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Recovery of Mercury-Contaminated Fisheries

In this paper, we synthesize available information on the links between changes in ecosystem loading of inorganic mercury (Hg) and levels of methylmercury (MeHg) in fish. Although it is widely hypothesized that increased Hg load to aquatic ecosystems leads to increases in MeHg in fish, there is limited quantitative data to test this hypothesis. Here we examine the available evidence from a range of sources: studies of ecosystems contaminated by industrial discharges, observations of fish MeHg responses to changes in atmospheric load, studies over space and environmental gradients, and experimental manipulations. A summary of the current understanding of the main processes involved in the transport and transformation from Hg load to MeHg in fish is provided. The role of Hg loading is discussed in context with other factors affecting Hg cycling and bioaccumulation in relation to timing and magnitude of response in fish MeHg. The main conclusion drawn is that changes in Hg loading (increase or decrease) will yield a response in fish MeHg but that the timing and magnitude of the response will vary depending of ecosystem-specific variables and the form of the Hg loaded.

INTRODUCTION

There is considerable evidence that natural background levels of mercury (Hg) in the environment have increased in the past 150 y. This evidence comes from studies of lake and ocean sediment profiles, peat bogs, and ice cores, in both remote and industrialized regions. These data point toward a general increase of Hg levels concurrent with industrialization as a result of both the intentional use of Hg and from combustion of Hg-containing fossil fuels, gold mining, and processing of ores. Because Hg is transported on hemispheric or global scales via the atmosphere, contamination is not limited to locations in the vicinity of emission sources and includes remote ecosystems. In remote regions, increases are in the range 2- to 5-fold over preindustrial values, whereas locally affected sites can have levels increased by a factor of 10 or more (1, 2).

Although most Hg in the environment is inorganic, some is converted to the highly toxic monomethyl mercury (MeHg), which bioaccumulates in fish. Fish are an important dietary source in many developed and developing nations. In some communities, fish, shellfish, birds, and marine mammals constitute critical components of the diet or local economies. MeHg concentrations in fish are commonly high enough to represent a risk to the health of the fish-eating communities (3–6). Elevated MeHg concentrations may impair the long-term sustainability of aquatic ecosystems and have deleterious effects on the behavior and health of associated wildlife. Recent studies suggest that trophic transfer of Hg also occurs in terrestrial food chains (7, 8).

A critical question currently being investigated is, "Will efforts to reduce inorganic Hg inputs and mobility in the environment reduce fish Hg concentrations?" Several confounding issues have historically complicated efforts to answer this question. Individual ecosystems have different abilities to translate a given Hg load into MeHg concentrations in biota due to a range of factors affecting Hg cycling and bioaccumulation (e.g., hydrology, water quality, trophic structure, temperature) and the uncertain effects of terrestrial ecosystems on the timing and magnitude of delivering atmospheric Hg deposition to waterbodies. As a result, waterbodies close to each other and receiving the same atmospheric Hg deposition loads often have very different MeHg concentrations in biota. The conversion of inorganic load to MeHg can also be affected not only by the absolute amount but also by the form of Hg loaded to an ecosystem in cases of point-source contamination.

Despite the above complexities, it is reasonable to expect that changes in inorganic Hg loading will affect MeHg concentrations in ecosystems. A few studies provide information on historical levels of MeHg in humans and aquatic top predators (whales, seals) (9) and show that MeHg levels have increased significantly since preindustrial times. Although associated with some uncertainty, these results indicate that anthropogenic emissions in the last century have caused general increases in not only total Hg in the environment but also MeHg in freshwater and marine food chains.

In this paper, our hypothesis is that the concentration of MeHg in fish will decrease in response to decreases in Hg loading. However, the magnitude and timing of this response will vary with the type of Hg contamination and ecosystem characteristics. To test this hypothesis, we explore the current knowledge on environmental cycling and levels of Hg in the environment focusing on:

- Controls on net methylation of inorganic Hg in the environment.
- Examples of documented fish responses to both increases and decreases in Hg loading in different ecosystems receiving direct input via industrial discharges.
- Results from spatial and temporal analyses of Hg contamination in ecosystems receiving mainly atmospheric Hg.
- Results from experimental ecosystem manipulations and loading experiments.

Finally, we discuss the critical factors affecting the magnitude and timing of fish Hg responses to changes in loading of Hg and attempt to explain the variability.

CONTROLS ON ECOSYSTEM SENSITIVITY TO MERCURY LOADING

In this section, we examine the main processes involved in transporting and transforming inorganic Hg to MeHg in fish, with a focus on freshwater ecosystems. We define ecosystem sensitivity to Hg loading as the ability of that ecosystem to transform inorganic Hg load into MeHg in biota. The evaluation thus includes the movement and net methylation of Hg in watersheds and the aquatic ecosystem, as well as uptake into food chains. Direct atmospheric input of MeHg is not considered in this synthesis, because with a few notable exceptions, *in-situ* production is the dominant MeHg source to watersheds. By analyzing components of this cycling separately, we aim to illustrate the complexity of ecosystem cycling of Hg and MeHg and to describe the main controlling factors in each step.

Factors Affecting Transport of Mercury Through Watersheds

In addition to direct atmospheric deposition, watersheds provide an input of Hg to freshwater ecosystems via runoff. Watershed characteristics control the retention of Hg in the terrestrial compartment and transport of Hg to sites of methylation via complex hydrologic, chemical, and biologic processes. Some of these key characteristics are:

- The size and topography of watersheds and the watershed-to-surface water area ratio (affecting the residence time and flow pathways of runoff and the total amount and yield of Hg to waterbodies).
- Land cover (affecting dry deposition rates and the degree of interaction between water and both methylating and non-methylating soils).
- Land use (affecting the transfer of particulate Hg load to sites of methylation and the transfer of MeHg to the aquatic compartment (10–14) and, in the case of forestry, increased transfer of MeHg from the terrestrial compartment to the aquatic (15, 16).

Watershed size and watershed-to-surface water ratios are perhaps the most important determinants of Hg retention and delivery to aquatic ecosystems. As watershed sizes increase, yields per unit area usually decline (17–19). Based on a synthesis of data from watersheds of varying size, Grigal (20) suggested that there is actually an overall decrease in Hg flux and concentrations in runoff with increasing watershed size due to less efficient transport and increased loss processes. Very large watersheds may be less responsive to changes in atmospheric Hg load than those in which the watershed area is smaller in comparison to the lake surface area. Land-use changes, anthropogenic disturbances, or local Hg sources may result in watershed Hg export many-fold larger than expected (20). Watersheds subject to significant transport of sediment in surface runoff, such as agriculturally dominated systems, contribute larger fractions of Hg to sites of methylation in aquatic sediments and wetland soils (14) than do forested watersheds (11, 21). However, because this Hg is largely associated with particulate matter, it may be less bioavailable than dissolved inputs of Hg. Other land-use impacts on Hg load and MeHg in fish have been noted, particularly in landscapes with erodable soils or developed landscapes (10), and controls on land-use practices may be used to reduce MeHg in biota. Using a modeling approach, Roué-Le Gall et al. (22) showed that when watershed characteristics such as those discussed above were coupled to information about the food web in 45 lakes, the sensitivity of biota to MeHg contamination could be predicted qualitatively (high to low).

