RECENT ANTHROPOGENIC IMPACTS ON THE GEOCHEMICAL COMPOSITION OF NORTHERN NEW ENGLAND LAKE SEDIMENTS

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Abstract

Nitrogen is an important component in the biogeochemical processes of freshwater systems. Likewise, it is unknown if, and to what magnitude, changes in land use in the watersheds of New England lakes have affected nitrogen availability. This study examines the effects of land-use change on the present and historic isotopic signatures of nitrogen in three New England lakes of varied histories, Lower South Branch Pond, Little Kennebago Lake, and Sennebec Pond. The histories of all three sites indicate minimal discernible disturbance before the onset of Euro-American-induced land use change. For two sites, the dominant mechanism of change was timber harvest, which began in the latter half of the 19th century. Sediment cores for each site were examined and variations in geochemical and sedimentological indicators were evaluated in the context of changes within respective basins. Statistical analysis indicates significant shifts in the means and variance of the geochemistry within the Little Kennebago Lake and Sennebec Pond watersheds after the incursion of Euro-American settlers, while the Lower South Branch Pond watershed displays similarities to a more widespread signal of anthropogenic nitrogen that has been deposited remotely. The record of magnetic susceptibility in Little Kennebago Lake displays the largest variation compared to the other two lakes, which

may indicate that the magnitude of land-use change within the basin was more impactful relative to Lower South Branch Pond and Sennebec Pond. This is significant in that all three sites experienced some level of land-clearance.

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1.0 Introduction

Since the incursion of Euro-American settlers, the northeastern United States has experienced a complex land-use history (Hall et al., 2002) including periods of extensive timber harvest, agriculture, and conservation-minded forest management, all while supporting a dense population. Today, understanding the geochemical response to land use change in a watershed is vital for developing informed policies that prioritize water quality and aquatic habitats. Lake ecosystems are sensitive to changes in water chemistry and landscape change (Brönmark & Hansson, 2002; Schindler, 2006). These changes can have effects on mixing patterns and trophic state (Makri et al., 2020), which can have negative consequences for the biodiversity of a lake (Brönmark & Hansson, 2002). Of particular importance is nitrogen, which is both a significant nutrient for organisms and a potentially harmful byproduct of anthropogenic disturbance (Vitousek et al., 1997). The amount and form of nitrogen in a lake is strongly influenced by processes within a lake basin (Abell et al., 2011; Cole et al., 2006; McLauchlan et al., 2007) and, more recently, large-scale anthropogenic processes outside a basin (Holtgrieve et al. 2011).

Lake sediments can provide an historical record of changes within a basin, which can be useful in determining the timing and magnitude of potential effects on geochemistry (Smol & Last, 2002). Much work has been done, particularly in the last twenty years, around the use of stable nitrogen isotope data ($\delta^{15}N$) as a proxy for past lake conditions (Zhuo & Zeng, 2020), productivity (Brenner et al., 1999), and to trace various sources of anthropogenic contributions (Ma et al., 2020). In the northeast United States, a complex, and well-documented, history of land use provides an excellent record against which to examine changes in geochemistry as recorded by lake sediments.

Additionally, the balance between local and regional geochemical signals continues to pose an issue in deciphering systems with complex land use histories (McLauchlan et al., 2007). Since the Industrial Revolution, an increasing amount of nitrogen has been supplied from anthropogenic sources such as fertilizers and emissions from fossil fuel combustion (Holtgrieve et al., 2011; McLauchlan et al., 2013). This anthropogenic nitrogen can be deposited directly into a lake through a variety of means, including atmospheric deposition, which is able to transport nitrogen great distances (Holtgrieve et al., 2011). Disturbances in the surrounding landscape, such as deforestation and human development, may also releases existing nitrogen, both natural and anthropogenic, into nearby lakes to be deposited in sediments. This can further increase the amount of nitrogen in a natural reservoir (McLauchlan et al., 2007).

Previous studies have sought to examine the effects of climate and land-use change on sediment yields on New England lakes (Cook et al. 2020), as well as the effects of local land use change and large-scale anthropogenic impacts on the geochemistry (Holtgrieve et al., 2011; McLauchlan et al., 2007). Despite this work, the long-term record of geochemical variability in lakes of the northeastern United States is not well established (McLauchlan et al., 2007) despite a well constrained historical record of deforestation and regional industrialization. It is unknown if, and to what magnitude, these anthropogenic processes have affected regional nitrogen geochemistry and how lake ecosystems have responded to different types of land use change. This study aims to build upon this previous work by: (a) examining the geochemical signatures of various types of land use through the examination of sediment cores from three different lakes; (b) quantifying the amount of geochemical variability pre- and post-Euro-American incursion; and, (c) comparing the influence of local processes versus regional processes.

1.1 Land Use in the Northeast United States

1.1.1 Pre-Euro-American

The record of land use change in New England begins before the intrusion of Europeans (pre-1492) and relies heavily on historic accounts and archeologic research to reconstruct. In the millennia preceding the arrival of Europeans, the Indigenous Peoples of New England consisted of tribal groups spread throughout the region that practiced both land clearing and agriculture (Cronon, 1983). These groups, whom had inhabited the area for approximately 12,000 years before present day (Bourque, 2004) were highly mobile, often moving their settlements to adapt to changing conditions in order to benefit most from the land around them (Bragdon, 1999; Fuller et al., 1998). Once established, they rotated agriculture to maximize productivity, and are well documented in using fire as a means to clear land for growing and for travel corridors (Cronon, 1983; Day, 1953; Meyer et al., 2014). It should also be stated that Cronon

(1983) identifies a potential bias in many of the historical records, introduced from European accounts that focus primarily on "merchandisable commodities," which most likely results in an exclusion of additional Indigenous land use.

Oswald et al. (2020) examined pollen and charcoal data derived from sediment cores from southern New England in order to observe the regional signal of Indigenous land use change in order to better inform contemporary conservation practices. After comparing these paleoclimate data with archaeological records, a signal of fire-based land clearing was not detected, which contradicted most historical accounts. This conclusion was met with considerable resistance. Roos (2020) suggests the magnitude of signals, based on historic estimates, may not appear on the regional scale. Additionally, there may have been variability among tribal group practices that would further dampen the signal. An earlier study by Abrams & Seischab (1997) suggests the methods used would fail to detect lower-intensity surface fires, which could have been a likely method used (Cronon, 1983). Leonard et al. (2020) notes that historically, burning occurred at focal points and travel corridors, which may not appear on a regional scale, and reference an earlier study that posits Indigenous practices may have coevolved with local ecosystems and cycles of succession, which would further limit signal detection (Lake & Christianson, 2019). While there is continuing debate over the extent of agriculture and fire use by indigenous peoples prior to the arrival of Europeans, there is limited evidence of a regional signal, and direct impact to the study areas was likely limited prior to the arrival of Europeans.

1.1.2 Pre-Industrial

At the onset of European colonization in New England during the mid- to late-17th century, settlements, and associated land use change, were primarily confined to coastal areas (Hall et al., 2002) and those areas of "urbanization" were estimated to be only 100 km^2 (Thompson et al., 2013). This meant that even with extensive *per capita* land use change, the human impact was restricted to a minuscule fraction of the region. However, accounts of the bountiful natural resources present in the area quickly spread back to Europeans, who aspired to benefit from the economic value of what they deemed a seemingly endless supply of timber fit for many uses (Cronon, 1983). Shortly thereafter, the clearing of land for lumber, agriculture, and livestock increased as the population grew and expanded away from urban centers and into the surrounding lands (Cronon, 1983).

These trends extended well into the 18th century as Euro-Americans spread farther inland. The clearing of land for lumber, agriculture, and livestock increased significantly as the population grew and expanded away from the once centralized centers and into the surrounding land. In the words of Cronon (1983),

> "…the replacement of an earlier village system of shifting agriculture and hunter-gatherer activities by an agriculture which raised crops and domesticated animals in household production units that were contained within fixed property boundaries and linked with commercial markets."

Indeed, the newly-established rural economy of New England had acquired a tendency towards expansion and consumption. These trends extended well in to the

18th century as peoples of European origins spread further inland, motivated by the promise of arable land, space for livestock, and timber with which to build their future (Cronon, 1983). Additionally, Foster (2009) notes high rates of ownership turnover throughout the 18th centuries, as land was acquired, harvested of its intrinsic worth, and sold again to restart the cycle. These trends, however, were not equivalent across the region, for the wilderness in northern New England was still relatively inaccessible, and would not see significant levels of timber harvest until the 20th century (Cronon, 1983).

The early 19th century saw a continuation of these trends until approximately 1830, when southern and central New England experienced a peak in both deforestation and agriculture (Foster et al., 1998; Hall et al., 2002). While the timing and mechanisms of change differed significantly between southern/central and northern New England, the trends in the former illustrates the changing patterns in land use at the time. The historical records of the period, however, contains uncertainty due to vague “unimproved” classification in reporting, leading to speculation around the extent of land use change. Hall et al. (2002) suggests a possible shift from passive pasturelands, in which livestock would be free to roam semi-forested areas, to a more “intensively-managed” pastureland; a significant change in patterns of land use that may have been absent from the historical record due a lack of detail in the classification scheme.

1.1.3 Post-Industrial

What is certain is that the 19th century brought with it a new age of technological advancement: the Industrial Revolution. No longer did people have to provide for themselves; rather food was grown and livestock raised in the Mid-West, and transported to New England. This led to the concentration of populations in urban centers, a decline in agriculture, and the return of forests in southern and central New England (Cronon, 1983). While this transition took place, northern New England lagged behind the rest of the region, as farming in Maine peaked in the late 1880s, when roughly 1/3 of the land in the state was used for agriculture (Ahn et al., 1997). Private companies began consolidating land holdings and investing in infrastructure to move farther up river drainages, establishing dams and camps to facilitate the hundreds of miles of log transport necessary to connect the remote stands of timber with the mills and yards near the coast (Smith, 1972).

Veritable cities emerged from the woods along the primary rivers. Small towns like Patten, Maine became the heart of extensive lumbering operations, hosting an estimated 350 horses and 4,050 men in support of lumbering around 1887 (Smith, 1972). By that year the East Branch of the Penobscot River, by most accounts a difficult place to lumber and drive, was producing 40,000,000 board feet per year. While this paled in comparison to larger operations like those on the Kennebec River, which produced well over 100,000,000 board feet per year many times in the preceding decade (Smith, 1972), it serves to illustrate the magnitude of change occurring in even the most remote areas.

1.1.4 Conservation Era

The 20th century was characterized by an increased dichotomy between land use policies in southern-central and northern New England. As population grew and development continued in the former, so did the rise of conservation. The Weeks Act, passed in 1911, allowed the government to purchase private land, which led to the creation of vast National Forests in Vermont and New Hampshire, which allowed for more restrictions on land use activities (Lilieholm et al., 2013). Private, non-profit groups began protecting land in Massachusetts for public enjoyment, and as the century wore on, major initiatives such as the Land and Water Conservation Fund of 1964 would serve to encourage the preservation of natural landscapes and limit land use change not only in New England, but around the country.

