

**Paleostratigraphy of Mercury in Branch Pond, Sunderland, VT**

Submitted to the Vermont Monitoring Cooperative

by

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## Executive Summary

In response to problems associated with mercury (Hg) in the freshwater environment, there is increasing interest in understanding the relative magnitude of present and historical atmospheric mercury deposition to north-temperate lakes. Improvement in the Hg situation on VT lakes will necessitate reductions in discharges and emissions of Hg both locally, and nationally. Paleolimnologically-derived estimates of atmospheric Hg deposition may serve as inexpensive monitors of anticipated reductions in Hg emissions. The purpose of this report is to present results of a paleolimnological analysis performed at Branch Pond, Sunderland, VT to identify the signature of atmospheric Hg deposition to the lake's watershed.

In August of 1999, a core was acquired from the sediments of Branch Pond, Sunderland, VT, in the Lye Brook Wilderness area. This core was dated by  $^{210}\text{Pb}$ , and analyzed for a variety of parameters relating to the deposition of mercury (Hg) to the lake sediments. This analysis was undertaken with support from VMC, in conjunction with the USEPA-sponsored REMAP Assessment of Mercury in Lake Sediments, Waters and Biota of VT and NH Lakes Project.

The general pattern of Hg fluxes to the sediments of Branch Pond indicate increasing atmospherically driven Hg deposition, beginning by the year 1850. This finding is consistent with other lakes studied by the REMAP project, as well as with findings of other studies. Hg fluxes to the lake continued to increase throughout the period 1850 to 1980. A decline in flux to the sediments is apparent in recent years.

The signals in fluxes of Hg to Branch Pond sediments are not as clear as for other lakes. There is evidence from the  $^{210}\text{Pb}$ -inferred sedimentation record, as well as from sediment organic content, that the delivery of Hg to the sediments of Branch Pond has been enhanced by land use activities which occurred during two distinct periods: 1825-1883; and 1905-1954.

Presently, disturbances to the Branch Pond watershed are near the lowest levels estimated by this analysis. It is likely that, given the designation of the area surrounding Branch Pond as the Lye Brook Wilderness area, watershed conditions will continue to stabilize, thus minimizing watershed-mediated Hg delivery to the lake.

Evidence from the Branch Pond core suggests that recent declines in Hg deposition rates to the sediments may be linked to reductions in atmospheric Hg deposition across the landscape. Over the long term, this may result in a reduced source of Hg available for methylation and subsequent bioaccumulation.

## Introduction:

In response to problems associated with mercury (Hg) in the freshwater environment, there is increasing interest in understanding the relative magnitude of present and historical atmospheric mercury deposition to north-temperate lakes. Measurements of direct atmospheric deposition of Hg are expensive, and are only being made at a handful of northeastern locations. Improvement in the Hg situation on VT lakes will necessitate reductions in discharges and emissions of Hg both locally, and nationally. Paleolimnological proxies for estimating atmospheric Hg deposition may serve as inexpensive 'monitors' of Hg deposition to the northeastern landscape. The purpose of this report is to present results of a paleolimnological analysis performed at Branch Pond, Sunderland, VT to identify the signature of atmospheric Hg deposition to the lake's watershed.

