

# FACTORS CONTROLLING MERCURY TRANSPORT IN AN UPLAND FORESTED CATCHMENT

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**Abstract.** Total mercury (Hg) deposition and input/output relationships were investigated in an 11-ha deciduous forested catchment in northern Vermont as part of ongoing evaluations of Hg cycling and transport in the Lake Champlain basin. Atmospheric Hg deposition (precipitation + modeled vapor phase downward flux) was 425 mg ha<sup>-1</sup> during the one-year period March 1994 through February 1995 and 463 mg ha<sup>-1</sup> from March 1995 through February 1996. In the same periods, stream export of total Hg was 32 mg ha<sup>-1</sup> and 22 mg ha<sup>-1</sup>, respectively. Thus, there was a net retention of Hg by the catchment of 92 % the first year and 95 % the second year. In the first year, 16.9 mg ha<sup>-1</sup> or about half of the annual stream export, occurred on the single day of peak spring snowmelt in April. In contrast, the maximum daily export in the second year, when peak stream flow was somewhat lower, was 3.5 mg ha<sup>-1</sup> during a January thaw. The fate of the Hg retained by this forested catchment is not known. Dissolved (<0.22 µm) Hg concentrations in stream water ranged from 0.5–2.6 ng L<sup>-1</sup>, even when total (unfiltered) concentrations were greater than 10 ng L<sup>-1</sup> during high flow events. Total Hg concentrations in stream water were correlated with the total organic fraction of suspended sediment, suggesting the importance of organic material in Hg transport within the catchment. High flow events and transport with organic material may be especially important mechanisms for the movement of Hg through forested ecosystems.

**Keywords:** biogeochemistry, catchment, input/output budget, Lake Champlain watershed, mercury, forest, stream

## 1. Introduction

Mercury contamination of fresh waters and their biota has become a widespread and serious problem in many parts of the world, including northeastern North America. There is widespread concern that atmospheric sources of Hg may be responsible for increasing Hg burdens in Lake Champlain (Scherbatskoy *et al.*, 1997; Watzin 1992; Vasu and McCullough, 1994), but mechanisms of transport and accumulation are not well understood. The Lake Champlain basin (Figure 1) is characterized by a large (19:1) ratio of land to lake surface area, of which 62 % is forested and 27 % is agricultural (Budd and Meals, 1994). This relatively large forested land area has the potential to capture atmospheric pollutants and transfer them to soils, surface waters and ground water by several mechanisms. Terrestrial ecosystems accumulate atmospheric pollutants and transfer them to the soil and surface waters by wet and dry deposition, throughfall (precipitation passing through the canopy), litterfall (deposition of senescent foliage), and interception of cloud water at higher elevations. Forest canopies in this region have foliage surface areas up to ten times greater than the land area they cover, providing Hg concentration in throughfall that averages twice that of precipitation and annual Hg flux in litterfall that is greater than annual wet deposition (Rea *et al.*, 1996).

Annual Hg deposition (wet + dry) in the Lake Champlain basin is over 125 mg ha<sup>-1</sup> (Scherbatskoy *et al.*, 1997). This Hg is primarily of anthropogenic origin, and if these emissions were to cease now, it would require 15–20 years to return to background Hg

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levels (Mason *et al.*, 1994). Examining the behavior of Hg in forested catchments is important to understanding the present and long-term role of atmospheric Hg in the Lake Champlain basin.

