Effect of Loading Rate on the Fate of Mercury in Littoral Mesocosms

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The effects of changes in atmospheric mercury (Hg) deposition on aquatic ecosystems are poorly understood. In this study, we examined the biogeochemical cycling of Hg in littoral mesocosms receiving different loading rates (7–107 μg Hg m⁻² year⁻¹). We added a ²⁰²Hg-enriched preparation to differentiate the experimentally added Hg from the ambient Hg in the environment. This approach allowed us to follow the distribution and methylation of the isotopically enriched (“spike”) Hg in the mesocosms. Within 3 weeks, spike Hg was distributed throughout the main environmental compartments (water, particles, periphyton, and sediments) and began to be converted to methylmercury (MeHg). Concentrations of spike total Hg and MeHg in these compartments, measured after 8 weeks, were directly proportional to loading rates. Thus, Hg(II) availability was the limiting factor for the major processes of the biogeochemical Hg cycle, including methylation. This is the first study to demonstrate a proportional response of in situ MeHg production to atmospherically relevant loading levels. On the basis of mass balances, we conclude that loading rate had no effect on the relative distribution of spike Hg among the main compartments or on the fraction of spike Hg converted to MeHg. Therefore, loading rate did not change the relative magnitude of biogeochemical pathways competing for Hg within the mesocosms. These data suggest that reductions of Hg deposition to lake surfaces would be equally effective across a broad range of deposition rates.

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Introduction

Mercury (Hg) is a naturally occurring element in the Earth’s crust and mantle, but anthropogenic activities have released large stores of this metal to the atmosphere (1). Long-range transport of atmospheric Hg has caused widespread contamination of aquatic ecosystems (2, 3). Methylmercury (MeHg), a potent neurotoxin, can accumulate in wild fish to levels that adversely affect humans and wildlife (4). Some countries have issued or proposed reductions in anthropogenic Hg emissions, but the effects of these reductions are insufficiently understood. The link between atmospheric Hg deposition and MeHg concentrations in fish remains a knowledge gap in predicting the effects of Hg emission reductions (5).

Atmospheric Hg is predominately deposited to aquatic ecosystems as inorganic mercury (Hg(II)). Several biogeochemical pathways compete for Hg(II) in aquatic ecosystems, predominately reduction and volatilization (6–8), sedimentation (9, 10), and methylation (11, 12). Several pathways also compete for MeHg once formed, including photochemical and biological demethylation (13, 14), burial (15), and bioaccumulation (16). The timing and relative magnitude of these pathways determine when and to what extent atmospheric Hg is ultimately available to food webs. It is unclear whether changes in Hg deposition will affect the relative magnitude of biogeochemical pathways competing for Hg in aquatic ecosystems.

In the Mercury Experiment To Assess Atmospheric Loading in Canada and the United States (METAALICUS), isotopically enriched Hg(II) is being applied to natural ecosystems, for the first time, to study the fate of newly deposited Hg. As part of this initiative, we conducted a dose–response experiment called MESOSIM (MESOcosm SIMulations of atmospheric Hg deposition to aquatic ecosystems). In this study, we added isotopically enriched Hg(II) to lake mesocosms at different loading rates. This experimental design simulated a broad range of atmospheric deposition rates within a single lake, which was not possible in the whole-ecosystem loading experiment of METAALICUS. Manipulating atmospheric deposition rates in mesocosms within a single lake minimized the influence of confounding environmental variables, thereby allowing us to examine the effect of Hg(II) loading. Our main objective was to examine the relationship between Hg(II) loading directly to a lake surface and MeHg bioaccumulation by fish. We also examined the component processes of the biogeochemical cycle to determine the response of the environment to increased Hg(II) loading.

