fish and shellfish, particularly piscivorous fish such as tuna. Dietary consumption of marine fish and other seafood is a major route of CH<sub>3</sub>Hg exposure among human populations with many populations dependent on fish for food, protein, and nutrients. The contribution of fish to global diets is increasing (17 kg cap<sup>-1</sup> yr<sup>-1</sup> in 2008).<sup>39</sup> In 2007, fish was 15.7% of the global intake of animal protein and 6.1% of all protein consumed. Globally, fish provides >1.5 billion people with ~20% of their average per capita intake of

animal protein, and 3.0 billion with at least 15%. China has the highest total food fish supply (38.4 million tonnes; 32.4% of the global supply) and Japan has the highest per capita consumption (56.7 kg cap<sup>-1</sup> yr<sup>-1</sup> in 2007).<sup>40</sup> The widespread contamination of fish by CH<sub>3</sub>Hg, therefore, is a threat to human health on a global scale. Further, recent studies from southeastern China suggest that rice grown in soils near artisanal Hg mining and receiving elevated atmospheric Hg deposition may also accumulate elevated CH<sub>3</sub>Hg.<sup>41</sup> More research is needed on CH<sub>3</sub>Hg in rice as an exposure pathway.

In addition to the global commercial fishery, CH<sub>3</sub>Hg exposure can be important at the “local consumer” scale. Local consumers include recreational anglers who eat their catch; people who rely on local marine fish and marine mammals for a majority of their protein and nutrition; immigrant communities who catch their own fish and may have different eating habits such as consuming whole fish instead of fillets; and consumers who prefer to eat local seafood. Indigenous peoples around the world that rely on local fish catch for subsistence living can also be at risk of elevated Hg exposure.<sup>42</sup> Studies have shown elevated exposure of local populations who consume local fish or animals. Either global Hg sources<sup>43</sup> or a combination of global and local sources<sup>44,45</sup> can be important in such locations.

Numerous studies and databases exist on total Hg or CH<sub>3</sub>Hg concentrations in fish.<sup>4</sup> As Hg in the muscle of piscivorous fish is predominately CH<sub>3</sub>Hg,<sup>46</sup> total Hg measurement is often used as a surrogate for CH<sub>3</sub>Hg in fish. A comprehensive review of both gray and published literature of Hg concentrations in 36 species of fish ( $n = 34\,300$ ) showed that (1) fish such as tuna and swordfish have the highest CH<sub>3</sub>Hg concentrations; (2) mackerel and sardines from the Mediterranean Sea contain more CH<sub>3</sub>Hg compared to other harvesting areas; (3) mollusks and crustaceans have relatively low CH<sub>3</sub>Hg concentrations (below 100 ng g<sup>-1</sup>);<sup>47</sup> and (4) within-species variability in concentration often exceeds the interspecies variability.<sup>48</sup> Note that fish Hg data are limited for most of the world, highlighting the need for local data to inform exposure assessments as is being done by international organizations.<sup>49</sup>

The toxicology of Hg and the environmental health implications of CH<sub>3</sub>Hg exposure have been extensively reviewed.<sup>50,51</sup> Recent research has advanced the understanding of the dose–response relationship between exposure to CH<sub>3</sub>Hg and its toxic effects. Information has been derived from observations of the CH<sub>3</sub>Hg poisoning incident in Minamata, Japan, where more than 2000 people consumed CH<sub>3</sub>Hg contaminated fish (total Hg concentrations >10 μg g<sup>-1</sup>) and suffered from neurological disorders collectively known as Minamata Disease.<sup>52</sup> Pregnant women in Minamata had exposure levels 27 times higher than reference areas,<sup>53</sup> equivalent to 40 μg g<sup>-1</sup> in hair. In comparison, the mean hair Hg concentrations among women in the U.S. was 0.2 μg g<sup>-1</sup> in 1999–2000<sup>54</sup> and the mean maternal hair concentration of the Hg exposure cohort in the Faroe Islands was 4.27 μg g<sup>-1</sup> in 1986–1987.<sup>55</sup> In Minamata, fetuses exposed to CH<sub>3</sub>Hg through the placenta displayed severe cerebral palsy-like symptoms, while their mothers had mild or no manifestations of poisoning. The high levels of pre- or postnatal exposure to CH<sub>3</sub>Hg were found to cause long-term psychiatric symptoms in adults including impairment of intelligence and mood, and behavioral dysfunction.<sup>56</sup>

Current global Hg exposure is at levels much lower than conditions at Minamata. However, there is convincing evidence

of adverse neurological/neurodevelopmental outcomes in infants and young children associated with CH<sub>3</sub>Hg exposure during fetal development as a result of maternal fish consumption during pregnancy. Three major epidemiological studies of neurobehavioral development in children have been conducted in populations consuming fish/seafood with no known local industrial contamination: (1) the Seychelles Islands (median maternal hair concentration at 6.6  $\mu\text{g g}^{-1}$  ( $n = 779$ ));<sup>57</sup> (2) the Faroe Islands (mean maternal hair 4.27  $\mu\text{g g}^{-1}$  ( $n = 900$ ));<sup>55</sup> and (3) the New Zealand study (mean maternal hair concentration at 8.3  $\mu\text{g g}^{-1}$  ( $n = 38$ )).<sup>58</sup> Adverse neurodevelopment effects were identified in the Faroe Islands at a benchmark dose (BMDL) of 12–15  $\mu\text{g g}^{-1}$  Hg in maternal hair. In contrast, the New Zealand study found a BMDL at 4–6  $\mu\text{g g}^{-1}$  Hg and for the Seychelles Islands at 17–25  $\mu\text{g g}^{-1}$  Hg.<sup>59</sup>

Long-term study of the Seychelles Child Development Study cohort at age 17 years found no consistent pattern of adverse associations between prenatal MeHg exposure and detailed domain specific neurocognitive and behavioral testing.<sup>60</sup> On the other hand, impact of prenatal CH<sub>3</sub>Hg exposure on neurobehavioral function was found to be multifocal and permanent among the children of the Faroe cohort as similar associations between CH<sub>3</sub>Hg exposure and deficits in motor, attention, and verbal tests were found in children at age 7 years and 14 years.<sup>61</sup> Sustained attention of the 14 year old children was found to be the neuropsychological domain that is particularly vulnerable to developmental CH<sub>3</sub>Hg exposure, indicating probable underlying dysfunction of the frontal lobes.<sup>62</sup> The discrepancies among studies may partly be explained by variability in long chained polyunsaturated fatty acids (LCPUFA) in the fish consumed which counter the adverse effects of CH<sub>3</sub>Hg.<sup>63</sup> A follow up cohort study of 229 mother-child pairs in the Seychelles Child Development Nutrition Study found that maternal fish consumption during pregnancy exposed the fetus simultaneously to CH<sub>3</sub>Hg and LCPUFA which had significant but opposite effects on child development at 9 and 30 months of age.<sup>64</sup> Moreover, at 5 years of age, the positive association of LCPUFA remained significant while the adverse associations between MeHg exposure and developmental outcomes was no longer observed.<sup>65</sup> These results demonstrate the importance confounding effects of the maternal nutritional status in determining the risk of prenatal CH<sub>3</sub>Hg exposure.