Conversely, the lands of northern New England, specifically Maine, continued to have a high percentage of private ownership (Meyer et al., 2014), with entities like the Great Northern Paper Company becoming the state's largest land owners (Smith, 1972). This area experienced an increase timber harvest in the early 20th century (Foster & Aber, 2006), peaking around 1909 when over a billion feet of wood was cut (Smith, 1972). After hovering around this extent, cut began to diminish in the succeeding decades until the mid-1930s, when a major expansion of logging roads, spurred in part by the Civilian Conservation Corps (Smith, 1972), led to an increase in timber harvest throughout the state, peaking around the beginning of the 21th century (Barton et al., 2012). Throughout the period there were indicators of the larger conservation movement, such as the creation of Baxter State Park in 1931, however, approximately 84% of land in Maine remains privately owned to this day (Butler et al., 2016). Baxter State Park, the vision of Maine Governor Percival P. Baxter, was created

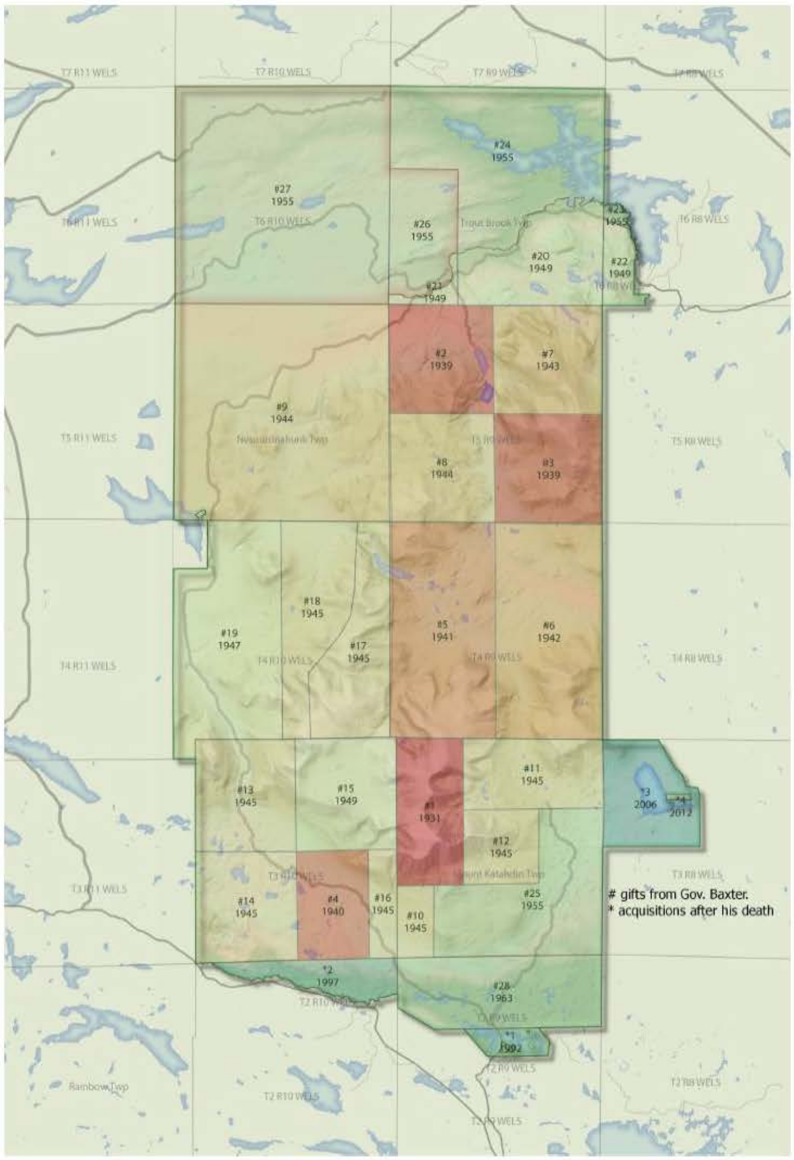

Figure 1 - A map of Baxter State Park, with parcel acquisition order and year acquired. Lower South Branch Pond watershed is located in parcel #s 2, 3, 7, and 8 (Baxter State Park, 2022).

after Baxter purchased and subsequently donated large parcels of land to the State of Maine on the condition that the land "shall forever be kept and remain in the natural wild state" (Baxter, 1931). The first parcel was donated in 1931 ("Baxter State Park: Organization," 2022), and by the time of his death in 1969, the park exceeded 800 km^2 (Figure 1).

Aside from Baxter's personal efforts, Maine was slow to set aside lands for conservation, largely driven by public acquisition of both parks and multiple-use forests (Meyer et al., 2014). Of today's protected lands, around 30% were acquired in the period preceding 1980, after which there was a rise in the creation of land trusts, spurred by the Uniform Conservation Easement Act of 1981 and subsequent tax legislation. This gave incentive for private landowners and environmental non-government organizations (ENGOs), like the Nature Conservancy and the Appalachian Mountain Club, to protect private property (Meyer et al., 2014).

1.1.5 Modern-Day

The later 20th century in New England has seen, for the most part, a decrease in forests, with most of the land being converted to agricultural lands, pasture, and low-density development, which makes up more than half of all conversion (Olofsson et al., 2016). The study also notes a distinct lack of existing forest expansion. The trends, however, are in contrast with a renewed rise in conservation. The period from 1999 to 2012 saw an increase in the rate of land protection 20 times that of the period from 1800 to 1979, as well as an increase in the average size of conservation easements, jumping to 469 ha per easement by 2010 (Meyer et al., 2014). This trend, again, is fueled in part by environmental non-governmental organizations (ENGOs), whom have created landscape-scale protected areas, though it should be mentioned that some of these combine ecosystem reserves with working forests, meaning the land is still subject to the effects of anthropogenic influence.

Continued innovation in conservation strategies and increase in regional conservation partnerships should continue to create large swathes of protected lands. Additionally, the rise of ecosystems services, carbon markets, and additional market-driven conservation incentives insinuate a continued interest, albeit largely financially-driven, in preserving the forests of northern New England (Meyer et al., 2014). Alternatively, while experiencing a slight downturn, timber harvest continues to be a significant in the region (Barton et al., 2012). Further still, the increase in both number and scale of hybrid lands, those practicing conservation while simultaneously working the forests, convolutes the potential impacts of the both actions. Consequently, it is of paramount importance that the full effects of changes in land use on the natural environment are understood.

1.2 Geochemical Indicators

1.2.1 Records of Land-use Change from Lakes

In the last few years, several lakes in the northeastern US have been the focus of multiple studies examining the effects of land use change on natural systems (Cook et al., 2015; 2020; Rich, 2021). Through the examination of sediment cores and land cover change analysis, these studies have improved constraints on the impacts of timber harvests and flood events on sediment yield, while compiling a high-resolution record of changes in land use. With such a record present, analysis of geochemical concentrations and isotopic composition can be a powerful tool in revealing the history of lake conditions and the isotopic signatures of changes within the catchments caused by human impacts.

1.2.2 Nitrogen

Nitrogen is one of the most important nutrients the natural world, and the effects of human activities on the nitrogen cycle have had a significant impact on its flux and storage (Barnes & Raymond, 2010). This anthropogenic influence has been linked to several environmental concerns including soil acidification, acute ground water pollution, and eutrophication (Burns et al., 2009). With this added influence comes increased uncertainty in our understanding of how nitrogen is introduced into a system, and how it moves and is transforms along the way. This understanding is important as we work to improve management methods intended to remediate the issues referenced above, especially as our current strategies fail to adequately address problems like eutrophication (Cecchetti et al., 2020). Therefore, a thorough understanding of how past conditions and events have affected nitrogen is critical towards advancing our understanding of how it may respond in the present and future.

Nitrogen availability is the supply of biologically available nitrogen (fixed nitrogen) to terrestrial plants and microorganisms relative to their nitrogen demand (McLauchlan et al., 2007). The processes controlling the availability of fixed nitrogen within a system can be simplified into two aspects: the processes that create or destroy fixed nitrogen; and, the processes that alter fixed nitrogen among its different forms, and move it around the system, while neither creating nor destroying it. The former can change the size of the overall reservoir while the latter cannot. The main outputs are losses to the atmosphere through denitrification, emissions of reactive nitrogen gases, and to the ocean through hydrologic transport (Zhang et al., 2020) (Figure 2).

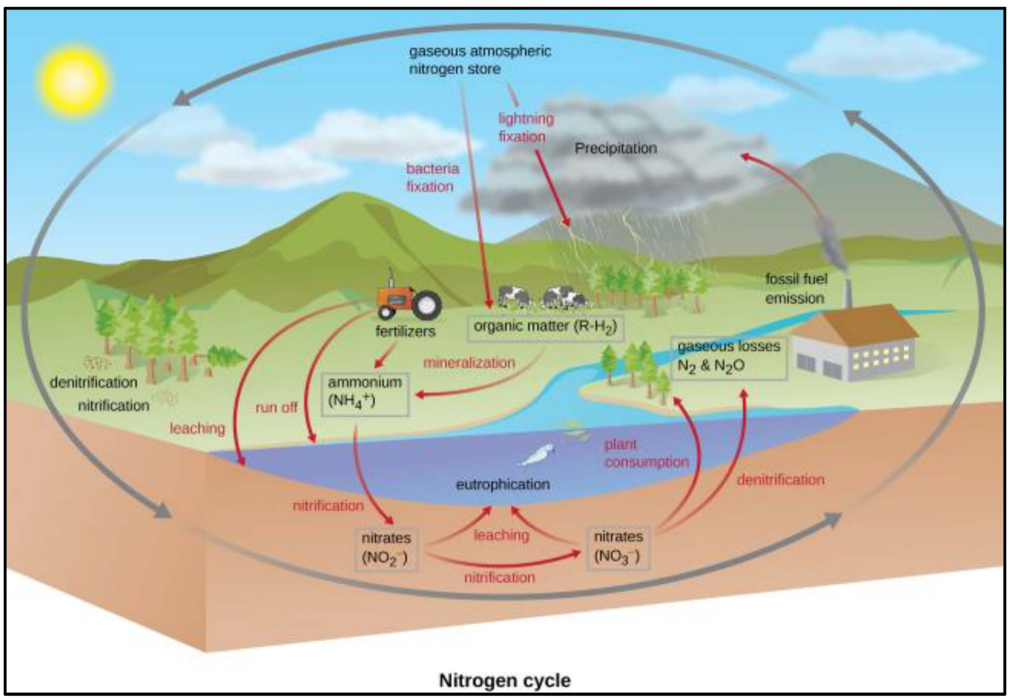

Figure 2- The (simplified) nitrogen cycle. Forms of nitrogen shown are: ammonium (NH_4+), nitrites (NO_2-), nitrates (NO_3-), gases (N_2 and N_2O), and organic nitrogen. Nitrogen altering processes include nitrification, denitrification, plant consumption, and mineralization. Sources of nitrogen include vegetation, anthropogenic emissions, atmospheric contributions, and fertilizers. (Lumen Learning; modification of work by NOAA)

Prior to the Industrial Revolution, most fixed nitrogen in a reservoir was sourced through biological N_2 fixation, a process by which nitrogen gas is incorporated into the tissue of a plant. Since the Industrial Revolution, fertilizers and cultivation of naturally N^2-fixing crops, combined with NO_x emissions from fossil fuel combustion, have dramatically increased nitrogen availability (Fowler et al., 2013). Deforestation, drainage of wetlands, and increased erosion can also increase N availability by freeing up biologically stored nitrogen (Vitousek et al., 1997). When these inputs are included, anthropogenic activities now outpace natural processes of nitrogen addition to terrestrial ecosystems (Zhang et al., 2020).

1.2.3 $\delta^{15}N$

There are two stable nitrogen isotopes, ^{14}N and ^{15}N, constituting 99.635 and 0.365% of the global nitrogen pool, respectively (Nier, 1950). The ratio between these two isotopes can be measured with respect to a reference standard, which is expressed using delta notation (δ) (Ryabenko, 2013). The equation for nitrogen is:

$$\delta^{15}N = \left(\frac{R_{sample}}{R_{standard}} - 1\right) \times 1000 \quad (1)$$

where R_{sample} is the measured ratio of nitrogen isotopes in a sample and $R_{standard}$ is the established ratio of nitrogen isotopes of a universal standard. For nitrogen, the standard typically used is atmospheric nitrogen. The resulting $\delta^{15}N$ value of the sample can then be compared to other values found in the literature. $\delta^{15}N$ is expressed in units permil (‰).

These $\delta^{15}N$ values can change through chemical and physical processes, known as fractionation. Different fractionation processes will result in a variety of isotopic values, or signatures (Mariotti et al., 1981; Xu et al., 2019). Using these signatures, fractionation processes, and therefore the corresponding biogeochemical and anthropogenic processes, can be identified and quantified (Figure 3).

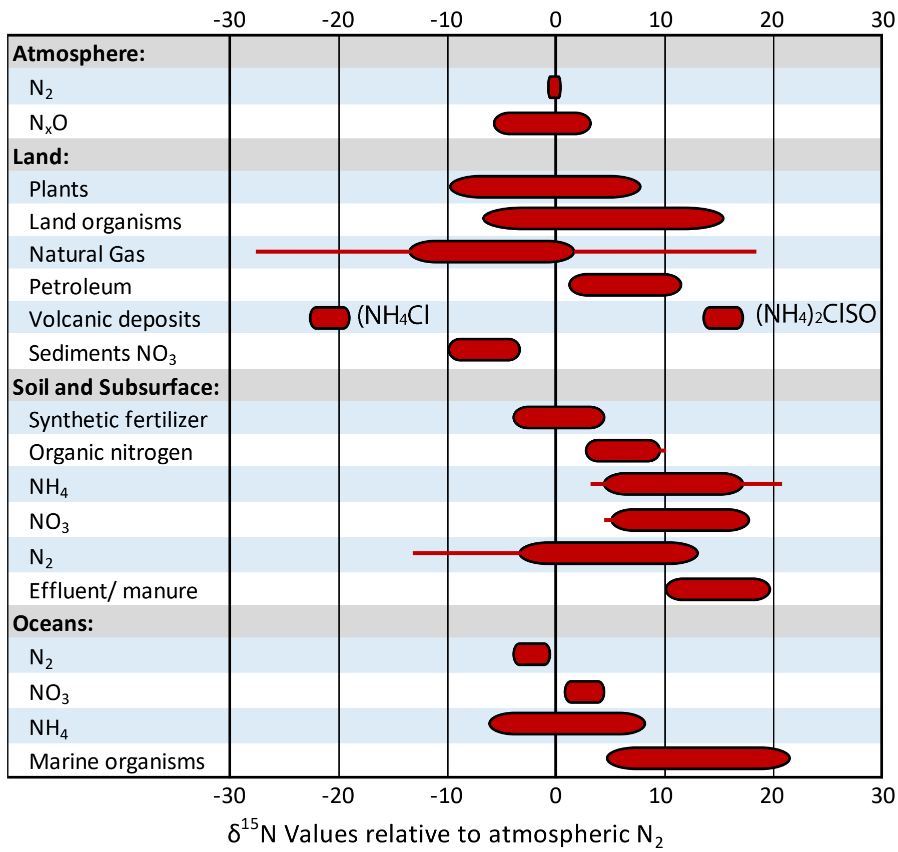

Figure 3- Ranges of $\delta^{15}N$ values relative to atmospheric N_2, by source. Solid bodies reflect established values, while thin bodies reflect possible values. Modified from Hoefs 1997 and Clark and Fritz 1997 with data from Amberger and Schmidt 1987, Böttcher et al. 1990, and Létolle 1980.