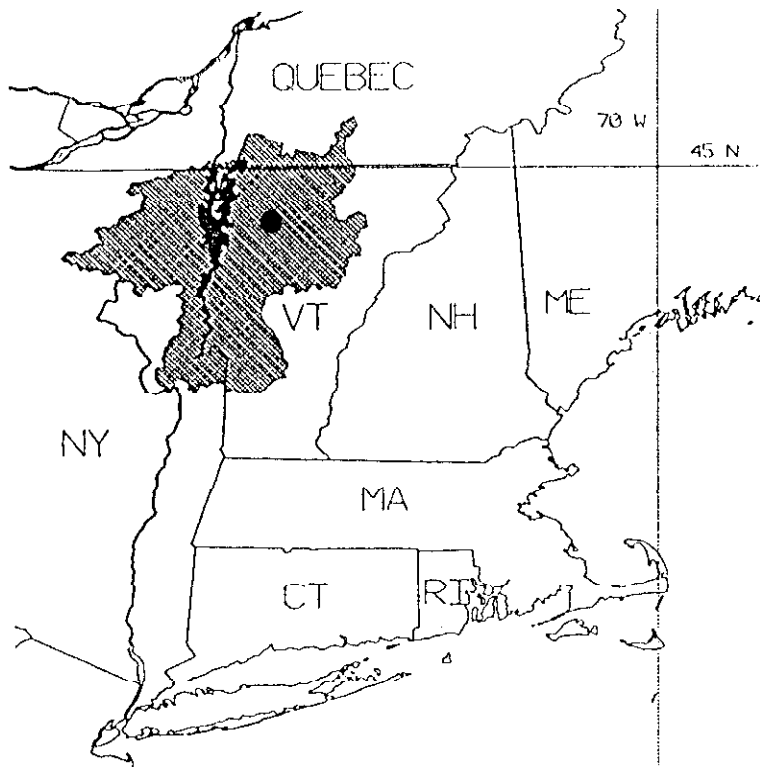


Fig. 1. Regional map of Lake Champlain and its drainage basin, with the study site (dot) shown in the eastern portion of the basin.

Information on levels or behavior of Hg in forested ecosystems in the northeastern United States is limited (Scherbatskoy *et al.*, 1997; Rea *et al.*, 1996). There is growing evidence that forested ecosystems have an important and complex role in the transport of atmospheric Hg to aquatic systems (Mierle and Ingram, 1991; Lee *et al.*, 1994; Bishop *et al.*, 1995; Johnson and Lindberg, 1995; Hurley *et al.*, 1995; Krabbenhft *et al.*, 1995). Recent studies of the behavior of Hg in terrestrial ecosystems have identified several processes that affect Hg cycling, including organic complexing in soils and water, foliar exchange and out gassing, and accumulation in plant tissues (Schuster, 1991; Lee *et al.*, 1994; Hanson *et al.*, 1995; Zhang *et al.*, 1995; Rea *et al.*, 1996). Swain *et al.* (1992) showed the importance of watershed:lake area ratio in Hg loading to lake sediments. From these indications, it appears that anthropogenic Hg deposition, organic acidic soils, forest wetlands, hydrologically important snowmelt dynamics, and the large catchment basin feeding Lake Champlain contribute to Hg transport in the terrestrial and stream components in this basin.

The research presented here is part of a larger program addressing concerns about Hg and other toxic metals in the aquatic, terrestrial and atmospheric environments of the Lake Champlain basin. Our goals in this paper were to characterize atmospheric inputs and stream export of Hg in a small upland forested catchment, and to identify mechanisms controlling Hg transport.

## 2. Site Description and Methods

This research was carried out at Nettle Brook, an 11 ha mixed hardwoods catchment on the western slope of Mt. Mansfield in the town of Underhill, Vermont, 34 km east of Lake Champlain (Figure 1). Nettle Brook is operated by the Vermont Forest Ecosystem Monitoring program (VForEM) as part of their long-term integrated studies on responses of forested ecosystems to environmental change (Wilmot and Scherbatskoy, 1994). Elevation in the catchment ranges from 445 to 664 m, aspect is southwest, and slope averages 35 % (range 5-75 %). Soils are moderately well-drained Peru extremely stony loams underlain by mica-albite-quartz schist bedrock. Average annual rainfall is 1,100 mm. January daily minimum temperature is -16 °C and July daily maximum temperature is 24 °C. Annual  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  deposition averages 21 and 16 kg ha<sup>-1</sup>, respectively.

Since October 1993, stream water samples have been collected at Nettle Brook at a continuously gauged V-notch weir for analysis of Hg, DOC, and suspended sediment. During spring snowmelt periods, daily or twice-daily water samples were collected at the weir; during the rest of the year, samples were collected approximately monthly.