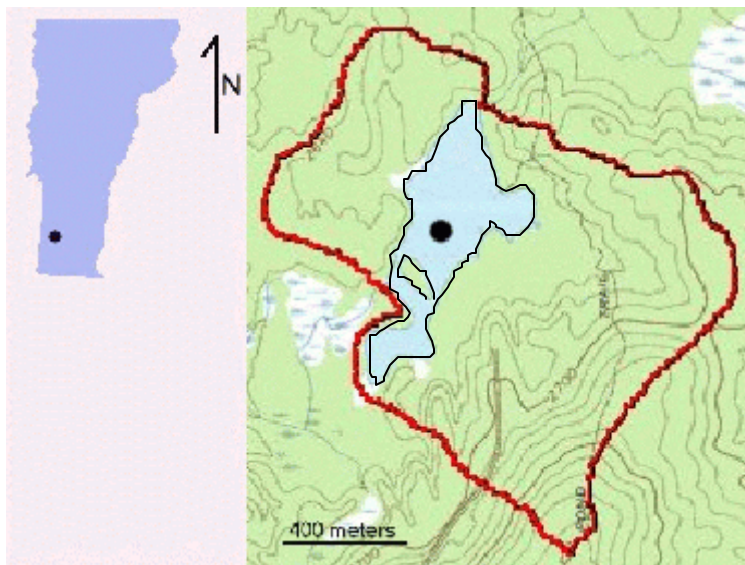
Paleolimnology is a tool which came of age largely in the 1980's, and which has been successfully used to infer the historical trophic state and acidification status of lakes (Ouellet and Jones, 1983, Oldfield and Appleby, 1984, Charles and Norton, 1986, Charles and Whitehead, 1986). Recent attention has been focused on inferring historical deposition of Hg to lakes, which has increased over the previous 100 years (Swain and Engstrom 1992). Results from large studies of lakes across North America suggest that declines have occurred in the previous 10 years in the midwestern lakes, but that deposition continues to increase for lakes west of the midwestern industrial centers, and in arctic regions (Engstrom and Swain, 1996, Hermanson, 1998, and Lockhart et al. 1998). Reductions are attributed to diminished Hg point source emissions. Most recently, archived Adirondack sediment samples from the PIRLA project (Charles and Whitehead, 1986) have been analyzed for Hg (Lorey and Driscoll, 1998). These analyses were used in part to compare sediment-inferred Hg deposition to measured atmospheric Hg deposition. The authors found that their core-derived estimates of modern atmospheric deposition of Hg were in good agreement with direct measurements, which gave confidence that core-derived estimates from deeper in the core could be used to estimate historical atmospheric deposition.

The Northeast States and Eastern Canadian Provinces Mercury Study (NESCAUM, 1998) recommends that "data on Hg emissions and deposition be statistically analyzed ... to enhance understanding of the air/water interrelationship." To this end, USEPA has entered into a cooperative agreement with the VTDEC to coordinate a Vermont-New Hampshire wide assessment of Hg and methylHg in sediments and waters of 90 lakes, following a regional EPA-EMAP (REMAP) sampling design. In conjunction with this project, a total of 12 paleolimnological sediment cores are to be collected, dated using  $^{210}\text{Pb}$ , and analyzed for total Hg at 15 depths downcore. The REMAP project is described in detail by Kamman et al. (1997).

In 1999, VMC supported the inclusion of a 13<sup>th</sup> lake, Branch Pond, in the Lye Brook Wilderness into the REMAP paleolimnology set. With respect to biogeochemistry, the critically acidic (Kellogg, 1998) Branch Pond has a variety of factors which the literature suggests should enhance methylation of Hg (Kellogg, 1998, N. Kamman, unpublished data). Among these are low pH, high dissolved organic carbon (DOC), anoxic hypolimnion, and large adjacent wetlands. Discussion regarding the specifics on how methylation is mediated by these parameters is beyond the scope of this report. A 19 year record of monitoring relating to acidification and biological conditions exists for Branch Pond (Kellogg, 1998; Kamman, 1998). Branch Pond was included in the early 1980's PIRLA project (Charles and Whitehead, 1986), but detailed results from that effort are not available as of this writing.

## Methods:

On August 19, 1999, a sediment core was acquired from the deep hole of Branch Pond (43°04.84'N, 073°01.19' W), in the main lake body (Figure 1), and in the southmost of two distinct hypolimnetic 'holes.' The core was acquired using a Glew-design gravity corer, equipped with a 70 cm x 6.5cm (i.d.) Lexan core tube, which was previously acidcleaned in 10% HNO<sub>3</sub>. The core was acquired from a depth of 10.1 meters, and was capped before breaching the surface to avoid any possible disturbance of the sediment-water interface.



**Figure 1.** Locator map for Branch Pond, Sunderland, VT, showing sediment coring location (!), and the watershed boundary.

Once in the boat, the core was de-watered by siphon and syringe to as close as practical to the sediment interface. The remaining overlying water was 'spilled' off without losing any surficial sediment. The core was then extruded in 1.0 cm intervals to the bottom of the core, with sediments transferred to lot-certified 125ml PETE bottles. A small aliquot of sediment was retained in 20ml PETE scintillation vials for elemental (%C, %N) and stable isotopic ( $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ ) analysis by other investigators. Throughout the extrusion and bottling procedures, USEPA method 1669 - 'Clean Hands - Dirty Hands' techniques were employed, using modifications developed for the REMAP study.