Nitrogen isotopes have been used for a multitude of analyses ranging from the comparison of the development of atmospheres on far away worlds (Mandt et al.,

2015) to provenance studies of illicit drugs (Mallette et al., 2017). However, the most widespread use of stable nitrogen isotope analysis is tracing sources of nitrogen. As previously stated, nitrogen moves through the biosphere taking on many forms. By using $\delta^{15}N$ values and mixing models, sources of nitrogen and processes it has undergone can be determined from stable nitrogen isotope analysis. Most importantly, the inherent sensitivity in stable nitrogen isotopic composition creates a powerful tool in detecting subtle changes in multifaceted systems (Robinson, 2001). This proves extremely useful when examining complex biogeological environments such as lakes.

1.2.4 $\delta^{13}C$, Total Nitrogen, and C:N

In addition to $\delta^{15}N$, several other geochemical values can be used in concert to detect changes in a natural system. Much in the same way we employ stable nitrogen isotopes, stable carbon isotopes may be used to detect changes in carbon sources and processes. Like nitrogen, carbon has two stable isotopes, ^{12}C and ^{13}C, and the isotopic signature is expressed as $\delta^{13}C$. It is calculated using the same equation (1) as nitrogen, and the standard used is Vienna Pee Dee Belemnite (VPDB). Due to its abundance in the natural world, $\delta^{13}C$ may vary as a function of source, productivity, and both biological and physical process (Faure & Mensing, 2004). Values can also be used to distinguish organic matter deposited during periods of high versus low primary productivity, as algae preferentially take up lighter ^{13}C isotopes. This yields organic matter with higher $\delta^{13}C$ during periods of very high primary productivity (Mizutani & Wada, 1982; Torres et al., 2012).

The total amounts of nitrogen and carbon, both individually and used as a ratio, are also important tools in comprehending geochemical systems in nature. While the importance of nitrogen concentrations has already been explained, carbon concentrations are similarly important when describing virtually all aspects of the natural world. The role of freshwater systems in the carbon cycle is significant, and can affect regional carbon balances (J. Cole et al., 2007). Biologically-productive lakes lead to greater amounts of respired carbon returning to the atmosphere as CO_2, and previous estimates may have underestimated this contribution to the global carbon sink (Raymond et al., 2013). Additionally, nearly 0.4 billion tons of carbon are buried in freshwater sediments, making them a significant reservoir (Battin et al., 2009). As we begin to understand more about the importance of freshwater systems in the global carbon cycle, it is important that we continue to gain an understanding how changes in these systems affect carbon (Biddanda & Koopmans, 2016).

The ratio of carbon to nitrogen (C:N) can also be useful in determining the source and fate of organic matter in aquatic environments (Gordon & Goni, 2003). Terrestrial organic matter will typically have a C:N ratio greater than 20, whereas algae typically have a C:N ratio between 4 and 10 (Meyers, 1994). Using this logic, periods of lower C:N ratios in lake sediment records have been used to identify of greater input from algal organic matter, and conversely, periods of a high C:N ratio indicate a greater input of terrestrial organic matter (Kansanen & Jaakkola, 1985).

<u>1.2.5 Applications in Lake Sediments</u>

The use of $\delta^{15}N$ signatures, total nitrogen and carbon, and C:N ratios to study lake ecosystems is a relatively recent advancement. Early examples include Brenner et al. (1999), who used $\delta^{15}N$ in sedimented organic matter as an indicator for past periods of trophic state changes in Florida lakes. Wolfe et al. (2001) attributed rapid ecological change in an alpine lake to excess nitrogen from agricultural and industrial sources.

More recently, Ma et al. (2020) used $\delta^{15}N$ data to define the sources and transformations of nitrogen to inform regulatory strategies for the restoration of Lake Okeechobee in Florida. Likewise, Zhuo & Zeng (2020) used the total concentration of nitrogen, $\delta^{15}N$, and carbon-nitrogen ratios in sediments from nine lakes to reconstruct the spatiotemporal variation trend of nitrogen deposition, determine sources of nitrogen, and compare the analysis to socioeconomic data from the lake basins. They concluded that since the 1950s, nitrogen content in the lakes has increased significantly, and that the rate of increase has accelerated in the last 20 years. They also attribute the increase to anthropogenic factors, specifically "intensive agriculture and urban expansion."

Most relevant to this study is the work of Kaushal & Binford (1999), McLauchlin et al. (2007), and Holtgrieve et al. (2011). Kaushal & Binford (1999) examined the C:N ratios of lake sediments from a 70-cm core taken from Lake Pleasant, Massachusetts to determine if variations were caused by changes within the watershed. They found a rapid increase in C:N following deforestation around 210 ± 50 years B.P., which they attributed to an increase in the proportion of terrestrial organic matter. They ascribed this increase to two possible factors: an increase in particulate matter loads (Hornbeck et al., 1986) and an increase in stream discharges following deforestation (Hedin et al., 1988). Finally, they noted the C:N of the lake sediments from the core declined as they approached present day concurrent to the reforestation of the watershed.

In looking at the response of hardwood forests to Euro-American perturbation in the northeast United States, McLauchlan et al. (2007) used $\delta^{15}N$ in tree rings and lake sediments taken from a 102-cm core from Mirror Lake, New Hampshire to demonstrate that nitrogen availability in a northeastern forest had declined in the latter half of the 20th century, likely due to ecosystem recovery from changes in land use. They found a sharp increase in $\delta^{15}N$ of roughly 1 ‰ that peaked shortly after 1900 and has been declining since. They observed that high terrestrial N availability leads to high $\delta^{15}N$ in lake sediments as ^{15}N-enriched organic matter and nitrate enter the lake. The authors attributed the decline of $\delta^{15}N$ in the early 20th century to ecosystem recovery from Euro-American land use. From these findings, they suggested that past anthropogenic disturbances such as logging and agriculture are major drivers of nitrogen cycling in forests today.

Holtgrieve et al. (2011) took a broader look at the anthropogenic influence on nitrogen by coring 25 remote lakes in the Northern Hemisphere and comparing the $\delta^{15}N$ signal from the last 400 years. They found a coherent signal of an isotopically distinct source of nitrogen to the lake ecosystems beginning around 1895 ± 10 years. They attributed this initial shift to increasing anthropogenic CO_2 emissions, and an acceleration in signal shift to widespread reactive nitrogen production associated with the Haber-Bosch process, an artificial nitrogen fixation process, which converts atmospheric nitrogen (N_2) to ammonia (NH_3). It is used primarily in the production of fertilizers. They posit that it is likely anthropogenic nitrogen has influenced nitrogen budgets in watersheds around the northern hemisphere for over a century.

1.3 Project Overview

With the exception of a few studies (Kaushal & Binford, 1999; McLauchlan et al., 2007), the impacts of land use change, specifically timber harvest, on the geochemistry of lake basins in northern New England since the onset of Euro-American settlement (~ 500 years BP) are not well understood. In utilizing a well-constrained historical record of land use change, high-resolution data from existing studies, and precise geochemical indicators, this study seeks to:

(1) Contribute to the long-term record of nitrogen availability in New England.

This study will contribute to the regional catalogue of biogeochemical data including nitrogen and carbon isotopes, total concentrations, and carbon to nitrogen ratios, with a focus on the most recent 500-year period.

(2) Determine the geochemical signature of various changes in land use in northern New England.

The study will identify isotopic and geochemical signatures of land use changes by exploring correlation between changes in nitrogen and carbon found in lake sediments with historical records and observations of the immediate area.

(3) Compare variations in nitrogen levels and sources through time in lakes with a variety of histories.

This study will compare how nitrogen availability varies between sites with known variables, determining both the differences in, and magnitude of, geochemical response in lakes to various mechanisms of land use change. These variables include the onset of anthropogenic influence, local geography, and the timing of land-use history. I hypothesize there are statistically significant differences in $\delta^{15}N$ pre- and post-Euro-American settlement.

As this study progressed, I also sought to test the following hypotheses:

(4) There is increased variability in geochemical indicators after the introduction of Euro-American settlers. This can be tested by comparing the standard deviations of geochemical records pre- and post-Euro-American settlement.

(5) The regional signal of anthropogenic nitrogen detected by Holtgrieve et al. (2011) is detectable in northern New England Lakes with more complex land-use histories. In comparing the magnitude and trends of $\delta^{15}N$, pre- and post-

Euro-American settlement, to the history of land use within a basin, I can consider whether the same signals seen in pristine, remote lakes in the northern hemisphere are detectable in my study lakes.

(6) Subtle changes in land use management, specifically in the style of timber harvest, are detectible in the geochemical records in lake sediments. In basins with a persistent, primary mechanism of land-use change in the historical record, such as timber harvest in the Little Kennebago Lake watershed, changes in $\delta^{15}N$ not consistent with other external forcings (e.g., Holtgrieve et al., 2011) may be attributed to the primary mechanism.

2.0 Study Areas

This study examined the geochemical signatures through time of three lakes – one site which has been under strict conservation management since the early 20th century, and two sites which have seen continued, and in some cases increased, land-use change. These sites have well-constrained histories of differing land uses and existing core data such as age-depth models and organic content to which I can compare our N analysis, such as an established age-depth model and sedimentological properties like organic content percentage. Additionally, the sites are in relatively close proximity within the state of Maine, so as to eliminate broad external factors such as variations in climate and atmospheric particulate load; in this case all the sites are located in the state of Maine. With this approach, the geochemical signature of two common watershed disturbances in New England may be compared. Such signatures have previously been studied in other areas (Ma et al., 2020; McLauchlan et al., 2007; Zhuo & Zeng, 2020), but these techniques have yet to be applied in concert in the region.

For the "control" site, the criteria set forth by Holtgrieve et al. (2011) serves as a template to select the optimum study area: a location with minimal disturbance in the watershed, including development and land-use change. For "impacted" sites, selecting a watershed with a well-defined record of land-use change and disturbance will allow me to examine the N-isotopic signature of these events on the sites. For that reason, it is advantageous to select several impacted sites with varying disturbances:

- a site that has been used for timber harvest, but is otherwise undeveloped;
- a site that has experienced "mixed" use – multiple land uses over time.

Two such sites, Little Kennebago Lake and Sennebec Pond, have already been studied by Cook et al. (2020) and Rich (2021), respectively, and provide complimentary data such as sediment properties and rates, as well as GIS analysis, to which I can compare N isotope data.

2.1 Lower South Branch Pond

For this study, Lower South Branch Pond satisfies the requirement for a "control" site, and is of comparable size and hydrological regime to previously studied lakes by Cook et al. (2020) and Rich (2021). An adjacent lake, Upper South Branch Pond, was cored previously by Anderson et al. (1986) for the purposes of examining late- and post-glacial vegetation and disturbance in the area. Though part of a pair of lakes, due to its lake area, watershed area, and depth, Lower South Branch Pond, the larger of the two, will serve as the focus in this comparison. The pond is located in Township 5, Range 9, in the northeast corner of Baxter State Park in Piscataquis County, Maine (Figure 4). Lower South Branch Pond has an area of 0.44 km^2 and a surrounding watershed area of 28.49 km^2, of which 61% drains into the upper lake and subsequently runs to the lower (Lake Stewards of Maine, 2021). It is fed by two main tributaries: Pogy Brook via Upper South Branch Pond, and Howe Brook. It drains out of a single outlet, South Branch Ponds Brook, which is a tributary of the East Branch of the Penobscot River.