The common factor among all of the watershed characteristics identified above is soil cover. Soils retain Hg in watersheds, and this retention is strongly coupled to organic matter (23, 20), where Hg is stored either through sorption of Hg deposited directly from the atmosphere or is associated with organic matter derived from the forest canopy or floor. Even in landscapes characterized by shallow and discontinuous soils, newly deposited Hg appears to be nearly completely retained in the short term (see The Gårdsjön Roof Study). The magnitude and timing of the release of Hg from this pool is controlled by the rate of decomposition of the soil organic matter pool and physical removal of the soil itself through erosion. The current lack of insight in the dynamics of Hg release from soils makes the greatest contribution to the uncertainty in the quantitative prediction of the magnitude and timing of the effects of a change in Hg load on MeHg in biota.

Factors affecting net mercury methylation. For Hg loads to ecosystems to result in MeHg in biota, inorganic Hg must be converted to MeHg. Net MeHg production is affected by the concentration of bioavailable Hg and by a complex system of environmental factors, most important:

- *Spatial controls:* the areal extent and connectivity of methylating and demethylating zones within the ecosystem.
- *Biogeochemical controls:* the bioavailability of Hg and MeHg to methylating and demethylating bacteria, and the relative activity of those organisms.

Spatial controls on net methylation. The ecosystem compartments that support net MeHg production are reasonably well defined, except for the oceans. Because net methylation is favored under anoxic conditions (24), the areal extent of wetlands and hydric (water saturated, partially anaerobic) soils (11, 21, 25) is a strong determinant of MeHg export from watersheds. Wetlands can be particularly active zones of MeHg production (e.g., the Florida Everglades [26], tidal marshes [27]). Methylation in bogs (28), fens (29, 30), and riparian zones (19, 31, 32) is highly dependant on flow paths and hydrologic connectivity that govern the location and extent of zones of microbial activity. Forest disturbance (clear cutting and mechanical damaging to soil cover by tractors) has been shown to increase the load of MeHg to the aquatic ecosystem (15, 16), including biotic concentrations (33, 34), thus providing a potential for more direct anthropogenic influence on MeHg levels in aquatic biota.

In freshwater aquatic ecosystems, warm, shallow, organic-rich lake sediments are often important zones of net methylation (35–37) and therefore a real extent of these sediments in a lake impacts the conversion of Hg to MeHg (38). MeHg concentrations and production rates in sediments are generally maximal at or near the sediment surface (39). The presence of anoxic bottom waters in stratified lakes significantly enhances net MeHg bioaccumulation, often through *de-novo* MeHg production in bottom waters (40, 41) and also through enhanced flux of MeHg from sediments to the anoxic water column.

Flooding increases the production of MeHg due to increased rates of decomposition of terrestrial organic matter, which stimulates development of poor oxygen conditions and activity of Hg-methylating bacteria (30, 42, 43). Soil drying and rewetting cycles also strongly affect Hg methylation (44). Oxidation of reduced sulfur during drying leads to a pulse of sulfate reduction and Hg net methylation when soils rewet (45). Changes in water levels in lakes and wetlands can significantly affect MeHg levels in fish (45, 46). Several examples of this behavior from reservoirs are also available (42, 47–51).

Although the MeHg contamination of ocean fish has the greatest impact on human health globally (52), information on the sources of MeHg to estuaries, coastal zones, and particularly the oceans is limited. In the limited number of estuaries studied, MeHg production in sediments appears to be its major source (28, 53–55). Although net methylation in salt marshes can be high (56, 57), their importance to estuarine and coastal MeHg budgets has not been quantified. In coastal zones, shallow sediments (54, 58–61) also appear to be the major sources of MeHg. There are also unidentified sources of marine MeHg in the high Arctic (62). The source of MeHg to open ocean fish remains a subject of debate (63), with little available information, although there is a documented accumulation of MeHg in deep ocean waters with an unknown source (64).

Factors affecting the uptake of MeHg by the food web. There are important physical, biological, and chemical controls on MeHg bioaccumulation that dramatically affect the transfer of Hg load into MeHg in fish and subsequent predators.

Box 1

Biogeochemical controls on net methylation in aquatic ecosystems

In addition to the impact of Hg loadings on MeHg production, some key biogeochemical cycles also have large effects on net MeHg production:

- **Sulfur.** The Hg and sulfur cycles are intimately linked, thus linking acid precipitation to the Hg cycle. The balance between sulfate and sulfide is a key control on Hg net methylation rate in many ecosystems. Sulfate stimulates Hg-methylating sulfate-reducing bacteria, while excess sulfide creates Hg complexes that are not bioavailable for methylation (57, 65, 66). Sulfate stimulation of methylation has been demonstrated in studies that range from pure culture (67–69), to sediment and soil amendments (70–72), to field amendments to lakes and wetlands (73–77). Among these studies, and in the field, the optimal concentration for methylation ranges from 10 to about 300 μM sulfate, while the optimal sulfide concentration is quite low, about 10 μM (26, 78). Factors such as iron and organic matter concentration that affect Hg and S complexation change these optima. Sulfate, along with pH and dissolved organic carbon, have been identified as parameters that relate Hg levels in fish among water bodies (75).
- **pH.** Many studies have linked lake acidity to increased MeHg bioaccumulation (75, 79–81). This observation has important implications for management of fish Hg contamination. Although several mechanisms have been hypothesized, the main process controlling this phenomenon is not evident. This pattern could be driven by effects on bioaccumulation, for example, decreases in aquatic productivity with decreases in pH. However, the effect could also be linked to MeHg production. Acidity linked to sulfate deposition may stimulate the activity of Hg-methylating sulfate-reducing bacteria. Additionally, uptake of Hg by micro-organisms that use facilitated transport for Hg uptake is enhanced with decreasing pH (82).
- **Organic matter.** Both the character and concentration of dissolved organic matter affect the complexation and potential bioavailability of Hg and MeHg for methylation/demethylation (83–85). Higher molecular weight dissolved organic matter can limit Hg availability to bacteria (86), probably through the formation of complexes that are too large to assimilate, whereas small organic ligands can enhance Hg uptake through facilitated transport (87). Recent work suggests that dissolved organic matter is an important ligand under sulfidic conditions, through interactions with Hg sulfur complexes (88, 89). The relationships between dissolved organic matter and methylation need to be further explored before they can be adequately modeled. Further, organic matter in sediments and soils often stimulates the activity of microorganisms, making it an important control on net methylation rates (90).
- **Iron.** Like sulfur and decayed organic matter, the impact of iron on net methylation appears to be concentration and environment dependent (14, 91, 92). Iron may affect net methylation through impacts on Hg complexation, solid phase partitioning, or microbial activity. A few strains of iron(III)-reducing bacteria are now known to be capable of Hg methylation (93, 94), but the impact of iron on Hg net methylation in the environment needs further study.
- **Hg "aging."** Recent Hg-amendment studies in lakes and wetlands show that the bioavailability for methylation of recently loaded Hg differs from that of the overall Hg pool in the ecosystem (95–97). Understanding these differences will be a key to modeling ecosystem responses to changes in Hg load. The "aging" effect could include both chemical changes in Hg complexation and partitioning with time and changes in the relative distribution of newly loaded Hg *versus* Hg that has been in the system for longer periods (e.g., newly loaded Hg could be more or less focused in zones of methylation).
- **Type and activity of bacteria.** Although Hg methylation can be measured in almost any soil or sediment under reducing conditions, only a few sulfate-reducing bacteria (24) and a few closely related iron-reducing bacteria (93, 94) have demonstrated methylation ability in pure culture. There are a variety of microbial demethylation mechanisms, including the *mer* operon detoxification system that is spread widely among microorganisms in contaminated environments, and may serve to limit MeHg accumulation at high Hg concentrations. The oxidative demethylation pathway (98) appears to be the dominant demethylation pathway in sediment and soil environments where the *mer* operon is not operating. It is anaerobic, driven by sulfate-reducing bacteria and methanogens, and linked to one-carbon metabolic pathways rather than to Hg levels. However, the processes of methylation and oxidative demethylation may be differently affected by temperature, organic substrates, and redox chemistry (24). For example, sulfide (57, 58, 99), organic matter (90), and redox (100) appear to affect methylation more than they do demethylation. A better understanding of the controls on demethylation is needed to improve assessments of net methylation in different environments.

Differences in these processes among water bodies may result in varying responses to Hg loading.

Biophysical controls. MeHg uptake is affected by the nature (pelagic *versus* benthic) and structure of the food web (101). MeHg bioaccumulation in lakes occurs through both benthic and pelagic routes. MeHg produced in sediments can be directly accumulated into benthic food webs or flux into overlying water and into pelagic food webs. Stratified lakes where MeHg accumulates in anoxic bottom waters (40, 41) often provide a pulse of MeHg to surface waters and plankton after fall turn over (102). In large lakes and coastal marine environments, MeHg accumulation appears to occur predominantly around near-shore areas of sedimentary MeHg production (55, 100, 103). Zones of MeHg production that are disconnected from aquatic food webs (e.g., isolated bogs) may have little impact on Hg in aquatic biota but can affect terrestrial food webs (104, 105).

Biological controls. Food web structure, fish population age structure, growth rates, and physiological controls on uptake all affect the bioaccumulation of MeHg. Any examination of

Table 1. Observations of fish mercury concentrations for selected point-source contaminated ecosystems.

Site	Primary Hg source	Fish species	Loading trend	Fish Hg response	Reference
Clay Lake, Canada	Chlor-alkali facility	Walleye *(Sander vitreus)*	10 tonnes of inorganic Hg released 80 km upstream, primarily from 1962 to 1969.	Clear increase in response to contamination and remediation. Fast then slower recovery after remediation (Fig. 1).	118
Haifa Bay, Israel	Chlor-alkali facility	Soldierfish *(Sargocentron rubrum)*	Tonnes of Hg released from 1950s to 1970s.	~80% decline from late 1970s to 1990.	120, 121
Princess Royal Harbour, Australia	Super-phosphate processing plant	Several marine fish species	14 kg y^{-1} for ~30 y, stopped in 1984.	50% decline from 1984 to 1993. Fast response at first, slower later.	122
Lake Vänern, Sweden	Chlor-alkali	Northern pike *(Esox lucius)*	Load ~3 tonnes per year from 1920s to 1960s, reduced to ~3 kg y^{-1} during 1970s to 1980s.	50% decline within a decade, then stabilized.	161
Onondaga Lake, NY, USA	Chlor-alkali	Small-mouth bass and several other freshwater species	75 000 kg discharged in the 1940s to 1970, followed by greatly reduced loading and closure.	No statistically significant decline after load reductions; fish concentrations remain elevated.	162
East Fork Poplar Creek, Oak Ridge, TN, USA	Inorganic Hg used in lithium isotope separation at national security complex in Oak Ridge	Red-breasted sunfish *(Lepomis auritus)*	250 tonnes of inorganic Hg released from 1950s to early 1960s, then an 80% decline in Hg load and concentrations in creek near facility.	After remedial actions, fish Hg decreased at a site near point of release but not a short distance downstream.	125

change in MeHg in fish through time must examine potential concomitant changes in food web and fish population structure. Changes in Hg bioaccumulation patterns after the invasion of a fish species provide an example of how food web structure affects Hg bioaccumulation (106). Food web structure and composition also affect bioaccumulation through food quality (42, 107) and gut chemistry (108) effects on uptake efficiency. Faster fish growth rates have the potential to reduce fish Hg levels when comparing concentrations standardized to a given size of fish (109, 110). Growth differences in multiple levels of the food web (e.g., plankton, prey fish, predatory fish) could result in cumulative growth effects across a food web.

Chemical controls. The bioavailability of MeHg to the plankton and benthic invertebrates at the bottom of food webs is affected by MeHg partitioning and complexation. Higher organisms, such as zooplankton (111) and fish (112), accumulate MeHg mainly from their diet. Thus, accumulation of MeHg by these organisms is affected strongly by bioavailability to lower trophic levels. The direct or indirect inhibition of MeHg accumulation by higher molecular weight dissolved organic matter has been well documented for fish and zooplankton (113). Particulate matter can play largely the same role, sorbing MeHg and making it less available for uptake by lower organisms through dissolved routes. The role of organic matter overall in Hg transport, methylation, and bioaccumulation is complex with competing influences but central to the physical and biogeochemical behavior of Hg in watersheds. For example, dissolved organic carbon in runoff may enhance the delivery of inorganic Hg and MeHg to waterbodies via binding and cotransport but may inhibit the biological uptake of MeHg within a waterbody. An inverse relationship between pH and MeHg bioaccumulation is also well documented, such as for loons (114), benthic invertebrates (115), and fish (116), although pH may affect both methylation and bioaccumulation.