In this paper, we examine the biogeochemical cycling of Hg in mesocosms receiving different loading rates. We present concentrations of isotopically enriched (“spike”) Hg in the main environmental compartments (water, particles, periphyton, and sediments) over one season of Hg(II) additions. Subsequent papers will describe bioaccumulation of MeHg by biota and compare spike Hg to other Hg in the mesocosms. Here, we first describe how quickly spike Hg was distributed throughout the environment and transformed to MeHg. We then examine the effects of loading rate on the distribution and methylation of spike Hg in the mesocosms. We test the hypothesis that spike Hg concentrations in the main environmental compartments were directly proportional to loading rates. We also construct mass balances to illustrate how loading rate affected the relative strengths of pathways competing for spike Hg in the mesocosms.
### Experimental Section

**Description of Mesocosms.** Eleven littoral mesocosms were installed in Lake 240 at the Experimental Lakes Area (ELA), Ontario, Canada (Figure S1, Supporting Information). Lake 240 (49°40’ N, 93°44’ W) is a remote, sixth-order lake surrounded by boreal forest. Each 10 m diameter mesocosm was suspended from a floating ring and sealed to the sediments with sandbags. The walls of the mesocosms were constructed from woven, poly laminated plastic. The average water depth in the mesocosms was 2 m. Mesocosm volumes were determined from a single addition of \(^{3}\text{H}2\text{O}\) and ranged from 130 to 147 m\(^{3}\) (Table S1, Supporting Information). On the basis of \(^{3}\text{H}2\text{O}\) concentrations over a 10 week period, water leakage was estimated to be minimal (<10%) (17). The mesocosms contained sandy, organic-poor sediments (loss on ignition, ≤3%) and oligotrophic, circumneutral water with comparatively high concentrations of dissolved organic carbon (Table S1, Supporting Information). Mesocosms differed somewhat in their water chemistry, but concentrations of standard chemical parameters did not deviate substantially from the surrounding lake and were not correlated with loading rates. Surface water temperatures ranged from 17.2 to 28.0 °C, and dissolved oxygen concentrations measured throughout the water column ranged from 7.3 to 10.6 mg L\(^{-1}\) (17).

Before the experiment began, total Hg (Hg\(_{T}\)) in filtered water (<0.7 μm) and suspended particles (>0.7 μm) ranged from 2 to 4 ng L\(^{-1}\) and from 0.05 to 0.6 ng L\(^{-1}\), respectively, while MeHg concentrations in filtered water were generally below the detection limit of 0.02 ng L\(^{-1}\). Hg concentrations in surface (0–2 cm) sediments were relatively low in comparison to those of other lakes, with concentrations ranging from 4 to 7 ng g\(^{-1}\) dry weight (dw) for Hg\(_{T}\) and from 0.03 to 0.1 ng g\(^{-1}\) dw for MeHg. Instantaneous methylation rates in sediments were measured at a reference site in Lake 240 by injecting enriched stable isotopes into intact sediment cores (18, 19). Methylation rates for the top 0–2 cm averaged 7.3 to 10.6 mg L\(^{-1}\) from 17% and 30% in sediments and biota, respectively, indicating this was a loading, not a tracer, experiment. Ambient Hg\(_{T}\) and MeHg concentrations in the mesocosms at the end of the experiment were similar to pre-experiment values (Table S1, Supporting Information).

**Mercury Additions.** To simulate atmospheric deposition, 10 mesocosms received multiple additions of isotopically enriched Hg(II). With a regression-based design, each mesocosm was randomly assigned a different loading rate: 1×, 2×, 3×, 4×, 5×, 6×, 7×, 8×, 12×, or 15× the wet deposition rate at the ELA in 1998–1999 (7.1 μg Hg m\(^{-2}\) year\(^{-1}\) (20)). Wet deposition rates were somewhat lower during the period of this study (21). The total mass of Hg added to each mesocosm is provided in Table 1.

Beginning on June 26, 2002, each mesocosm received eight equal weekly additions of Hg(II). This is appropriate because the majority of atmospheric Hg deposition at the ELA occurs during the open water season (21). Mercuric chloride (90.9% Hg; Trace Sciences International, Richmond Hill, Canada) was dissolved in 5% HNO\(_{3}\) to make a stock solution. Approximately 12 h before addition, the appropriate volume of stock solution for each mesocosm was added to 500 mL of Lake 240 water. At dusk, this solution was released below the water surface and mixed into the water column with a trolling motor. A control mesocosm (0×) received no experimental Hg(II) additions.