The lake has an elevation of 299 m and the watershed relief is 781 m (U.S. Geological Survey, 2021; Vaux & Entwood, 2010). The bedrock is Devonian age Traveler rhyolite (Rankin, 1961) and is exposed in the steep slopes and upper elevations surrounding the lake. The remaining area is covered with shallow till deposited during the Pleistocene and retreat of the Laurentide Ice Sheet. The area receives an average of 114 cm of precipitation annually, based on the period of 2011-2022, and is distributed evenly throughout the year (Maine Climate Office, 2022).

Pollen data from the adjacent lake indicates the area to be similar to the regional pattern of vegetation after the recession the ice sheet, and for the last ~5,600 years BP has been composed of mixed hardwood-conifer forest, with an increase in boreal conifers for the last ~1700 years (Anderson et al., 1986). As previously stated, due to both the remoteness of the area as well as the ruggedness of the terrain, the area was not significantly impacted by Euro-Americans until the later 19th century. Timber harvest, whose impact had slowly crept up the Eastern Branch of the Penobscot River and into nearby areas such as Wassataquoik Stream in the preceding decades, began in the area in the early 1880s (Smith, 1972). This is corroborated by pollen data from Upper South Branch Pond, which show an increase in pollen types indicating settlement (such as grasses) and changes in arboreal pollen percentages, which reflect lumbering (Anderson et al., 1986). Compared to the other sites, the magnitude and extent to timber harvest is difficult to ascertain as the active period predates aerial imagery and the extent of land use was not sufficient to generate relic indicators such as stone walls, which could help quantify the degree of alteration (Johnson & Ouimet, 2016).

In 1903, a major fire swept through the area, burning more than 340 km^2, and more than 92% of the Upper South Branch Pond watershed, which falls completely within the Lower South Branch Pond watershed (Spring, 1904). This event, again, is corroborated by a significant layer of charcoal found in the sediment core from Anderson et al. (1986). Post-fire, common species in the area include paper birch, quaking aspen, red maple, balsam fir, white and spruce, and eastern white pine (Anderson et al., 1986). There is little evidence, both in the historical and sedimentary record, for any significant local anthropogenic influence after the fire of 1903.

In 1939, much of the Lower South Branch Pond basin was included in the second parcel of land to be donated to the State of Maine from Percival Baxter as part of the creation of Baxter State Park (Figure 1). By 1944, the entirety of the watershed was within the protection of the State of Maine and managed, as per Baxter's wishes, to be kept in a wild state in perpetuity (Baxter, 1931). Practically, this meant to manage the land so as to minimize human impacts. Today, the area fulfills this goal with two exceptions: an underwater ledge near the outlet was dynamited by Maine officials to discourage non-local fish from entering from the stream below (*Lower South Branch Pond*, 1963); and the former logging camp on the northern shore of the lake was converted to a campground with several permanent structures and toilets. It is currently classified as an oligotrophic lake, indicating it has low nutrient levels and generally "good" water quality (Lake Stewards of Maine, 2021).

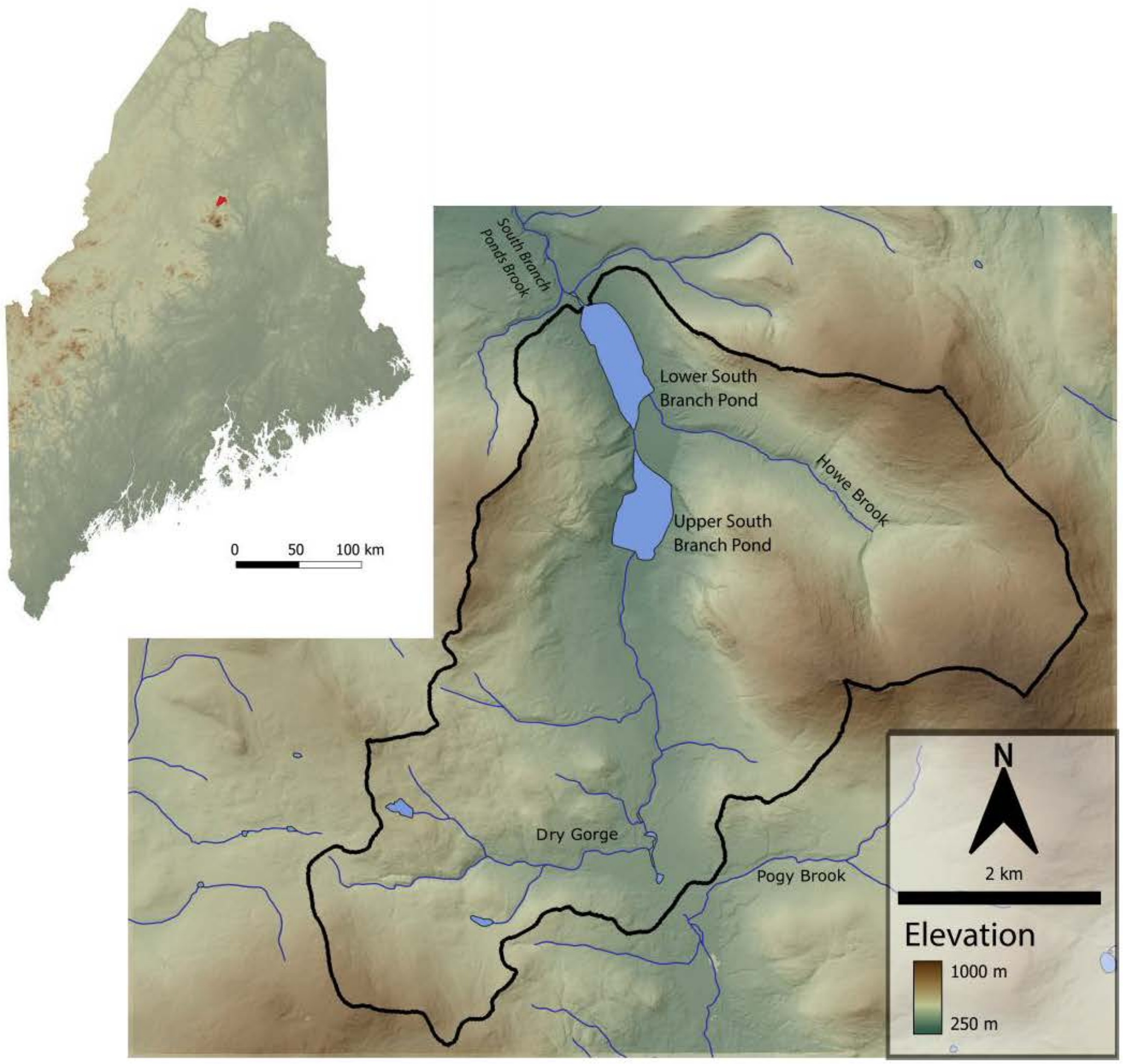

Figure 4- Elevation map from a 2-meter, LiDAR-derived USGS DEM of the Lower South Branch Pond watershed (black line), showing locations and features mentioned in the text. Inset displays location of watershed (red) within Maine.

2.2 Little Kennebago Lake

For a site that has experienced timber harvest as the sole category of land-use change, Little Kennebago Lake has both an appropriate history and has been previously studied in detail by Cook et al. (2020). Little Kennebago Lake is located in Stetsontown Township, Franklin Country, Maine (Figure 5). It has an area of 0.67 km^2, with a watershed area of 135 km^2. It is fed primarily by the Kennebago River, as well as by the smaller Sol Brook. Its main outlet is the continuation of the Kennebago River, which drains through a series of lakes that, ultimately, run into the Androscoggin River. The lake has an elevation of 543 m and the watershed relief is 666 m (U.S. Geological Survey, 2021; Vaux & Entwood, 2010). The bedrock in the area is composed of Devonian intrusives, mafic volcanics, the Hurricane Mountain formation (composed of variably metamorphosed sedimentary and volcanic rocks of greenschist to granulite facies), and gneiss. However, sections of the area, subject to the same regional glaciation as Lower South Branch Pond, are covered with ~5-10 m of basal till, with localized deltaic and kame terrace deposits (Borns & Calkin, 1977). The area experiences an annual average of 135 cm of precipitation, distributed evenly throughout the year (Cook et al., 2020).

Similar to Lower South Branch Pond, the remoteness of the Little Kennebago Lake inhibited human impacts until the end of the 19^{th} century (Cook et al., 2020). Timber harvest likely began shortly after the creation of the Kennebago Improvement Corporation in 1891 (State of Maine, 1893) and has continued to present day, though the magnitude of harvest and associated impacts such as the creation of logging roads has varied through time (Cook et al., 2020). In 1931, the 4.6 m high Upper Station Dam was constructed to facilitate log drives, and increased water levels upstream as far as Little Kennebago Lake (Kaufman & Paradis, 1992). The dam was fitted for hydroelectric power generation in 1952, the same year the last log drive on the river occurred (Palmer, 2004). Afterwards, timber was transported by truck, increasing the length of logging roads in the watershed, with the most active expansion occurring between 1958 and 1966 (Cook et al., 2020). From that point, the intensity of harvest within the watershed fluctuated until present day, with the highest rate of 4.1 km^2/year occurring between 2015 and 2018. From the of 1958 to 2018, 46.3% of the area of the watershed was harvest (Cook et al., 2020). It is currently classified as a mesotrophic lake, indicating it has moderate nutrient levels and an intermediate level of productivity (Lake Stewards of Maine, 2021).

Ultimately, Cook et al. (2020) suggested the landscape was resilient to natural hydrologic disturbance, though they did note the potential for large natural variability in sediment yield as shown by the pre-Euro-American timing of the largest event deposit in the core record. Perhaps more significant to the goals of this study, they found that human activities can influence suspended sediment yield in a moderately impacted watershed, and that changes in forestry practices may have effectively reduced the occurrence of the most severe erosional events.

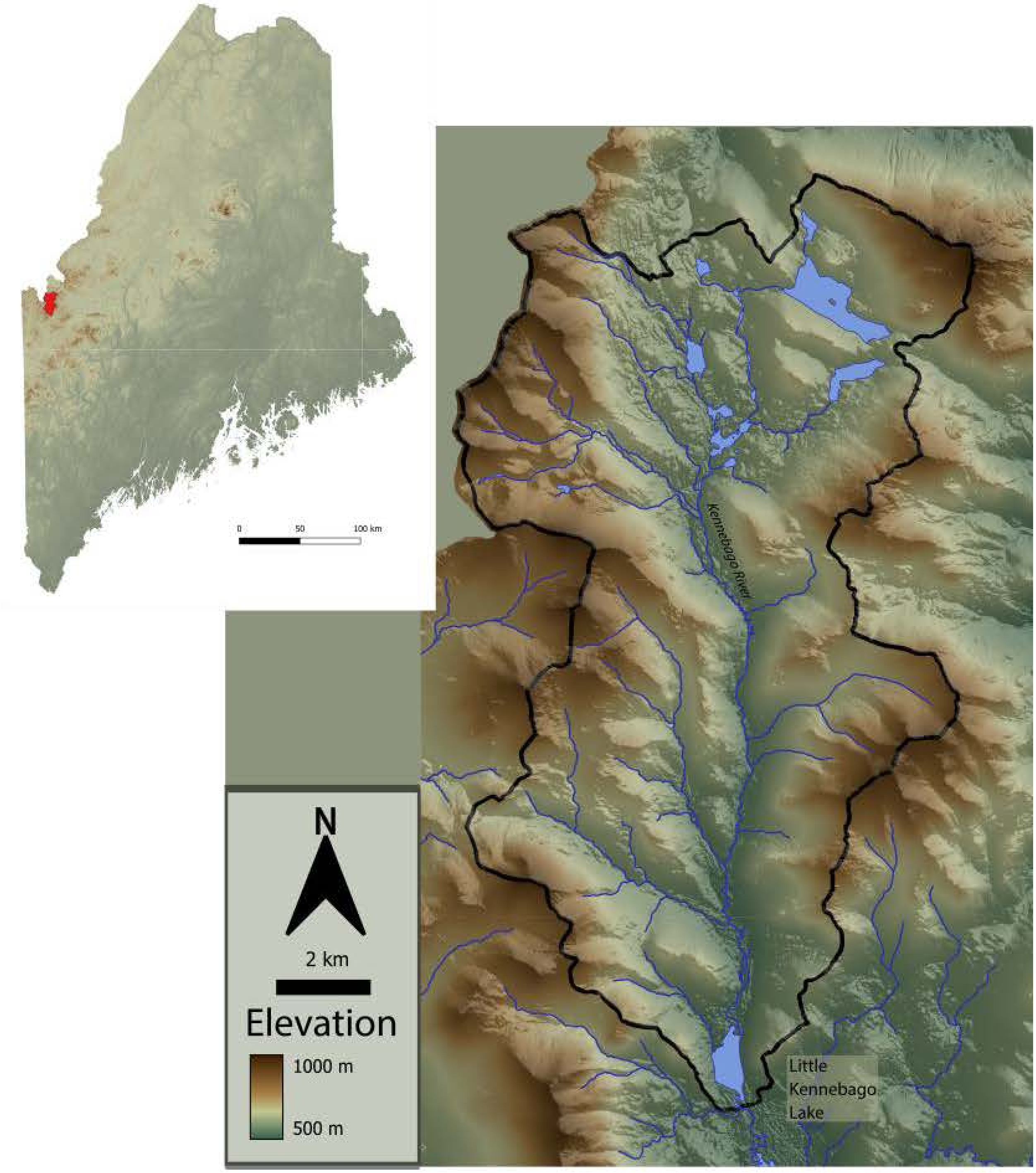

Figure 5– Elevation map from a 10-meter USGS DEM of the Little Kennebago Lake watershed (black line), showing locations and features mentioned in the text. Inset displays location of watershed (red) within Maine.