Using the data from these three studies, a dose–response relationship between maternal hair Hg concentrations and child IQ was established suggesting a loss of 0.18 IQ points (95% confidence interval, –0.378 to –0.009) for each  $\mu\text{g g}^{-1}$  increase of maternal hair Hg.<sup>66</sup> Based on the three studies, the U.S. Environmental Protection Agency (EPA) derived a reference dose (RfD) for CH<sub>3</sub>Hg of 0.1  $\mu\text{g kg body weight}^{-1} \text{d}^{-1}$  as the daily intake likely to preclude appreciable risk of deleterious effects during a lifetime, assuming a total uncertainty factor (UF) of 10 to account for variability and uncertainty.<sup>59</sup> Between 3 and 15% of women of child bearing age in the U.S.<sup>67</sup> and 27.7% in Korea<sup>68</sup> exceed this RfD indicating the scale of the public health problem.

**Risks and Benefits of Fish Consumption.** A review of epidemiological studies on effects of low levels of CH<sub>3</sub>Hg exposure (maternal hair Hg less than 4  $\mu\text{g g}^{-1}$ ) conducted in Poland, Spain, Korea, France, the U.S., Austria, Canada, and the United Kingdom found possible effects on fetal growth among susceptible subgroups and on infant growth in the first two years of life.<sup>69</sup> There was also evidence of early childhood

neurological effects at low levels of prenatal CH<sub>3</sub>Hg exposure that may differ by age, gender, and timing of exposure. The authors found no conclusive evidence on effects on the risk of cardiovascular disease or immune functions.

The dose response relationship between low dose CH<sub>3</sub>Hg exposure and effects in adults is not as clear. The lowest observable effect was reported in the Tapajós River basin (Brazil), a major Amazon tributary impacted by traditional gold mining since the mid 1980s. Alteration of motor performance and visual dysfunction was found in adults whose mean Hg hair concentration was above 10  $\mu\text{g g}^{-1}$ .<sup>70</sup> Interestingly, it appeared that selenium from dietary sources could mitigate the adverse effects of CH<sub>3</sub>Hg on motor functions in some Amazon populations.<sup>71</sup>

Health effects such as cardiac function from CH<sub>3</sub>Hg exposure have received recent attention. Mechanistic evidence and experimental animal toxicological studies suggest that CH<sub>3</sub>Hg can be a risk factor for cardiovascular disease (CVD).<sup>72</sup> However, epidemiological evidence is inconclusive.<sup>73</sup> Roman et al.<sup>74</sup> considered the current epidemiological literature sufficiently robust to support the development of a dose–response function between CH<sub>3</sub>Hg exposure and acute myocardial infarction. However, results of two major cohort studies in the U.S. found no evidence of any clinically relevant adverse effects of Hg exposure on coronary heart disease, stroke, or total CVD disease in adults.<sup>75</sup> The public health implications are potentially significant, as CVD is the leading cause for mortality in most developed countries. For example, the expected monetary value of the annual health benefits associated with decreases in fatal heart attacks due to a 10% decrease in Hg exposure of the U.S. population would be 4 times higher than that associated with IQ gains.<sup>76</sup> Therefore, it is important to improve characterization of the potential linkage between CH<sub>3</sub>Hg exposure and the risk of CVD.

Nutritional benefits are an important consideration in risk assessment of fish consumption.<sup>77</sup> Essential polyunsaturated fatty acids, such as docosahexaenoic acid (DHA), confer benefits to brain and visual system development in infants and reduce risks of certain forms of heart disease in adults which mitigate against CH<sub>3</sub>Hg effects. Advisories issued with the aim of decreasing Hg exposure can also result in substantial reductions in healthful DHA.<sup>78</sup> Facing this dilemma of risk and benefit of fish consumption, the FAO and WHO held an Expert Consultation Workshop in 2011.<sup>48</sup> Using multiple national databases of Hg and fatty acids concentrations in commercial fish species, they compared the benefits of n-3 polyunsaturated fatty acids with the risks of CH<sub>3</sub>Hg among women of childbearing age, concluding that maternal fish consumption lowers the risk of suboptimal neurodevelopment in their offspring compared with the offspring of women not eating fish in most circumstances evaluated. This integrative approach of benefits and risks assessment has been successfully applied to estimate optimal fish consumption level in The Netherlands,<sup>79</sup> Finland,<sup>80</sup> and Sweden.<sup>81</sup> Given the nutritional benefits of fish, it is important to continue to decrease Hg sources with a goal of achieving fish Hg concentrations for commonly consumed species of <0.3  $\mu\text{g g}^{-1}$ . In the interim, it is important to work with public health agencies to improve fish consumption advisories and communication strategies to minimize risks and maximize benefits from consumption of low-Hg fish.

**Hg Exposure to Wildlife.** While concern of exposure of CH<sub>3</sub>Hg is focused on human health, research is increasingly demonstrating that elevated CH<sub>3</sub>Hg also impacts the health of

fish and other wildlife. Studies in the mid-1990s suggested that concentrations of CH<sub>3</sub>Hg in the range of 5–10 μg g<sup>-1</sup> (wet weight) were necessary for sublethal and lethal effects on fish.<sup>82</sup> However, recent studies have shown that whole body concentrations at 0.3 μg g<sup>-1</sup> and muscle tissue concentrations of 0.5 μg g<sup>-1</sup> compromise fish reproduction, embryonic development, change biochemical process, and cause damage to cells and tissues.<sup>83</sup> These levels are in the range commonly reported for top predator fish<sup>84</sup> and suggest that the health of fisheries may be impaired due to CH<sub>3</sub>Hg contamination.

Elevated concentrations of CH<sub>3</sub>Hg have been observed in piscivorous birds (common loon, *Gavia immer*;<sup>85</sup> wading birds;<sup>86</sup> bald eagles, *Haliaeetus leucocephalus*<sup>87</sup>) and mammals (mink, *Mustela vison*; river otter, *Lontra Canadensis*<sup>88</sup>). Although a relatively insensitive species, there is considerable understanding of Hg effects on the common loon. Exposure to elevated CH<sub>3</sub>Hg causes hormonal changes, impairment of motor skill and decreases in reproduction. As a result, a wildlife criterion of 3.0 μg g<sup>-1</sup> has been established for common loon blood.<sup>89</sup> This blood concentration value is comparable to that observed in eastern North America.<sup>85,90</sup> In addition to piscivores, insectivorous songbirds and bats, particularly those associated with wetland habitats, have been shown to have elevated Hg.<sup>91,92</sup> Using the Carolina wren (*Thryothorus ludovicianus*) as a model system of CH<sub>3</sub>Hg effects of Hg songbirds, Jackson et al.<sup>93</sup> found that nesting success (i.e., the ability to fledge at least one offspring) decreased as the parents CH<sub>3</sub>Hg exposure increased, with a 10% or more nest failure when females had blood Hg of 0.7 μg g<sup>-1</sup>, 20% failure at blood Hg of 1.2 μg g<sup>-1</sup> and 30% failure at blood Hg of 1.7 μg g<sup>-1</sup>.