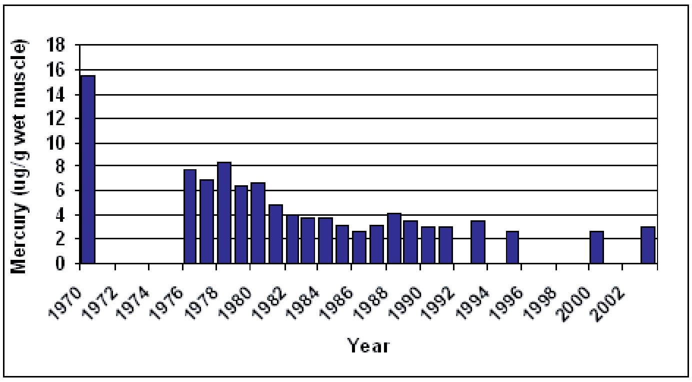

Figure 1. Observed decline in walleye Hg concentrations in Clay Lake, Canada, after reduced releases of Hg from a chlor-alkali facility 80 km upstream (results standardized to 50 cm fish) (118).

Summary of controls. Mercury concentrations in fish can vary by an order of magnitude or more in regions with similar atmospheric Hg deposition rates. This is because there are many factors that affect the movement, methylation, and bioaccumulation of Hg in different environmental settings. The form of Hg being loaded into an ecosystem can also affect the production and bioaccumulation of MeHg. These considerations often obscure a relationship between Hg loading and fish Hg levels when looking at multiple sites, even if the fish Hg levels are standardized to a particular species and size. In the next section, however, we present evidence suggesting that although each ecosystem has its own efficiency to translate an inorganic Hg load into MeHg in biota, Hg loading is still an important determinant of fish Hg concentrations.

EVIDENCE

Here we examine data from remote areas and point-source–contaminated sites that help characterize the response of fish Hg concentrations to changes in Hg loading rates. Studies can be grouped into four categories according to Carpenter (117), three of which are used in this synthesis: i) time series measurements, *ii)* spatial analysis/gradient studies, and *iii)* experimental manipulations.

Time Series Measurements

Temporal data from point-source–contaminated sites. Freshwater and marine sites with large inputs of Hg from local industrial discharges clearly show that Hg loading rates can affect fish Hg concentrations (Table 1). Clay Lake, Canada, for example, is downstream of a chlor-alkali facility that released approximately 10 tonnes of inorganic Hg to receiving waters, primarily from 1962 to 1969 (118). Mercury concentrations in walleye in Clay Lake in 1970 exceeded 15 μg g^{-1} wet muscle (50 cm standard fish), far higher than concentrations measured in systems without point-source Hg inputs. Walleye Hg concentrations quickly declined when Hg releases were curtailed, although the rate of recovery slowed in later years (Fig. 1). These observations can only be explained if Hg inputs associated with the chlor-alkali facility contributed to higher fish Hg concentrations.

Minamata Bay, Japan, is a very well-known case of Hg release to a marine environment in the 1950s but atypically involved the direct industrial release of MeHg (119). Other examples of Hg concentrations changing in marine fish in response to Hg releases or remedial actions associated with point sources include Haifa Bay, Israel (120, 121); Princess Royal Harbour, Australia (122); Dutch Wadden Sea and Ems Estuary (123); and Liverpool Bay, UK (124) (Table 1).

The response of MeHg concentrations at point-source–contaminated sites is not always proportional to the increase in inorganic Hg concentrations and, by inference, the rate of inorganic Hg loading. At East Fork Poplar Creek, Tennessee, USA, 250 tonnes of inorganic Hg were released to water and soils from a U.S. Department of Energy facility in the 1950s and early 1960s. Total Hg concentrations in surface waters and sediments were two to three orders of magnitude higher than natural levels (e.g., >1 μg L^{-1} in surface waters during the early 1990s), while MeHg concentrations in surface waters and Hg concentrations in fish increased less, by roughly one order of magnitude (125). This suggests that at a sufficient level of contamination either Hg concentration is no longer a limiting factor for gross MeHg production or rates of demethylation are enhanced to a greater degree than methylation, resulting in a less than proportional increase in net methylation. Similarly, decreases in Hg loading are not always followed by proportional decreases in MeHg levels. Elevated discharge of Hg from a chlor-alkali facility to Onondaga Lake, New York, USA, in the 1940s through 1970 contributed to elevated concentrations of Hg in the water column and fish (126, 127). However, cessation of Hg discharge from the facility has not resulted in changes in fish Hg concentrations after 30 y. This condition is likely due to continued high Hg loading in this urban lake due to a combination of external ongoing sources and resuspension of contaminated sediments, high natural sulfate loading, high loading of labile carbon, and highly anoxic conditions in the lower waters that promote high rates of methylation.

In contrast, significant reductions in fish Hg concentrations (50% or greater) have been observed at freshwater and marine sites with point-source contamination within a decade after remedial actions (118, 122–124). This initial response can be followed by a slower second phase with the potential to last decades. Multiple-phase temporal responses may occur due to residual loading (e.g., leaking equipment) or because some compartment(s) in the ecosystem accumulates Hg during the contamination phase (e.g., sediments or terrestrial system) and feed Hg back to the water column and biota for long periods after the original source of contamination is curtailed.

Fish Hg concentration trends in grossly contaminated areas may also be influenced by bioaccumulation of inorganic Hg. Although fish Hg burdens are primarily in the form of MeHg at remote sites (128), it is reasonable to question whether fish in ecosystems receiving very large inputs of inorganic Hg may accumulate larger proportions of inorganic Hg (59, 129). This would result in more rapid declines after load reductions, because inorganic Hg is depurated from fish more quickly than MeHg (130).

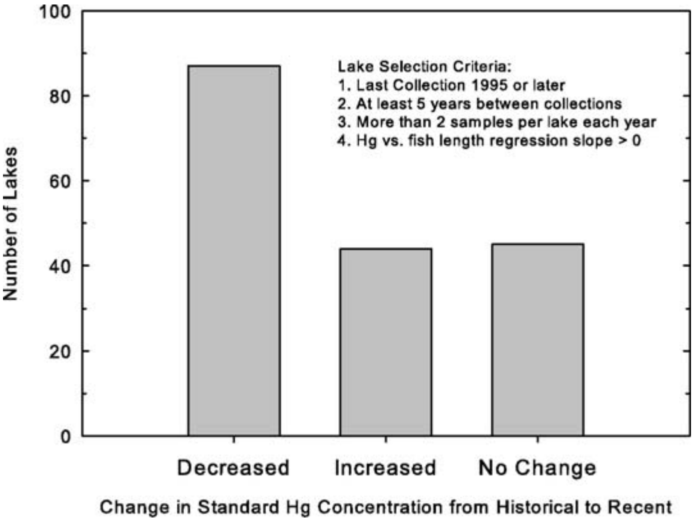

Figure 2. Comparison between recent and historical fish Hg concentrations in 176 lakes in Minnesota. Fish are northern pike *(Esox lucius)* and walleye *(Sander vitreus)* standardized to 55 and 40 cm, respectively (138).