2.3 Sennebec Pond

For a site with a more complex history of land use change, Sennebec Pond, previously studied by Rich (2021) satisfies the requisite requirements. Sennebec Pond is located in Appleton and Union Townships, in Knox County, Maine (Figure 6). It has an area of 2.17 km^2, with a watershed area of 292.7 km^2. The primary inlet and outlet is the St. Georges River, and the pond is also fed by several smaller streams, most notably Allen Brook. The watershed contains two other notable bodies of water: Saint George Lake and Quantabacook Lake. The lake has an elevation of 27 m and the watershed relief is 319 m (U.S. Geological Survey, 2021; Vaux & Entwood, 2010). The local bedrock includes Precambrian-Ordovician metamorphic rocks, Cambrian-Ordovician metamorphic rocks and Silurian-Devonian volcanic rocks (Osberg et al., 1985). Differing from the previous two sites, the surficial geology of the Sennebec Pond watershed is composed of both basal till, as well as glacial-marine silt and clay deposits (Thompson & Borns, 1985). The area receives an average of 124 cm of precipitation annually, based on the period of 2011-2022, and is distributed evenly throughout the year (Maine Climate Office, 2022).

Due to its lower elevation, gentler relief, and proximity to larger population centers, Sennebec Pond was more susceptible to earlier influence from Euro-Americans than both the South Branch Ponds and Little Kennebago Lake. Though initially hampered by a harsh physical environment and conflicts with the native Abenaki, by the mid-18th century, Euro-American families began settling the area

(Ghere, 1995; Gould, 1950). By 1800, nearby towns like Brunswick and Augusta were developing into industrial centers as large rivers like the Kennebec and Penobscot became the highways and power supplies for the burgeoning timber and textile industries (Köster et al., 2007). The areas surrounding Sennebec Pond, seemingly in support of this nearby urbanization, saw land clearing for timber harvest and

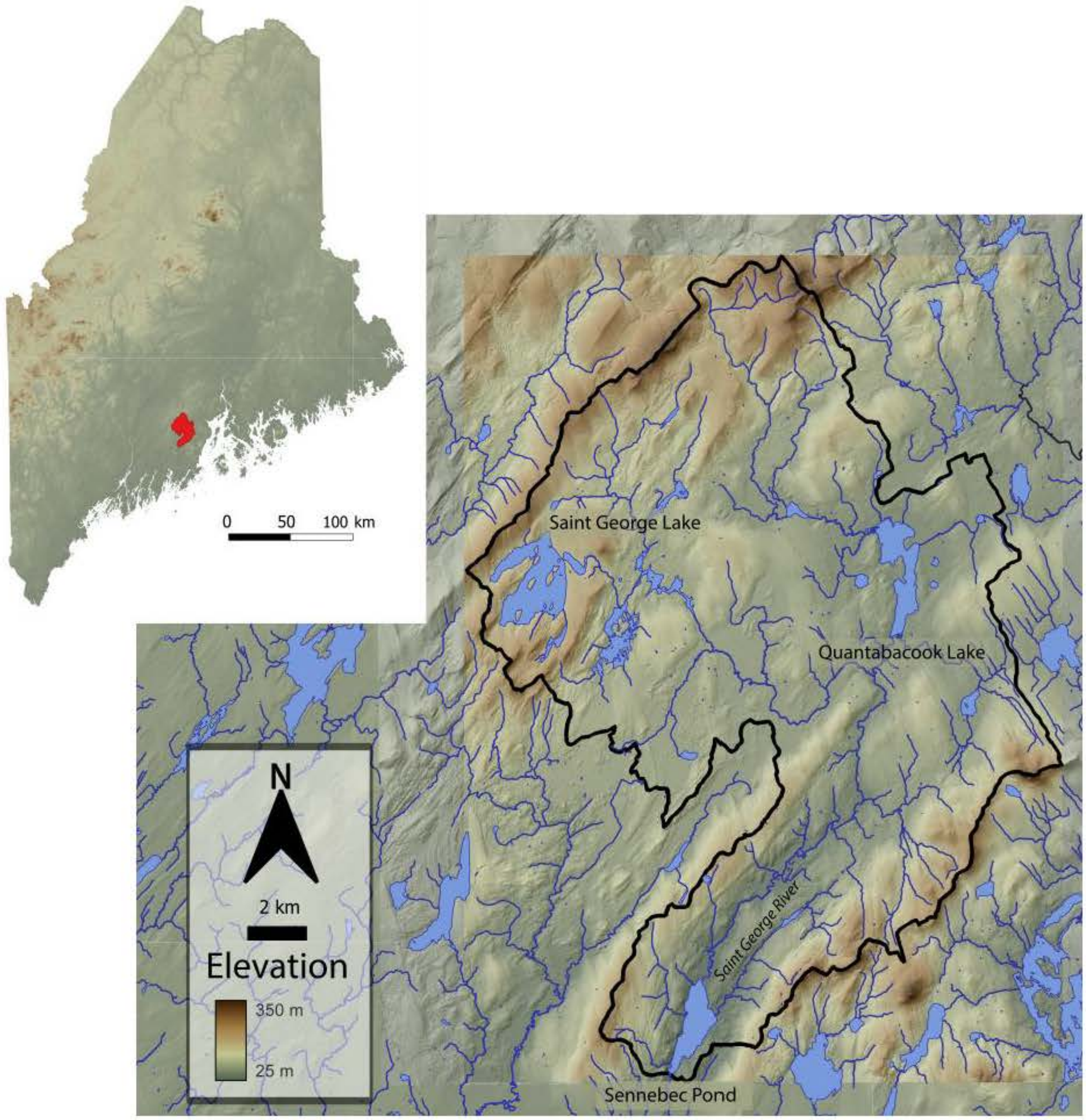

Figure 6– Elevation map from a 2-meter, LiDAR-derived USGS DEM of the Sennebec Pond watershed (black line), showing locations and features mentioned in the text. Inset displays location of watershed (red) within Maine.

subsequent conversion to agriculture and pastureland, much like the rest of southern and central New England (Ahn et al., 1997).

This conversion peaked in the later 19th century, after which the effects of the industrial revolution and improved transportation shifted the burden of food production to other parts of the United States, leading to a decline in agricultural lands and reforestation (Cronon, 1983). Following the trend of the state, this decline in agriculture continued into the 20th century, and the area has seen a general trend of reforestation of altered land in the last few decades, as supported by GIS analysis of aerial imagery (Rich, 2021).

2.4 Acknowledgment of Indigenous Peoples

It must be stated that all three sites lay within the traditional lands of the Penobscot, Passamaquoddy, Nanrantsouk, Wawenock, and Abenaki Peoples ("Native Land Digital," 2021), all of whom belong to the larger Wabanaki Confederacy. I would like to acknowledge their historical and present impacts on the land, and in no way wish to diminish, or contribute to the erasure of, their impacts both past, present, and future.

3.0 Methods

3.1 Field Collection

Two sediment cores from Lower South Branch Pond were collected in May 2021; a surface-sediment core (core 21-1; 60 cm) and a longer, deep core (core 21-2; 171 cm) with partial overlap of core 21-1. To collect sediment samples that most represent processes occurring within an entire lake basin, I collected cores from a deep, central, basin within the lake (17.5 m water depth, 46.102067, -68.895826). Cores were taken using a multiple-meter, single-drive, piston-percussion corer. Once the cores were capped and visual inspected to ensure an intact sediment-water interface and undisturbed layers, they were drained of any excess water, filled with foam to reduce movement, and secured for transportation back to a storage facility. The long core (core 21-2) was split in the field to facilitate transportation, and further divided into core sections 21-2-1 and 21-2-2). Cores were kept at low temperatures, around 5° C, to preserve organic matter and isotopic values. Upper South Branch Pond was cored in the same manner and archived for future analysis. Little Kennebago Lake and Sennebec Pond were previously cored by Cook et al. (2020) and Rich (2021) using the same methods.

3.2 Laboratory Analysis

Cores recovered from the Lower South Branch Pond were split, photographed, and described prior to analysis. The cores were scanned at fine resolution (1.0 mm) using an ITRAX scanning x-ray fluorescence (XRF) core scanner to obtain continuous, down-core elemental abundance (Croudace et al., 2006). The scanner produces measurements of 36 elements and a suite of other properties, which provides preliminary geochemical data, as well as identifies horizons of interest for additional analysis. Additionally, magnetic susceptibility, which reflects the content of clastic materials, was measured using a Mo X-ray source operating at 30 kV and 55 mA with a 10 s exposure time, measured at 0.5 cm intervals.

Percent loss on ignition (LOI) and dry bulk density (ρ_{db}) were measured on 1 cm^3 subsamples removed from the core at 1 cm intervals following standard procedure (Dean, 1974). Post-removal, these samples were weighed, dried for ~16 hours at 100° C to eliminate all water content, and weighed again to determine dry-bulk density (ρ_{db}) (Equation 2).

$$\rho_{db} = \frac{mass\ dry\ sediment}{volume\ of\ sample} \qquad (2)$$

Afterwards, they were combusted at 550° C in a muffle furnace to eliminate all organic matter, and weighed again to determine percent loss on ignition (Equation 3).

$$LOI = \frac{mass\ dry\ sediment - mass\ combusted\ sediment}{mass\ dry\ sediment} * 100 \qquad (3)$$

Finally, the offset between cores 21-1 and 21-2 (split into 2 sections) was determined using sections of overlapping XRF data. The composite sequence used the upper 29 cm of core 21-1 spliced to the section from 5 cm through 171 cm within core 21-2, resulting in a total length of 195 cm.

3.3 Age-Depth Model

Chronological control of the core is based on radiocarbon (^{14}C) analysis of terrestrial macrofossils. These macrofossils, mainly pine needles, were extracted, measured, and sent to the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility in Woods Hole, MA for analysis. There, the amount of ^{14}C in the sample was measured and used to calculate a radiocarbon age – the duration of time since the sample ceased exchanging carbon with the environment. This could be when an organism died, or was buried in sediment. Due to the varied concentration of ^{14}C in the atmosphere through time, the radiocarbon age was then calibrated to a calendar age using the IntCal20 radiocarbon calibration curve in open-sourced R software packages (Blaauw & Christeny, 2011; Reimer et al., 2020) (Table 2). The resulting calendar age was then used to create a composite sequence from which the comprehensive age-depth model was derived (Figure 10) (Cook et al., 2020). Rich (2021) used a linear interpolation from ^{14}C dates to establish an age model for Sennebec Pond, while Cook et al. (2020) supplemented ^{14}C with ^{137}Cs and ^{210}Pb to create a model for Little Kennebago Lake.

3.4 Geochemical Analysis

To obtain $\delta^{15}N$, $\delta^{13}C$, and total nitrogen and carbon, samples were extracted from the core at 1 cm intervals and freeze dried, rather than oven dried, to preserve isotopic signatures (Bosley & Wainright, 1999; Kaehler & Pakhomov, 2001). After the samples were dried, approximately 4500 μg of sample was weighed, and placed in a small tin capsule. This mass was used based on the estimated nitrogen content of the samples, with the goal of bracketing the measured values within the range of standard values. The tin capsules were then sealed, rolled into a round pellet, and loaded into an autosampler to freely fall when released.

The isotopic and elemental concentrations were then measured by an EA IsoLink CN-Flash isotope ratio mass spectrometer (IRMS). Briefly, the IRMS works by ionizing the sample material in a furnace and accelerating the ions around a magnetic sector. As the ions are accelerated around the magnetic sector, slight differences in mass will separate the paths of ions. These ions are then counted by several collectors aligned to these different paths. The collectors register peaks, with associated amplitudes and areas, from which isotopic ratios are then calculated. As configured, the IRMS measures the concentrations of the two nitrogen isotopes, ^{14}N and ^{15}N, and two stable carbon isotopes, ^{12}C and ^{13}C. Additionally, elemental concentrations of nitrogen and carbon were measured using the integrated thermal conductivity detector. This detector senses changes in the thermal conductivity of a sample, which can then be corrected to a standard of known concentrations.