At the VTDEC LaRosa Environmental Laboratory, sediment subsamples from 15 one-cm strata were analyzed for Hg, percent solids, and loss on ignition (LOI). Hg was analyzed by aqua-regia digestion with detection by cold vapor atomic absorption spectroscopy on a Leeman<sup>®</sup> mercury analyzer. Percent solid was analyzed by dessication (80 °C) and gravimetry. LOI was analyzed by combustion (550 °C) and gravimetry.

At the St. Croix Watershed Research Station of the Science Museum of Minnesota, <sup>210</sup>Pb activity was measured for the core intervals by the method of Oldfield and Appleby (1984). Sedimentation rates and inferred dates were modeled from the <sup>210</sup>Pb activity using the 'Constant Rate of Supply' model by Dr. D. Engstrom. Fluxes of Hg to the lake sediments were calculated as the Hg concentration in each sediment interval multiplied by the sedimentation rate for the same interval. Flux rate units were scaled to  $\mu\text{g}\cdot\text{m}^{-2}\cdot\text{yr}^{-1}$  for consistency with other published studies.

References and detailed analytical procedures for all phases of collection, processing, and analytical chemistry are provided in Kamman et al. (1997).

## Results:

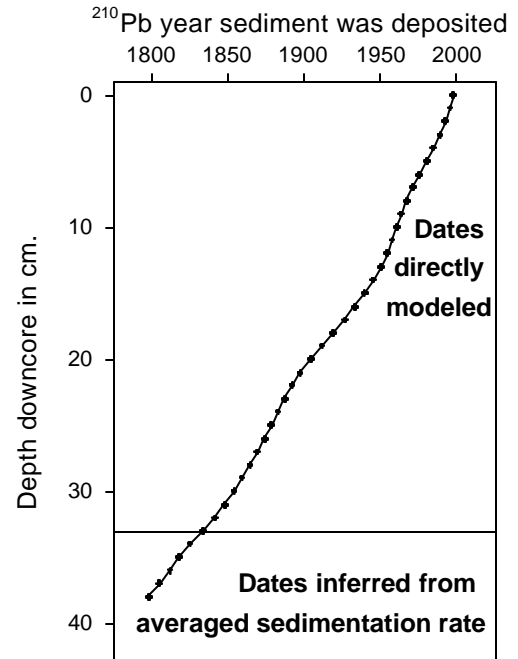
A clean core was acquired, which was 39 centimeters in total depth. The core displayed remarkable consistency in appearance, with no notable variation in lithology apparent by visual inspection. Sediments were brown gyttja. No laminations were apparent in the core. Throughout this text, each sediment interval identified (e.g. 5 cm to 12 cm) refers to the measurement, in centimeters, from the sediment-water interface.

Date values were estimated from measured  $^{210}\text{Pb}$  activity for even-numbered sediment intervals (e.g. 0-1 cm, 2-3 cm) to the maximum depth of 'unsupported'  $^{210}\text{Pb}$ , which represents the oldest sediment for which sufficient  $^{210}\text{Pb}$  remains to accurately estimate age. Date values for odd-numbered intervals were interpolated from measured intervals. For this core, the limit of unsupported  $^{210}\text{Pb}$  occurred at the 34 cm interval. For sediments below this, an averaged sedimentation rate for the 28 cm to 34 cm intervals was used to extrapolate date values for the remaining intervals. The relationship of sediment depth and  $^{210}\text{Pb}$  date on Branch Pond is shown in Figure 2.

