

**Paleolimnology of Lakes in the Vicinity of the Lye Brook Wilderness,  
Vermont, USA.**

**<sup>210</sup>Pb Dating, Mercury and Carbon Isotopes**

**prepared for:**

**United States Forest Service  
Green Mountain and Finger Lakes National Forest**

**and**

**Vermont Monitoring Cooperative**

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Prepared by:

Neil Kamman, Environmental Scientist  
Vermont Department of Environmental Conservation  
103 S Main 10N  
Waterbury VT 05671-0408

Additional data, interpretation, and information supplied by:

Dr. Daniel Engstrom, Science Museum of Minnesota  
Dr. Andrea Lini, University of Vermont, Department of Geology  
Mr. Ken Oldrid, University of Vermont, Department of Geology

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## 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 Hg contamination of VT lakes will necessitate reductions in discharges and emissions of Hg both regionally and nationally. Paleolimnological estimation of atmospheric Hg deposition to lakes provides a long-term, reproducible indicator of Hg deposition to the northeastern landscape. The purpose of this report is to present results of a paleolimnological analysis performed on four lakes in the vicinity of the Lye Brook Wilderness (LBW), of southern Vermont, and to relate these findings to a designed paleolimnological study of lakes across Vermont and New Hampshire (Kamman and Engstrom, 2002).

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 since approximately 1875 (Swain and Engstrom 1992). Results from several studies show recent declines in Hg fluxes to Midwest and northeast lakes (Engstrom and Swain, 1996; Landers et al.; 1998, Kamman and Engstrom, 2002), although these declines are not noted for lakes which are not downgradient of Midwestern Hg emission sources (Engstrom and Swain, 1996; Hermanson, 1998; Lockhart et al. 1998). Locally, archived Adirondack sediment samples from the PIRLA project (Charles and Whitehead, 1986; Lorey and Driscoll, 1998), and a series of 10 VT and NH lakes (Kamman and Engstrom 2002) have been analyzed for historical Hg deposition. 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.

Elemental and stable isotopic ratios of carbon and nitrogen in sediments have shown promise in interpreting the trophic history of lakes (Brenner et al., 1999). Elemental carbon-nitrogen ratios have been used to infer the source of organic sediment constituents (Meyers and Lallier-Vergès, 1999), and stable isotopic ratios of  $^{13}\text{C}/^{12}\text{C}$  have been successfully used to interpret long-term trophic histories of Lakes Erie and Ontario, and of several Florida lakes (Hodell and Schelske, 1998; Schelske and Hodell, 1991 and 1995). The use of elemental and stable isotopic ratios is developing in Vermont.

Current research into Hg across the VT-NH region in conjunction with on-going Regional Environmental Monitoring and Assessment Program (REMAP) studies (Kamman et al., 1997, 2002, 2003) and state fish tissue monitoring activities (Langdon, 1994 and revs.) suggests that a large Hg signal is evident across the southern portion of Vermont, particularly in the vicinity of LBW. Specifically, The surficial total Hg concentration in Branch and Stratton Pond sediments are elevated above VT-NH averages. In addition, wet deposition of Hg is modeled to be among the highest observed across the VT-NH region (Miller, 2002). The high elevation plateau of the Lye Brook wilderness supports numerous ponds and wetlands, which act as excellent sites for the production of methylmercury. Finally, Hg is elevated in several species of fish in several ponds, including Grout Pond and Somerset Reservoir. This combined weight of evidence suggests that ponds within LBW are at enhanced risk of Hg contamination relative to other parts of Vermont. The purpose of the present study is therefore to quantify, using paleolimnological techniques, the historical and current Hg loading to these lakes, and to use isotopic analyses to shed light on the individual lake trophic histories.

Reported in the present document are the analytical results of sediment cores collected from Bourn, Branch, Grout, and Stratton Ponds in the vicinity of LBW. Results are provided for sediment solids,  $^{210}\text{Pb}$ -inferred dates and sedimentation rates, C and N isotopic analyses, and Hg concentrations and fluxes. Limited interpretation of the profiles for each lake is provided, and the Hg fluxes observed in these lakes are related to those measured from other minimally-disturbed lakes in VT-NH. Sediment phosphorus concentrations for Stratton Pond are provided by request of USFS, and are not interpreted in light of the paleolimnological results. A brief historical sketch is provided for Bourn, Grout, and Stratton Ponds.

## Method Summary

### *Site characteristics*

The LBW occupies a relatively high-elevation plateau situated between routes VT30 and VT9 to the north and south, and VT100 and US7 to the east and west. LBW and the nearby vicinity contains numerous small ponds, and several larger ones, including Branch, Bourn, Grout, Stratton, Beebe and Lost Ponds, Lye Brook Meadows, and the northern branches of the Somerset Reservoir. Branch and Bourn Ponds (Sunderland, VT), and Grout and Stratton Ponds (Stratton, VT) were included in the present study. Coring locations, and basic lake information are provided in Table 1. The geographic locations of the lakes and sampling locations are displayed on Figure 1.

**Table 1. Characteristics and sampling locations of four lakes in the vicinity of the Lye Brook Wilderness included in this study.**

Lake Name	Coring location Lat., Long. (DD MM SS)	Coring date	Mean long-term alkalinity (mg/l CaCO <sub>3</sub> )*	Lake elevation (ft, m.s.l.)	Lake size (ac)	Watershed:lake area ratio
Bourn Pond	43 06 19 -073 00 16	8/8/2001 7/12/02	0.46	2,552	34	8.54
Branch Pond	43 04 56 -073 01 10	8/8/1999	-0.62	2,632	48	9.71
Grout Pond	42 02 34 -072 56 34	8/8/2001	1.44	2,225	84	4.43
Stratton Pond	43 06 17 -072 58 10	8/9/2001 7/13/2002	1.14	2,555	46	5.78