The use of Hg in artisanal gold mining can be treated as a point-source situation with large amounts of Hg being released to the aquatic ecosystem, but very little data exist on temporal trends of Hg in fish in areas affected by Hg releases from areas affected by artisanal gold mining. This use of Hg is undoubtedly responsible for marked increases of Hg concentrations in soils, water, sediments, and air near mines located in the Brazilian Amazon, but available information on fish Hg provide variable results (131–134).

Response to changes in atmospheric Hg deposition. The response of fish Hg levels to changes in atmospheric Hg deposition is typically more difficult to demonstrate than for point-source contamination because of the smaller degree of change in Hg loading, natural year-to-year variability, and changes to other environmental factors that can affect MeHg cycling to a similar extent as changes in Hg deposition (e.g., water quality, land use, climate, hydrology, trophic structure). Relationships between Hg deposition and fish Hg may also be confounded by delays in delivery of Hg deposition to waterbodies via terrestrial systems. It has long been postulated that terrestrial ecosystems may impose time lags on the transport of atmospheric Hg deposition to lakes. This hypothesis has been corroborated in recent years by forest roof experiments in Sweden (16) and initial results from a whole-watershed Hg-loading experiment (METAALICUS) in Canada (135, 136) (see also Spatial/Gradient Analysis). Finally, there are generally limitations on data describing overall Hg loading to a waterbody, including wet deposition, dry deposition, and terrestrial export (where relevant), in terms of what was measured and for how long.

Recognizing the above challenges, there are examples of Hg concentrations systematically declining in fish for remote individual waterbodies (137) or on average for groups of waterbodies (138, 139), where associated data indicate a decline in wet or bulk Hg deposition. Hrabik and Watras (137) studied a seepage lake not affected appreciably by terrestrial Hg sources (Little Rock Lake, Wisconsin, USA) and concluded that Hg levels in yellow perch (30 g standard) decreased by roughly 30% between 1994 and 2000 due to decreased atmospheric Hg loading (bulk Hg deposition). Johansson et al. (139) reported a 20% average decline in Hg concentrations for northern pike (*Esox lucius,* 1 kg standard) from 42 remote Swedish lakes sampled at least 3 y apart, initially between 1981 and 1987, and again between 1988 and 1995. Wet Hg deposition declined by more than 50% in southwest Sweden (less in central and northern parts) during this period. Although fish Hg concentrations declined on average for the study lakes, increases and decreases were both observed for fish Hg levels in individual waterbodies. Although declines in fish Hg and wet Hg deposition were observed for these studies, other factors were also changing that could affect MeHg production (e.g., sulfate loading), confounding the interpretation of the effects of Hg loading. In Sweden during the same period as the above study, sulfate loading and to some extent pH levels in many lakes changed. Hrabik and Watras (137) accounted for pH changes in their analysis, but the extent to which sulfate effects were considered was not clear. In another study, Hg in predatory fish was determined in 176 Minnesota lakes over a period when deposition was known to decrease (138, Fig. 2). A range of trends in fish Hg concentrations (increase in 44, no change in 45, and decreases in 87 lakes) suggested a general link to atmospheric input but also that multiple factors likely affect fish Hg concentrations. These studies demonstrate the need for carefully designed monitoring programs that monitor not just Hg but also sufficient environmental information to help explain very different trends that might emerge among individual sites (140, 141).

The timing and magnitude of the response of fish Hg concentrations in open oceans to changes in Hg loading are not clear. Trends in Hg loading, primary sources of MeHg to the mixed surface layer of oceans, and temporal patterns for marine biota are matters of ongoing investigation and debate. There are no statistically robust datasets documenting temporal trends for Hg levels in marine fishes in the open oceans at warmer latitudes. Kraepiel et al. (63) reported no increase in yellow-fin tuna caught near Hawaii in 1998 relative to 1971 values, although many factors that could have affected Hg in fish, such as growth rates and food chain structure, were not studied, and changes could have occurred in the intervening years. Monteiro and Furness (142) reported increases in Hg concentrations of 65 to 397% for samples collected before 1931 and after 1990 in feathers of seabirds breeding in Azores, Madeira, and Salvages (subtropical northeast Atlantic, 30–40°N, 8–32°W). Temporal datasets exist for Hg levels in beluga, ringed seals, moose, seabirds, and fish from the Canadian Arctic marine environment during the past 20 to 30 y but show conflicting trends (143, 144). Some animal populations exhibited significant increases in Hg concentrations while others did not.

From this analysis of temporal studies we can conclude:

- Mercury loads can affect fish Hg concentrations in cases of point-source contamination, and reductions in Hg loading in such situations usually reduces fish Hg concentrations.
- The long-term magnitude of the response of fish Hg concentrations to changes in atmospheric deposition is not yet clear.

It is also very likely that different environmental settings affect the response time of fish Hg concentrations after changes in atmospheric Hg deposition, but this has not been shown clearly using datasets from natural settings. It has been observed experimentally however, as discussed in the next section.

Spatial/Gradient Analysis

The Nordic countries. Previous studies have demonstrated a link between spatial patterns of atmospheric deposition of Hg and MeHg levels in Swedish freshwater fish (145, 146). An examination of a larger dataset of Hg in fish from more than 1500 lakes in Sweden, Finland, and Norway revealed decreasing levels from south to north but with the high concentrations also found in central Sweden and along the Swedish coast of the Gulf of Bothnia (147). The elevated concentrations in central and northern coastal Sweden may be influenced by historical emissions of Hg to both water and air. However, specific ecosystem characteristics also contribute to the variability in fish Hg between regions. This variation is particularly evident for the southernmost part of Sweden, where very low concentrations of Hg were found in fish despite a larger influence of atmospheric pollution. This region of Sweden has mainly agricultural land use. This land disturbance typically increases lake productivity, decreasing fish Hg concentrations through biodilution (148).

Northeast Ecosystem Research Cooperative. A number of spatial datasets including atmospheric deposition, surface waters, sediments, and biota were recently compiled for eastern North America as part of a Northeast Ecosystem Research Cooperative initiative (149). These observations include 831 surface water, 570 surface sediment, more than 15 000 fish, more than 4,700 bird, and more than 900 mammal records. The spatial patterns and relationships evident for this analysis are similar to others reported elsewhere. None of the organisms studied showed direct relationships with spatial patterns in atmospheric Hg deposition, except blood Hg of Bicknell's thrush, for which Rimmer et al. (8) found a strong relationship with estimates of Hg in litter fall.