**Sample Collection.** Water, particles, periphyton, and surface sediments were sampled from all 11 mesocosms between June and September 2002. Samples were collected more frequently in three “intensive” mesocosms (2×, 5×, and 12×). Mercury-clean techniques were followed to minimize contamination of samples (22).

**Water and Particles.** At the center of each mesocosm, water was pumped from a depth of 1 m and filtered through a precleaned quartz microfiber filter (Whatman QM-A, nominal 2.2 μm pore size). Filtered water was collected in precleaned 250 mL glass bottles. Usually, 1250 mL of water was passed through a filter. Each water sample was preserved with 1 mL of concentrated HCl (≤0.0008 ng Hg mL\(^{-1}\)), and filters were kept frozen until analysis. Unfiltered water was collected for standard chemical analyses. Hereafter, “water” and “particles” refer to the filtered (<0.7 μm) and particulate (>0.7 μm) phases, respectively.

**Periphyton.** This term is used here to represent the algal mat growth on the walls of the mesocosms. At the start of the experiment, strips of wall material (0.1 m × 0.9 m) were hung vertically in each mesocosm from untreated wood floats. Strips were naturally colonized with periphyton. Periphyton films removed from each strip were diluted with lake water, homogenized, and filtered through a precleaned quartz microfilter (Whatman QM-A, nominal 2.2 μm pore size). Filters were stored frozen until analysis. Filters were also assayed for particulate carbon.

**Sediments.** At each sampling event, sediments were sampled from three random locations in each mesocosm. Intact cores were manually collected in clear polycarbonate tubes (4.8 cm diameter) and transported on ice and in darkness. The 0–2 cm layer was sectioned from each core and stored frozen.
until analysis. Bulk density, wet-to-dry weight ratios, and loss on ignition were measured separately for each section.

**Mercury Analyses.** All samples were analyzed by a high-resolution, quadrupole ICP-MS coupled to a plasma torch. The relationship between two variables is directly proportional; i.e., a percentage change in the independent variable results in the same percentage change in the dependent variable. An F-test was used to determine whether there was a significant relationship between Hg(II) loading rates and spike Hg concentrations. Two-tailed t-tests were then used to determine whether the slopes of the regression lines were significantly different from 1. All analyses were performed with STATISTICA 6.1 (StatSoft, Inc.).

A mass balance of spike HgT was calculated for each intensive mesocosm through time and for all mesocosms after 8 weeks. Each mass balance contained five components: (i) Hg in the filtered phase of the water column, (ii) Hg on particles in the water column, (iii) Hg in periphyton on the mesocosm walls, (iv) Hg in surface (0–2 cm) sediments, and (v) Hg evaded to the atmosphere. The mass in biota was small and did not contribute meaningfully to the overall budget and, consequently, was not included. Hg evasion was only determined for the intensive mesocosms and was calculated with a thin boundary layer model. Dissolved gaseous Hg concentrations and flux calculations are detailed in ref 25. The masses of spike MeHg in particles, periphyton, and sediments were calculated for all mesocosms after 8 weeks.

**Results**

**Mercury Concentrations.**

**Temporal Dynamics.** Spike HgT concentrations in water usually increased immediately after each Hg(II) addition and then decreased over the following week (Figure 1; solid circles). Concentrations of spike HgT on particles exhibited a similar saw-tooth pattern over time (Figure 1; open circles). Spike HgT concentrations in both water and particles declined substantially after the last Hg(II) addition. Spike HgT was detected in periphyton collected after 3 (July 17) and 8 (August 18) weeks, but the change in concentration over time was not consistent among mesocosms (Figure 2; left panels, white bars). Spike HgT was not detected in sediments collected during the first week, but increasing concentrations were observed after 3 and 8 weeks (Figure 2; right panels, white bars).