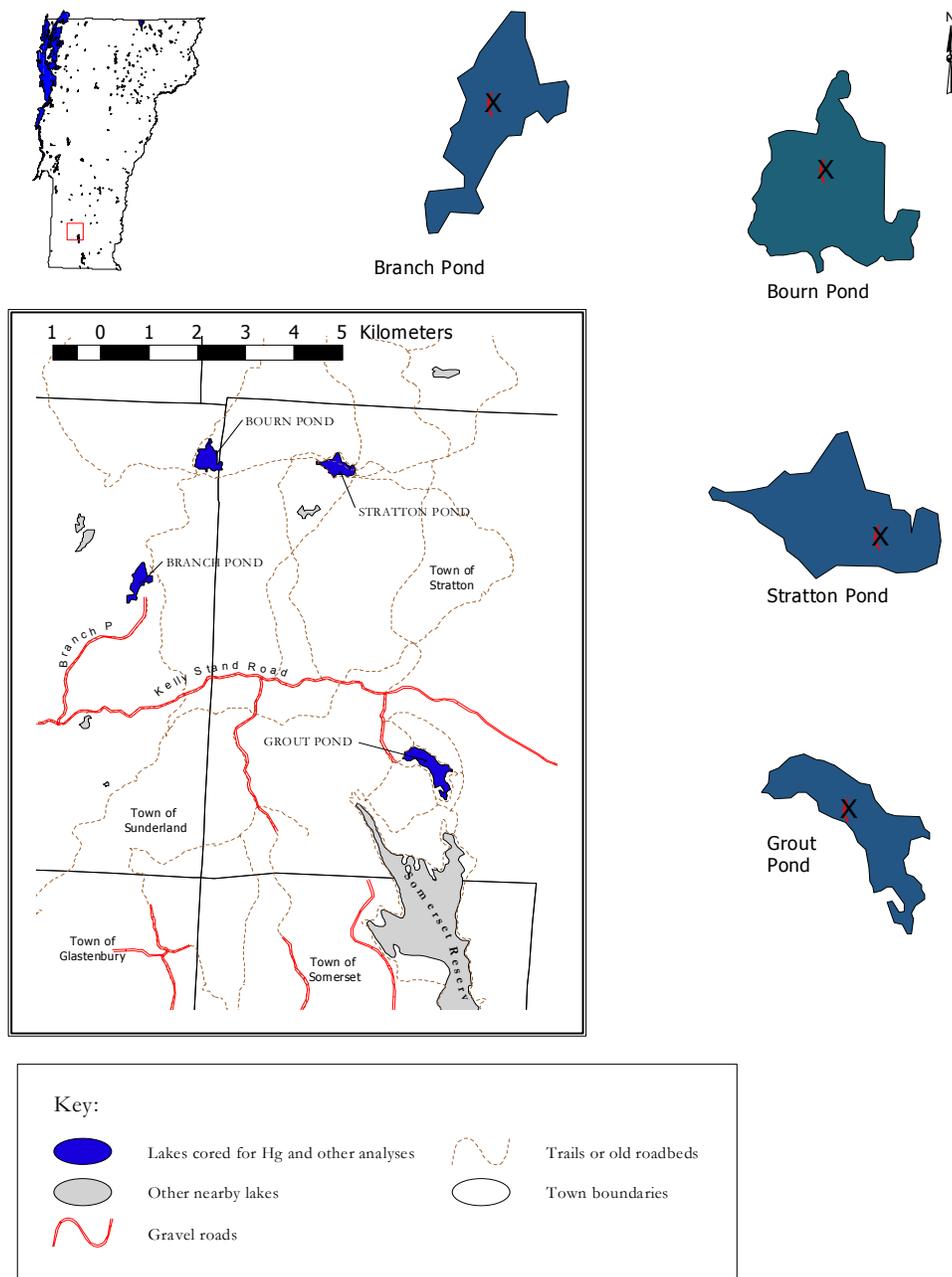
\* VTDEC (2001).

### *Field methods*

In the field, sediment cores were acquired from the known deep hole, using a gravity corer equipped with a 6cm by 60cm lexan tube. Sampling equipment including core tubes and extrusion tools were prepared as per Kamman (1997) prior to use. Paired cores were acquired at the deep location of each lake, and that core reflecting the least apparent disturbance and greatest depth was retained for further processing and analysis. Sediments were extruded at one-cm intervals from the core top, split, and stored in lot-certified, 250 ml polyethylene Nalgene™ bottles, at 4°C. Sample aliquots were split for analysis of Hg, <sup>210</sup>Pb, and elemental and isotopic analysis. On Stratton Pond, additional surficial samples were collected for total sediment phosphorus analysis, to support specific USFS needs.

### *<sup>210</sup>Pb dating*

Sediment dating was performed at the St. Croix Watershed Research Station, Science Museum of MN, under the direction of Dr. Dan Engstrom. In summary, dried sediments were counted radiometrically using alpha- and gamma counters, and resultant pCi values were converted to sedimentation rate and date values following the Constant Rate of Supply dating model. Complete detail for sediment dating procedures is provided in Kamman and Engstrom, 2002.



**Figure 1. Site map, and location and plan views of four lakes cored for historical Hg analysis in the vicinity of the Lye Brook Wilderness.**

Sediment total sediment Hg was analyzed at the VTDEC LaRosa Environmental Laboratory. In brief summary, dried (60 °C) sediment aliquots were digested in aqua-regia, with resulting Hg oxidized to Hg<sup>0</sup>, reduced using SnCl<sub>2</sub>, and carried by Ar gas into a Leeman™ cold-vapor atomic absorption mercury analyzer. This procedure follows USEPA Method 245.1 (USEPA, 1983), and is also described in detail by Kamman and Engstrom (2002).

### ***Isotope geochemistry***

To remove inorganic carbon (e.g. carbonates), dried sediments were treated with 6ml 0.1M HCl, homogenized by vortexing, and allowed to sit uncovered in a hood for 24 hours. Percent total organic carbon (%Corg) and percent total nitrogen (%N) were measured using a CE Instruments™ NC2500 elemental analyzer. The Corg/N ratio was calculated as %Corg / %N for each interval.

For C isotopic ratio determinations, no more than 10 mg of sediments from 24 core intervals were sealed into quartz tubes containing 600 mg cupric oxide, quartz wool, and 500 mg elemental copper. The tubes were combusted at 800°C for two hours. CO<sub>2</sub> gas in isotopic equilibrium with sediment Corg was cryogenically isolated from these tubes on a vacuum line, and introduced into a VG-Isogas™ SIRA mass spectrometer. Isotopic composition was determined by comparison with internal reference CO<sub>2</sub> gas. Isotopic carbon ratios in the sediments are expressed in the delta (δ) notation, as the parts-per-thousand, or per mil (‰) deviation from the international standard PeeDee Belemnite limestone.

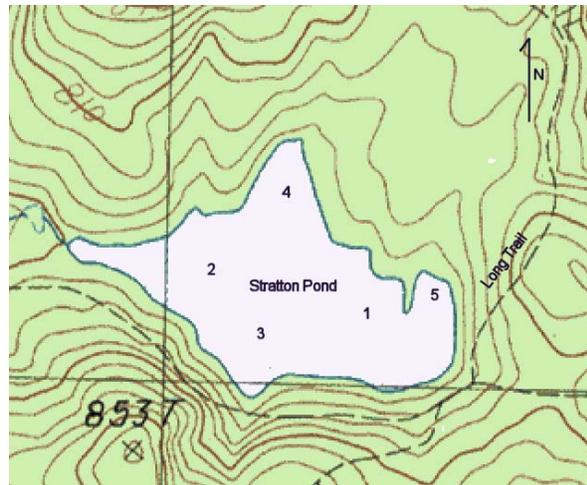
### ***Phosphorus***

Sediment total phosphorus was analyzed from wet-sediment aliquots by persulfate digestion following USEPA Method 365.1 (USEPA, 1983). Total phosphorus concentrations were adjusted to dry-weight values using percent solid values for Stratton Pond. Figure 2 shows sampling locations for sediment phosphorus locations.