Biological Hg hotspots were geographically identified using this large dataset of biotic Hg concentrations. Yellow perch and common loon were chosen as indicator species for human and ecological effects of Hg, respectively (150). Impact thresholds of 0.30 μg/g (wet weight) for yellow perch fillets and 3.0 μg/g (wet weight) in blood for common loons were used to determine the location of biological Hg hotspots. These biological Hg hotspots reflect conditions that influence ecosystem and associated biological response to atmospheric Hg deposition. In particular, three factors were identified that control the formation of biological Hg hotspots, including *i)* local elevated atmospheric Hg deposition due to proximity to emission sources; *ii)* landscape sensitivity associated with forest cover, shallow hydrologic flowpaths, the abundance of wetlands, and unproductive surface waters; and *iii)* water level manipulations.

Links between Hg in fish and Hg wet deposition across the US. Two recent studies by Hammerschmidt and Fitzgerald (151, 152) reported strong relationships between rates of wet Hg deposition, as measured by the Mercury Deposition Network (153), and Hg concentrations in mosquitoes and largemouth bass in North America. Mosquito Hg is relevant to aquatic ecosystems because these animals have aquatic life stages and accumulate primarily MeHg. Hammerschmidt and Fitzgerald (143) found that the state-wide average concentration of MeHg in largemouth bass was significantly correlated with wet atmospheric Hg deposition among many American states. Only the accumulation of large, long-term data sets over wide geographic areas made this analysis possible.

From these studies we can conclude that a growing body of studies show significant correlations between Hg deposition rates and Hg levels in fish.

Experimental Manipulations

There have been a number of recent manipulation experiments that help resolve the relationship between Hg loading and the response of Hg in fish. These experiments have been conducted in freshwaters (and associated watersheds). Most are addition experiments. None of the addition experiments has been conducted long enough to observe a recovery phase.

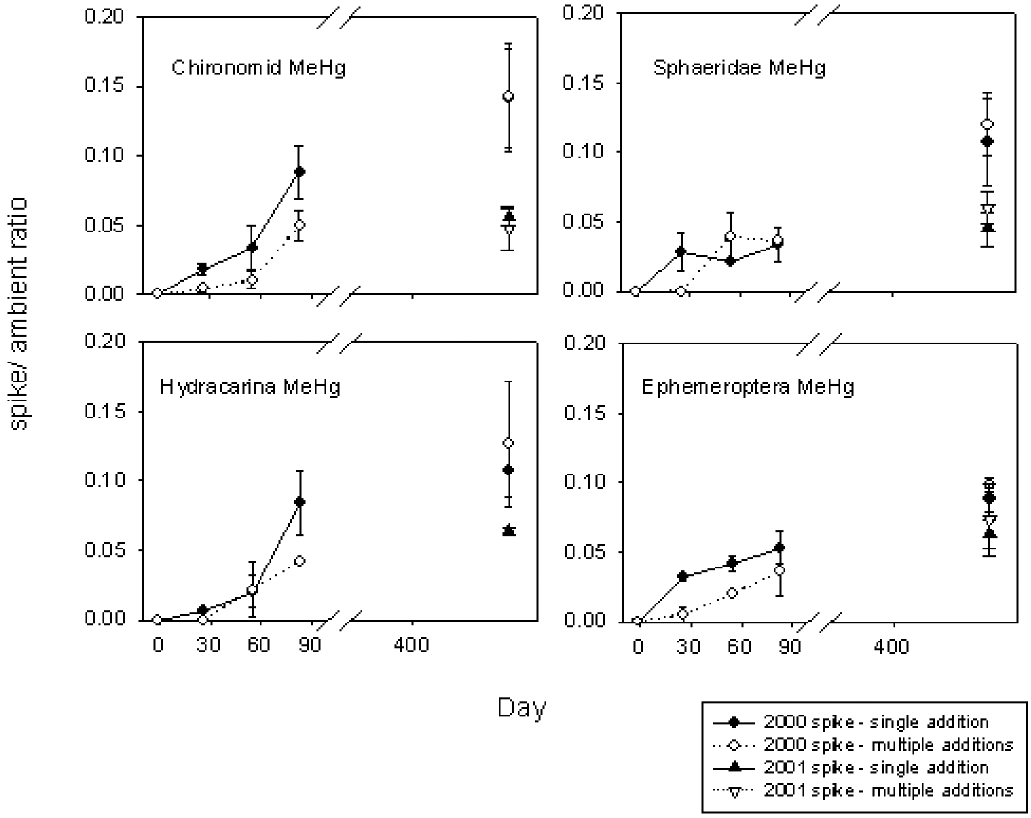

Figure 3. Response in MeHg in invertebrate biota in Lake 239 enclosures at the Experimental Lakes Area, northwestern Ontario, Canada, after additions of stable Hg isotopes in enclosures. Response is given as the ratio of spike (enriched stable isotopes) to ambient Hg (157).

Stable isotope tracer and loading experiments. The Mercury Experiment to Assess Atmospheric Loading in Canada and the US (METAALICUS) is a whole-ecosystem, Hg-loading study being conducted in a boreal, oligotrophic lake and its watershed at the Experimental Lakes Area in northwest Ontario, Canada (154, 155). The lake, upland, and wetland of the Lake 658 ecosystem are receiving Hg enriched with different stable Hg isotopes to determine the relative importance of each as sources of Hg to aquatic biota. Annual Hg additions of approximately 22 μg m^{-2} yr^{-1} (5-fold greater than wet deposition) began in 2001 and are ongoing (through 2006).

Mercury applied to the upland and wetland catchments was strongly retained in the watershed and not observed in the lake in appreciable amounts after 3 y of treatment (136). Results to date indicate that the overall Hg load to the lake increased by nearly a factor of two due largely to the Hg isotope added directly to the lake surface ("lake spike"). This lake spike Hg began to be methylated within weeks after the addition of Hg and was quickly accumulated in biota (156). Throughout the study, the fraction of lake spike observed as MeHg in both sediment and water has been higher than for ambient Hg, although the differences have declined over the years since spikes began. This pattern demonstrates a decline in the availability of Hg deposition for methylation with time in lakes (155).

Increases in the amount of MeHg in water and the food chain may eventually match the percent increase in Hg load to the lake, but this had not occurred within 3 y. After the second year of additions, MeHg concentrations in zooplankton, benthic invertebrates, and small fish were more than 25% higher than would have occurred in the absence of experimentally added Hg (156). By the third year of the study, MeHg concentrations in zooplankton, benthic invertebrates, and small fish were approximately 30 to 40% higher than would have occurred in the absence of experimentally added Hg. METAALICUS clearly illustrates that biota in aquatic ecosystems respond to changes in Hg load at rates comparable to atmospheric deposition presently experienced in some regions and that different components of the ecosystem respond at different rates (96). The implication of this work is that the rate of response to changes in Hg deposition depends on ecosystem-specific characteristics.