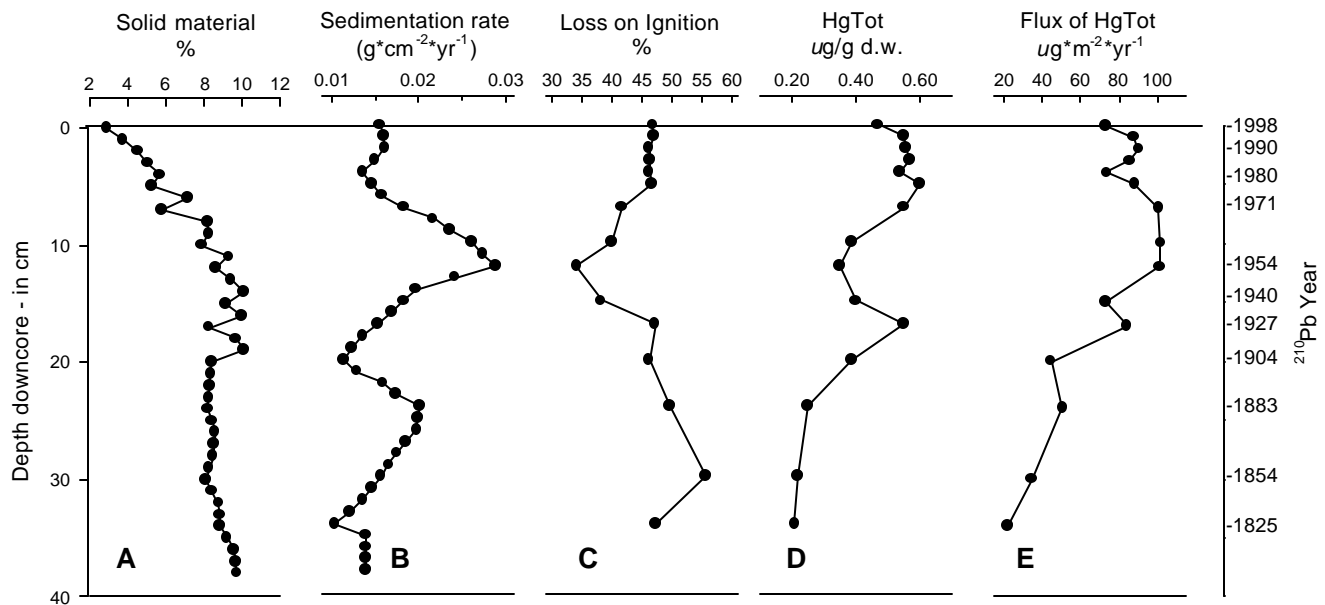
Percent solids ranged from near 3% at the surface-most muds, to approximately 8% in the core-bottom sediments (Figure 3A). There was an expected diagenetic increase in percent solid with depth from the core top to 7 cm. From 8 cm to 20 cm, the percent of solids was variable, clearly alternating between 8% and 10% by approximately 10-year cycles. From 20 cm to the core bottom, percent solids were remarkably homogenous with a slight rising trend from 30 to 38 cm.

Sedimentation rates directly reflect the degree to which material is deposited to the sediments. Increases in sedimentation rates can be attributed either to increased in-lake productivity, or to increases in watershed sediment loading. The sedimentation profile for Branch Pond is quite interesting (Figure 3B). Captured in the record provided by this sediment core are three periods of relatively low sedimentation, punctuated by two episodes during which sedimentation became quite high. In 1998, (the core top), the sedimentation rate was nearly as low as at any time in the record. Between 0 and 4 cm (to 1984), sedimentation was stable, averaging approximately  $0.0152 \text{ g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$ . From 5 cm to 12 cm (1984 to approx. 1954), sedimentation rate increased to the core maximum value of  $0.0289 \text{ g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$ . From 12 cm to 20 cm, sedimentation rates declined again to a low value of  $0.0114 \text{ g}\cdot\text{cm}^{-2}\cdot\text{yr}^{-1}$  at approximately 1904. A similar cycle is apparent in the 20cm to 34 cm interval. The peak sedimentation rate for this older cycle occurred at 24 cm (1883), and the subsequent decline ended at 34 cm (approx. 1825). This horizon marks the bottom of the unsupported  $^{210}\text{Pb}$  dating profile.

LOI is highly correlated to percent total carbon existing in the sediments, and is used in many paleolimnological investigations. Periods characterized by low sediment LOI are typically those during which land clearing or land management was occurring in the lakes watershed, since these activities result in a greater proportion of mineral sediments arriving into the lake relative to organic matter. This is due



**Figure 2.**  $^{210}\text{Pb}$  dating profile for a sediment core from Branch Pond, Sunderland, VT. Date estimates prior to the year 1825 are based on averaged core-bottom sedimentation rates.