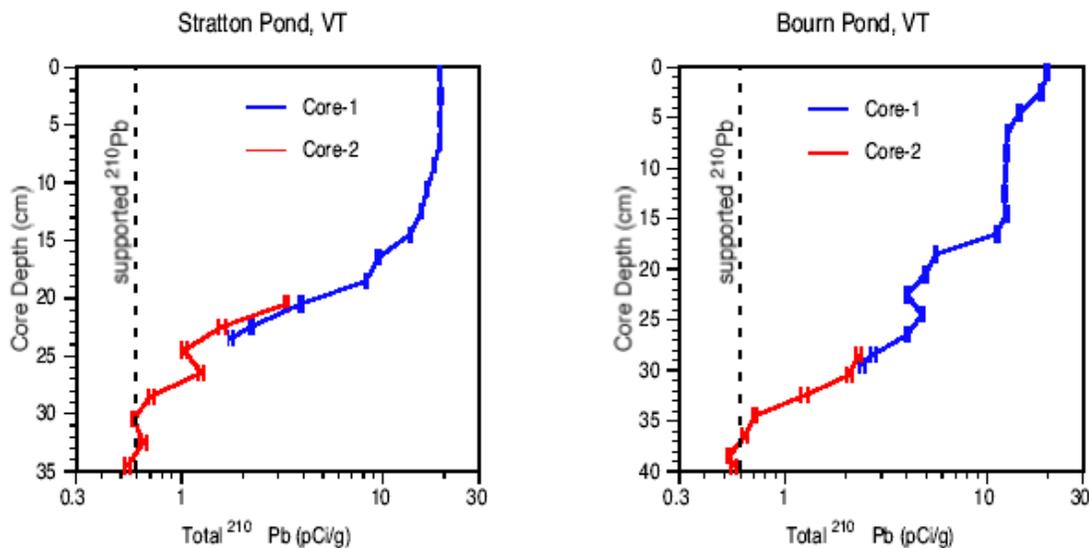
## **Data Results**

### ***<sup>210</sup>Pb dating and sedimentation rates***

All four cores reached stable supported <sup>210</sup>Pb with depth, and show the anticipated exponential decay with time. Between two and four supported strata are available at depth for each core. Initial <sup>210</sup>Pb results indicated that for two of the lakes (Bourn and Stratton), the cores were not sufficiently deep to achieve supported levels of <sup>210</sup>Pb. As an estimate of supported, or baseline <sup>210</sup>Pb is critical to establishing a date profile for a sediment core, these lakes were revisited during 2002 with the goal of acquiring deeper sediments which would permit measurement of the background <sup>210</sup>Pb levels. These additional samples were acquired in July of 2002, and resulting deep-sediment <sup>210</sup>Pb profiles were plotted with those developed from the 2001 cores (Figure 3).



**Figure 2. Locator map for surficial total phosphorus sampling locations in Stratton Pond**



**Figure 3. Total  $^{210}\text{Pb}$  activity plotted from cores collected in 2001 and 2002, for Stratton and Bourn Ponds.**

Fortunately, the 2001 and 2002 cores matched up well, and data from both cores were thus pooled to calculate depth-age profiles. Unsupported fluxes ranged from 0.41 to 0.68  $\text{pCi cm}^{-2} \text{yr}^{-1}$  (Table 2). These values are consistent with regional atmospheric  $^{210}\text{Pb}$  deposition rates, implying that core-specific sedimentation rates are not significantly amplified by sediment focusing (Kamman and Engstrom, 2002). In summary, all four cores provided reliable date estimates from 1850 or before to present, including quantifiable sediment accumulation rates and errors. Lead-210 fluxes and other pertinent information regarding sediment dating are provided in Table 2. Age-depth profiles for all four lakes are shown in Figure 4.

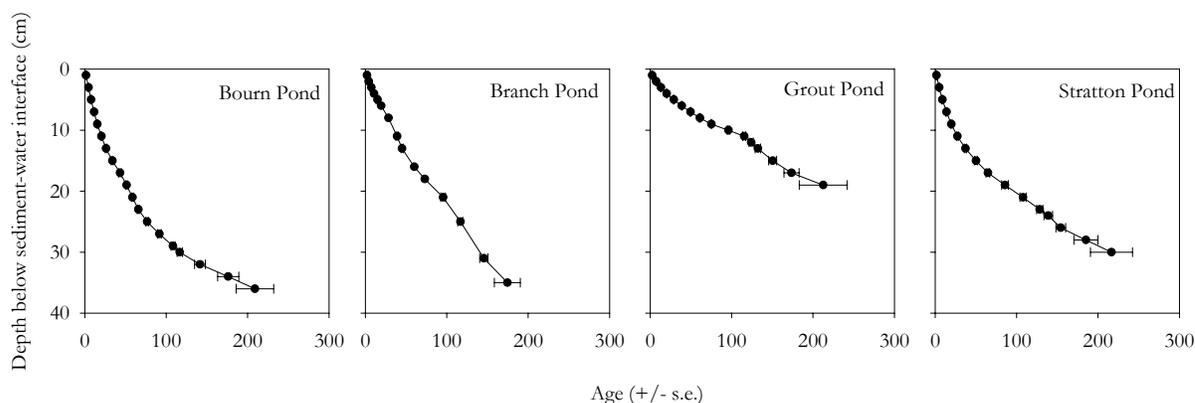
**Table 2. Supported and unsupported  $^{210}\text{Pb}$  fluxes: four Lye Brook Wilderness vicinity lakes.**

Lake	Supported $^{210}\text{Pb}$ $\text{pCi g}^{-1}$	Supported $^{210}\text{Pb}$ error	N supported samples	Cumulative unsupported $^{210}\text{Pb}$ $\text{pCi cm}^{-2}$	Unsupported $^{210}\text{Pb}$ flux $\text{pCi cm}^{-2} \text{yr}^{-1}$
Bourn Pond	0.5657	0.0298	3	21.1412	0.6819
Branch Pond	1.1411	0.0219	2	12.7846	0.4103
Grout Pond	0.2903	0.0278	4	16.4215	0.5299
Stratton Pond	0.5832	0.0321	3	20.9571	0.6762

Sedimentation rates for the four lakes ranged between 4.3 to 10.4  $\mu\text{g cm}^{-2} \text{yr}^{-1}$  prior to 1850, and ranged between 13.6 and 31.4  $\mu\text{g cm}^{-2} \text{yr}^{-1}$  at the core tops. These rates are in excellent agreement with values published for 10 VT-NH lake cores in Kamman and Engstrom (2002).