Lake enclosure Hg addition studies at the Experimental Lakes Area. A number of mesocosm-scale studies associated with METAALICUS were also conducted to examine the timing and magnitude of responses to Hg loading. In the first mesocosm studies, Hg was added at a single loading rate of 30 μg m^{-2} (157). In another lake enclosure study, the MESOSIM project, the dose response was examined over a range of Hg loadings (95), something that could not be done at a single loading rate and single site in the whole watershed study. Stable isotopes of Hg were added to 11 enclosures (10-m diameter) at 1 to 15x ambient wet deposition (7.1–106.5 μg m$^{-;2}$ yr^{-1}). In both mesocosm studies, newly added Hg was rapidly (i.e., weeks) detectable as MeHg in sediments and biota (Fig. 3). Importantly, isotopically labeled Hg and MeHg concentrations in sediments, water, and biota were all linearly related to loading, across concentrations representative of range of atmospheric Hg deposition (Fig. 4). These results suggest that changes in Hg loading affect MeHg concentrations in fish and other biota and that the response is linear and proportional at least in the early stages (95, 157).

Wetland Hg addition studies at the Experimental Lakes Area. A small-scale Hg loading experiment to an Experimental Lakes Area wetland also demonstrated the link between Hg deposition, methylation, and transport to surface waters (158). Over 3 mo, added isotopic Hg migrated below the water table and transported several meters horizontally to the lake margin. At one location, 6% of aqueous isotopic Hg was detected as MeHg 1 d after application. These results indicated that new inorganic Hg in atmospheric deposition could be readily methylated and transported towards the lake by shallow groundwater flow, confirming the importance of wetlands in the supply of Hg to surface waters.

Small-scale upland plot Hg addition experiment. A boreal upland subcatchment was also amended with a stable isotope of

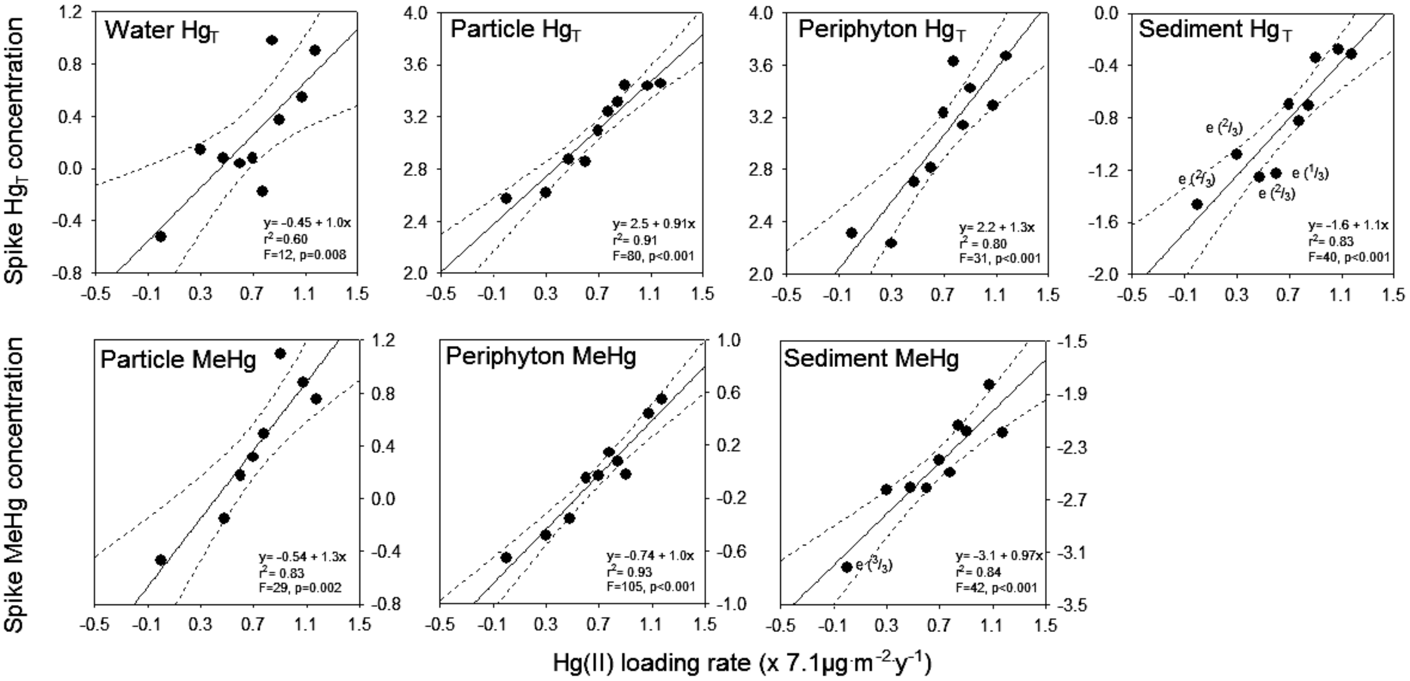

Figure 4. Dose-response relationships for the MESOSIM study, an enriched stable-isotope Hg loading study in lake enclosures at the Experimental Lakes Area, Ontario. Relationship between Hg(II) loading rate and spike HgT (top panels) or spike MeHg (bottom panels) concentration of water (ng L^{-1}), particles (ng g C^{-1}), periphyton (ng g C^{-1}), and sediments (ng g^{-1} dry weight). All variables are on a log10 scale. The regression lines are shown with 95% confidence intervals. The samples were collected approximately 2 mo after dosing. The symbol "e" indicates that one or more samples was estimated (in brackets is the number of samples with estimated concentrations/total number of samples). Reprinted with permission from (95), Copyright 2006 American Chemical Society.

Hg (135). The new Hg was found to be more reactive than old Hg for both volatilization to the atmosphere and methylation in soils. However, the yield of new Hg from the catchment was very low, less than 1% in the first year. The fraction of new Hg that was bound to vegetation was much greater than for old Hg, suggesting that the canopy delays the delivery of some atmospheric deposition to soils until after needle and leaf fall occurs.

Everglades studies. The Florida Everglades is a large, subtropical, predominantly freshwater wetland with strong spatial gradients in trophic condition, biogeochemistry, and net methylmercury production rates (24, 26, 159). Hg stable isotopes were added to small enclosures corresponding to 0.5, 1, and 2x the average annual wet deposition of Hg to the Everglades (160). At four sites, chosen to represent a wide range of water chemistry and productivity, Me^{200}Hg concentrations in surface sediments, water, and fish responded linearly to Hg dose. However, the rate of the response varied significantly among the sites (Fig. 5). The results demonstrate both the response of ecosystems to changing Hg load and the variability in the magnitude of that response among ecosystems of different chemical characteristics.