Fish eating mammals such as mink are affected by Hg exposure at dietary concentrations as low as 0.1 μg g<sup>-1</sup>. Concentration-dependent decreases in N-methyl-D-aspartic acid (NMDA) receptors were found in the basal ganglia, cerebellum, brain stem, and occipital cortex of wild and captive mink in Canada.<sup>94</sup> Similar effects on NMDA receptors were found in the brain stem of polar bears (*Ursus maritimus*).<sup>95</sup> High Hg concentrations were reported in the brain of other apex predators in the Arctic ecosystem such as beluga whale (*Delphinapterus leucas*).<sup>96</sup> These findings are of physiological and ecological concern because they demonstrate that at ambient concentrations, Hg can affect the health of key species of wildlife in diverse ecosystems.

## ■ AIR

The atmosphere is a major pathway for the biogeochemical cycle of Hg. Background atmospheric concentrations typically range from 1.3 to 1.7 ng m<sup>-3</sup> in the Northern Hemisphere and from 1.1 to 1.3 ng m<sup>-3</sup> in the Southern Hemisphere.<sup>9,13,17,97</sup> Data from lake sediments, peat cores, and ice cores all indicate that atmospheric Hg deposition has increased globally by about a factor of 3 since preindustrial times (~1850), with larger increases in regions receiving elevated deposition from regional sources.<sup>13,98</sup> On a global scale net atmospheric Hg deposition is determined by the overall emissions, whereas deposition at the local scale is controlled by atmospheric processes and the speciation of regional and local emissions. Understanding these processes is therefore critical for the development of accurate predictive models that inform global Hg policy. Models of atmospheric Hg must accurately depict four basic processes: emissions, transport, redox chemistry, and deposition.

**Emissions.** Estimates of Hg emissions to the atmosphere are depicted in Figure 1, based on Holmes et al.<sup>99</sup> Primary

emissions include natural geogenic sources (volcanism and erosion; estimated at 500 Mg a<sup>-1</sup>) and anthropogenic sources (fuel combustion, waste incineration, and mining; 2100 Mg a<sup>-1</sup>). Primary emission sources due to human activity have increased over geogenic sources by a factor of 2–15, which is larger than the aforementioned factor of 3 average increase in Hg deposition (Figure 1). This discrepancy reinforces the fact that the Earth system (including recycling through surface reservoirs) is not in steady-state, and that Hg in surface reservoirs will continue to increase even if future anthropogenic emissions remain constant.<sup>100</sup>

Noncombustion processes emit Hg as Hg(0), but combustion processes (including fuel use, waste incineration, open fires) emit about half of their Hg as Hg(II).<sup>22</sup> This partitioning of emissions is of considerable importance because the emitted Hg(II) can be deposited regionally downwind of the source, resulting in a local deposition enhancement in contrast to the global influence associated with emissions of Hg(0). Emission inventories also generally include a particulate component of Hg (Hg(p)) in addition to Hg(0) and RGM. Hg(p) is best viewed as part of Hg(II), with gas-particle partitioning of Hg(II) occurring in the atmosphere on the basis of local particle concentrations and temperature.<sup>101</sup>

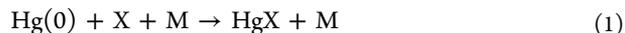
**Transport.** Hg transport in the atmosphere is rapid compared to other environmental reservoirs, enabling the dispersal and exchange of Hg in the global environment. The time scale for global mixing of the troposphere is about 1 year, limited principally by exchange of air between the two hemispheres. Surface air observations of total gaseous Hg (TGM) show little variability in general, and data from ship cruises indicate 30% higher concentrations in the northern than in the southern hemisphere. The observed atmospheric variability of TGM implies an atmospheric lifetime of 0.5–1 year.<sup>99,102</sup> While Hg(0) emitted to the atmosphere is transported efficiently on a hemispheric scale and may be deposited anywhere in its hemisphere of origin, transport between hemispheres is less efficient.<sup>103</sup> Aircraft observations show that Hg(0) is well-mixed through the depth of the troposphere, as would be expected since vertical mixing in the troposphere occurs on a time scale of about a month. Concentrations decrease sharply above the tropopause,<sup>104</sup> implying rapid oxidation of Hg(0) in the lower stratosphere.

**Redox Chemistry of Atmospheric Hg.** There is considerable uncertainty regarding the atmospheric redox chemistry of Hg. Until about 10 years ago, the conventional view based on experiments and expressed in atmospheric models was that oxidation of Hg(0) would principally occur by OH and O<sub>3</sub>, and that reduction of Hg(II) would occur by reaction with HO<sub>2</sub>(aq) in clouds. However, recent laboratory experiments and thermodynamic and kinetic constraints indicate that these reactions are unlikely to be significant in the atmosphere.<sup>105</sup> This understanding has led to a fundamental reassessment of the chemical processes involved in Hg(0)/Hg(II) redox chemistry.

Atmospheric observations place some constraints on the processes involved. The diurnal cycle of RGM in surface air, especially in the marine boundary layer (MBL), indicates that the oxidation of Hg(0) to Hg(II) must be photochemical.<sup>106,107</sup> Observations of a weak summer minimum in Hg(0) concentrations at northern midlatitudes are consistent with this photochemical sink. Mercury depletion events (MDEs) are frequently observed in the Arctic boundary layer in spring.<sup>18</sup> There is good evidence that MDEs are due to rapid oxidation

of Hg(0) by Br atoms released by photochemical reactions following the debromination of sea salt, either in the aerosol or at sea ice surfaces.<sup>108</sup> Similar MDEs have been observed in surface air over the Dead Sea.<sup>109</sup>

The halogen atoms are potentially important Hg(0) oxidants in the atmosphere because the overall oxidation reactions are exothermic, involving a two-step process:<sup>105,110,111</sup>



where X  $\equiv$  Cl, Br, I is a halogen atom and Y is another radical, either a halogen atom or another species such as OH, OH<sub>2</sub>, or NO<sub>2</sub>.<sup>111</sup> Both reactions 1 and 2 are three-body processes. The binding energies of the Hg(I) compounds produced in reaction 1 are in the order HgCl > HgBr > HgI.<sup>110</sup> Cl is thus a particularly efficient oxidant, but its tropospheric concentration is low because reaction with methane results in stable HCl. Oxidation by I atoms may be limited by the stability of HgI.