### *Hg*

Total Hg concentrations in the sediment cores averaged between 0.134 to 0.229  $\mu\text{g g}^{-1}$  (d.w.) prior to 1850, and ranged between 0.266 and 0.468  $\mu\text{g g}^{-1}$  at the core tops. These concentrations also agree very well with the results of Kamman and Engstrom (2002), although concentrations from Branch Pond can be considered high (peak concentration of 0.599  $\mu\text{g g}^{-1}$  for the sediment interval corresponding to 1980).



**Figure 4. Age-depth profiles derived using  $^{210}\text{Pb}$  fluxes. Bars indicate standard errors of age estimates.**

### *C/N ratios and $\delta^{13}\text{C}$*

Elemental Corg/N ratios in sediments are informative as to whether the source of organic sediment fractions is derived from terrestrial (e.g., foliage) or in-lake (e.g., algal) primary production (Meyers and Lallier-Verges, 1999). In general Corg/N ratios in excess of 20 indicate primarily terrestrially-derived primary production, while values below 10 indicate that the organic fraction of sediments is derived primarily from in-lake algal production. Intermediate values are common in Vermont lakes, rendering interpretation challenging. However, the direction of sedimentary Corg/N change also provides key insight into the trophic trend for individual lakes. Declining trends are typically a signature of reduced watershed-based inorganic sediment delivery, increased in-lake algal productivity, or a combination of both phenomena. Corg/N ratios varied within a range of 9.5 to 12.5, and displayed declines with time.

Algae and terrestrial plants also display differences in their isotopic composition. Photosynthetic organisms exhibit a preference for  $^{12}\text{C}$ , which is the naturally-occurring stable carbon isotope of least mass. This preference for the lighter carbon isotope is due to its preferential incorporation by a key enzyme which mediates photosynthesis. Terrestrial plants utilize atmospheric  $\text{CO}_2$  for photosynthesis, and due to the abundance and high rate of mixing of this carbon source, land plants exhibit a relatively constant isotopic composition of around  $-27\text{‰}$  (Boutton, 1991). By contrast, aquatic biota such as algae and aquatic macrophytes utilize dissolved inorganic carbon in the water as their source of carbon, and thus they exhibit a wider range of isotopic compositions which tend to reflect in-lake carbon sources.

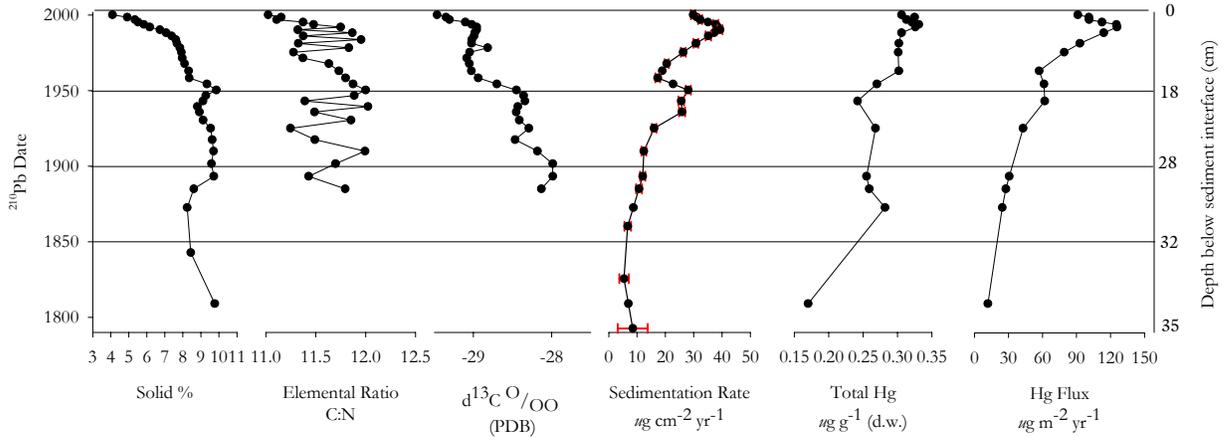
Because of the differences in  $^{12}\text{C}$  and  $^{13}\text{C}$  utilization by aquatic algae and plants, isotopic carbon ratios (called  $\delta^{13}\text{C}$ ) also provide insight into the direction of trophic change in lakes. Algae in oligotrophic lakes are more enriched in  $^{12}\text{C}$  than are terrestrial plants, whereas in eutrophic lakes they are more depleted in  $^{12}\text{C}$ . These chemical differences act as tracers of the source of organic matter in lake sediments, whereby declining values of  $\delta^{13}\text{C}$  are indicative of increasingly productive (e.g., eutrophic) conditions. For the four LBW lakes,  $\delta^{13}\text{C}$  ranged from  $-28.0$  to  $-31.0\text{‰}$  (PDB).

## **Individual Lake Profiles**

### ***Bourn Pond:***

The core acquired from Bourn Pond was 35 centimeters deep, and yielded a well resolved dating profile back to approximately 1792. Percent solids in the core increased with depth to 18cm (1950), and were relatively stable thereafter, ranging between 9% and 11% (Figure 5). This pattern is common and consistent with many other lake cores, and reflects a normal diagenetic signal of increasing sediment density with depth. Elemental Corg/N ratios were highly variable. Increasing %C accompanied by decreasing C/N ratios and  $\delta^{13}\text{C}$  values indicate a moderate increase in algal productivity. This change is associated with deforestation events that occurred between 1850 and 1950. Isotopic  $\delta^{13}\text{C}$  was stable from 1925 to 1950, and again from 1960 to 1990. Recent  $\delta^{13}\text{C}$  are the lowest of the entire core. Sediment accumulation ranged from a stable baseline of  $\sim 10$  to  $\sim 15\ \mu\text{g cm}^{-2}\ \text{yr}^{-1}$  from 1792 to  $\sim 1925$ , and