Precipitation was collected daily in a MIC-B wet-only precipitation collector (MIC Co., Richmond Hill, Ontario) modified for Hg sampling (Landis and Keeler, 1997), at the VForEM air quality monitoring station at the Proctor Maple Research Center (400 m elevation), 3.1 km northwest of Nettle Brook. Ambient vapor and particulate samples were collected every sixth day for 24 hr (8 AM cycle) on gold-coated bead vapor traps and glass fiber particulate filters, respectively. Dry deposition flux of Hg vapor to this forest was estimated according to Scherbatskoy *et al.* (1997) as amended by Lindberg (1993), providing an approximation of the downward flux within a factor of 2 (Lindberg *et al.*, 1992). Upward flux (Hg evasion from soils and vegetation) is unknown for this site (indeed is poorly quantified in general), but could be significant (Hanson *et al.* 1995). Snowmelt was collected from a 1 m<sup>2</sup> Teflon-coated lysimeter installed on the ground beneath the snowpack. Meltwater was collected at mid-day from 24 March-14 April, 1994.

Samples were shipped by overnight courier to the University of Michigan Air Quality Laboratory in Ann Arbor, MI, where analysis for total Hg was performed by cold vapor atomic fluorescence spectrometry. Stream samples were filtered through 0.22 µm nitrocellulose filters (Millipore MF) to separate operationally defined dissolved and particulate fractions. Ultra-clean sampling, handling and storage techniques were used throughout all procedures. Field and laboratory analytical blanks were analyzed (20 % of all samples) for quality assurance purposes. Analytical accuracy was within 5 % of known standards and precision error was less than 8 %. The Hg loading error was ±5 % for wet deposition and the uncertainty of the modeled dry deposition estimates was about a factor of two (Lindberg *et al.* 1992). Analytical and quality assurance details are given in Burke *et al.* (1995). No Hg speciation was performed.

DOC was determined by ultraviolet persulfate oxidation with infrared detection at the USGS laboratory in Albany, NY. Suspended sediment concentration was determined gravimetrically from the filtrant on a sample replicate passed through a 0.7  $\mu\text{m}$  glass fiber membrane. The detection limit was approximately 2  $\text{mg L}^{-1}$ . The organic fraction of sediment was determined by loss-on-ignition at 550  $^{\circ}\text{C}$ .

Stream Hg flux was calculated from hourly average flows and modeled Hg concentrations. Dissolved Hg was weakly correlated with flow ( $r^2 = 0.13$ ,  $p = 0.0022$ ) and this relationship was used to derive hourly dissolved Hg concentrations throughout the period. Total Hg flux was calculated by flow class. For flow less than 0.5  $\text{mm hr}^{-1}$ , the concentration:discharge relation was significant ( $r^2 = 0.21$ ,  $p = 0.0002$ ), and was used to derive total Hg concentrations. For flows greater than 0.5  $\text{mm hr}^{-1}$ , geometric means of sample total Hg concentrations for each of three flow classes were used in flux calculations (Figure 2). Upper and lower error limits for the fluxes were determined from the 95 % confidence limits of the regression for the  $<0.5 \text{ mm hr}^{-1}$  flow class, and from the high and low sample Hg concentrations for the higher flow classes. Sampling was frequent enough during the 1994 snowmelt period (March 14-April 30) to determine fluxes directly from sample total Hg concentrations, with linear interpolation of total Hg concentrations between sampling times. Error limits for this period were calculated as above, but adjusted as necessary to keep the lower error limit between 20 % and 50 % less than the interpolated flux, and the upper limit within 20 % of the interpolated flux.

### 3. Results

#### 3.1. HYDROLOGY

Precipitation inputs were similar in the two study years; in each year slightly more than half of the precipitation input ran off as streamflow (Table I). The distribution of precipitation and runoff was quite different in the two years, however. The first year (March 1994 - February 1995) was dominated by a major snowmelt event in April (Figure 3), as an unusually large snowpack melted quickly. Moderate spring rains sustained relatively high flows into the early summer, but low rainfall thereafter touched off a moderate drought, which extended into the second year of the study. The 1995 snowpack was unusually small and led to a small and early snowmelt in March. Below-average precipitation produced June streamflow that was more typical of late summer. A series of heavy convective storms in July and August of 1995 generated several high flow events. Autumn rainfall was well above average and produced high sustained flow and some notable peak flows. A deep snowpack developed early in the winter of 1995-96, but most of it left the catchment in a major January thaw (Figure 3).

