# The chemistry and transport of mercury in a small wetland in the Adirondack region of New York, USA

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**Abstract.** The biogeochemistry of Hg was evaluated in a small wetland in the Adirondack region of New York. Concentrations of total Hg (Hg<sub>T</sub>) in streamwater draining the wetland showed little temporal variation. The annual areal watershed flux of Hg<sub>T</sub> (2.2  $\mu$ g/m²-yr) was considerably smaller than regional inputs of atmospheric deposition of Hg<sub>T</sub>, indicating that the terrestrial environment is a net sink for atmospheric deposition of Hg<sub>T</sub>. Drainage inputs of Hg<sub>T</sub> were conservatively transported through the beaver impoundment. The annual flux of total methylmercury (CH<sub>3</sub>Hg<sub>T</sub>+) was greater than literature values of atmospheric deposition suggesting that the watershed is a net source of CH<sub>3</sub>Hg<sub>T</sub>+. Stream concentrations of CH<sub>3</sub>Hg<sub>T</sub>+ increased during low-flow summer conditions in a riparian wetland, and particularly at the outlet of the beaver impoundment. Net production of CH<sub>3</sub>Hg<sub>T</sub>+ occurred in the beaver impoundment (0.45  $\mu$ g/m²-yr). Rates of net methylation for the beaver impoundment were comparable to values reported in the literature for wetlands.

### Introduction

Mercury (Hg) is a toxic trace metal and has been linked to human illness and environmental damage. It appears that industrial activities have resulted in substantial emissions of Hg to the atmosphere and long-distance transport (Engstrom et al. 1994; Mason et al. 1995; Hudson et al. 1995). Atmospheric emissions have apparently caused widespread contamination of Hg in aquatic environments, and elevated concentrations in biota (Zillioux et al. 1993). In the U.S., the principal anthropogenic sources of Hg include coal-fired power plants, municipal and medical waste incinerators, and commercial/industrial boilers, which together contribute nearly 98% of the total anthropogenic emissions of Hg to the atmosphere (EPRI 1994).

The mechanisms and rates of the supply of methylmercury  $(CH_3Hg^+)$  to lakes and ultimately to fish is a critical uncertainty in assessments of the effects

of atmospheric deposition of Hg (EPRI 1994; Rudd 1995). Although small amounts of CH<sub>3</sub>Hg<sup>+</sup> enter ecosystems by atmospheric deposition (Fitzgerald et al. 1991; Lee & Iverfeldt 1991; St. Louis et al. 1994), there is increasing evidence that CH<sub>3</sub>Hg<sup>+</sup> supply from wetlands is due to internal methylation of ionic Hg (Hg<sub>T</sub><sup>2+</sup>; St. Louis et al. 1994; Bishop et al. 1995; Krabbenhoft et al. 1995). Concentrations of CH<sub>3</sub>Hg<sup>+</sup> increase in waters draining wetlands (Krabbenhoft et al. 1995; Branfireun et al. 1996). Large increases in CH<sub>3</sub>Hg<sup>+</sup> also occur when upland watersheds are flooded (Hecky et al. 1991; Rudd 1995). Previous studies of lakes in the Adirondack region of New York indicate that wetlands have a critical role in the transport, cycling and bioavailability of Hg (Driscoll et al. 1994, 1995). Concentrations of total Hg (Hg<sub>T</sub>) and CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> in lakewater were strongly related to concentrations of dissolved organic carbon (DOC). Lakewater DOC appeared to be largely controlled by the fraction of this catchment that is wetlands. In addition, organic acids produced in wetlands complex both  $Hg_T^{2+}$  and  $CH_3Hg_T^+$ , decreasing the bioavailablity of these species (Driscoll et al. 1994).

The objective of this study was to evaluate concentrations and fluxes of  $Hg_T$  and  $CH_3Hg_T^+$  in streamwaters draining a small wetland in the Adirondack region of New York.