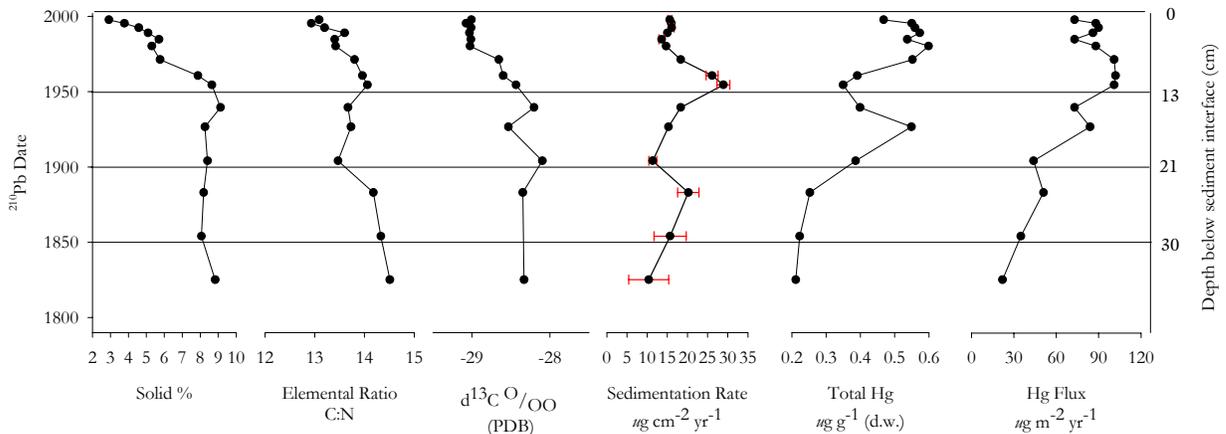
then increased to a peak of  $\sim 30 \mu\text{g cm}^{-2} \text{yr}^{-1}$  at 1950. This increase was coincident with an increase in sediment percent solid, suggesting a significant sedimentation event in the Bourn Pond watershed at that time. Records indicate that between 1953 and 1956, 3.5 million board-feet of timber were logged around Bourn Pond. From  $\sim 1960$  to 1985, sedimentation increased to the core maximum of  $38.7 \mu\text{g cm}^{-2} \text{yr}^{-1}$ , declining thereafter to present values. Although  $\delta^{13}\text{C}$  varied only within a narrow range, the negative trend which corresponds closely to reductions in sedimentation rate suggests that Bourn Pond recovers quickly from sedimentation events. Hg fluxes increased slowly from a baseline of  $\sim 15 \mu\text{g m}^{-2} \text{yr}^{-1}$  to a peak of  $125.7 \mu\text{g m}^{-2} \text{yr}^{-1}$  in 1992, and showed a decline from 1992 to present.



**Figure 5. Profiles of percent solid, C:N ratio,  $\delta^{13}\text{C}$ , sedimentation rate, and Hg concentration and flux rates, for Bourn Pond, Sunderland, VT.**

***Branch Pond:***

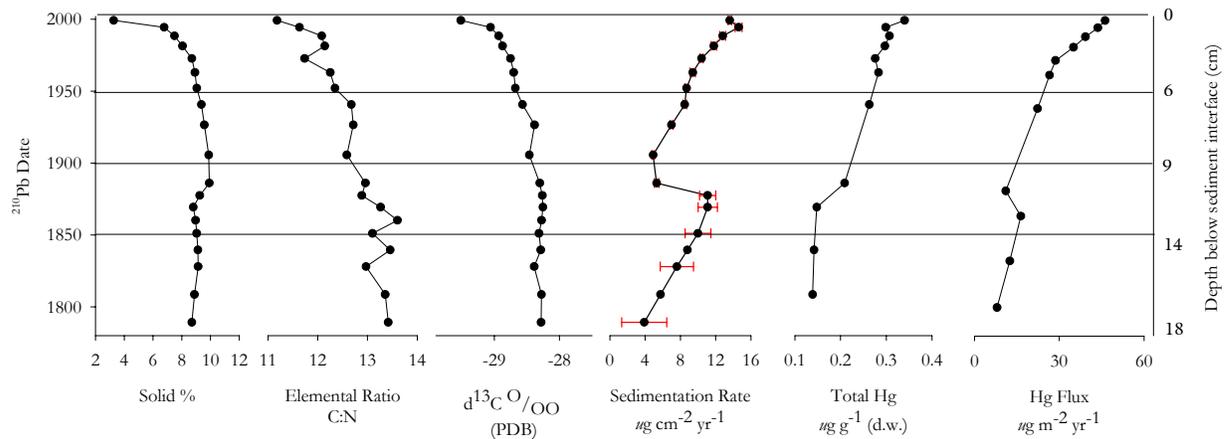
The core acquired from Branch Pond was 35 centimeters deep, and yielded a well resolved dating profile back to approximately 1825. Percent solids in the core increased to 15cm (1940), and were relatively stable thereafter, ranging between 9% and 10% (Figure 6). This pattern is common and consistent with many other cores, and reflects a normal diagenetic signal of increasing sediment density with depth. Elemental Corg/N ratios were slightly variable, but showed a distinct decline with time, with a pulse ca. 1940. Isotopic  $\delta^{13}\text{C}$  was stable from 1825 to 1900, declined from 1940 to  $\sim 1975$ , and remained remarkably stable from 1975 to present. Sediment accumulation showed baseline rates of  $10 \mu\text{g cm}^{-2} \text{yr}^{-1} \sim 1825$ , then increased to a peak  $\sim 1880$ , then another significant peak at 1955 ( $28.9 \mu\text{g cm}^{-2} \text{yr}^{-1}$ ). This latter peak was the result of logging activities (Kamman, 1999), and the influence of this event is evident in the profiles of Hg concentrations and fluxes.



**Figure 6. Profiles of percent solid, C:N ratio,  $\delta^{13}\text{C}$ , sedimentation rate, and Hg concentration and flux rates, for Branch Pond, Sunderland, VT.**

## Grout Pond

The core acquired from Grout Pond was only 19 centimeters deep, but yielded a well resolved dating profile back to approximately 1789. Percent solids in the core were stable at  $\sim 9\%$ , from 1789 to 1975, and declined thereafter. (Figure 7). The stability of percent solids in this core is greater than what is commonly observed. Elemental Corg/N ratios declined slowly but steadily from  $\sim 1850$  to present, suggesting an increase of algal productivity. The  $\delta^{13}\text{C}$  was very stable as well. Overall, these show a lake that has experienced very little trophic change over time. Sedimentation rates in Grout Pond were also low, ranging from  $4 \mu\text{g cm}^{-2} \text{yr}^{-1}$  to near  $14 \mu\text{g cm}^{-2} \text{yr}^{-1}$  presently. Although logging communities were prevalent in the vicinity of Grout Pond, little is known about the duration and extent of the logging operation, nor of any alterations to the lake hydrology (e.g. dams) that may have accompanied the logging. The shape of the sedimentation profile indicates that an event, circa 1880 resulted in a minor but rapid decline in sediment accumulation, and this is reflected by small adjustments in all other profiles. However, following that short decline, sedimentation rates have continued to increase to the present time, with Hg concentrations and fluxes showing concordant trends. This change in sedimentary characteristics could be related to the building and operation of a sawmill just north of Grout Pond in 1874. Hg fluxes in Grout Pond show no decline at the most recent core-intervals, which is at odds with Bourn and Branch Ponds, and with the lakes described by Kamman and Engstrom (2002). Current Hg flux rates to Grout Pond sediments are, however, relatively low at  $46.2 \mu\text{g m}^{-2} \text{yr}^{-1}$ .