#### 3.2 Hg IN ATMOSPHERIC DEPOSITION

Atmospheric concentrations and deposition of Hg are summarized in Table II for 1993-95. Wet and dry deposition fluxes of Hg are summarized on a seasonal basis for 1994-95 in Figure 4. Vapor phase Hg (which dominates dry deposition) tends to be relatively constant throughout the year, but greater deposition velocities in the summer months lead to greater

dry deposition. Both precipitation amount and Hg concentration in precipitation tend to be greatest in the summer months, resulting in the highest wet deposition of Hg during this period. Additional information on Hg deposition at this site is in Scherbatskoy *et al.* (1997). These data are comparable in magnitude and trend to values for rural northern Michigan (Burke *et al.*, 1995; Hoyer *et al.*, 1995; Keeler *et al.*, 1995).

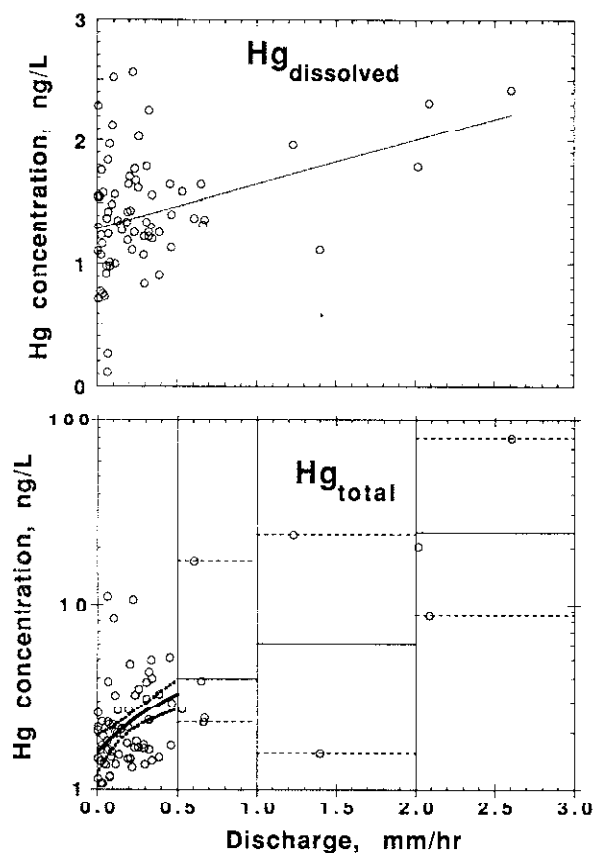


Fig. 2. Concentration:discharge relations for dissolved Hg (upper panel) and total Hg (lower panel) for two years of study. Regression line in upper panel is for all samples. Regression line in lower panel is for 1995 snowmelt period only.

TABLE I

Inputs and outputs of water and mercury at Nettle Brook catchment for the two study years.

Date	Precipitation (mm)	Streamflow (mm)	Hg input (mg ha <sup>-1</sup> yr <sup>-1</sup> )			Hg export (mg ha <sup>-1</sup> yr <sup>-1</sup> )			Retention
			Wet	Dry	Total	Particulate	Dissolved	Total	
Mar 94 - Feb 95	1139	587	74.5	350	425	23.6	8.4	31.9	92 %
Mar 95 - Feb 96	1109	568	92.7	370	463	13.4	8.2	21.7	95 %

