

Atmospheric Deposition, Transformations, and Fate of Mercury Across a Northern Forest Landscape

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Abstract

Mercury (Hg) is a toxic trace metal, that has been linked to human illness and degradation of ecosystems. Atmospheric emissions have caused widespread contamination of Hg in terrestrial and aquatic environments, including dangerously high concentrations in Adirondack lake biota. However, the linkage between atmospheric deposition and contamination of aquatic biota is not well established. Additional research is necessary to determine the ultimate fate of Hg within remote ecosystems. A quantitative understanding of atmospheric Hg deposition and the factors regulating the fate, transformation, and bioavailability of such Hg within terrestrial environments and downstream aquatic ecosystems are critical research needs.

The current research examines Hg deposition and cycling within the Arbutus Lake watershed (Newcomb, NY) to clarify the atmospheric, geochemical, and biological mechanisms regulating Hg concentrations in soft-water lakes of the northeastern U.S. The effects of deposition pathway, soil retention, and landscape characteristics are being examined. Data obtained within the Arbutus watershed will provide information regarding Hg contamination and bioavailability of Adirondack ecosystems. The data may be extrapolated regionally via modeling to predict the effect of Hg deposition within similar watersheds of the Adirondacks.

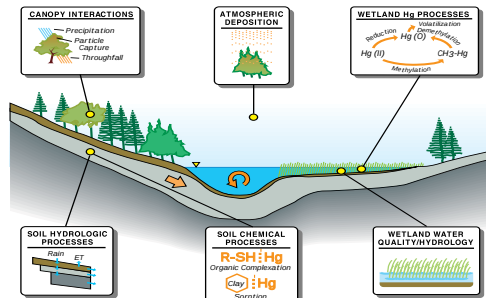


Figure 1. Processes affecting the fate of Hg within a watershed. Input is represented through direct deposition and canopy interactions. Representative hydrologic and transformation processes are included for upland, wetland, and lake compartments. The model which depicts these processes is the Mercury Cycling Model for Headwater Drainage lakes (MCM-HD).

Research Objectives

The purpose of this study is to examine the inputs and fate of Hg within a forest-wetland-lake watershed ecosystem. Results of analyzing individual fluxes/compartments will be integrated to examine the processes affecting Hg fate within the watershed. For the examination of the various processes, instrumentation was installed at various sites within the watershed to:

1. Measure atmospheric interactions, including direct deposition, throughfall, evasion, and canopy Hg profiles.
2. Evaluate the transport and speciation of deposited Hg within an upland terrestrial system, including the application of stable isotope to track Hg fate.
3. Conduct Hg budgets on Arbutus Lake and an associated inlet stream wetland system. The budget for Arbutus (high rT, low DOC) is being compared with Sunday Lake (low rT, high DOC).



Figure 2. Arbutus Lake from near the outlet. The Archer Creek watershed is in the background at the far northern end of the lake. The upland plots are to the right of the picture location, approximately 300 m upslope.

Atmospheric Interactions

Atmospheric deposition is the predominant mechanism for Hg input to remote forest watersheds. Therefore, air-canopy and air-soil interactions are particularly important for defining the total Hg loading to the watershed. Soil-air flux is critical for determining the fate of newly-applied Hg. To investigate loading and air-canopy fluxes, experimentation is being conducted to quantify:

- Deposition:
 - Direct deposition (Mercury Deposition Network – MDN site)
 - Upland plot throughfall (Arbutus)
- Measure canopy concentration profiles
- Quantify the evasive flux of Hg from upland soil.
- Effect of event (rainfall) on air-canopy-soil interactions

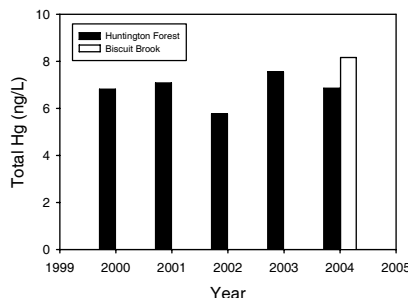


Figure 4. Annual volume-weighted concentrations of total Hg for MDN station Huntingdon Forest (Arbutus Lake) compared to values at Biscuit Brook in the Catskills. Note that the MeHg concentration in precipitation is typically <1% of total Hg.

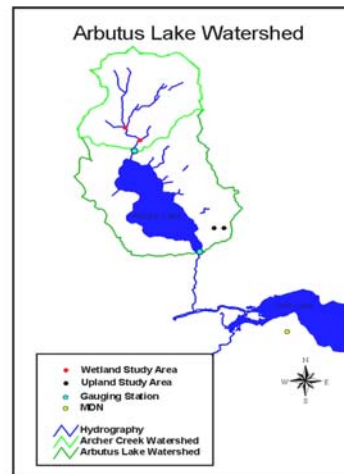


Figure 3. Intensive study areas for Hg cycling within the Arbutus Lake watershed. Shown are the locations for lake inlet/outlet sampling, the two wetland study locations, the upland plots for soil fate and atmospheric interaction, and the Mercury Deposition Network (MDN) site.