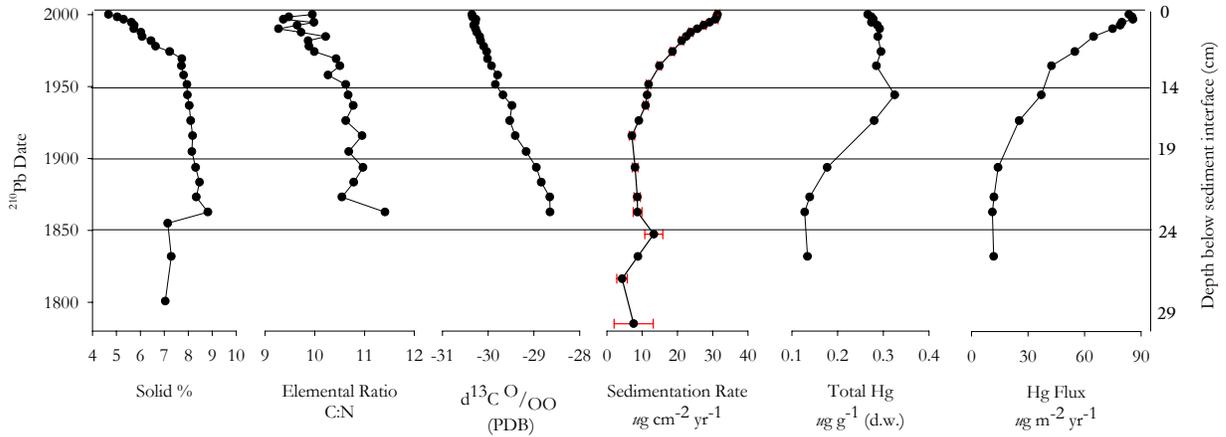


**Figure 7. Profiles of percent solid, C:N ratio,  $\delta^{13}\text{C}$ , sedimentation rate, and Hg concentration and flux rates, for Grout Pond, Stratton, VT.**

## Stratton Pond

The core acquired from Stratton Pond was 30 centimeters in length, and provided a well resolved dating profile to 1785. Percent solids were stable throughout the core from 1785 to 1970, at which time they declined rapidly to a core minimum value of 5% showing a standard diagenetic signal (Figure 8). The offset in percent solids prior to 1860 is an artifact of the splicing of the 1999 and 2000 cores discussed above (Figure 2), and should not be interpreted to represent any change in the lakes water quality status. Elemental Corg/N ratios declined steadily throughout the lakes recent history, punctuated by minor oscillations. The core minimum values of  $< 10$  indicate that sediments laid down after  $\sim 1970$  are mostly derived from algal particles, suggesting an increase in this lake's trophic status. The consistent, nearly linear decline in  $\delta^{13}\text{C}$  to the most negative values ( $-31^{0}/_{00}$  P.D.B) in the present set of lakes corroborates this inference. Around 1830, the Kelley Stand Road was built to connect the towns of Stratton and Arlington. Stratton Pond is located approximately 4 miles north of the road, and is adjacent to a secondary, but well-established spur of the Kelley Stand Road known as the "I.P. Road." These roads were built to allow the harvest of timber from the previously inaccessible wilderness, among other uses. Sedimentation rates in Stratton Pond were low (mean of  $8.8 \mu\text{g cm}^{-2} \text{yr}^{-1}$ ) for the period 1785-1950, but then increased rapidly to a core maximum value of  $31.4 \mu\text{g cm}^{-2} \text{yr}^{-1}$  presently. This value is the highest contemporaneous sedimentation rate for the present set of lakes, and taken in concert with the Corg/N and  $\delta^{13}\text{C}$  profiles, demonstrates an increase in productivity and therefore trophic status of this lake in the last 50 years. The major logging event of 1953 (see the description for Bourn Pond) by the Sweezy Company could have extended into the watershed of Stratton Pond, which would have contributed to the uninterrupted increase in

sedimentation rate observed there beginning after 1900. Total Hg concentrations increased to a modest peak value of  $0.32 \mu\text{g g}^{-1}$  in 1945, and have stabilized since then. Hg fluxes, however, continue to increase to a current rate of  $85 \mu\text{g m}^{-2} \text{yr}^{-1}$ , owing to continuing increases in sediment accumulation in this pond



**Figure 8. Profiles of percent solid, C:N ratio,  $\delta^{13}\text{C}$ , sedimentation rate, and Hg concentration and flux rates, for Stratton Pond, Stratton, VT.**

Wet and estimated dry-weight sediment total phosphorus for composited surficial sediments (0cm-5cm) of Stratton Pond ranged from 38,157 to 52,213  $\mu\text{g l}^{-1}$  and from 704 to 991  $\mu\text{g g}^{-1}$ , respectively, as reported in Table 3.

**Table 3. Total phosphorus concentrations measured from the top five centimeters of sediments, from five locations in Stratton Pond.**

Location	TP $\mu\text{g l}^{-1}$	TP $\mu\text{g g}^{-1}$ d.w.	Remark
01	42,928	815	Dry-weight values estimated by percent solid of sediments run as wet-aliquots.
02	44,232	839	
03	52,213	991	
04	37,098	704	
05	38,157	724	

## Hg Flux Ratios and Hg Deposition to Lakes in the Vicinity of the Lye Brook Wilderness

Background Hg fluxes for the study lakes (Table 4) ranged from 10.24 to 28.5  $\mu\text{g m}^{-2} \text{yr}^{-1}$ , and the average baseline flux of 15.6 was only slightly elevated over the mean of 11.4 calculated for 10 VT-NH lakes reported by Kamman and Engstrom (2002). The average for the present study lakes is skewed by the high baseline value from Branch pond of 28.5  $\mu\text{g m}^{-2} \text{yr}^{-1}$ . The difference between the average baseline fluxes for the present study lakes and those reported by Kamman and Engstrom (2002) was not statistically significant. Current fluxes for the study lakes ranged from 46.24 to 91.19  $\mu\text{g m}^{-2} \text{yr}^{-1}$ , and the average current flux was significantly elevated ( $p=0.033$ ) by 31  $\mu\text{g m}^{-2} \text{yr}^{-1}$  over the average value reported by Kamman and Engstrom (2002).