The Gårdsjön Roof Study

To determine the relationship between reduced atmospheric deposition and runoff of Hg from a subcatchment, an experimentally covered catchment and a reference catchment in Sweden were monitored for total Hg and MeHg yield (16). The roof limited the atmospheric wet deposition of total Hg and MeHg (but not litterfall) to the catchment. There was no statistically significant change in the output of total Hg or MeHg as a result of the roof treatment after 10 y. In the catchments around the Lake Gårdsjön, stream Hg export (2–3 g km^{-2} yr^{-1}) and wet plus dry deposition (30–40 g km^{-2} yr^{-1}) are only small fractions in comparison to the total soil pool (10–20 kg km^{-2}). Clearly, factors other than deposition (e.g., decomposition of organic matter in soils) appear to be controlling runoff over the short term (i.e., years).

From these experiments we can conclude:

- All aquatic ecosystems that have been subjected to direct experimental additions of Hg to the surface waters begin to respond rapidly, with MeHg increases in the food web, including fish.
- Increased Hg loading directly to aquatic ecosystems at rates relevant to changes in atmospheric deposition results in increased uptake of labeled Hg in fish that is initially proportional to loading rates (i.e., experiments were not run long enough to observe steady-state responses).
- Terrestrial compartments are slow to respond to changes in Hg loading, with this response governed by the size and biogeochemistry of the soil pool and watershed hydrology. The implications of this response for the recovery of Hg-contaminated fisheries are dependent on the characteristics of the watershed. No studies have been conducted for a long enough period to understand soil Hg dynamics.

SYNTHESIS AND FINAL CONCLUSIONS

This synthesis clearly shows that fish MeHg concentrations respond to changes in Hg load. The magnitude and the timing of the response will vary depending on case-specific factors, including the timing and extent of loading, chemical form of the added Hg, and ecosystem characteristics (e.g., the efficiency of delivery of Hg load to zones of methylation, controls on net methylation, availability of MeHg to the food web, trophic transfer of MeHg), and the role of the terrestrial system in delivering atmospheric deposition to receiving waters. The main body of evidence comes from cases in which the Hg loading originates from point sources. For responses to changes in atmospheric loading, evidence is limited to a few cases that

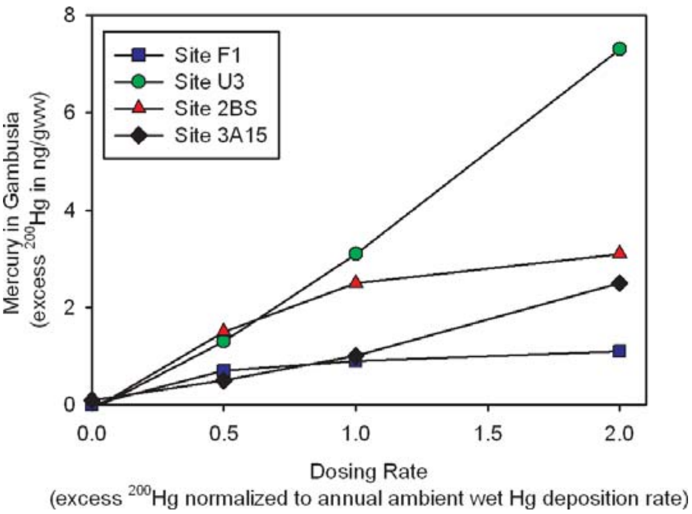

Figure 5. Response of MeHg in *Gambusia* 40 days after the addition of enriched stable Hg isotopes to the water column of 0.7 m diameter enclosures placed at four sites in the Everglades. Dosing rates are normalized to an average annual wet deposition rate of 22 μg m^{-2} y^{-1}. The sites cross a wide range of sulphur and dissolved organic matter concentrations (160).

currently limit the possibilities to draw firm conclusions. This is partly due to a lack of long-term, consistent monitoring data (Hg loading and fish Hg) and partly an expected consequence of the complex biogeochemistry of Hg, with multiple chemical, physical, and chemical factors besides Hg load that control the MeHg levels in fish. Based on the synthesis presented here, the following conclusions can be drawn:

- Numerous examples exist in freshwater and coastal marine systems where cessation of point source Hg loading have resulted in lowered Hg levels in biota, mainly from ecosystems contaminated by industrial releases.
- For freshwater ecosystems affected by atmospheric Hg deposition, a growing body of temporal and spatial data are generally consistent with a relationship between Hg deposition and MeHg in fish. Some evidence of the relationship between Hg loads and MeHg in fish exists for coastal regions. However, data for the open ocean areas are far from sufficient.
- In most ecosystems, large pools of Hg are present in soils and sediments. Much of this Hg may not be readily available for methylation. Additions of relatively small amounts of Hg that can reach sites of methylation in bioavailable forms can lead to increased MeHg production and bioaccumulation.
- All aquatic systems that have been subjected to experimental increases in direct Hg loading have begun to respond rapidly and significantly to increased Hg, as reflected in increased MeHg levels in the food web, including fish.
- Experimental studies suggest that the magnitude of fish responses to changes in direct Hg loading (within the range relevant to atmospheric Hg loadings) is initially proportional. However, experimental dose-response studies have not been conducted for long enough periods to assess the steady state conditions in response to changes in loading.
- Terrestrial ecosystems are slow to respond to increases and decreases in loading rates. Hg that has accumulated in soils from previous atmospheric Hg deposition is a source that may mask or delay the recovery of fish Hg levels when emissions of Hg decline. Mobilization of Hg from soils as well as MeHg production can be enhanced by land-use changes, such as agriculture, forestry, and reservoirs.

In summary, existing field and experimental evidence clearly indicates that Hg loading rates affect fish Hg concentrations. The magnitude and timing of the dose-to-response relationship are affected by the form of Hg loaded and by a wide range of environmental conditions. Current information suggests a very wide range of ecosystem response times to changes in Hg loading, ranging from months to decades or longer. Current knowledge is also consistent with a long-term proportional response to changes in atmospheric Hg deposition but not for cases of extreme point-source contamination. We use "consistent" rather than "clearly demonstrates" because existing data are not sufficient to draw firm conclusions regarding the timing and magnitude of the response to changing Hg loads, particularly for the oceans. The many factors affecting Hg cycling and bioaccumulation can mask the influence of atmospheric Hg deposition across a range of environmental settings, highlighting the need for carefully designed national and international monitoring programs (132), currently lacking, to detect trends for Hg levels in the environment and the response to changes in Hg emissions.

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