Upland Transport and Fate

The transport and fate of Hg within upland forest ecosystems is important as inputs are largely via throughfall and leaf litter. Leaves have been shown to accumulate THg, likely via canopy air exchange. As leaves are deposited and biodegrade, THg enters the soil-water-plant system. The fate, transformations, and fluxes of Hg within the forest system are being assessed through instrumentation and measurements focused on two plots (Reference and Treatment).

- Soil pits
 - Soil collected from Oa, Bh, E, Bs1, Bs2, Bs3, and C horizons
 - Lysimeter pairs – Oa and Bh (both plots) and Bs2 (Reference)
- Plant tissue and litterfall collection – beech, sugar maple, birch
- Soil evasion

The Treatment plot will be dosed with stable isotope, ²⁰⁰Hg, to investigate fate of newly-applied THg. Each of the above samples will be analyzed for Hg speciation and isotopic distribution.

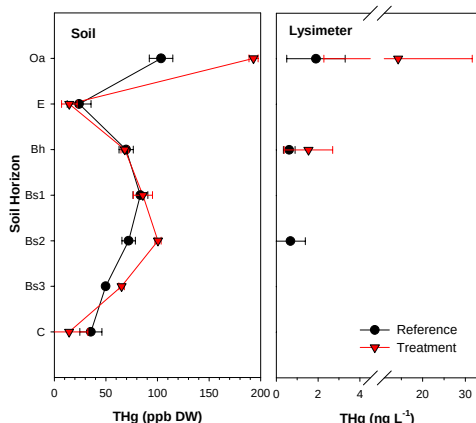


Figure 5. THg concentration profile for soil horizons and soil waters. Soil was collected through pit installation. Soil solutions reflect monthly collections between September 2004 and April 2005. Concentration data for duplicate lysimeters were aggregated.

Summary

The fate and interaction of Hg within upland systems is influenced by air-canopy-soil, and watershed processes. Specific activities are:

- Monitoring Hg deposition and throughfall; examine canopy profile
- Examining evasive flux from upland systems
- To determine soil horizon THg profile
- To install and measure THg in lysimeter pairs for upland plots
- Collecting wetland stream and lake system water samples

Future Work

- Continue current monitoring
 - Deposition, throughfall, and evasion
 - Lysimeter soil water, leaf tissue, and litterfall
 - Wetland and lake water sampling, including piezometers
- Event sampling
 - The measurement of Hg mobility during hydrologic events
- Upland plot dosing
 - Add ²⁰⁰Hg to Treatment plot
 - Track fate of newly-applied Hg

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Lake Watershed / Wetland Budgets

The effect of landscape characteristics on MeHg production and bioavailability is being investigated via stream water and shallow groundwater sampling within the Archer Creek wetland system. Comparisons are also being conducted between the Arbutus Lake data (high rT, low DOC) and that obtained from Sunday Lake (low rT, high DOC). Water chemical parameters such as DOC and SO4²⁻ concentrations have been found to affect Hg speciation and transport.

- Examine Hg transport and speciation within wetland system
 - Nested series of stream water sampling points – includes wetland and upland sites
 - Piezometer transects (n=4) across two wetlands, with two depths represented at each location
- Lake watershed budget – monitor Hg content and speciation for lake inlet/outlet

Table 1. Comparison of different chemical species concentrations for upland, wetland, and lake waters. Data represents range of values from August 2004 to February 2005. Wetland and higher DOC waters experience higher Hg concentrations and fluxes.

	DOC (mmol L ⁻¹)	SO4 ²⁻ (mmol L ⁻¹)	THg (ng L ⁻¹)	MeHg (ng L ⁻¹)
Upland Waters	160 - 263	61 - 76	0.50 - 2.00	0.008 - 0.15
Wetland Waters	188 - 1338	43 - 75	0.51 - 5.50	0.01 - 0.77
Sunday Lake	197 - 918	29 - 56	0.28 - 4.30	0.06 - 0.57
Arbutus Lake	267 - 670	52 - 73	1.13 - 4.42	0.04 - 0.40

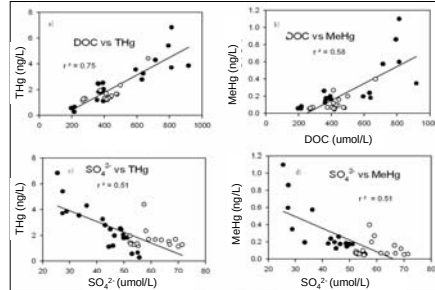


Figure 6. Relationships between MeHg and THg for Arbutus (o) and Sunday Lake (•) and DOC and SO4²⁻, demonstrating their effect on Hg speciation and mobility. Upland waters displayed weaker patterns while wetland stream waters exhibited stronger patterns. Data represent values from August 2004 to February 2005.