Holmes et al.<sup>112</sup> suggested that Br atoms could provide the dominant global atmospheric oxidant for Hg(0). Known sources of Br atoms to the troposphere include photolysis and oxidation of bromoform (CHBr<sub>3</sub>), dimethylbromide (CH<sub>2</sub>Br<sub>2</sub>), and methyl bromide (CH<sub>3</sub>Br), as well as debromination of sea salt aerosol. CHBr<sub>3</sub> and CH<sub>2</sub>Br<sub>2</sub> are emitted to the marine biosphere, while CH<sub>3</sub>Br has both biogenic and anthropogenic sources. This ensemble of sources produce inorganic bromine (Br<sub>2</sub>), which then cycles among its different forms in a manner that is well established from earlier studies of stratospheric chemistry.<sup>113</sup> Unlike HCl, HBr has a lifetime of only a few days against oxidation by OH, providing a sustained pool of reactive Br. Satellite observations indicate a tropospheric background of 0.5–2 ppt BrO<sup>114</sup> and this can be reproduced in models including the above sources.<sup>115,116</sup> Holmes et al.<sup>112</sup> found that the implied Br atom concentrations yielded a global atmospheric lifetime of 0.5–1 years for Hg(0). However, the kinetics of the two-step oxidation of Hg(0) by Br have recently been revised<sup>111,117</sup> and these revisions need to be incorporated in atmospheric Hg models.

Even more uncertainty is associated with atmospheric reduction of Hg(II). Indeed, there is enough uncertainty in the rate of Hg(0) oxidation for global model simulations to account for observational constraints even without reduction of Hg(II).<sup>99</sup> The depletion of Hg(0) above the tropopause<sup>104</sup> indicates that oxidation cannot take place only by a gas-phase photochemical reaction. Reduction could occur by a condensed-phase photochemical process in lower tropospheric clouds, although there is no evidence to support this process. Observations of a diurnal cycle of Hg(0) in Antarctica by Dommergue et al.<sup>118</sup> offer indirect evidence of photochemical reduction of Hg(II) on snow surfaces.

There is some evidence for rapid Hg(II) reduction taking place in power plant plumes. Atmospheric observations in plumes and in polluted regions show much lower Hg(II)/Hg(0) concentration ratios than inferred from emission inventories,<sup>101,119,120</sup> and observed atmospheric deposition patterns do not show the regional enhancements that would be expected from power plant Hg(II) emissions.<sup>121</sup> The mechanism for reduction is unknown. It cannot be a simple first-order process since there is no indication of rapid Hg(II) reduction in the background atmosphere.

**Deposition.** Mercury can be removed from the atmosphere by wet or dry deposition. Wet deposition involves scavenging of

gas-phase and aerosol-phase Hg(II). Dry deposition involves surface uptake of both Hg(0) and Hg(II).<sup>122</sup>

Observations of the diurnal cycle of Hg(0) and RGM on land imply rapid nighttime removal by dry deposition.<sup>123</sup> Such data suggest that dry deposition of Hg(0) is a major atmospheric sink of Hg, although the underlying mechanisms and its reversibility remain poorly characterized and quantified. The rapid dry deposition of RGM can be understood in terms of its high water solubility. Dry deposition of Hg(p) depends on particle size, being slow for submicrometer particles but fast for supermicrometer particles. Observations indicate that Hg(p) in continental air is mostly in the submicrometer size range, while Hg(p) in marine air is mostly in the supermicrometer size range of sea salt particles.<sup>124,125</sup> Efficient uptake of RGM by sea salt particles is expected due to formation of HgCl<sub>3</sub><sup>-</sup> and HgCl<sub>4</sub><sup>2-</sup> complexes.<sup>126</sup>

Wet deposition provides an efficient sink for Hg(II). However, there is uncertainty regarding the efficiency of scavenging by cold precipitation (snow and ice). Field observations indicate that snow does not effectively scavenge RGM.<sup>127</sup> Wet deposition flux observations from the Mercury Deposition Network (MDN) in the eastern U.S. show a winter minimum that has been attributed to inefficient scavenging of RGM.<sup>101</sup> By contrast, aerosol particles are expected to be scavenged by snow more effectively than by rain.<sup>128</sup>

The phase partitioning of Hg(II) thus has important implications for removal by deposition. Observations of RGM and Hg(p) suggest that Hg(II) behaves as a semivolatile compound, partitioning between the gas and aerosol phases in a manner dependent on local aerosol concentration and composition and ambient temperature.<sup>129,130</sup> As limiting cases, Hg(II) is mainly in the gas phase in warm environments with low aerosols and mainly in the particle phase in cold environments with high aerosols.<sup>101</sup> Efficient uptake of Hg(II) by sea salt particles makes sea salt deposition a major process for delivery of Hg to the surface ocean.<sup>126</sup>

**Source-Receptor Relationships and Long-Term Trends.** The atmospheric behavior of Hg affects source-receptor relationships. Early work focused on Hg deposition “hot spots” associated with sources upwind. Very high Hg deposition rates in Florida in the 1990s were attributed to local sources from waste incineration.<sup>131</sup> However, high deposition remained even after those sources were controlled. It is now well established that the maximum in Hg wet deposition observed along the Gulf Coast of the U.S. in summer is due to scavenging of the Hg(II)-rich pool in the upper troposphere by deep convection.<sup>121,132,133</sup> Thus the maxima in Hg wet deposition in the southeast U.S. does not appear to be associated with local sources of Hg(II).

Sediment cores indicate a 3-fold increase in total Hg atmospheric deposition from the mid-19th century to present,<sup>134</sup> but with substantial variability indicative of local and regional impacts.<sup>98</sup> Longer records show evidence of human influence extending back several millennia.<sup>135,136</sup> Amos et al.<sup>100</sup> thus estimated that present-day Hg deposition associated with past and current anthropogenic emissions is enhanced 7-fold globally relative to preanthropogenic values.

Compilations of atmospheric observations from ship cruises and long-term monitoring sites around the Atlantic suggest decreasing trends in atmospheric Hg(0) in recent decades.<sup>102,123,137,138</sup> These decreasing trends appear at odds with global inventories indicating constant or increasing primary anthropogenic emissions (Figure 2).<sup>22,139</sup> One possible

explanation is a decline in secondary terrestrial Hg emissions<sup>102</sup> or changes in ocean evasion. For example, Mason et al.<sup>140</sup> suggest that subsurface concentrations in the North Atlantic have decreased markedly since the 1980s (see below), which should drive decreases in evasion from the ocean surface. Soerensen et al.<sup>141</sup> modeled the impact of the resulting decrease in Hg(0) evasion from the North Atlantic, demonstrating that it could explain the observed atmospheric decline and its spatial variability. Possible explanations for the decrease in North Atlantic subsurface Hg include a decrease in near-field Hg(II) deposition from sources in North America and Europe,<sup>140</sup> changes in the Hg(0) oxidation rate in the MBL, and decreases in Hg product use and in wastewater and riverine inflows to coastal zones.<sup>141</sup> Recent decreases measured in estuarine sediment cores and in coastal watershed Hg mass balances in the North Atlantic suggest that coastal inputs have decreased dramatically.<sup>142–144</sup> A better understanding of coastal Hg inputs to the open ocean and their historical trends is needed, particularly for the North Atlantic and the Arctic Ocean where the relatively large riverine contributions are thought to be a significant Hg source.<sup>145</sup>