## Methods and site description

The northern hardwood forest landscape of the northeastern U.S. contains a relatively small area of wetlands (usually < 10% of the landscape), but these wetlands appear to be important in regulating the supply of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> to surface water (St Louis et al. 1994). Wetlands are distributed across the glaciated landscapes of North America in a discontinuous complex mosaic. Two of the most common wetland types are riparian wetlands and wetlands created by beaver impoundments. Riparian wetlands usually occupy the 5–10 m wide zone adjacent to first and second order streams in northern forests and are often dominated by alders (*Alnus* sp.) and herbs. Beavers (*Castor canadensis*) build dams across streams; hence, most of the beaver ponds in northern forests develop over riparian wetlands. Our study site at Pancake-Hall Creek (43°50'N, 74°52'W) was selected because of the presence of a riparian wetland and a beaver impoundment within the drainage basin.

Pancake-Hall Creek is located in the Big Moose drainage basin in the Adirondack region of New York (Figure 1). The morphometry and basin characteristics of Pancake-Hall Creek are similar to other headwater streams in the Adirondacks impacted by beaver (Cirmo & Driscoll 1993). The beaver pond associated with this stream has a major perennial second-order inlet and several ephemeral inlets along its western and northern edges. The pond has

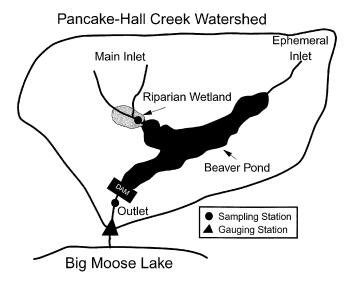


Figure 1. Location of Pancake-Hall Creek Watershed, gauging station and water sampling sites.

a volume of 12,500 m³, a surface area of 30,000 m² and a mean depth of 0.4 m. Hydrologic discharge through the wetland is highly seasonal, with high flow occurring during late fall and spring. Low flow conditions are typically evident during winter and summer. The hydrologic gauging station for this site includes a flume to accommodate relatively high flow conditions and a V-notch weir for low flow conditions. A detailed description of the hydrology and biogeochemistry of Pancake-Hall Creek is given by Cirmo and Driscoll (1993).

The catchment is primarily a second-growth deciduous forest. Major tree species include American beech (*Fagus grandifolia* Ehrh.), yellow birch (*Betula alleghaniensis* Britton), sugar maple (*Acer saccharum* Marsh.), red maple (*Acer rubrum* L.), and red spruce (*Picea rubens* Sarg.). Balsam fir (*Abies balsamea* (L.) Mill.) and hemlock (*Tsuga canadensis* (L.) Carr.) are evident along the shoreline and in saturated zones adjacent to the pond. The shallow area near the inlet to the pond is dominated by sedges (*Carex* sp.) and mosses (*Sphagnum* sp.). The sediments are organic and peaty, and covered with *Sphagnum* in shallow zones.

To investigate temporal patterns in the chemistry of Hg at Pancake-Hall Creek, water samples were collected at the major inlet (riparian wetland) and outlet of the beaver impoundment from October 1993 to April 1995 (Figure 1). Clean techniques were used in all phases of sample collection and handling. Streamwater was collected into 1 L Teflon bottles for Hg analysis, and into 1 L polypropylene bottles for the analysis of major solutes. Bottles were

opened and closed beneath the water surface during sample collection. Each bottle was rinsed three times with water prior to collecting the final sample. The bottles were hermetically sealed and double-bagged immediately after sample collection.

For analysis of Hg<sub>T</sub>, samples were first oxidized with BrCl. After oxidation, NH<sub>2</sub>OH·HCl was used to reduce excess BrCl. Samples were then reduced with SnCl<sub>2</sub>, purged to Au traps, and thermally desorbed as Hg<sup>0</sup> with He carrier gas for detection by cold vapor atomic fluorescence spectrometry (CVAFS). Total CH<sub>3</sub>Hg<sup>+</sup> was processed by first extracting samples from a HCl/KCl matrix into CH<sub>2</sub>Cl<sub>2</sub> followed by back extraction into pure water (by solvent evaporation). Determination of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> was accomplished by aqueous phase ethylation with NaB(C<sub>2</sub>H<sub>5</sub>)<sub>4</sub>, cryogenic gas chromatograph separation and CVAFS detection. Details of the procedure used to fractionate aqueous samples for Hg are provided by Bloom (1989). Water samples collected were also measured for concentrations of major solutes using the procedures summarized in Driscoll and van Dreason (1993). All chemical analyses were conducted at Syracuse University. Solute mass balance calculations for the wetland were calculated from 1 March 1994 to 1 March 1995 using the approach described by Cirmo and Driscoll (1993).