These outputs were then corrected to the standard and blank values, as well as for drift and linearity. For the isotopic values, I used five standards over the course of multiple runs (Table 1). The acetanilide standard was also used for elemental concentration corrections. For each run, triplicates of at least three different standards were used. The measures standard values were then compared to the known standard values to create a calibration slope and intercept, which were used to correct the measured sample values.

Standard	δ^{15}**N**	δ^{13}**N**	**%N**	**%C**
Acetanilide	-5.06	-28.23	10.36	71.09
USGS 64	1.76	-40.81	[redacted]	[redacted]
USGS 65	20.68	-20.29	[redacted]	[redacted]
USGS 66	40.83	-0.67	[redacted]	[redacted]
Glycine	0.675	-45.02	[redacted]	[redacted]

Table 1 - Standards used, with δ15N, δ13C, % Nitrogen, and % Carbon listed (United States Geological Survey, 2020).

The drift correction is to ensure the precision of the IRMS is maintained throughout a run. It was determined by placing 3-6 samples of acetanilide standard evenly spaced throughout the run, comparing the measured levels, and correcting for any deviation based on run order if necessary. The linearity correction is to ensure the precision of the IRMS is maintained regardless of the mass of samples, and ergo the amount of nitrogen and carbon in a sample. It was determined by including a group of acetanilide standards with increasing mass, making sure to bracket the expected nitrogen and carbon content found in the samples, and correcting for any deviation based on mass.

For the elemental concentrations, acetanilide standard measured values were compared to known values and a correction factor was calculated and applied to sample values. When determining carbon concentrations, samples included both organic carbon and inorganic carbon, as the samples were not acidified prior to analysis.

Standard error was calculating using the standard deviation from a triplicate of samples, run in succession. This method was used for the isotope ratios as well as elemental concentrations. In the case of C:N, error was calculated by propagating the percent content error using equation 4:

$$\frac{\Delta CN}{CN} = \sqrt{\frac{\Delta C}{C}^2 + \frac{\Delta N}{N}^2} \tag{4}$$

where CN is the ratio of carbon to nitrogen, ΔCN is the error of ratio of carbon to nitrogen, C is the total concentration of carbon in mg/g of sample, ΔC is the error of the total concentration of carbon, N is the total concentration of nitrogen in mg/g of sample, and ΔN is the error of the total concentration of nitrogen.

3.5 Watershed Characteristics

A watershed geomorphic analysis of Lower South Branch Pond was performed using ArcGIS to provide information on the characteristics and spatial distribution of historical and modern processes that may influence geochemistry. The analysis used a 0.7 m lidar-derived digital elevation model (DEM) from the State. The drainage basin was defined using *StreamStats*, an online USGS application that uses 10 m DEMs to delineate user-specified drainage areas. Similar analysis was performed by Cook et al. (2020) and Rich (2021) for Little Kennebago lake and Sennebec Pond, respectively.

To construct records of land use for each site, a combination of historical records, photographs, aerial and satellite imagery, and ArcGIS analysis were used. For periods before aerial and satellite imagery, I relied on historical accounts and proxies to determine the extent and magnitude of land use change in an area. Though difficult to quantify, these data sources are valuable in that they provide some context when interpreting results, though this context comes with increased uncertainty, much as it does with climate data discussed earlier. In some cases, like timber harvests and conversion to pastureland, these effects can be quantified using relic indicators, such as forest composition or stone walls (Johnson & Ouimet, 2016), though these techniques have yet to be broadly applied.

The earliest aerial imagery I could find for the Lower South Branch Pond area is from 1969, well after the establishment of the park and, therefore, and basin-scale changes in land use. As such, the record of land use of the area was constructed entirely by historical accounts. Cook et al. (2020) and Rich (2021) use similar historical records to determine the extent and type of land use change before available aerial imagery.

To quantify more recent land use change, Cook et al. (2020) created orthophotomosaics from aerial photographs sets ranging from 1958 to 2018 to calculate the total areas of timber harvest within the Little Kennebago Lake watershed using ArcGIS. Additionally, they quantified changes to the road network within the watershed by digitizing USGS topographic maps from 1931 to 2018. Similarly, Rich

(2021) used historical aerial single frame imagery from 1953-1956 as well as 2018 National Agriculture Imagery Program (NAIP) orthoimagery to determine past and recent land cover information.

3.6 Statistical Analysis

In order to determine the statistical significance of observed trends both pre- and post-Euro-American settlement, sediment records were divided into two groups based on estimated arrival date: Lower South Branch Pond – 1876; Little Kennebago Lake – 1891; and Sennebec Pond – 1700. Then, for each variable, the mean and standard deviation were determined for respective pre- and post-populations. Finally, two tests were employed to determine the statistical significance of the differences between populations: a two-sample t test to test the hypothesis that the pre- and post-samples have equal means; and a two-sample f test to test the hypothesis that the pre- and post-samples have the same variance. The tests were run in *Matlab* using the ttest2 and vartest2, respectively.

From these tests, p-values were calculated. A p-value is a statistical measurement used to validate a hypothesis against observed data. For the means, a $p < 0.05$ indicates that the hypothesis can be rejected, and that the pre- and post-samples have significantly different means. For standard deviations, a $p < 0.05$ also indicates that the hypothesis can be rejected, and that the pre- and post-samples have significantly different variances, which in the context of this study would suggest that variance increases significantly after Euro-American disturbance.

4.0 Results

4.1 Lower South Branch Pond

4.1.1 Core Description

The surface-sediment core (core 21-1) recovered from Lower South Branch Pond in May, 2021 is 60 cm long and is composed primarily of dark brown to brown

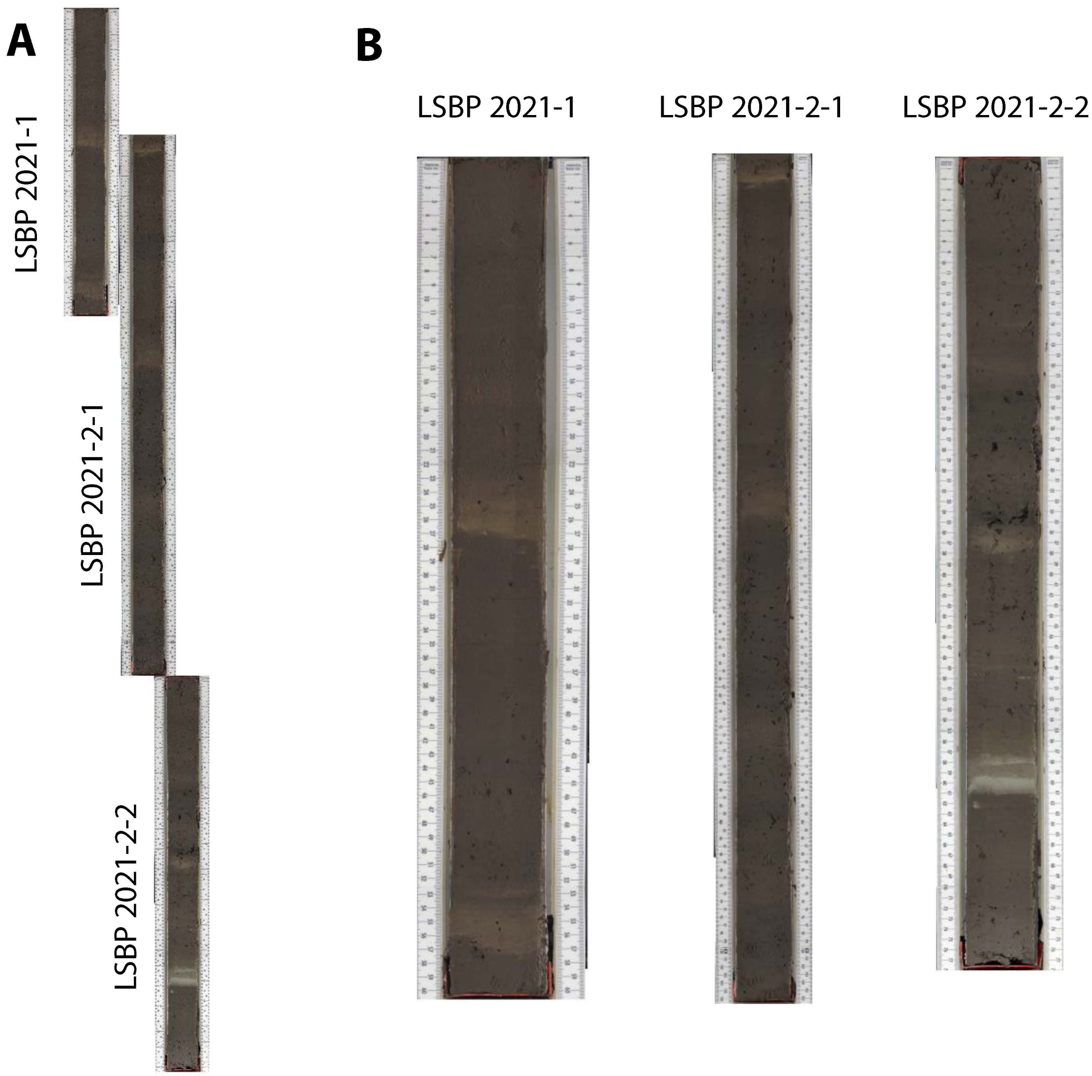

Figure 7- (A) Composite optical images of split Lower South Branch Pond cores 21-1, 21-2-1, and 2021-2-2, oriented to show overlap and correlation. (B) Enlarged, individual core sections photographs. Clastic event layers are clearly visible in the split core.

gyttja and two distinct tan layers of clastic, inorganic material that range in thickness from 1 to 3 cm (Figure 7). The clastic layers visible in the split core are characterized by reduced LOI, and elevated magnetic susceptibility, ρ_{db}, and potassium content (Figure 8). The lower transition of both clastic layers is abrupt, while the upper transition is gradual. The lower bounds of the clastic layers can be found at a depth of 27 cm and 57 cm in the core.

The longer, deep core (cores 21-2-1 and 21-2-2), recovered at the same time and location as core 21-1, was divided into two core sections in the field to facilitate transportation, and are 106 and 77 cm long, respectively. Core 21-2-1, the upper section, is composed primarily of dark brown to brown gyttja with sporadic light grey or tan layers of clastic material ranging in thickness from 1 to 4 cm (Figure 7). Like core 21-1, the clastic layers in the split core are characterized by reduced LOI, and elevated magnetic susceptibility, ρ_{db}, and potassium content (Figure 8). Similarly, the lower transition of the clastic layers tends to be abrupt, while the upper transition is gradual. The characteristics and location of the top two clastic layers in core 21-2-1 suggest they correspond to the two clastic layers found in core 21-1, and the two cores overlap in this section.

Core 21-2-2, the lower section, is composed primarily of dark brown to brown gyttja with sporadic light grey or tan layers of clastic material ranging in thickness from 0.25 to 4 cm (Figure 7). Like core 21-1, the clastic layers in the split core are characterized by reduced LOI, and elevated magnetic susceptibility, ρ_{db}, and potassium content (Figure 8). Similarly, the lower transition of the clastic layers tends to be abrupt, while the upper transition is gradual. The most significant clastic layer, both in terms of appearance and clastic proxies, found in all cores is located at a depth of 57 to 61 cm, or at a composite depth of 188 to 192 cm.