## ■ LAND AND FRESHWATERS

**Processes Controlling Hg Dynamics on Land and Freshwaters.** Atmospheric Hg deposition is the dominant input of Hg to most watersheds.<sup>146</sup> Weathering of Hg-bearing minerals in soil also contributes natural Hg inputs. Note legacy or ongoing Hg supply from mining, industrial activities, or wastewater can overwhelm inputs from atmospheric deposition and natural sources resulting in localized areas of elevated contamination.<sup>35,147</sup>

Atmospheric inputs of Hg readily partition with living and detrital biomass influencing global and local Hg dynamics and ultimately its bioavailability in terrestrial and freshwater ecosystems. There are three mechanisms by which inputs of Hg to terrestrial ecosystems are released as secondary emissions. Following atmospheric deposition, Hg can be reduced from foliage, soil, surface waters or snowpack, and evaded back to the atmosphere.<sup>148</sup> Second, Hg(0) entering the stomata of foliage is subsequently deposited to soil as litterfall Hg inputs. This input, supplemented by wet and dry Hg(II) deposition and throughfall, is immobilized by soil organic matter probably through binding by reduced sulfur functional groups.<sup>149,150</sup> Conceptually detrital organic matter and associated Hg has been viewed as fractions of varying reactivity and residence times (i.e., from months, years, decades, to centuries<sup>148,150</sup>). As these “fractions” of soil organic matter are mineralized, a byproduct of organic matter “respiration” is Hg(0), which is evaded back to the atmosphere. Globally, these two pathways resupply from 1700 to 2800 Mg yr<sup>-1</sup> of Hg back to the atmosphere (Figure 1). Third, Hg is emitted to the atmosphere from biomass burning (300–600 Mg yr<sup>-1</sup>; Figure 1<sup>20</sup>) and other nonpoint sources. Overall, a considerable fraction of the Hg deposited to land is evaded back to the atmosphere as Hg(0) (Figure 1) and can be subjected again to transport, oxidation and deposition.

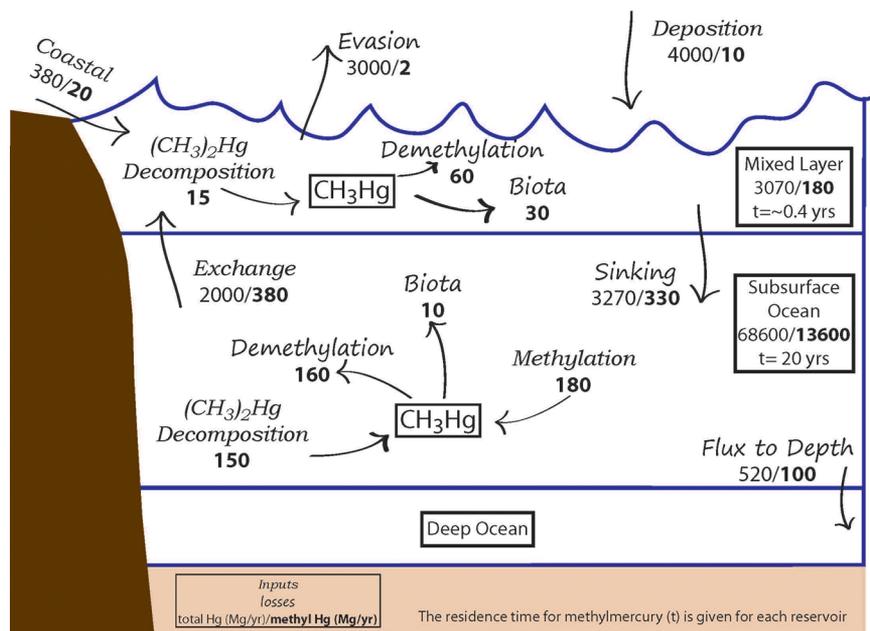
The balance of Hg input to terrestrial ecosystems largely accumulates in soil organic matter. Reconstructions of past Hg emissions suggest that although deposition has increased by about a factor of 3, soil Hg pools have increased by only about 20%, and like the ocean, are not at steady-state with respect to current emissions.<sup>22,100,148</sup> The fraction of soil organic matter that degrades rapidly is largely associated with surface soil and a

relatively small Hg reservoir (~10–15 Gg). This pool is thought to respond rapidly to changes in Hg emissions. In contrast, slow decaying (~50 Gg) and highly recalcitrant (~250 Gg) soil Hg pools are large reservoirs that turnover slowly. These soil pools are important legacy reservoirs of historical Hg emissions, which return Hg to the atmosphere over long time scales. On a global basis, a small fraction of soil Hg is transported by riverine fluxes largely in association with dissolved or particulate organic matter (Figure 1), but this can be an important pathway to coastal areas and to certain portions of the open oceans.<sup>43,151,152</sup>

Considerable research on terrestrial and freshwater ecosystems has focused on (1) transport of Hg(II) via drainage waters to reducing zones where net CH<sub>3</sub>Hg production is high; (2) the net methylation of Hg(II); and (3) the trophic transfer of CH<sub>3</sub>Hg. A host of characteristics and disturbances can accelerate or diminish each of these processes. Watershed characteristics such as size, topography and elevation, land cover, and use influence the net deposition of Hg to the land surface and transport to the aquatic ecosystems.<sup>153,154</sup> Hydrologic characteristics and conditions regulate the fluvial transport of Hg(II) from uplands to reducing zones, which exhibit high rates of net methylation and from which large quantities of CH<sub>3</sub>Hg are supplied to downstream and adjacent aquatic environments.<sup>152</sup>

A variety of factors alter net rates of CH<sub>3</sub>Hg in methylation zones such as wetlands, riparian zones and surface water sediments, including temperature, hydrology, and hydrologic perturbations (i.e., changes in discharge, hydroperiod, reservoir management), the supply of Hg(II) in a bioavailable form, labile organic carbon, and reducing conditions.<sup>35</sup> The formation of CH<sub>3</sub>Hg is largely microbially mediated by SO<sub>4</sub><sup>2-</sup><sup>155</sup> and to a lesser extent Fe<sup>156,157</sup> reducing bacteria. Thus, the supply of electron acceptors to zones of methylation impact rates of methylation. Inputs of oxygen and NO<sub>3</sub><sup>-</sup> slow net CH<sub>3</sub>Hg supply either by the suppression of SO<sub>4</sub><sup>2-</sup> reduction or maintaining oxidizing conditions that allow for the formation of Fe and Mn oxides and the partitioning of Hg to these surfaces.<sup>158</sup> At low concentrations of SO<sub>4</sub><sup>2-</sup>, further addition of SO<sub>4</sub><sup>2-</sup> stimulates production of CH<sub>3</sub>Hg until a maximum value of CH<sub>3</sub>Hg<sup>+</sup> production is reached.<sup>155,159</sup> At higher concentrations, further addition of SO<sub>4</sub><sup>2-</sup> diminishes production of CH<sub>3</sub>Hg due to the formation of charged mercuric sulfide complexes that are less available and/or the immobilization of Hg(II) due to the formation of HgS<sub>(s)</sub>.<sup>160</sup> Note that ecosystem accumulation of CH<sub>3</sub>Hg is the net effect of the production of CH<sub>3</sub>Hg by methylation and the loss by demethylation. Therefore any processes that increase methylation or diminish demethylation will increase the accumulation of CH<sub>3</sub>Hg.