**Figure 3.** Solid material (A), sedimentation rate (B), loss on ignition (C), Hg concentration (D), and Hg flux (E), inferred from a sediment core of Branch Pond, Sunderland, VT.

to increased erosion. Conversely, periods during which LOI is high would indicate times when the watershed was more stable.

Results for LOI in Branch Pond are shown in Figure 3C. From the core-top to 5 cm (1975), LOI averaged a consistent 46%. From 5 cm to 12 cm (1984 to approx. 1954), LOI decreased to the core minimum value of 34.1%. From 12 cm to 30 cm (1854), there was a steady increase in LOI, to the core maximum value of 55.8%.

The concentration of Hg in the sediments is presented in Figure 3D. The concentrations were near their peak levels at the core top, and rose to the core maximum value of  $0.6 \text{ ug@g}^{-1} \text{ d.w.}$  at 6 cm (approx. 1980). From 5 cm to 17 cm (1980 to approx. 1927), concentrations declined to a low of  $0.35 \text{ ug@g}^{-1} \text{ d.w.}$  and subsequently returned to a high of  $0.55 \text{ ug@g}^{-1} \text{ d.w.}$  Below 17 cm, concentrations trended towards baseline values of  $0.21 \text{ ug@g}^{-1} \text{ d.w.}$  at the core bottom.

The concentration profile for Hg in lake sediment cores will be directly affected by the rate of sedimentation which existed when the Hg was deposited into the lake. Given a constant atmospheric supply of Hg to the lake watershed, the concentration in the sediments will be reduced under periods of enhanced sedimentation, and increased under periods of diminished sedimentation (Dr. D.R. Engstrom, St. Croix Watershed Research Station, pers. comm.). This phenomenon is apparent in the concentration trend which occurs between 5 cm and 17 cm. This artifact can be corrected by calculating flux rates of Hg to the sediments, which are presented in Figure 3E. The resulting curve shows the estimated sedimentation-corrected Hg accumulation (flux) rates to sediments over time.

As with the concentration profile, fluxes were near their maximum value at the top of the core, and increased to the core maximum flux rate of  $102 \text{ ug@m}^2\text{@yr}^{-1}$  at 10 cm (approx. 1960). From that point, fluxes declined to baseline rates at the core bottom (at and presumably before 1825). In the middle of this

decline, there was a small spike at 17 cm (1927). This spike was clearly reflected in the concentration profile as well.

### Comparison with other Studies:

The flux rates measured in this analysis are elevated in relation to the remaining REMAP lakes, and are also elevated in relation to those measured for a set of Adirondack lakes by Lorey and Driscoll (1998), and for Wisconsin and Minnesota lakes cored by Engstrom and Swain (1996). This likely reflects the very high concentrations of Hg existing in the lake sediments as compared to lakes in these other studies. At face value this might call into question the validity of the measurements made for Hg. However, internal QC checks (including standard reference material) performed by the DEC laboratory in the course of this study produced excellent results. These high concentrations are probably the result of a high delivery of Hg to the lake from the very poorly buffered watershed, and nearby littoral wetlands, phenomena shown by many studies which are reviewed by Kamman (1998b). Table 1 provides a comparison of Branch Pond Hg concentrations and fluxes to the remainder of the REMAP study set.

In addition, the modern and peak flux ratios (ratio of modern or peak to baseline fluxes) for Branch Pond are 3.36 and 4.66 respectively, and these compare very well with the above mentioned studies as well as for the remaining REMAP set.

Table 1. Comparison of Hg concentrations and fluxes between Branch Pond and lakes in the REMAP paleolimnology study set.