Initial analysis of the Lower South Branch Pond cores (cores 21-1, 21-2-1, and 21-2-2) using the ITRAX scanning x-ray fluorescence (XRF) core scanner produced magnetic susceptibility and potassium (K) counts, both proxies for clastic input (Figure

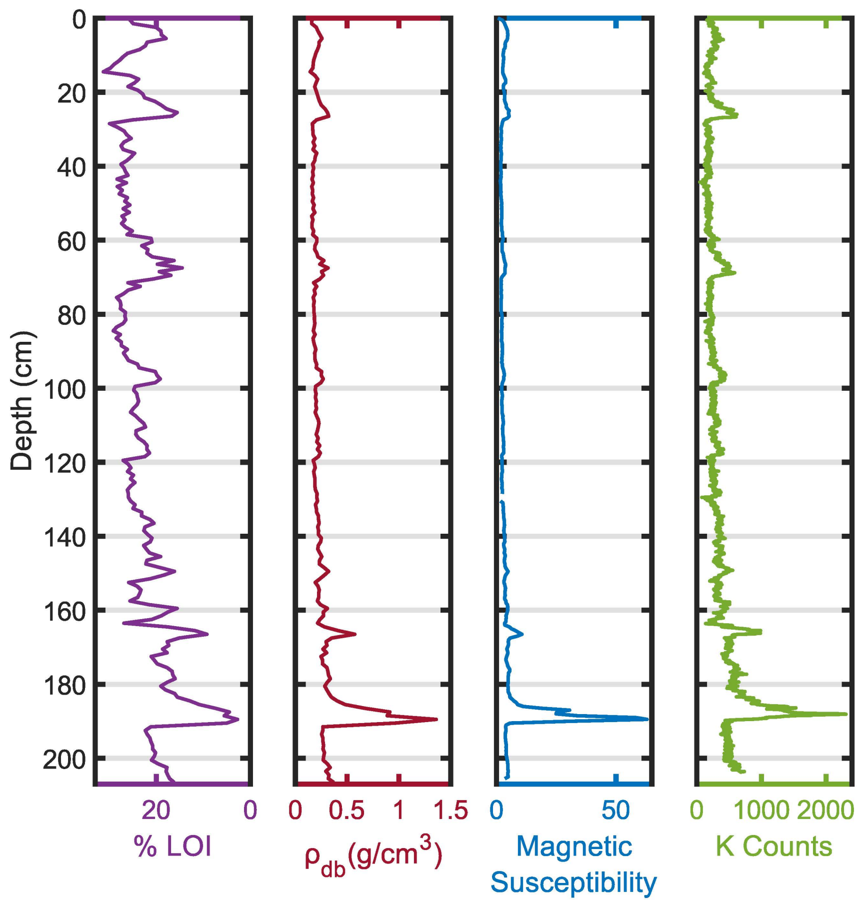

Figure 8- Downcore variations in percent loss on ignition (LOI; note reversed scale for X axis), ρ_{db}, magnetic susceptibility, and relative abundance of potassium (K) for Lower South Branch Pond.

8). These data were supported by measured percent loss on ignition (% LOI) and dry-bulk density (ρ_{db}), also proxies for clastic input (Figure 8). All four datasets indicate the most significant event layer, as indicated by clastic content, is found at a composite

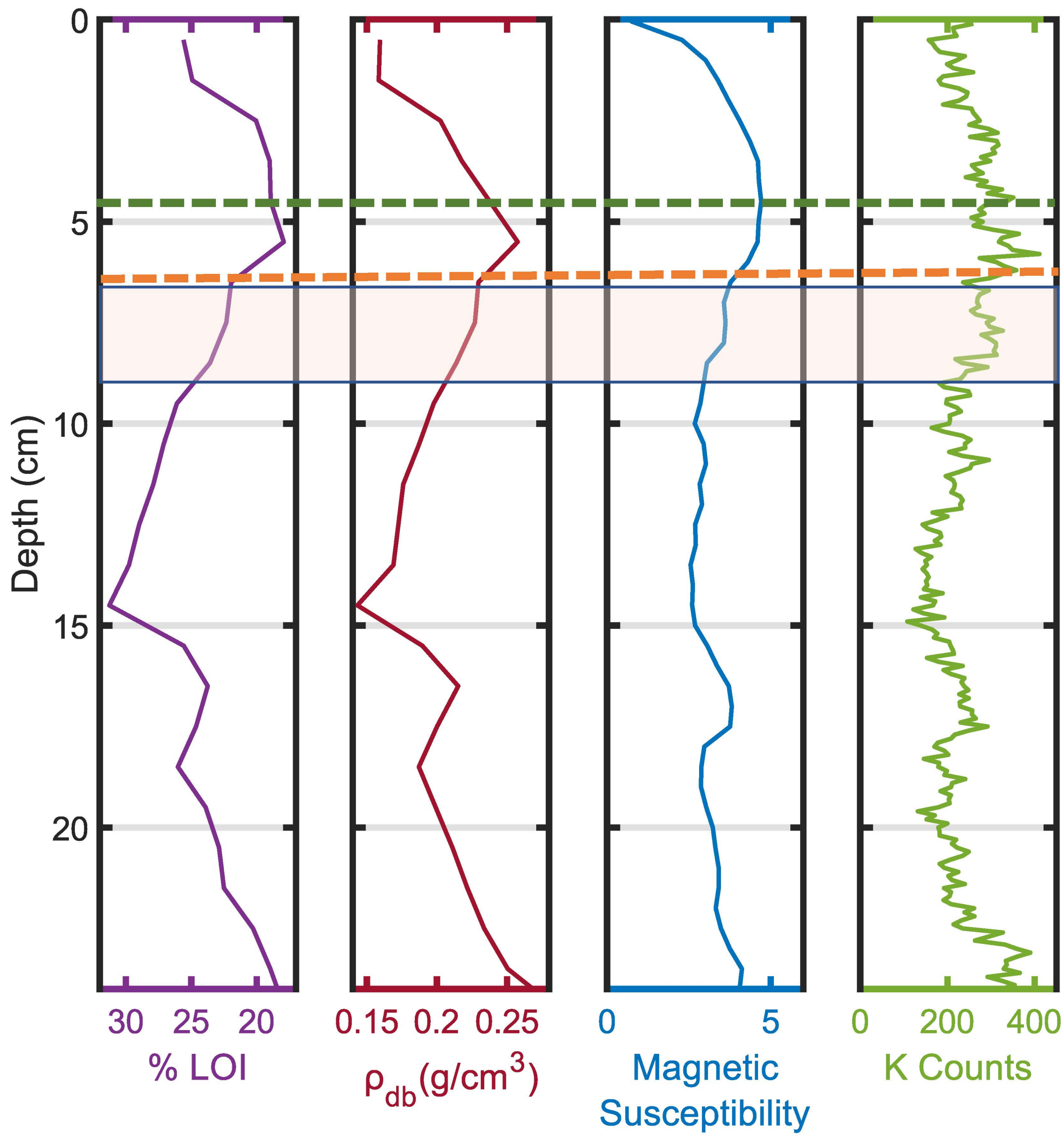

Figure 9- Downcore variations for the most recent ~500 years in percent loss on ignition (LOI; note reversed scale for X axis), pdb, magnetic susceptibility, and relative abundance of potassium (K) for Lower South Branch Pond. Orange shaded region represents estimated period of Euro-American incursion; orange dashed line represents approximate date of a large forest fire in the watershed (1903); green dashed line represents approximate date of the establishment of Baxter State Park (1938).

depth of approximately 190 cm, or an age of approximately 6,000 years BP. Excluding this event, % LOI displays the greatest variability, ranging from ~32% to ~9% over the record. Significant event layers appear to correspond across all four proxies, though the magnitude of events is most pronounced in % LOI. Focusing on the study period of the last 500 years, there is, again, strong agreement between all four proxies as to periods of increased clastic input (Figure 9).

These datasets were used to create the composite core by comparing and aligning prominent "clastic peaks" (Cook et al., 2020). The resulting composite uses the top 29 cm of the surface core, 21-1, and then transitions to core 21-2-1, beginning at a section depth of 5 cm. After scaling the short core by 1.67 its original length to account for some compression during coring, this method yields the fewest discrepancies between cores 21-1 and 21-2 relative to other interpretations (Cook, T., personal communication, October 22, 2021).

4.1.2 Age-Depth Model

Chronology results indicate that the record, consisting of core sections 21-1, 21-2-1, and 21-2-2, roughly spans the last 6600 years (Figure 10). Chronological control is provided by six radiocarbon ages (Table 2; Figure 10). The sediment accumulation rate, based on these ages, can be interpreted as having remained relatively stable across the entirety of the record.

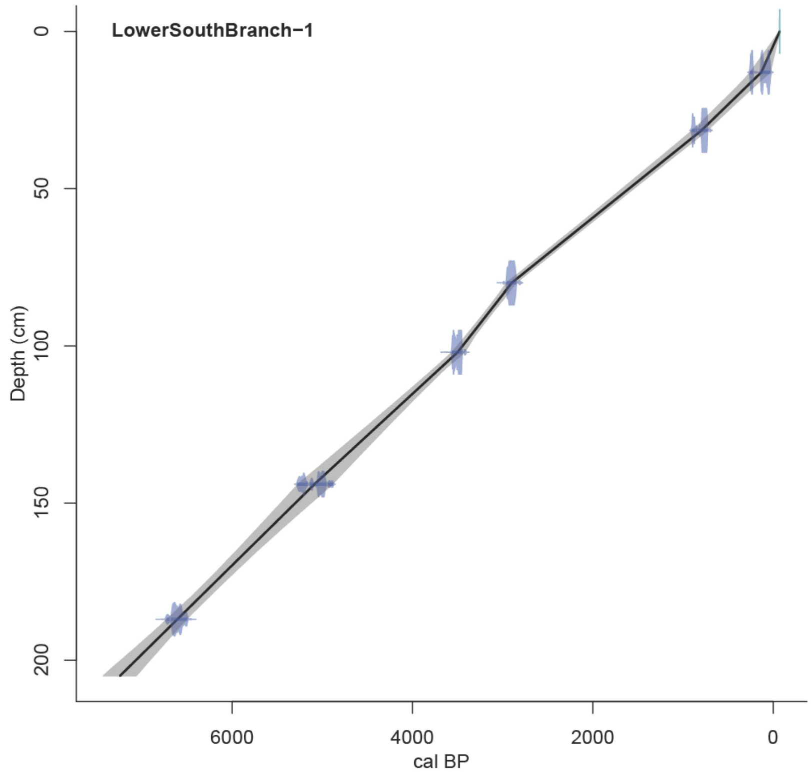

Figure 10- Age-depth model derived from radiocarbon ages based on terrestrial macrofossils (plant/wood). 1 sigma probability distributions are given for each calibrated age (shaded blue). Gray shaded region represents the potential envelope of age-depth models that could fit through available radiocarbon ages.

It should be acknowledged that only one control point lays within the most recent 500-year section of the core. Due to this lack of data for the period of focus, as well as the inherent uncertainty associated with calibrated ages, there exists considerable uncertainty in the precise timing of events over this interval. While the exact date of events or trends in the data cannot be confidently determined, the sequence of relative changes in variable can be stated with some certainty. Alternative dating techniques, such as ^{210}Pb, would have provided for better control on the last 200 years, but were cost-prohibitive.

Lab Number	Composite Depth (cm)	Source Material	Radiocarbon Age (Years BP)	Age Error	Calibrated Age Min (Years BP)	Calibrated Age Max (Years BP)	Calibrated Range Probability
OS-165301	13	Plant/Wood	90	25	257	224	25.8
					140	32	69.1
OS-165302	31.5	Plant/Wood	885	30	905	866	20
					856	846	2
					828	727	72.9
OS-162303	80	Plant/Wood	2800	20	2957	2852	94.9
OS-162227	102	Plant/Wood	3280	25	3562	3452	94.8
OS-162228	144	Plant/Wood	4440	30	5279	5166	32.9
					5136	5101	7
					5077	4957	47.6
					4936	4880	7.4
OS-162229	187	Plant/Wood	5810	40	6730	6698	6.3
					6678	6497	88.5

Table 2- Radiocarbon sample information. Included are all possible calibrated age ranges. The reported age error is an estimate of the precision (repeatability) of measurement for a single sample (National Ocean Sciences Accelerator Mass Spectrometry Facility, 2023). The calibrated range probability is based on 95% (2-sigma) confidence interval (Blaauw et al., 2022).

4.1.3 Isotope Geochemical Analysis

Geochemical analysis of Lower South Branch Pond sediments includes $\delta^{15}N$, $\delta^{13}C$, total nitrogen, total carbon, and C:N records (Figure 11). For $\delta^{15}N$, values range from 1.44 to 3.05 ‰, with an average of 2.65 ‰, and an average error of 0.05 ‰. The signal is stable from the onset of the record around 1500 CE to shortly after 1900 CE, when $\delta^{15}N$ begins to rise, with a peak roughly 40 years later, though the peak falls just short of pre-1875 CE values. The values then continue to fall to a minimum around 2000 CE.

For $\delta^{13}C$, values range from -32.07 to -25.94 ‰, with an average of -28.65 ‰, and an average error of 0.36 ‰. The signal peaks at the onset of the record at approximately 1500 CE. It then slowly declines until around 1960 CE, at which point the rate of decline increases and the values drop suddenly to a minimum around 2000 CE.