Recent research has demonstrated the complex and pivotal role of dissolved organic matter (DOM) in net CH<sub>3</sub>Hg production.<sup>161</sup> DOM can serve as a Hg “carrier” facilitating transport and has the ability to chelate Hg(II) altering bioavailability. DOM can also stabilize or destabilize colloids altering transport, and inhibit or promote metal precipitation thereby altering the immobilization by HgS<sub>(s)</sub> formation, through the formation of Hg-containing nanoparticles.<sup>162,163</sup> Furthermore, CH<sub>3</sub>Hg can be produced from Hg associated with colloids.<sup>164,165</sup> Under reducing conditions, low concentrations of DOM can reduce Hg(II) to Hg(0).<sup>166</sup> However with increasing DOM, binding of Hg(II) limits the production of Hg(0).



**Figure 3.** Overall budget for the sources and losses of total and methylated (in bold) mercury to the mixed layer and the subsurface ocean (defined as waters above the permanent thermocline) using data and information discussed throughout the paper. Fluxes are in  $\text{Mg yr}^{-1}$  and reservoirs are given in Mg.

Several factors have been shown to enhance trophic transfer of  $\text{CH}_3\text{Hg}$ . Mercury burdens increase with the age and size of individuals within given fish populations, in part due to the slower rates of elimination and longer exposure in larger individuals, and due to the consumption of higher trophic level foods by older and larger individuals.<sup>82</sup> Mercury concentrations in top predator fish are higher in food webs with longer chain lengths and less omnivory.<sup>167</sup> Both experimental and field studies show that nutrient enrichment diminishes  $\text{CH}_3\text{Hg}$  bioaccumulation in phytoplankton via the biodilution under algal bloom conditions.<sup>168</sup> Mercury concentrations in zooplankton also decrease with increasing zooplankton densities that, in turn, are correlated with lower  $\text{CH}_3\text{Hg}$  concentrations in fish.<sup>169</sup> Growth dilution in fish, also under conditions of high productivity and food availability, may also result in lower  $\text{CH}_3\text{Hg}$  concentrations.<sup>170</sup> Many studies have shown that fish Hg concentrations increase with decreases in pH.<sup>84,171</sup> Most importantly the magnitude of fish Hg concentrations appears to be set at the base of the food chain and maintained through to top predators.<sup>172</sup>

**Spatial and Land Cover Patterns.** Land cover and land use appear to be critical controllers of Hg in terrestrial and freshwater ecosystems. For example, Denkenberger et al.<sup>154</sup> synthesized rates of Hg(0) evasion across land cover types, finding elevated rates from agricultural and pasture lands, which are characterized by greater incident solar radiation and rates of soil organic matter turnover, and lower values in forests. Rates of net Hg deposition can be enhanced under forest cover due to greater precipitation quantity coupled with enhanced scavenging of atmospheric Hg by the forest canopy and lower evasive losses.<sup>149,154</sup> These patterns suggest that in regions remote from local sources that net Hg deposition would generally be greater under forest cover than agricultural land cover. Obrist et al.<sup>150</sup> found that soil Hg concentrations across the continental U.S. were not strongly related to Hg deposition but rather increased with soil C, latitude, precipitation quantity and clay content. They hypothesize

that over large spatial scales soil Hg accumulation is strongly linked with organic C accumulation. These spatial and land cover patterns are generally consistent with fish Hg concentrations. Fish Hg concentrations for a given region are typically higher in waters draining areas with greater forest and wetland cover, and lower in waters draining agricultural lands.<sup>33,34</sup>

**Temporal Patterns.** There are few comprehensive studies of long-term response of ecosystems to Hg deposition, which has limited our quantitative understanding of the time-scale and extent of ecosystem response to changes in atmospheric Hg deposition.<sup>35</sup> Multidecadal insight has been mostly obtained through paleolimnological studies involving lake sediment cores and time series of fish Hg concentrations. Sediment studies show enrichment in Hg deposition for approximately the past century with the extent of increases in Hg deposition from preanthropogenic values decreasing with increasing latitude from the midlatitudes ( $41\text{--}50^\circ\text{N}$ )<sup>173</sup>. Studies have indicated that remote regions receiving Hg inputs from largely global sources of Hg generally show long-term increases in Hg inputs. In contrast, in regions experiencing recent controls on elevated Hg emissions have been decreases in sediment Hg deposition over recent decades.<sup>98</sup>

Fish Hg measurements are available over shorter time scales but in some cases extend back 40 years or more. Generally these studies have shown declines in fish Hg concentrations consistent with regional declines in Hg deposition,<sup>174</sup> although patterns of changes in fish Hg have been linked with other disturbances including land use, nutrient supply, climate change effects, and decreases in acid deposition.<sup>33</sup> Drevnick et al.<sup>175</sup> observed decreases in fish Hg in lakes on Isle Royale in Lake Superior, which they attributed to decreases in atmospheric  $\text{SO}_4^{2-}$  deposition. Decreases in acid deposition could also drive decreases in fish Hg due to increases in surface water pH.<sup>171</sup> In contrast, recent studies in Scandinavia report increases in fish Hg associated with increases in DOM resulting from watershed recovery from decreases in acid deposition.<sup>176,177</sup> These

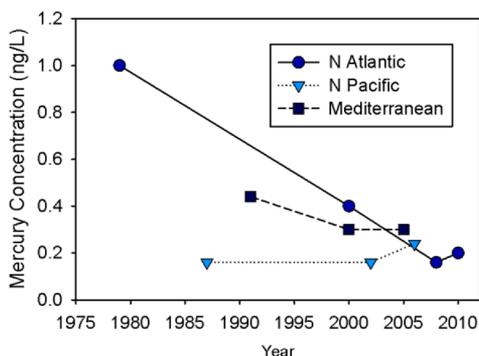
variable spatial and temporal patterns indicate that while controlling Hg inputs are essential to decrease Hg exposure, the response of biota will be highly variable in space and time due to the residence time of Hg associated with processing through detrital organic pools, and watershed processes/disturbances that affect Hg transport, net methylation and trophic transfer.

## ■ OCEANS

**Ocean Hg Transport and Long-Term Changes.** Empirical and modeling studies<sup>16,99,178–183</sup> provide a reasonable constraint on external Hg inputs to oceans, including the magnitude of air-sea exchange of Hg (wet and dry deposition and gas evasion). Atmospheric deposition is the dominant input of Hg(II) to the open oceans (3600 Mg yr<sup>-1</sup>; Figures 1,3).