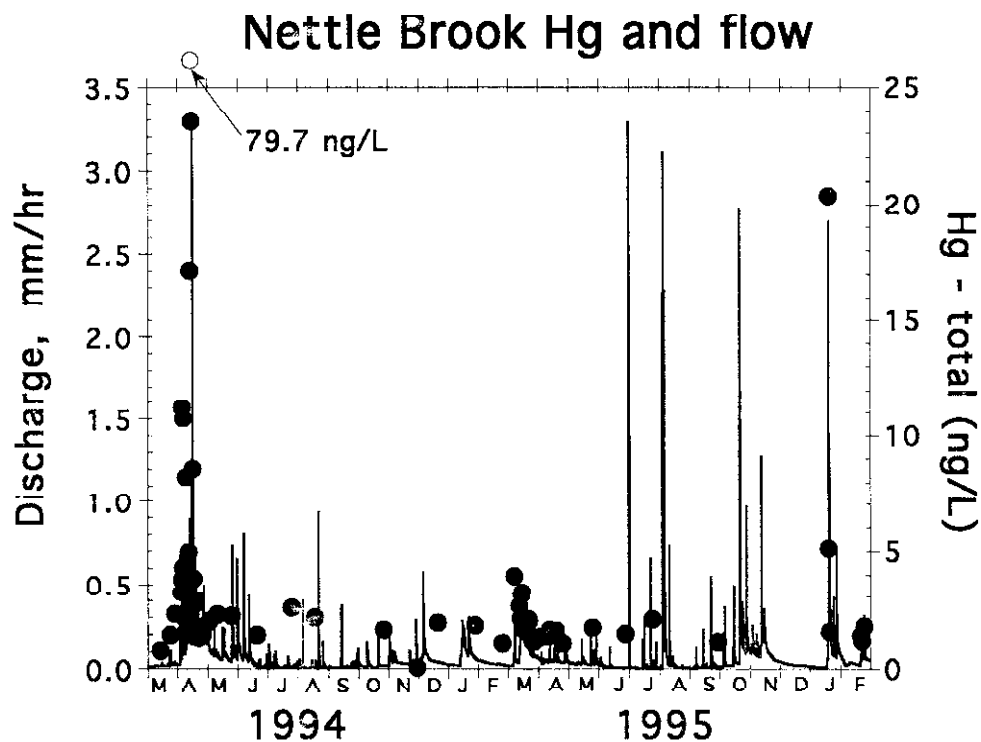


Fig. 3. Flow ( $\text{mm hr}^{-1}$ ) and stream water total Hg concentration ( $\text{ng L}^{-1}$ ) at the Nettle Brook weir from March 1994 through February 1996. Water was sampled for Hg analysis on a daily basis during spring snowmelt and periodically during the remainder of the year.

TABLE II

Descriptive statistics for concentrations of Hg in the atmosphere and precipitation at the air quality monitoring site in Underhill, VT for the period January 1993 through December 1995.

	Mean	Std. Dev.	n	Minimum	Maximum	Median
Vapor ( $\text{ng m}^{-3}$ )	1.72	0.55	204	0.51	6.86	0.05
Aerosol ( $\text{pg m}^{-3}$ )	10.21	6.75	210	0.89	43.20	7.70
Precipitation ( $\text{ng L}^{-1}$ )	6.49 <sup>1</sup>	6.08	410	0.09	59.09	6.51

<sup>1</sup>volume-weighted

### 3.3. Hg IN SNOW MELTWATER

Snow meltwater collected during the 1994 spring melt period had an average total Hg concentration of  $4.8 \text{ ng L}^{-1}$ , with a range of 2 to  $9.2 \text{ ng L}^{-1}$ , similar to the Hg concentration of incoming snowfall (Scherbatskoy *et al.*, 1997). These concentrations were in the same range as stream water samples collected during this period (Figure 5), except for peak snowmelt, suggesting that stream water Hg chemistry could be explained by meltwater inputs.

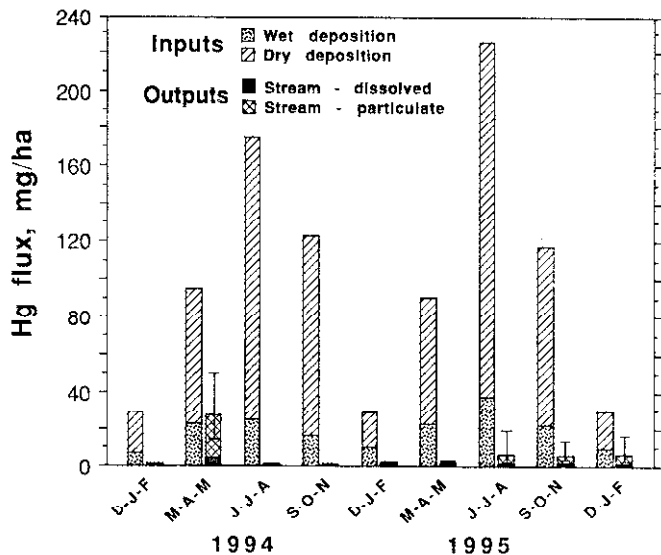


Fig. 4. Seasonal input (wet and dry deposition) and stream export (dissolved and particulate) of total Hg in Nettle Brook catchment from December 1993 through February 1996, expressed as  $\text{mg ha}^{-1}$ . Error bars on export fluxes represent the upper and lower error limits for the Hg fluxes (see Methods); where no bar is shown, error was  $< +0.7 \text{ mg ha}^{-1}$ . The wet deposition error was  $\pm 5\%$ , while the uncertainty of the modeled dry deposition estimates was about a factor of two (Lindberg *et al.* 1992).