Lake	Mean Hg ( $\mu\text{g g}^{-1}$ d.w.)	Mean sedimentation rate ( $\text{g cm}^{-2} \text{yr}^{-1}$ )	Mean HgT flux ( $\mu\text{g Hg m}^{-2} \text{yr}^{-1}$ )
All lakes (N= 11)	0.258	0.016	40.08
Branch Pond	0.44	0.018	74.06

### Discussion:

The literature indicates that a large component of the increases in flux observed for lake sediment profiles is attributable to atmospheric deposition. This signal is superimposed over that of Hg released from land use activities in the watershed (Engstrom and Swain, 1996; Hermanson et al., 1998; Lockhart et al., 1998; Lorey and Driscoll, 1998). Given data from only one lake, it is difficult to separate the effect of atmospheric deposition from that of watershed loading due to land use activities (S. Norton, Univ. of Maine, pers. comm.). Therefore it is useful to compare Hg flux profile to those within other similar lakes.

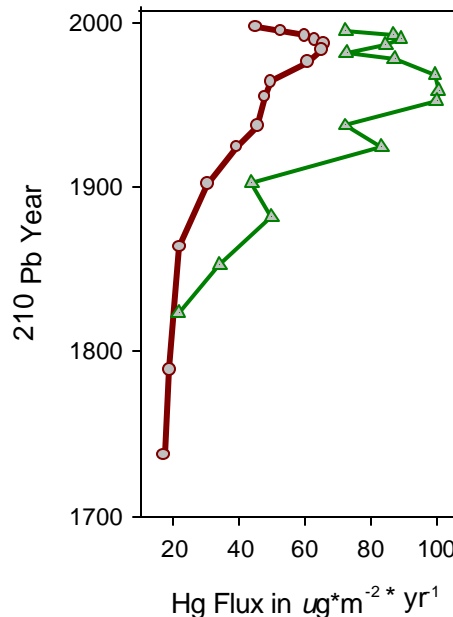
A generalized pattern of Hg flux to sediments is apparent in north-temperate lakes, as described by Engstrom and Swain (1997), Lorey and Driscoll (1998) and others. This pattern is also evident for nearly all lakes analyzed for the REMAP paleolimnology study set (N. Kamman, unpublished data), and for several lakes in Maine (S. Norton, pers. comm.). For lakes which have experienced only minimal watershed disturbance, there exists a pattern of slowly increasing fluxes above baseline levels, trending towards a peak at the period 1975 to 1980. Many cores show a subsequent decline, which Engstrom and Swain (1996) attribute to real reductions in atmospheric Hg deposition.

Lakes which occupy poorly buffered watershed experience a greater watershed delivery of Hg, and thus higher sediment Hg concentrations, than do lakes which are well buffered Kamman (1998b). This is due to binding of the Hg onto watershed-delivered DOC (Driscoll et al., 1994). This may be accentuated during periods of high runoff due to the binding of Hg to particulate matter, as suggested by Hg stream streamflow monitoring data from the Lake Champlain Basin of Shanley et al. (1999). While the generalized signal of atmospheric contribution is clearly apparent in the sediments of Branch Pond, the pattern appears punctuated by episodes of sedimentation due to land use in the watershed (Figure 4).

The Branch Pond sediment record identifies two periods of disturbance in the watershed. The older period began approximately in 1825. Historically, the area in the vicinity of what is now the Lye Brook Wilderness was in farm ownership (Dr. C. Cogbill, pers. comm.), and the land use at that time could be characterized as low-intensity agricultural / pasture use. No specific information is available about land use in the Branch Pond watershed at that time. From 1825-1854 LOI values were high and increasing, which suggests a stabilizing watershed. Thus, it could be reasonably hypothesized that the first cycle of sedimentation to the lake, which peaked at 1887, was related to low-intensity agriculture, and that the enhanced sedimentation rate was the manifestation of enhanced in-lake productivity as opposed to erosion from the watershed. Accumulation of atmospherically deposited Hg, as documented in lake sets throughout North America, began between 1850 and 1875 (Engstrom and Swain, 1996; Hermanson et al., 1998; Lockhart et al., 1998; Lorey and Driscoll, 1998; N. Kamman, unpublished data; S. Norton, pers. comm.). Thus, the signal of increasing Hg flux to Branch Pond sediments, clearly in evidence by 1854, is most likely an atmospheric one, but may be enhanced by the localized land use in the watershed.

During the period between 1880 and 1905, the record indicates that Branch Pond was in a period of reducing sedimentation. Forest regrowth may have been occurring at this time. During this period, HgT concentrations and fluxes continued to rise, suggesting a continual contribution of atmospherically derived Hg. Indeed, in 1927, there is a strong peak in the concentration of Hg. This might indicate an atmospheric pulse, or may be related to the onset of forest management activities as discussed below.