MeHg was formed from the Hg(II) additions within 3 weeks of the first addition. Concentrations of spike MeHg in water were low (<8 pg L−1), often below detection limits, and accounted for <0.5% of spike HgT. After 8 weeks, spike MeHg concentrations on particles ranged from <0.02 to 11 pg L−1 and accounted for an average of 0.2% (range, <0.01–0.5%) of spike HgT. Periphyton and sediments accumulated spike MeHg between July and August (Figure 2; hatched bars). On the latter date, spike MeHg accounted for an average of 0.1% (range, 0.03–0.2%) of spike HgT in periphyton and 3% (range, 0.2–11%) of spike HgT in sediments. After 8 weeks, spike MeHg and spike HgT concentrations were strongly correlated with each other across all mesocosms in particles, periphyton, and sediments (Figure S2, Supporting Information).

**Effect of Loading Rate.** Loading rate had a significant effect on spike HgT concentrations measured after 8 weeks (Figure 3; left panels). More than 80% of the variation in spike HgT concentrations of particles, periphyton, and sediments among mesocosms was explained by loading rate. This relationship was somewhat weaker for water, but loading rate still explained the majority of the variation among mesocosms. The slopes of these log–log relationships were not significantly different from 1 (water; t = 0.05, p = 0.96; particles, t = −0.89, p = 0.40; periphyton, t = 1.2, p = 0.26; sediments, t = 0.54, p = 0.60). Therefore, spike HgT concentrations in environmental compartments were directly proportional to Hg(II) loading rates. These relationships held true regardless of whether the estimated values (i.e., samples with concentrations below detection limits) were included.

Similarly, spike MeHg concentrations of particles, periphyton, and sediments measured after 8 weeks were significantly related to loading rate (Figure 3; right panels). This relationship could not be assessed for spike MeHg in particles.
water because most values were below detection limits. Loading rate explained between 83% and 93% of the variation in spike MeHg concentrations of particles, periphyton, and sediments among mesocosms. The slopes of these log-log relationships were not significantly different from 1 (particles, t = 1.2, p = 0.27; periphyton, t = 0.28, p = 0.78; sediments, t = −0.18, p = 0.86). Therefore, like spike HgT concentrations, spike MeHg concentrations in environmental compartments were also directly proportional to Hg(II) loading rates.

**Mercury Mass Balances.**

**Temporal Dynamics.** Most of the spike HgT mass was in water at the beginning of the experiment but was efficiently removed from this compartment over time (Figure 4; solid circles). Spike HgT associated with particles accounted for a maximum of 16% of the amount added and also decreased over time (Figure 4; open circles). Only a small percentage (<2%) was associated with periphyton on the mesocosm walls (Figure 4; solid squares), and between 8 and 16% was associated with surface sediments (Figure 4; open squares). Hg evasion was an important loss mechanism throughout the experiment (Figure 4; open triangles). After 8 weeks, the largest sink for spike HgT was the atmosphere, followed by water and sediments (Table 1). After 3 weeks (July 16–17), we accounted for 73%, 92%, and 87% of the Hg added to mesocosms 2×, 5×, and 12×, respectively. After 8 weeks (August 18–20), we accounted for 67%, 81%, and 59% of the Hg added to mesocosms 2×, 5×, and 12×, respectively. We estimate that the error associated with these totals is approximately 30%, based on the RSDs of replicate samples (see Experimental Section) and assuming an error of 40% for the evasion term. The relative sources of error in the mass balance were: evasion > sediments > water > particles > periphyton. By the end of the experiment, less than 1% of the Hg added to each mesocosm was methylated, and the largest pool of spike MeHg was in surface sediments (Table 1).

**Discussion**

We simulated changes in atmospheric Hg deposition by adding isotopically enriched Hg(II) to large mesocosms in a boreal lake. The use of isotopically enriched Hg(II) allowed
us to differentiate the experimentally added Hg ("spike Hg") from the ambient Hg in the mesocosms. We observed a rapid decline in spike HgT concentrations in water after each Hg(II) addition, with a half-time of approximately 11 days. Similar loss rates have been reported by studies in which a single pulse of radioactive Hg was added to enclosures (26, 27) or a whole lake (28). In our experiment, evasion was the major loss mechanism of spike Hg from water. Over 8 weeks, 38%, 59%, and 33% of the spike Hg added to mesocosms 2×, 5×, and 12×, respectively, were lost to the atmosphere (Table 1, ref 25). Other loss mechanisms included partitioning to particles and sediments and, to a lesser extent, to periphyton. Spike Hg partitioned to main environmental compartments within days to weeks, which is also in agreement with previous studies (26, 29).