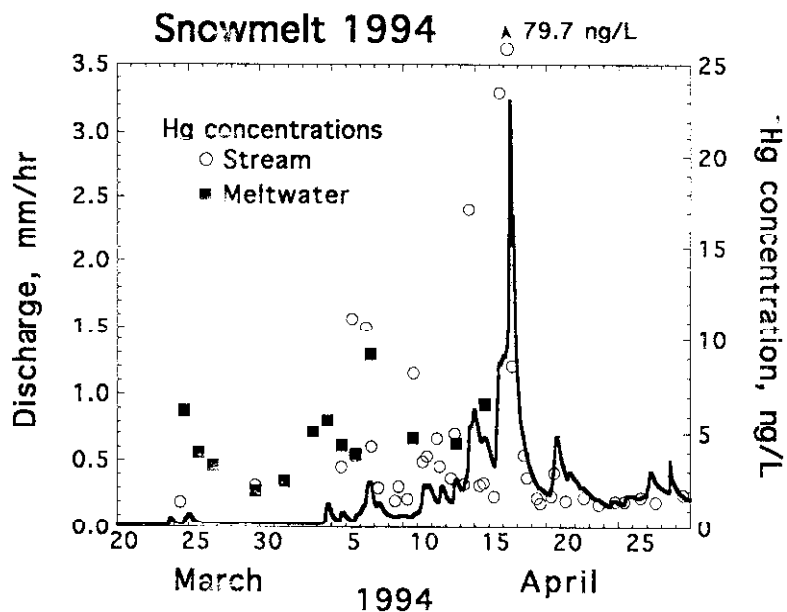


Fig. 5. Flow ( $\text{mm hr}^{-1}$ ) and stream water total Hg concentration ( $\text{ng L}^{-1}$ ) for the 1994 snowmelt period, with meltwater Hg concentrations from snowmelt lysimeter samples.

## 3.4. Hg IN STREAM WATER

Dissolved Hg concentrations in stream water were consistently low, ranging only from 0.5 to 2.6 ng L<sup>-1</sup>, and there was no apparent seasonal pattern. Total Hg concentrations ranged from 1.1 to 79.7 ng L<sup>-1</sup> (Figure 3). The highest Hg concentrations occurred in the 1994 spring snowmelt, reaching 10 ng L<sup>-1</sup> during the initial minor daily flow increases, and reaching a maximum at peak snowmelt. The only other period sampled where Hg concentrations exceeded 5 ng L<sup>-1</sup> was during high flow in the 1996 January thaw, when total Hg concentration increased to greater than 20 ng L<sup>-1</sup>. During the smaller than average 1995 snowmelt event, total Hg concentration approached 5 ng L<sup>-1</sup>. At all other sampling times, total Hg generally remained less than 3 ng L<sup>-1</sup>. Note, however, that we sampled only two of the five high-flow events and only once each year in autumn (Figure 3).

## 3.5. Hg AND SUSPENDED SEDIMENT

Total Hg concentration increased with increasing suspended sediment concentration ( $r^2 = 0.41$ ,  $n = 28$ ,  $p = 0.0002$ ). The  $r^2$  improved to 0.50 when one outlier was removed. The particulate fraction (total - dissolved) of total Hg in stream water was highly correlated with the organic fraction of suspended sediment ( $r^2 = 0.85$ ,  $p < 0.0001$ ; Figure 6). A single high Hg value exerts leverage, but the regression remains strong if that point is removed ( $r^2 = 0.53$ ,  $p < 0.0001$ ). All but three of the samples that had non-detectable suspended sediment (<2 mg L<sup>-1</sup>) had less than 1 ng L<sup>-1</sup> particulate Hg.