Flux ratios are defined as the ratio of modern Hg fluxes to background fluxes, and reflect the degree to which Hg accumulation to lake sediments is augmented over baseline, or background levels. Flux ratios ranged from 3.32 to 7.72 for the present study lakes, indicating that on average, the sediments to these lakes currently accumulate Hg at a rate nearly 6 times background levels (Table 4). The difference in average flux ratios between the present lakes and those reported by Kamman and Engstrom (2002) is only marginally significant statistically ( $p=0.07$ ), but suggests that Hg deposition to the present lakes is augmented by a factor of 1.5X over flux ratios measured from the 10 VT-NH lakes (Table 4).

**Table 4. Background, peak, and current Hg fluxes, and modern flux ratios**

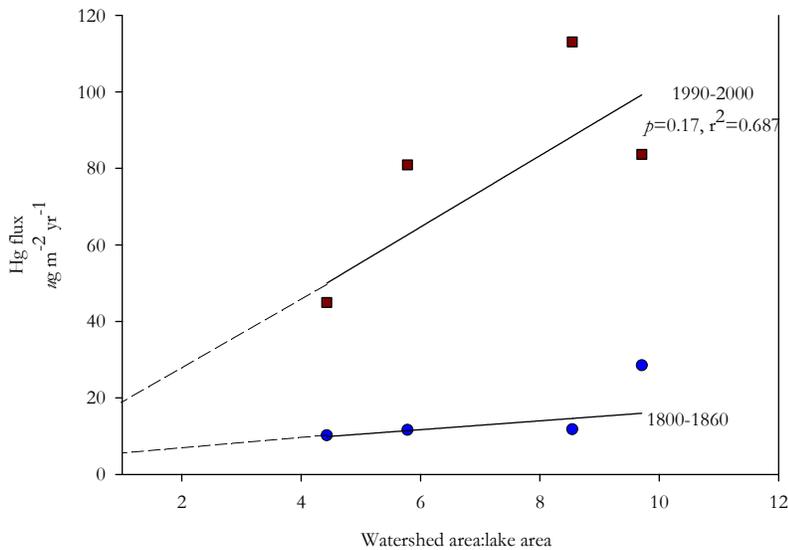
Lake	Background flux ( $\mu\text{g m}^{-2} \text{ yr}^{-1}$ )	Years used for background	Peak flux ( $\mu\text{g m}^{-2} \text{ yr}^{-1}$ )	Year of peak	Current flux ( $\mu\text{g m}^{-2} \text{ yr}^{-1}$ )	Flux ratio
Bourn	11.82	1809	125.67	1992	91.19	7.72
Branch	28.50	1825-1854	73.00	1961	73.00	3.32
Grout	10.24	1809-1839	46.24	2000	46.24	5.79
Stratton	11.63	1850-1873	85.62	1996	83.52	7.08
Mean for all lakes	15.55	-	82.63	-	73.49	5.98
Mean for 10 VT-NH lakes*	11.4	-	52.9	-	42.5	3.9

\*) from Kamman and Engstrom, 2002

## Watershed Influence on Hg Fluxes

Several studies (Swain et al., 1992; Engstrom and Swain, 1996; Lorey and Driscoll, 1998; Kamman and Engstrom, 2002) have used the relationship between watershed area:lake area ratio (e.g., “watershed ratio”) and Hg flux to: 1) assess the influence of watershed size on Hg flux retention with time; and 2) generate estimates of direct atmospheric Hg deposition to lake surfaces. Figure 9 provides regressions for background (<1850) and current averaged (1999-2000) fluxes in relation to watershed ratio, for the LBW lakes.

If back-casted to a watershed ratio of one, regression estimates provide an estimate of Hg flux derived from a watershed of the same size as the lake, which can be interpreted as net direct Hg deposition to the lake surface, minus

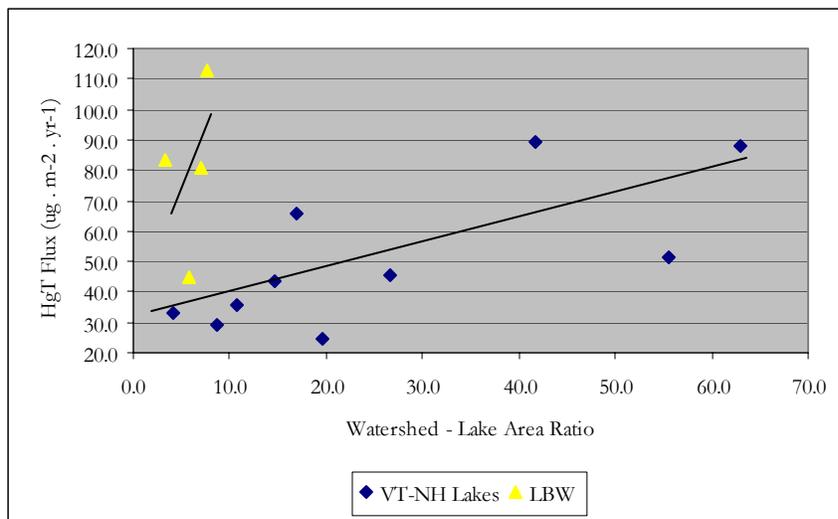


**Figure 9. Relationship between watershed-lake area ratio and Hg flux in four Lye Brook Wilderness Lakes. Dotted lines provide back-casted estimates of Hg flux at a watershed ratio of 1.**

(Swain et al., 1992).

any Hg which may be re-evaded from the lake surface (Kamman and Engstrom, 2002). While neither regression is statistically significant, the relationship suggested by the data for the period 1990-2000 should not be rejected from further consideration, because the small number of datapoints necessarily yields a low-power statistical test. Several important pieces of information are apparent in Figure 9. First, the flux of Hg attributed to direct atmospheric deposition for the present time period is approximately  $19 \mu\text{g m}^{-2} \text{ yr}^{-1}$ , a value remarkably similar to the  $21 \mu\text{g m}^{-2} \text{ yr}^{-1}$  estimated for the 10 VT-NH lakes of Kamman and Engstrom (2002). Second the value estimated for background fluxes is approximately  $6 \mu\text{g m}^{-2} \text{ yr}^{-1}$ , which is consistent with the value of 5 estimated by Lorey and Driscoll (1999) for eight Adirondack lakes. Finally, and most importantly, the slope of this relationship ( $8.7 \mu\text{g m}^{-2} \text{ yr}^{-1}$  per unit increase in watershed ratio) is very high relative to those published for the lakes in VT-NH (slope = 1.12), the Adirondack Mountains (slope = 1.93)(Lorey and Driscoll, 1999), and the upper Midwest (slope = 3.27)

The differences in these slopes are most easily observable on Figure 10, where it is clear that Hg fluxes are much higher in the LBW vicinity lakes than would be expected given the relationships developed for the greater VT-NH lake set. In the LBW lakes, small increases in watershed ratio produce large increases in Hg fluxes (e.g., enhanced delivery to downstream lakes). This is likely attributable to the humic-rich nature of LBW waters (Kellogg and Pembroke, 1997). Generally speaking, humic-rich watersheds have a higher capacity carry Hg from watersheds to downstream lakes than other watershed types, owing to the preferential binding of Hg on humic and fulvic acid complexes (Driscoll et al., 1994; Kamman et al., 2003).