### **Results and discussion**

The time-series of water chemistry at Pancake-Hall Creek showed remarkably constant concentrations of  $Hg_T$  throughout the study period (Figure 2). The mean concentrations ( $\pm$  standard deviation) of  $Hg_T$  was 2.3 ( $\pm$  0.26) ng/L in the inlet stream and 2.5 ( $\pm$  0.22) ng/L in the outlet to the beaver impoundment. This relatively small variation in concentrations is surprising in view of the large changes in discharge that occurred over the annual cycle. Given the strong relationship between concentration of  $Hg_T$  and DOC in Adirondack lakes (Driscoll et al. 1995), we anticipated concentrations of  $Hg_T$  to follow temporal variations in DOC at Pancake-Hall Creek. The inlet stream showed little temporal variation in concentrations of DOC (and  $Hg_T$ ), with the exception of a high concentration in July (7.6 mg C/L). Pond outflow, however, exhibited marked increases in DOC during the summer period, with no corresponding increase in  $Hg_T$ .

In contrast to  $Hg_T$ , concentrations of  $CH_3Hg_T^+$  showed seasonal variations. Concentrations of  $CH_3Hg_T^+$  were relatively low in October when monitoring was initiated, increased during the summer in both the inlet and outlet waters, decreased in the early fall and remained relatively constant through the winter. Concentrations of  $CH_3Hg_T^+$  were consistently higher in the outlet of the beaver impoundment than the inlet stream draining the riparian

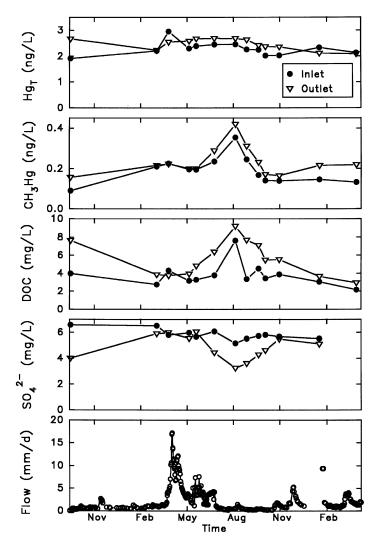


Figure 2. Time-series of concentrations of  $Hg_T$ ,  $CH_3Hg_T^+$ , DOC and  $SO_4^{2-}$ , and discharge for streamwaters draining the riparian wetland (inlet to the beaver impoundment) and the beaver pond at Pancake-Hall Creek.

wetland. Only during the high flow spring period were concentrations of  $CH_3Hg_T^+$  similar in the inlet and outlet waters. The magnitude of the summer increase in  $CH_3Hg_T^+$  was greater in the outflow of the beaver impoundment than the inlet stream. The increase in  $CH_3Hg_T^+$  concentrations in the pond outflow was coincident with a depletion in concentrations of  $SO_4^{2-}$  in the outlet waters during the summer compared to inlet concentrations. We observed