Within 3 weeks, we observed production of MeHg from the Hg(II) added to the mesocosms. This is particularly important because, although instantaneous methylation rates have been measured in water or sediment samples (12, 30), the time required for Hg in aquatic ecosystems to reach sites of methylation and be converted to MeHg has seldom been studied. The timing of methylation can also be deduced from studies in the English-Wabigoon River system, in which 203Hg added to enclosures was detected as Me203Hg in fish within 1–2 months (31, 32). Some of the MeHg produced in our experiment was also incorporated into the food web, which will be presented in a subsequent paper.

We can infer that surface sediments were the primary site of de novo MeHg production in the mesocosms. Although periphyton communities can be important contributors to MeHg production in some ecosystems (33), they did not seem to be important contributors here. Surface sediments contained the largest pool of spike MeHg, which was 2 orders of magnitude more than the pool in periphyton (Table 1). Further, the percentage of spike HgT as MeHg in sediments (3%, on average) was higher than the percentage in any other environmental compartment. The low concentrations of MeHg in the water column suggest that water phase methylation was likely not important in the mesocosms, as expected, considering the availability of oxygen throughout the water column. Less than 1% of the Hg(II) added to the mesocosms was methylated, presumably because only a small fraction of spike Hg was delivered to sites of methylation. The fraction of spike Hg delivered to sediments and the subsequent amount methylated in this experiment were similar to those observed in Lake 658 (the site of the whole-

![FIGURE 2. Temporal changes in spike HgT (white bars; left axes) and spike MeHg (hatched bars; right axes) concentrations in periphyton (left panels) and sediments (right panels), shown for each intensive mesocosm. Each bar is the value of one periphyton sample (ng Hg (g C)−1) or the mean (± standard error) of three sediment samples (pg Hg g−1 dry weight). The symbol “e” indicates that the concentration of one or more samples was estimated (see text for details). The number of samples with estimated concentrations/total number of samples is in brackets. Y-axes are scaled as in Figure 1.](image-url)
ecosystem METAALICUS experiment) during the first summer of Hg(II) additions.

Most importantly, we found that the relationship between Hg(II) loading rate and concentrations of spike Hg_T and MeHg in the environment was highly significant and linear (Figure 3). Furthermore, all relationships were directly proportional within the loading rates examined in this experiment, which represent a wide range of atmospheric Hg deposition. For example, the spike Hg concentration in a mesocosm receiving a particular loading rate was 2 times higher than the concentration in a mesocosm receiving half that loading rate. We conclude that Hg(II) availability was the limiting factor for the main components of the biogeochemical Hg cycle, including methylation.

Only a few previous studies have examined the dose–response of MeHg production to Hg(II) loading experimentally, and none has examined the full suite of biogeochemical processes. Observational studies have sometimes shown non-linear relationships between Hg_T and MeHg concentrations, potentially representing saturation of some component of the methylation process, but these occurred at Hg(II) loadings well above the range observed for atmospheric deposition. For example, Benoit et al. (30) examined the relationship between Hg_T and MeHg in surface sediments across various
ecosystems; MeHg and Hg\textsubscript{T} concentrations were correlated at concentrations below 0.5 \(\mu\)g Hg g\textsuperscript{-1}, which are typical of ecosystems with no point sources of Hg pollution, but at more contaminated sites, increasing concentrations of Hg\textsubscript{T} had little impact on MeHg production. Nonetheless, a number of studies in mildly to severely contaminated ecosystems have shown linear relationships between Hg\textsubscript{T} and MeHg concentrations. For example, in the highly contaminated English–Wabigoon River system, MeHg and Hg\textsubscript{T} concentrations in water were found to be highly correlated (35). Additions of \textsuperscript{203}Hg(II) to sediment samples from this river system increased MeHg production up to, but not above, \(\sim 5 \mu\)g g\textsuperscript{-1} (26). In four Arctic lakes, gross methylation potentials of sediments were correlated with porewater Hg\textsubscript{(II)} concentrations (36). Further, net rates of MeHg production in these lakes were positively related to atmospheric Hg deposition, as inferred from sediment Hg loadings. The experimental evidence from MESOSIM and prior observational studies, taken together, suggest that MeHg production responds linearly to Hg\textsubscript{(II)} loading, except under extreme levels of contamination. It is important to acknowledge that factors other than Hg\textsubscript{(II)} availability can influence the biogeochemical cycling of Hg in aquatic ecosystems, and consequently, a linear response in MeHg production to changes in Hg\textsubscript{(II)} availability will only occur if other factors remain constant.