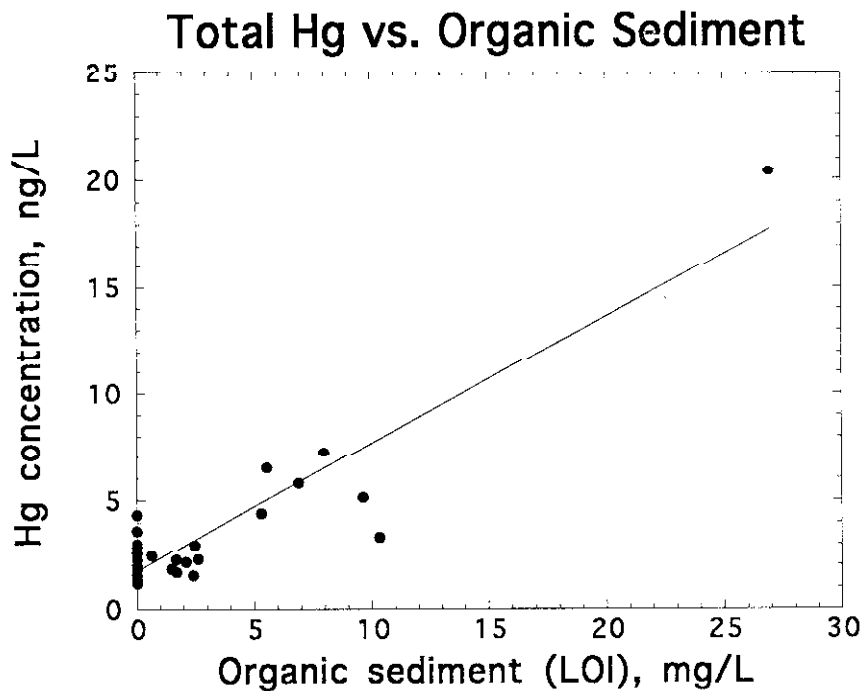


Fig. 6. Total Hg concentration as a function of organic matter fraction of suspended sediment for all samples analyzed for suspended sediment.



### 3.6. Hg EXPORT

The flux of total Hg in stream water was strongly linked to high flow events. In the first year of study, about half the annual Hg flux occurred on the single day of peak snowmelt. About 84 % of the annual flux of total Hg occurred with only 56 % of the annual stream-flow during the 3-month period of March-May (Figure 4). Mercury flux was minimal through most of the 1995 snowmelt because of drought conditions and associated low snowpack. High-flow events in July and August and the January thaw of 1996 were responsible for a significant share of the Hg flux in the second year.

Dissolved Hg flux in stream water varied less than the highly episodic total Hg flux. However, because dissolved Hg concentration was positively correlated with flow, dissolved Hg flux also was dominated by high flow periods (e.g., March-May 1994). During low flow periods, dissolved Hg flux accounted for most of the Hg flux from the basin. On an annual basis, dissolved Hg accounted for 26 % of the total Hg flux in the first year of study and 38 % in the second year (calculated from Table I).

### 3.7. Hg BUDGET

Total Hg input from wet and dry deposition substantially exceeded Hg export in stream water (Figure 4). During spring 1994, export was 30 % of deposition due to the large snowmelt event in April. By contrast, Hg export was only 2 % of Hg input during the summer fall period in 1994. On an annual basis, the forested ecosystem retained 92 % of atmospheric Hg during the first year of the study and 95 % the second year (Table I).

## 4. Discussion

The forested Nettle Brook catchment is a net sink for atmospherically deposited Hg. Within the large uncertainties associated with our flux determinations because of limited event sampling, we found that between 60 and 90 % of deposited Hg is retained. This retention rate is similar to the 80 % to 90 % retention found in Sweden (Aastrup *et al.*, 1991; Johanssen *et al.*, 1991), but less than the 92 % retention determined by Krabbenhoft *et al.* (1995) at a Wisconsin site. Of the Hg exported in streamflow, the low but consistent dissolved Hg concentration accounts for about one-third of the annual flux. A large portion of the annual Hg export in stream water occurs as particulate Hg during a short time interval in extreme hydrologic events.

Hydrologic events at Nettle Brook have generated some extremely high total Hg concentrations. The two largest events sampled had greater than 20 ng L<sup>-1</sup> Hg, and one had nearly 80 ng L<sup>-1</sup> Hg. Among several catchments in Sweden, by contrast, maximum Hg concentrations did not exceed 15 ng L<sup>-1</sup> (Johanssen *et al.*, 1991; Lee and Iverfeldt, 1991; Hultberg *et al.*, 1995), except for 21 ng L<sup>-1</sup> in runoff from a peat bog (Westling, 1991).