Over 80% of the Hg deposited in marine ecosystems is reemitted to the atmosphere (as Hg(0) predominantly, but some as (CH<sub>3</sub>)<sub>2</sub>Hg) (Figures 1,3), increasing the residence time of Hg cycling through the reservoirs of the surface biosphere.<sup>148,181</sup> Net biotic and photochemical reduction of Hg(II) (photochemical oxidation and reduction, as well as biological reduction occur in surface waters) drives evasion of the Hg(0)<sup>181,184–187</sup> While evasion may decrease the pool of potentially bioavailable Hg(II) for methylation and bioaccumulation over the short-term, model calculations suggest that because of the overall recycling of Hg (deposition and re-emission) between both the surface ocean and the land and the atmosphere, the ocean is the dominant long-term sink for primary anthropogenic Hg emissions to the atmosphere.<sup>22,100</sup>

An evaluation of the cumulative impact of historical anthropogenic emissions from the last century on ocean Hg concentrations and distribution suggests an increase in the surface ocean (~200% increase on average) and subsurface waters (~25% increase<sup>140</sup>). In contrast, the effects of historical emissions on the deep ocean are small (~10%) and are confined to recent deep water formation, such as the deep North Atlantic (due to sinking North Atlantic Deep Water) and the deeper waters of the Southern Ocean (where Antarctic Deep Water sinks from the surface). If one further considers inputs from historic precious metal mining in the Americas and elsewhere dating back from the 1500s,<sup>22,188</sup> the anthropogenic impact of Hg on the surface and intermediate ocean waters is greater, increasing by a factor of 2–3. Time series data indicate an enrichment in Hg concentrations in the upper waters of the North Atlantic Ocean with the first reliable measurements in the 1980s, followed by decreases in recent years<sup>140</sup> (Figure 4).



**Figure 4.** Long-term trends in total Hg concentrations in the surface waters of the oceans and Mediterranean Sea.<sup>140</sup>

Similar decreases have also been reported for the Mediterranean.<sup>189</sup> In contrast, in the Pacific there have been increases in Hg concentrations in the upper waters since 2000.<sup>32,190</sup> These data confirm that Hg in the upper ocean responds on a decadal time scale to changes in the atmosphere, and reflects regional signals associated with anthropogenic inputs. The recent trends in ocean water column Hg appear consistent with decreasing Hg emissions in North America and Europe and rapidly increasing emissions in Asia<sup>6,8</sup> (Figure 2).

**Methylated Hg Dynamics.** In contrast to an improved understanding of the dynamics of Hg inputs to the oceans, there is limited information on how methylated Hg concentrations (both CH<sub>3</sub>Hg and (CH<sub>3</sub>)<sub>2</sub>Hg; collectively termed  $\sum$ CH<sub>3</sub>Hg) have responded to these changes. Nevertheless, past and ongoing changes in the extent and distribution of anthropogenic emissions and Hg inputs to ocean waters<sup>6,8,22,191</sup> have and will lead to changes in ocean fish concentrations.<sup>183,192,193</sup> Ocean circulation patterns and the presence of a subsurface zone of maximum methylation<sup>140</sup> contribute to lags between the onset of changes in atmospheric Hg deposition and the response of fish CH<sub>3</sub>Hg concentrations in the open ocean.

Concentrations of  $\sum$ CH<sub>3</sub>Hg are low in ocean surface waters and increase with depth, reaching a maximum in the subsurface/subthermocline waters (>100–1000 m) depending on location. Concentrations of  $\sum$ CH<sub>3</sub>Hg are generally low in the deep ocean. The higher concentrations of  $\sum$ CH<sub>3</sub>Hg are found in zones of the upper ocean where sinking organic matter is rapidly remineralized.<sup>194</sup> Open ocean profiles are generally consistent with the formation of  $\sum$ CH<sub>3</sub>Hg in conjunction with the decomposition of organic matter.<sup>32,195–198</sup> Stable Hg isotope incubation experiments in the Arctic also indicate that methylation of Hg(II) occurs in the water column producing both CH<sub>3</sub>Hg and (CH<sub>3</sub>)<sub>2</sub>Hg.<sup>199</sup> In sum, field observations and modeling demonstrate that in situ methylation, rather than external inputs, is the predominant source of CH<sub>3</sub>Hg that accumulates in ocean fish consumed by humans.

The formation of  $\sum$ CH<sub>3</sub>Hg in the open ocean contrasts with patterns of freshwater and coastal ecosystems (see Land and Freshwaters above). First, while measurable (CH<sub>3</sub>)<sub>2</sub>Hg has generally been found in open ocean studies, it is rarely detected in coastal and freshwater ecosystems. Moreover, while CH<sub>3</sub>Hg is generally formed in oxygen deficient or anoxic zones in coastal and freshwaters, this is not a prerequisite for formation in the open ocean.<sup>140,160</sup>

A recent mass balance suggests riverine/coastal inputs of CH<sub>3</sub>Hg are small relative to the production of  $\sum$ CH<sub>3</sub>Hg within the ocean water column<sup>140</sup> (Figure 3). Riverine inputs of CH<sub>3</sub>Hg from the coastal zone to offshore waters are approximately 20 Mg yr<sup>-1</sup> (range 14–24 Mg yr<sup>-1</sup>) with about 30 Mg yr<sup>-1</sup> (20–40 Mg yr<sup>-1</sup>) deposited into the coastal zone due to net particulate settling.<sup>140</sup> The return flux of dissolved CH<sub>3</sub>Hg due to diffusional and advective transport from coastal (shelf and slope) sediments is estimated to be on the same order as the net depositional input (range 4–40 Mg yr<sup>-1</sup>).<sup>140,200</sup> Groundwater inputs are not considered substantial given the generally low concentrations of CH<sub>3</sub>Hg and the estimated water flux.<sup>140,201</sup>

The major ocean sink for  $\sum$ CH<sub>3</sub>Hg is photochemical and biotic demethylation (Figure 3). Estimates of demethylation of  $\sum$ CH<sub>3</sub>Hg in the surface waters (<50 m) and in the subsurface waters (50–1000 m) are 60 and 160 Mg yr<sup>-1</sup>, respectively, based on an assessment of the literature.<sup>184,185,187,198,202</sup> While

these values are average rates, demethylation rate constants vary by a factor of 5 depending on incident solar radiation and other factors. The net removal of  $\sum\text{CH}_3\text{Hg}$  to deep ocean sediments is a secondary sink ( $<20\text{ Mg yr}^{-1}$ ;  $<20\%$  of the inputs to the deep ocean; Figure 3). The rate of decay of  $\sum\text{CH}_3\text{Hg}$  even in deeper, colder intermediate depth waters is sufficiently rapid that both  $\text{CH}_3\text{Hg}$  and  $(\text{CH}_3)_2\text{Hg}$  should not persist over the time scales of decades (i.e., the time scale of offshore water mass transport), suggesting continued in situ formation in subsurface ocean waters in concert with organic matter degradation.<sup>197,198,203</sup>