The second period of sedimentation, which began in 1904 and peaked by 1954, was likely attributable to forest management within the watershed. Inspection of aerial photographs taken in 1942 identify heavy logging in the Northwest portion of the Branch Pond watershed. This photo clearly shows a network of skid roads and small tracks which approach very near to the lakeshore from the northwest. During this time, Hg fluxes increased steeply, peaking in 1954, co-incident with the record-maximum sedimentation rate, and core minimum LOI. Thus, some of the Hg entering the lake sediments at this



**Figure 4** Historical Hg flux rates for Branch Pond (Δ) in relation to the generalized flux pattern expected when accumulation of Hg to sediments is largely due to atmospheric deposition to a minimally disturbed watershed (•). The profile used for comparison purposes is from Wallingford Pond, Wallingford, VT, which was analyzed in conjunction with the REMAP paleolimnology study set.



time was likely flushed from the watershed during cutting.

From 1954 to 1971, Hg fluxes remained stable during a period of decreasing sedimentation, indicating continually increasing atmospheric flux. Aerial photography from 1962 shows the watershed as being completely forested, with the network of logging roads evident on the 1942 photography regrown. The 1974 photography identifies the construction of the first major improved dirt road to penetrate the Branch Pond watershed and allow public access to the pond from the Arlington-Stratton ('Kelly Stand') Road. This road was drastically widened and improved in recent years, and this activity is evident in the slight sedimentation pulse which occurred in the mid 1980's. This activity and resulting watershed disturbance likely released some Hg which was stored in that section of the watershed, thus masking the expected reduction in fluxes of atmospherically deposited Hg which are idealized by the Wallingford Pond flux profile (Figure 4). Finally, in present times, there are decreases in Hg fluxes based on the top centimeters, but these should be interpreted cautiously since surficial core sediments are prone to disturbance during sampling.

### Conclusions:

Fluxes of Hg to Branch Pond are driven largely by atmospheric deposition, but mediated by watershed-based activities.  $^{210}\text{Pb}$ -inferred sedimentation rates suggest that prior land-use activities played a large part in accelerating the delivery of Hg to the sediments of Branch Pond. Based on this analysis, it can be reasonably suggested that Branch Pond is biogeochemically sensitive to Hg accumulation, and accordingly, to bioaccumulation of Hg up the trophic chain.

Hg concentrations in and fluxes to the sediments have been elevated above what would be considered baseline since at least 1850. Across the REMAP set, and to a lesser degree in Branch Pond itself, there exists evidence of declines in modern Hg flux rates. These declines may be linked to the reduction in atmospheric deposition of Hg across the landscape. Evidence from the Branch Pond core suggests that, should these atmospheric improvements continue, surficial sediment Hg concentrations would continue to decline. Over the long term, this may result in a reduced source of both hypolimnetic and watershed-Hg available for methylation and subsequent bioaccumulation.

### Recommendations:

This analysis shows that Hg fluxes to the sediments of Branch Pond are high in relation to other lakes in the REMAP paleolimnology study set. This is most likely attributable to enhanced delivery of Hg from the Branch Pond watershed. Branch Pond is the only lake to be cored and analyzed for Hg in the Lye Brook Wilderness area. Several lakes exist in and near the Lye Brook Wilderness which would serve as excellent sites to perform additional Hg core analyses. Bourn Pond, Stratton Pond, Grout Pond, and Beebe Pond are all presently forested, and given their proximity to each other, were all very likely subjected to similar historic land uses.

The present research would be enhanced by coring these lakes, dating the sediments, analyzing them for Hg and for the stable isotopes  $^{13}\text{C}$  and  $^{15}\text{N}$ . Such analyses would permit more robust inferences as to the provenance of the materials which constitute the sediments, and thus clarify the relative importance of land use in interpreting the signal of atmospheric Hg deposition across the Lye Brook region. From the standpoint of developing a paleolimnological proxy for estimating atmospheric Hg contributions to the Lye Brook region, the combined results of several lake cores would be extremely useful, since they would permit direct estimates of current and historical localized Hg deposition rates.

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