On the basis of mass balances constructed after 8 weeks, loading rate had no effect on the relative distribution of spike Hg among environmental compartments or on the relative amount of spike Hg converted to MeHg. Hence, we found no evidence that loading rate affected the relative strength of pathways competing for Hg in the mesocosms. This is the first study to examine this question at this scale. The mass balances could not fully account for the total mass of Hg added to the mesocosms (Table 1). We did not include spike Hg accumulated in biota or adsorbed to the mesocosm walls in the mass balance because these components accounted for \(\ll 1\%\) of the amount added. The amount of spike Hg lost by leakage and transported to deep sediments was also relatively unimportant to the mass balance. Therefore, a component of the mass balance was likely underestimated, probably evasion because it was the most uncertain estimate. Although we could not completely close the mass balances, our conclusions as to the relative distribution and methylation of Hg under different loading rates remain unaffected.

Before the implications of these findings on proposed changes in Hg reductions are discussed, several issues need to be considered. The first is the ability of MESOSIM to simulate atmospheric Hg deposition. We added Hg\textsubscript{(II)} to the mesocosms in multiple small doses to simulate a realistic frequency of rain events. We also added Hg\textsubscript{(II)} during the

*FIGURE 4. Temporal changes in the mass of spike Hg\textsubscript{T} (as a percentage of mercury added to date) in the different compartments of each intensive mesocosm. Evasion data are from ref 25. Vertical lines are as in Figure 1.*
summer, which is when most Hg deposition occurs at the ELA (21). The Hg(II) added to the mesocosms was bound to relatively weak ligands. In remote areas, most of the Hg in wet deposition is reactive (to SnCl2) (37, 38), suggesting that it too is likely complexed to relatively weak ligands. A second issue is the ability to translate results from littoral mesocosms to whole lakes. This study captured many, but not all, of the important biogeochemical processes in natural lakes. In particular, mesocosms were unstratified and isolated from watershed inputs. Therefore, our findings are most applicable to shallow, unstratified lakes and the epilimnion of stratified lakes and are more relevant to seepage lakes than drainage lakes. Third, a steady state was not reached within the time frame of the experiment, and therefore, our results represent an initial rather than a long-term response to changes in loading. Longer-term responses and a more complex landscape are being examined in the whole-ecosystem METAALICUS experiment.

On the basis of this study, we suggest that Hg(II) deposited by precipitation to lake surfaces is quickly dispersed in the environment and acted on by methylation. Further, the fate of Hg deposited to lake surfaces, including MeHg production, appears to be proportional to dosage throughout the range of observed wet deposition rates. Hence, we predict that changes in deposition will not affect the relative magnitudes of biogeochemical pathways competing for Hg in littoral environments, at least not within the time frame and range in loading rates examined here. The importance of this becomes more apparent if the alternative is considered. Hypothetically, if MeHg production were saturated at high deposition rates, then a greater proportion of newly deposited Hg would be methylated if deposition were decreased. Consequently, Hg emission reductions would be less effective than expected. In contrast, our findings suggest that the same proportion of newly deposited Hg would be methylated if deposition were decreased. Consequently, Hg emission reductions should be equally effective across a broad range of deposition rates.

Acknowledgments

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Supporting Information Available

Water chemistry and sediment properties, ambient Hg, and MeHg concentrations, map of Lake 240, and correlations between spike Hg- and spike MeHg concentrations. This material is available free of charge via the Internet at http://pubs.acs.org.

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