**Mercury in Marine Biota.** As with freshwaters,  $\text{CH}_3\text{Hg}$  concentrations in ocean fish generally track trophic position and age, but important differences are evident in concentrations for the same species found in different ocean basins. Generally, patterns in fish Hg concentrations are consistent with variations in total Hg concentrations among oceans, although there are differences in the fraction of Hg as  $\sum\text{CH}_3\text{Hg}$  across marine ecosystems.<sup>31,32,195,204</sup>

Feeding depth and location are important considerations. Many commercial and noncommercial fish species that feed in subsurface waters to 600 m have higher  $\text{CH}_3\text{Hg}$  levels than species of similar trophic status feeding at the surface. Positive correlations have been found between the concentrations of  $\text{CH}_3\text{Hg}$  in piscivorous fish and marine birds and their feeding depth or the depth of their prey.<sup>204–209</sup> Studies using stable isotopes of Hg, C, and N in marine fish indicate that while estuarine and coastal fish may obtain  $\text{CH}_3\text{Hg}$  generated within the coastal watersheds and sediments, offshore ocean fisheries derive  $\text{CH}_3\text{Hg}$  from the open ocean.<sup>210,211</sup>

As for freshwater ecosystems, it is unlikely that future  $\text{CH}_3\text{Hg}$  concentrations in ocean fish will solely be controlled by changes in atmospheric Hg emissions and deposition to the oceans. The future impacts of changing climate on ocean Hg deposition, transformations and bioaccumulation of  $\text{CH}_3\text{Hg}$  will likely be manifested through changes in (1) the efficiency of net reduction in surface waters and net methylation in the subsurface; (2) the rate of gas exchange driven by changes in physical characteristics; (3) the rate of oxidation of  $\text{Hg}(0)$  in the atmosphere; and/or (4) changes in wet deposition patterns.<sup>212,213</sup> Moreover, changes in Hg removal from the surface ocean by particle scavenging as a result of changes in productivity also could impact the extent of re-emission to the atmosphere, and therefore net methylation. The impacts of climate change, eutrophication,<sup>214</sup> and other secondary factors (e.g., changes in species composition, overfishing) on the net transfer of atmospheric Hg deposition into the aquatic food chain as  $\text{CH}_3\text{Hg}$  are often ignored in model projections of future changes in the global Hg cycle. However, these secondary effects will likely be important drivers of future Hg exposure.

## ■ CONCLUDING REMARKS

**Policy Implications.** The major policy implications from this synthesis of existing Hg science are (1) atmospheric emissions and deposition dominate anthropogenic Hg inputs and controlling these inputs remain an important priority for national and international policy; (2) releases to land and water are very high in some locales and more information is needed on the extent to which these inputs circulate globally and contaminate global fisheries via re-emissions and transport from nearshore to offshore waters; (3) human consumption of marine fish and shellfish is a globally important exposure

pathway and should be more fully integrated into cost-benefit analyses; (4) health effects to humans beyond IQ decrement and at low levels of exposure, and the widespread effects to wildlife suggest the need for expanding the end points evaluated in risk assessments; and (5) effectiveness evaluation for both national policy and the international Hg treaty should include not only fish, but also supporting measurements of Hg deposition, water column  $\text{Hg}/\text{CH}_3\text{Hg}$ , and sediment  $\text{Hg}/\text{CH}_3\text{Hg}$ , for a range of systems and geographic locations.

Mercury is a local, regional, and global pollutant; while we have focused on Hg pollution at the global scale, it is important to emphasize that Hg also has regional and local dimensions. Regional/local releases and/or exposure of the local consumer and wildlife can be mitigated by strategies that are geared toward the characteristics of a particular location. In parallel with discussions to limit and manage Hg releases through an international treaty, there is a critical need for coordination of local, national and international health and environmental agencies to develop (1) strategies and technologies to limit releases and exposure from local/regional sources;<sup>4,45,215,216</sup> and (2) quantitative risk-benefit assessment paradigms for local/regional fish consumption to protect local populations.

**Monitoring and Effectiveness Evaluation.** Documenting the response of Hg in ecosystems following the implementation of Hg policies and management activities and the confounding effects of other actions that affect Hg dynamics (e.g., nutrient control, controls on atmospheric S and N deposition, land use and climate change, changes in water management) at both local and global scales is a critical element of the international Hg treaty and most national policies.<sup>4,17,217,218</sup>

To support evaluation of these national and international policies, there is a need for coordinated and detailed global, multimedia Hg monitoring network (air, water, sediment and soils, and biota). Better coordination among current and future measurement campaigns should focus less on a particular media or ecosystem type (e.g., air, water) and more on cross boundary transport, such as land–atmosphere exchange or riverine transport to the coastal ocean.<sup>13,97,219</sup> There have been few studies of changes in Hg transport and contamination following recent industrialization of developing countries which are likely to have increasing future emissions.<sup>6,8,220</sup> There is also a critical need for observations in understudied areas such as the Southern Hemisphere, the major oceans, and within the free troposphere.

Further model development will be required to validate processes and hypothesis testing; probe the quality of empirical observations; and evaluate future scenarios of the impacts of changing Hg emissions and/or global change on the levels of  $\text{CH}_3\text{Hg}$  in aquatic organisms. While monitoring is useful to assess trends and validate models, it often fails to elucidate the processes and factors which influence the net conversion of  $\text{Hg}(\text{II})$  into  $\text{CH}_3\text{Hg}$ . A quantitative understanding of pathways and mechanisms that affect the transport of  $\text{Hg}(\text{II})$  from sources to ecosystems, the conversion of  $\text{Hg}(\text{II})$  to  $\text{CH}_3\text{Hg}$ , and its bioaccumulation in food webs is fundamental to evaluating and managing human and wildlife health risks. Therefore, scientific synthesis should be periodically supported.

**Future Research Priorities.** While research over the past few decades has provided considerable insight on Hg dynamics and effects ranging from local to global scales, this synthesis and the associated literature have identified several focused questions for future Hg research:

- What is the redox chemistry of atmospheric Hg and how does it determine source-receptor relationships between emissions and deposition?
- To what extent and how rapidly will changes in anthropogenic Hg emissions at one location affect local, regional and global concentrations of Hg and ultimately CH<sub>3</sub>Hg in fish elsewhere?
- How do climatic variability and anthropogenic disturbances (e.g., climate change, population increases, perturbations to food chains, changes in other air pollutants, land disturbance) affect CH<sub>3</sub>Hg concentrations in fish and other top predators?
- What are the low dose chronic effects of CH<sub>3</sub>Hg on the fish eating populations?

These questions should continue to drive future Hg research that will improve understanding of the response to anticipated controls on anthropogenic Hg emissions and releases.

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### Notes

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