**Figure 10. Relationship of watershed ratio to Hg flux for lakes in the vicinity of Lye brook Wilderness compared to that for 10 VT-NH lakes, for the period 1990-2000.**

Disturbance in the LBW watersheds necessarily releases such complexes, and thus any new land use alterations in the LBW could significantly enhance Hg accumulation in downstream lakes. Any proposed land use changes or disturbances to lands in and near the LBW should be carefully considered in light of the potential increases to the already considerable Hg contamination signal. A fruitful avenue of inquiry would be to investigate the physicochemical attributes of the lakes and lake watersheds of the Kamman and Engstrom (2002) study and the present study, to determine the extent to which watershed geochemistry alone may cause the differences observed between these two study lake sets.

## Summary

Mercury accumulation and sediment isotopic chemistry was studied in four lakes in the vicinity of the Lye Brook Wilderness of southern Vermont, using <sup>210</sup>Pb-dated sediment cores. The four lakes investigated are relatively high-elevation, forested waters occupying small watersheds, and characterized by similar water chemistry.

The four studied lakes all have a history of prior land clearing and logging, and this is reflected by short-term increases observed sediment accumulation rates, and by longer-term shifts in lake trophic status inferred using C/N ratios and stable carbon isotope ratios. Specifically, historical logging appears to have increased sedimentation rates at all ponds circa 1850-1880, and again at Bourn, Branch, and perhaps Stratton Ponds during the 1950's. Bourn, Branch and Grout Pond sediments showed re-establishment of relative stability in both sedimentation and trophic status following logging events. By contrast, sediments from Stratton Pond indicated that sedimentation and eutrophication have increased continually lake since approximately 1950.

Two lakes (Bourn and Grout Ponds) showed generally linear increases in Hg accumulation with time. The other lakes (Branch and Stratton Ponds) showed significant declines in Hg fluxes in recent time periods. Hg accumulation to Branch Pond, and to a lesser degree to the remaining lakes, was influenced by land-use induced changes in sediment accumulation rates. In Grout Pond, a very low overall sedimentation rate resulted in rather low Hg accumulation rates to that pond. The average flux ratio (ratio of modern to baseline Hg flux) of 5.98 was elevated by approximately 1.5X over that derived from a separate similar study of 10 VT-NH lakes. The relationship between relative watershed size and Hg accumulation was anomalously high in the Lye Brook Wilderness lakes, indicating that geochemical watershed processing and release of Hg is very important in controlling Hg accumulation in the lakes.

## **Historical Sketch of Bourn, Grout, and Stratton Ponds**

### ***Bourn (Muskrat) Pond***

The Beers Atlas of 1869 depicts a small logging hamlet in the northeast corner of the Lye Brook Plateau, north of Bourn Pond. This area, called Rootville after Henry Root, contained three sawmills, a shop, and five other buildings. Although there is little historical information regarding these abandoned sites, the presence of the mills makes it clear that a considerable amount of logging took place in the Lye Brook area during the mid-nineteenth century. From 1912 to 1919, the Rich Lumber Company operated a railroad they built from Manchester up Lye Brook Hollow and past the western and northwestern shores of Bourn Pond. The company took a contract for 12,000 acres, with boundaries that almost match that of the current wilderness area. They cleared the contracted land in only five years. Although logging had been taking place for almost a century, this was the first time Lye Brook was deforested. In 1953, Chris Swezey Jr. won a bid to log 3.5 million board-feet around Bourn Pond from the National Forest Service, which he logged in only three years.

### ***Branch Pond***

A prior analysis of Branch Pond sediments (Kamman, 1999, Appendix A) provides interpretation of the sediment record in light of known historical events. The Swezey Logging operation noted above clearly extended into areas quite close to Branch Pond, as evidenced by aerial photography dating back to the mid-1950's, and certainly is responsible for changes observed in the sediment isotopic signatures and Hg flux patterns.

### ***Grout Pond***

The Abel Grout Farmstead is believed to be one of the first settlements of the area. Abel, a 48 year old Revolutionary War veteran, came here with his family of 10 children in 1806. This farm was located 1.3 miles north of Grout Pond. This site remained a pasture as late as 1921. In 1835, Hudson Grout moved into a farm on Grout Pond with his family. The Kelley Stand Road crosses the East Branch of the Deerfield River at the current AT/LT parking lot. This is the location of the Grout Job sawmill and community. People settled here as early as 1830; it was operated by the Grouts between 1874-1901. In 1874, Joel F. Grout Sr. built a new mill that produced shingles, chair stock, and planed and unplaned lumber. A brick mill foundation and remnants of a stone dam on the Deerfield is all that remains of the operation. This now wooded area was heavily cut during the mill's operation. In 1883, Joel F. Grout Jr. and brother Irving Grout built a 2-family house near the sawmill.

### ***Stratton (North or Jones) Pond***

The first road across the "wilderness" between the towns of Arlington and Stratton was the Stratton Turnpike, built between 1827-1831 (Kelley Stand Road Improvement Project). This road was later named Kelley Stand Road after a hotel on the road, which bore the same name. At its prime, 300 people were thought to have lived in the settlement around the Kelley Stand Hotel. This location housed the loggers and their families, and included a post office, schoolhouse, barns and a blacksmith shop. During the 1850's, there were 9 mills on the Roaring Branch, two of which were edge tool mills. There were at least 3 known logging settlements off of Roaring Brook. A mill was in operation on the South Fork confluence as early as 1827. The Battenkill Lumber Company built a six mile flume up Lye Brook Hollow to transport whole logs from the plateau. A series of dams and holding ponds were constructed near the brook. The company went bankrupt in 1907.

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## Appendix A

[Kamman, N.C. 1999. Paleostratigraphy of Mercury in Branch Pond, Sunderland, VT](#)