an increase in the percentage of Hg<sub>T</sub> as CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup>: from 6 to 10% during the fall/winter/spring to 16% during summer. These patterns are expected because methylation of Hg is largely mediated by SO<sub>4</sub><sup>2-</sup> reducing bacteria (Gilmour et al. 1992) and the percent of Hg<sub>T</sub> occurring as CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> increases under reducing conditions (Driscoll et al. 1994). The small increase in concentrations of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> in the inlet stream during summer suggests that the riparian wetland may also be an environment for Hg methylation. Subtle decreases in  $SO_4^{2-}$  and increases DOC concentrations in the inlet stream during the low flow summer conditions may be suggestive of reducing conditions in the riparian wetland. In contrast to concentrations of Hg<sub>T</sub>, concentrations of  $CH_3Hg_T^+$  were correlated with concentrations of DOC,  $(CH_3Hg_T^+(ng/L) =$  $0.025*DOC (mg C/L) + 0.10, r^2 = 0.41)$ . Summer increases in concentrations of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> coincided with increases in DOC, particularly for the outflow of the beaver impoundment. It is unclear if the correlation between CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> and DOC is causal (i.e., due to organic complexation of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup>) or due to the increased production of both solutes in the beaver impoundment during summer. However, these observations illustrate the importance of wetlands in regulating Hg<sub>T</sub> and CH<sub>3</sub>Hg<sub>T</sub> fluxes in this ecosystem and ultimately the supply to the downstream lake (i.e., Big Moose Lake), and are consistent with the findings of Bishop et al. (1995) and Krabbenhoft et al. (1995) in Sweden and Wisconsin, respectively.

Annual watershed areal fluxes of  $Hg_T$  for the inflow and outflow from the beaver impoundment were 2.2  $\mu g/m^2$ -yr at both sites. This terrestrial flux of  $Hg_T$  was in the upper part of the range of values reported for remote watersheds (Table 1). The similar fluxes of  $Hg_T$  for drainage from the riparian wetland and the outflow of the beaver impoundment indicate that inputs of  $Hg_T$  were largely transported through the pond. This apparently steady-state transport of  $Hg_T$  may reflect the older age of the impoundment (> 10 yr.). Pond sediments do not appear to be an important source of  $Hg_T$ . Rather, drainage inflow seems to be the major input of  $Hg_T$  to the pond. If these watershed areal fluxes are compared to estimates of atmospheric deposition of  $Hg_T$  from Lake Champlain (9  $\mu g/m^2$ -yr; Burke et al. 1995), it appears that the terrestrial environment is a sink for atmospheric deposition of  $Hg_T$ .

The annual watershed flux of  $CH_3Hg_T^+$  from the upland/riparian wetland was  $0.17 \, g/m^2$ -yr. This value is high in comparison to other watershed studies, and similar to values reported by St. Louis et al. (1994) for wetland drainage in Ontario (Table 1). Unfortunately, measurements of atmospheric deposition of  $CH_3Hg_T^+$  are not available for the study site or the region. Fitzgerald et al. (1991) measured wet deposition of  $CH_3Hg_T^+$  at Wisconsin obtaining a value of  $0.09 \, \mu g/m^2$ -yr. The range of  $CH_3Hg_T^+$  reported for wet/bulk deposition in the literature is 0.04– $0.4 \, \mu g/m^2$ -yr (St. Louis et al. 1995; Hultberg et al.

*Table 1.* Comparison of watershed fluxes of  $Hg_T$  and  $CH_3Hg_T^+$  obtained in this study with values reported in the literature (in  $\mu g/m^2$ -yr).

Location	Hg <sub>T</sub> Flux	CH <sub>3</sub> Hg <sub>T</sub> <sup>+</sup> Flux	Reference
Sweden	0.8-5.9		Johansson et al. (1991)
Southern Sweden	2.3 - 3.5	0.12	Lee and Hultberg (1990)
Northern Sweden	1.2 - 1.8	0.08 – 0.16	Lee et al. (1995)
Wisconsin	0.8	0.06-0.15	Krabbenhoft et al. (1995)
Ontario	0.6 - 2.1		Mierle and Ingram (1991)
Ontario upland	0.3 - 2.3	0.007 – 0.098	St. Louis et al. (1994)
Ontario wetland		0.18-0.55	St. Louis et al. (1994)
New York	2.2	0.17	this study