The tendency of Hg concentration to increase with flow also was found by Bishop *et al.* (1995) during snowmelt in Sweden, although their stream water Hg concentration increased to only 6 ng L<sup>-1</sup>. Bishop *et al.* (1995) found that 37 % of total annual Hg flux occurred in a three week spring snowmelt period. Other studies have given little attention to event sampling, so Hg behavior during high-flow events cannot be compared. Event-

related Hg transport clearly lowers overall Hg retention at Nettle Brook. Some studies suggest that wetlands are responsible for Hg or Me-Hg (methyl Hg) mobilization (Driscoll *et al.*, 1994; Hurley *et al.*, 1995; Branfireun *et al.*, 1996). Nettle Brook has relatively little wetland area, but this may be compensated for by the high particulate transport of Hg resulting from the steep, mountainous slopes.

What becomes of Hg retained by the forested catchment? Some Hg may return to the atmosphere by reduction to  $\text{Hg}^0$ , transport of  $\text{Hg}^0$  in the plant transpiration stream, and evasion from soils (Siegel and Siegel, 1988; Hanson *et al.*, 1995; Johnson and Lindberg, 1995). Some Hg must accumulate in forest soils and biota, where it is actively cycled. Litterfall adds an estimated  $130 \text{ mg ha}^{-1}$  annually to the forest floor at this site (Rea *et al.*, 1996). Leaf litter decomposes over a few to many years, releasing the Hg for potential aqueous transport, most likely as organically bound complexes (e.g., humic acids).

Just how is Hg mobilized during large events? If Hg is sequestered primarily in the forest floor in association with organic matter (Mierle and Ingram, 1991; Aastrup *et al.* (1991), one would expect that flushing of the soil would deliver Hg to the stream as a dissolved organically bound complex. However, we observed little or no increase in dissolved Hg during events. Moreover, Hg and DOC were poorly correlated, in contrast to the positive Hg-DOC relation found elsewhere (Driscoll *et al.*, 1994; Bishop *et al.*, 1995). The episodic Hg increase in stream water was dominated by particulate Hg and thus appears to be linked to sediment transport. The process supplying sediment may be soil erosion by overland flow in riparian areas, streambank erosion, or mobilization of in-stream sediments.

The association between total Hg concentration and the organic fraction of suspended sediment can be explained by the high affinity of  $\text{Hg(II)}$  for organic substances in forest soils, as reported by several authors (Lindqvist, 1991; Mierle and Ingram, 1991; Schuster, 1991). Krabbenhoft *et al.* (1995) found that total Hg in stream water increased sharply with releases of DOC, while Me-Hg did not, in a small forested stream system in Wisconsin. Bishop *et al.* (1995) found that the increase in Hg concentration during the spring snowmelt was associated with total organic carbon.

Episodic fluxes of Hg during large events are not limited to upland catchments. Results of synoptic sampling of some larger streams in the Lake Champlain basin (Scherbatskoy *et al.*, 1997) also suggest a strong relation of increasing Hg with increasing flow. As at Nettle Brook, there appears to be a low but constant supply of dissolved Hg in the larger streams, but the dominant Hg loading to Lake Champlain from the rivers probably occurs in large events, such as the 1994 snowmelt or the August 1995 flood. It is not known whether these elevated particulate Hg concentrations in the larger rivers reflect transport from steep upland headwater catchments such as Nettle Brook or more local sources such as runoff from agricultural or disturbed lands.

## 5. Conclusions

The Nettle Brook catchment exports less than one tenth (8 % and 5 % during two study years) of the Hg that enters in atmospheric (wet + dry) deposition. Of the Hg that is exported in streamflow, between one-quarter and two-fifths (26 % and 38 %) is in the dissolved form, representing a slow but continual export. The remaining Hg is in particu

late form and is exported mostly on an episodic basis; fully one half of the total annual export in the first year of study occurred on the single day of peak snowmelt. Particulate Hg in stream water is strongly associated with the organic fraction of suspended sediment. Episodic pulses of particulate Hg are evident at downstream reaches of larger rivers as well, suggesting that mobilization of Hg during large events may be the dominant source of Hg loading to Lake Champlain.

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