1994). Hultberg et al. (1994) and St. Louis et al. (1994) have reported that upland watersheds are sinks for atmospheric deposition of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup>. Because atmospheric deposition of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> is well in excess of drainage losses, it seems likely that the upland/riparian wetland portion of the Pancake-Hall Creek Watershed is a net source of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup>. Based on these studies of upland watersheds, the riparian wetland of Pancake-Hall Creek watershed is likely an important area of CH<sub>3</sub>Hg<sub>T</sub> production. The annual watershed areal flux of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> was greater in the outflow (0.20  $\mu$ g/m<sup>2</sup>-yr) than entering the beaver impoundment (0.17  $\mu$ g/m<sup>2</sup>-yr), indicating that net production of  $CH_3Hg_T^+$  occurs within the pond (0.45  $\mu$ g/m<sup>2</sup>-yr based on the pond surface area). Drainage inputs of Hg<sub>T</sub> were probably methylated within the beaver impoundment. Rates of in-pond methylation, DOC production and  $SO_4^{2-}$ reduction were greatest during the summer, coinciding with conditions of maximum temperature, anoxia and hydraulic residence time in the beaver impoundment (Cirmo & Driscoll 1993). The areal rate of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> production for the beaver impoundment is comparable to rates reported for wetlands  $(0.3 \mu \text{g/m}^2\text{-yr})$ , at the low end of the range reported for lakes  $(0.5\text{--}3 \mu \text{g/m}^2\text{--}$ yr) and well below values reported for flooded terrestrial areas (13  $\mu$ g/m<sup>2</sup>-yr; Rudd 1995). A critical question regarding the supply of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> to downstream lakes is whether beaver impoundments function as wetlands or recently flooded terrestrial environments. Based on rates of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> production, it appears that net  $CH_3Hg_T^+$  production in this older beaver impoundment is more comparable to a wetland than a recently flooded terrestrial area. However, over the lifetime of a beaver impoundment it seems likely that rates of net CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> production would be initially high, in response to flooding, and decline with time as the impoundment approaches steady-state with respect to Hg<sub>T</sub> inputs.

Changes in land-use and anthropogenic disturbance (e.g., forest cutting and regrowth, acidification from atmospheric deposition) may alter the transport, fate and bioavailablity of Hg. It seems plausible that changes in the distribution and abundance of wetland area and, in particular, beaver ponds may contribute to the supply of CH<sub>3</sub>Hg<sub>T</sub> to Adirondack drainage lakes. As described above, wetlands are an important landscape position for CH<sub>3</sub>Hg<sup>+</sup> formation. Our biogeochemical studies of beaver impoundments in the Adirondacks (Driscoll et al. 1987a; Cirmo & Driscoll 1993; Cirmo & Driscoll 1996) have shown that these systems greatly enhance the supply of DOC to downstream surface waters. Transport of Hg<sub>T</sub> and CH<sub>3</sub>Hg<sup>+</sup> are strongly linked to transport of DOC (Driscoll et al. 1995). In the northeastern U.S., the distribution and abundance of wetlands has varied through time. Agents such as beaver are responsible for establishing an ontogeny of ponds and wetlands from creation to maturation to decline. It seems likely that the production of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> and supply to surface waters will vary over the life-history of beaver ponds, with initially very high rates, similar to values reported for recently established reservoirs ( $\sim 10 \ \mu g/m^2$ -yr), which declines over time to values reported for wetlands ( $\sim 0.5 \mu g/m^2$ -yr). Even for older ponds which are at steady-state with respect to Hg<sub>T</sub> inputs, these environments are net sources of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup>, which contrasts with freely draining upland catchments which exhibit net retention of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup>. The overall abundance of beaver ponds on the landscape is tied to changes in the population of beavers. Studies have shown that beaver populations in North America (and in Scandinavia), which declined to very low levels by 1900 as a result of over-exploitation (trapping), irrupted about 30 years after the re-introduction. In the boreal forest of northern Minnesota, a major episode of beaver pond construction occurred between 1940 and 1960, which still dominates the landscape today (Johnston & Naiman 1990). In Scandinavia, re-introduction started about 1920, with a major episode of pond building in the mid 1960's (Hartman 1994). It is likely that similar changes have occurred in the Adirondack region of New York, although quantitative information is lacking (Hammerson 1994). Increases in wetland area through the 1900's may have enhanced the transport of CH<sub>3</sub>Hg<sub>T</sub><sup>+</sup> to downstream lakes and contributed to elevated concentrations of Hg observed for fish in the Adirondack region (Driscoll et al. 1995).

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