For total nitrogen, values range from 5.77 to 9.80 mg g^{-1}, with an average of 7.32 mg g^{-1}, and an average error of 0.23 mg g^{-1}. The signal peaks shortly after 1800 CE,

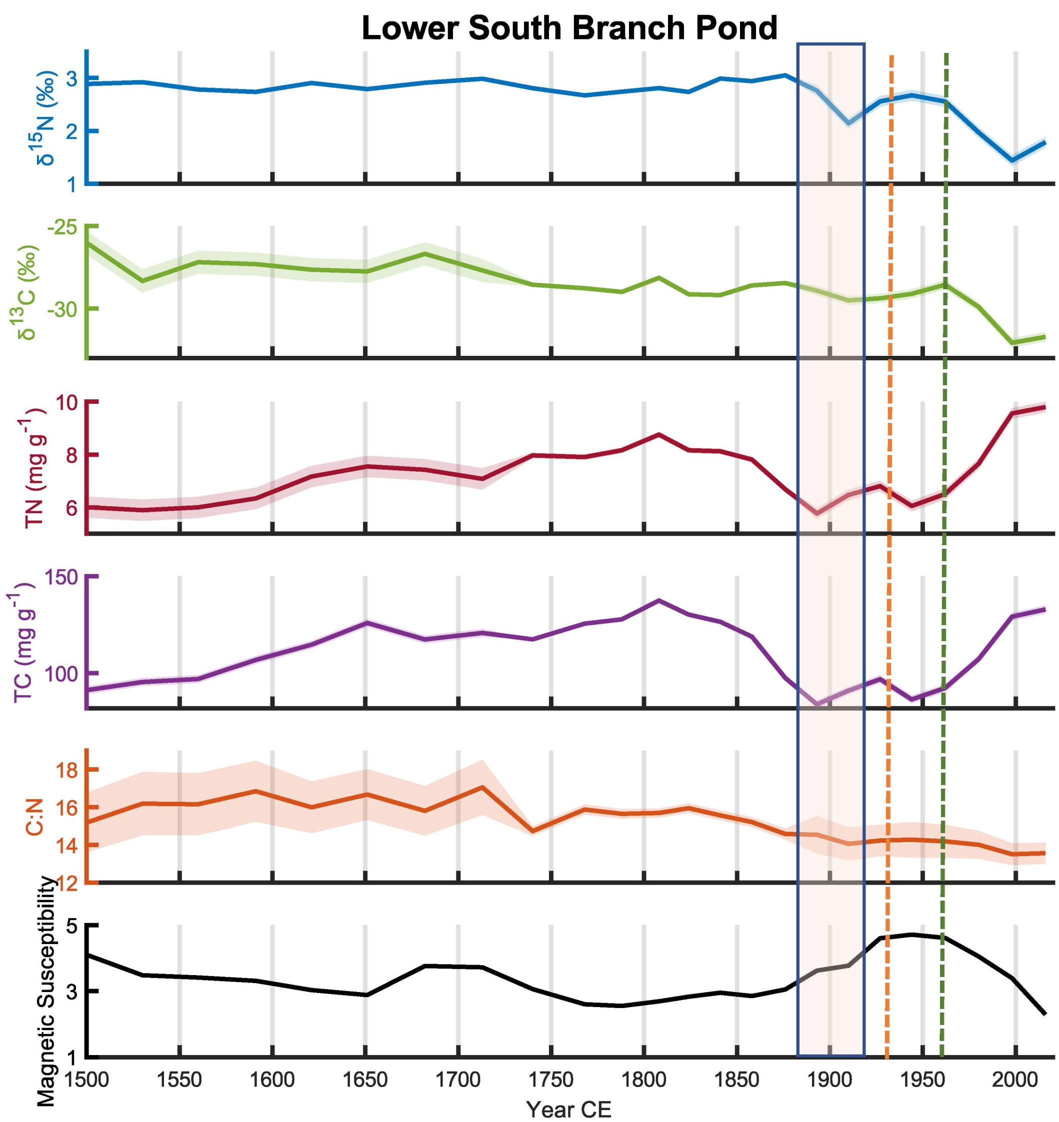

Figure 11- Lower South Branch Pond $\delta^{15}N$, $\delta^{13}C$, total nitrogen, total carbon, C:N, and magnetic susceptibility from 1500 to 2020. Orange shaded region represents estimated period of Euro-American incursion; orange dashed line represents approximate date of fire (1903); green dashed line represents approximate date of the establishment of Baxter State Park (1938).

at which point it falls to the pre-peak minimum shortly before 1900 CE. The signal then rises briefly, falls close to the minimum again, the rises rapidly in the last 70 years of the record.

For total carbon, values range from 83.88 to 137.42 mg g^{-1}, with an average of 111.23 mg g^{-1}, and an average error of 1.40 mg g^{-1}. The signal peaks shortly after 1800 CE, and subsequently decline, at times rapidly, reaching a minimum shortly before 1900 CE. Like total nitrogen, total carbon then rises briefly, falls close to a minimum again, and rises rapidly in the last 70 years of the record.

For C:N, which is calculated from total nitrogen and carbon, values range from 13.51 to 17.05, with an average of 15.23, and an average error of 0.85. The signal remains relatively stable from the onset of the record until approximately 1730 CE. This period has the highest possible error in the signal. After 1730 CE, C:N declines slightly and, again, remain relatively stable until approximately 1830, at which point the signal slowly declines, reaching a minimum around 2000 CE.

4.2 Little Kennebago Lake

4.2.1 Geochemical Analysis

Geochemical analysis of Little Kennebago Lake sediments produced $\delta^{15}N$, $\delta^{13}C$, total nitrogen, total carbon, and C:N records (Figure 12). For $\delta^{15}N$, values range from 1.33 to 3.05 ‰, with an average of 2.65 ‰, and an average error of 0.05 ‰. The signal, while experiencing some minor variability, remains relatively consistent from 1500 CE until approximately 1900 CE. Shortly thereafter, there is a sharp spike in $\delta^{15}N$, immediately followed by a sharp decline, taking place within a 10–20-year period. Afterwards, the signal shows an increased variability, falling to a minimum around 1950 CE, and slowly rising from then on.

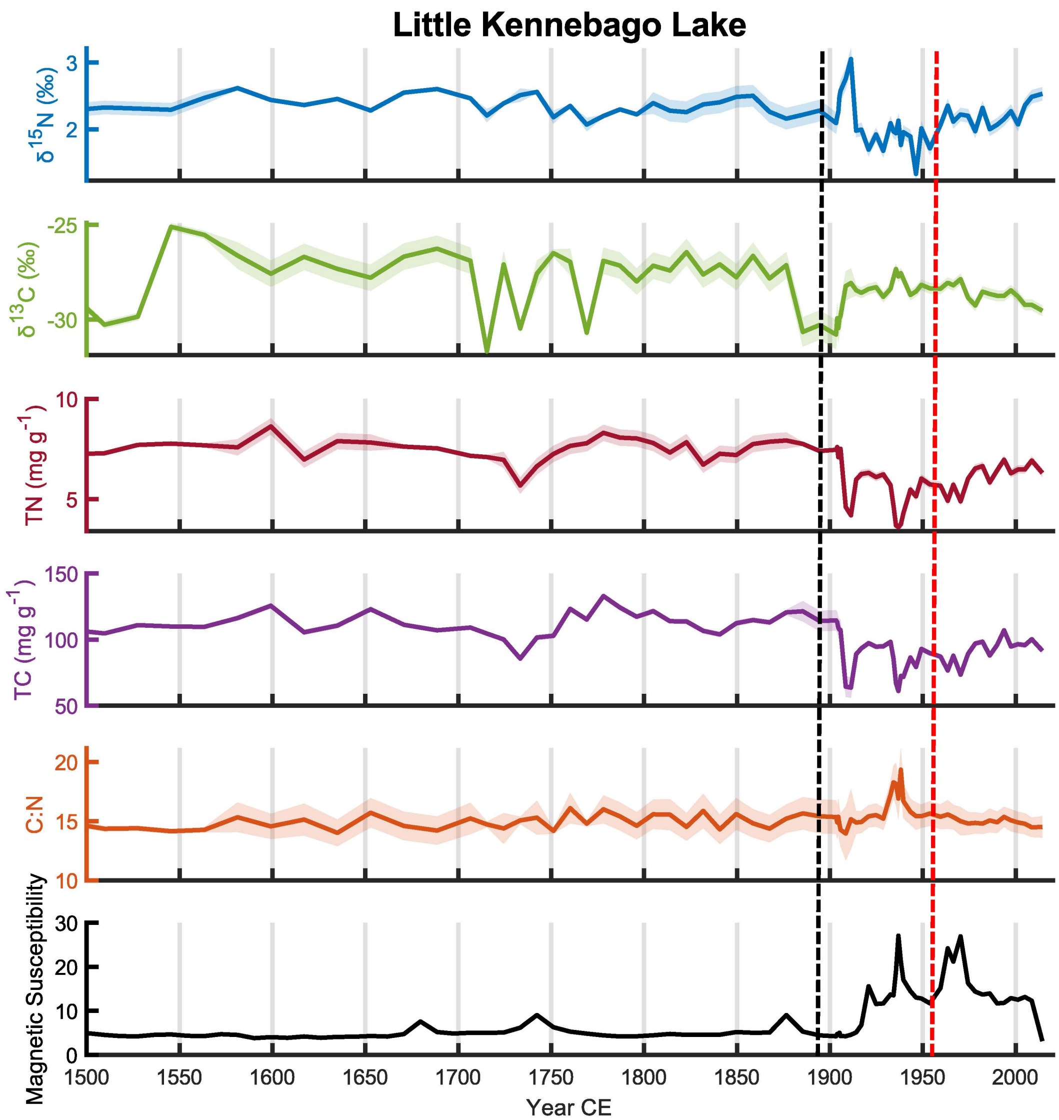

Figure 12- Little Kennebago Lake $\delta^{15}N$, $\delta^{13}C$, total nitrogen, total carbon, C:N, and magnetic susceptibility from 1500 to 2020. Black dashed line represents estimated date of Euro-American incursion; red dashed line represents a shift in the primary method of log transport.

For $\delta^{13}C$, values range from -31.68 to -25.11 ‰, with an average of -28.24 ‰, and an average error of 0.46 ‰. The signal is markedly more varied than the other geochemical markers, displaying little in the way of trends pre- and post-Euro-American settlement.

For total nitrogen, values range from 3.61 to 8.63 mg g^{-1}, with an average of 6.65 mg g^{-1}, and an average error of 0.24 mg g^{-1}. The signal, while experiencing some minor variability, remains relatively consistent from 1500 CE until approximately 1900 CE. At that point signal variability increases significantly, and total nitrogen values fall dramatically, approaching the minimum, after which they rise briefly and suddenly, and fall steeply again around 1940 CE to the minimum. This increased variability continues over the most recent 70 years of the record, displaying a slight increase to modern-day.

For total carbon, values range from 61.12 to 132.90 mg g^{-1}, with an average of 100.65 mg g^{-1}, and an average error of 2.49 mg g^{-1}. The signal is almost identical to that of total nitrogen, highlighted by a sharp decline shortly after 1900 CE, the same double minima structure in the subsequent 50 years, and a variable, slight increase over the most recent 70 years of the record.

For C:N, values range from 13.95 to 19.34, with an average of 15.23, and an average error of 1.10. Though is experiences some minor variability, the C:N signal is perhaps the most stable of all geochemical indicators, displaying no significant deviations from the average value except for a single peak around 1940 CE.

4.3 Sennebec Pond

4.3.1 Geochemical Analysis

Geochemical analysis of Sennebec Pond sediments produced $\delta^{15}N$, $\delta^{13}C$, total nitrogen, total carbon, and C:N (Figure 13). For $\delta^{15}N$, values range from 1.70 to 3.81 ‰, with an average of 2.71 ‰, and an average error of 0.13 ‰. The signal shows relative stability throughout the record, with some interspersed, mild variability. The signal reaches a minimum around 1770 CE, and a maximum around 1950 CE. In the most recent 70 years of the record, the signal displays a slow decline.

For $\delta^{13}C$, values range from -30.93 to -25.51 ‰, with an average of -27.64 ‰, and an average error of 0.30 ‰. The signal begins low, hovering around the minimum until shortly after 1700 CE, at which point it increases sharply. It remains elevated, consistently approaching the maximum value, until around 1900 CE, after which a slow decline, taking place over the most recent 70 years of the record, is discernable. For total nitrogen, values range from 4.81 to 7.02 mg g^{-1}, with an average of 5.81 mg g^{-1}, and an average error of 0.06 mg g^{-1}. The signal experiences a period of elevated values from approximately 1700 to 1830 CE. It subsequently drops, experiencing a minimum shortly before 1950 CE, and slowly rises in the most recent 70 years of the record to the average value.

For total carbon, values range from 48.83 to 80.11 mg g^{-1}, with an average of 64.85 mg g^{-1}, and an average error of 2.40 mg g^{-1}. The record begins with elevated values that remain somewhat stable until around 1780 CE. The signal then slowly declines to the minimum value around 1860 CE, where it remains consistent until the

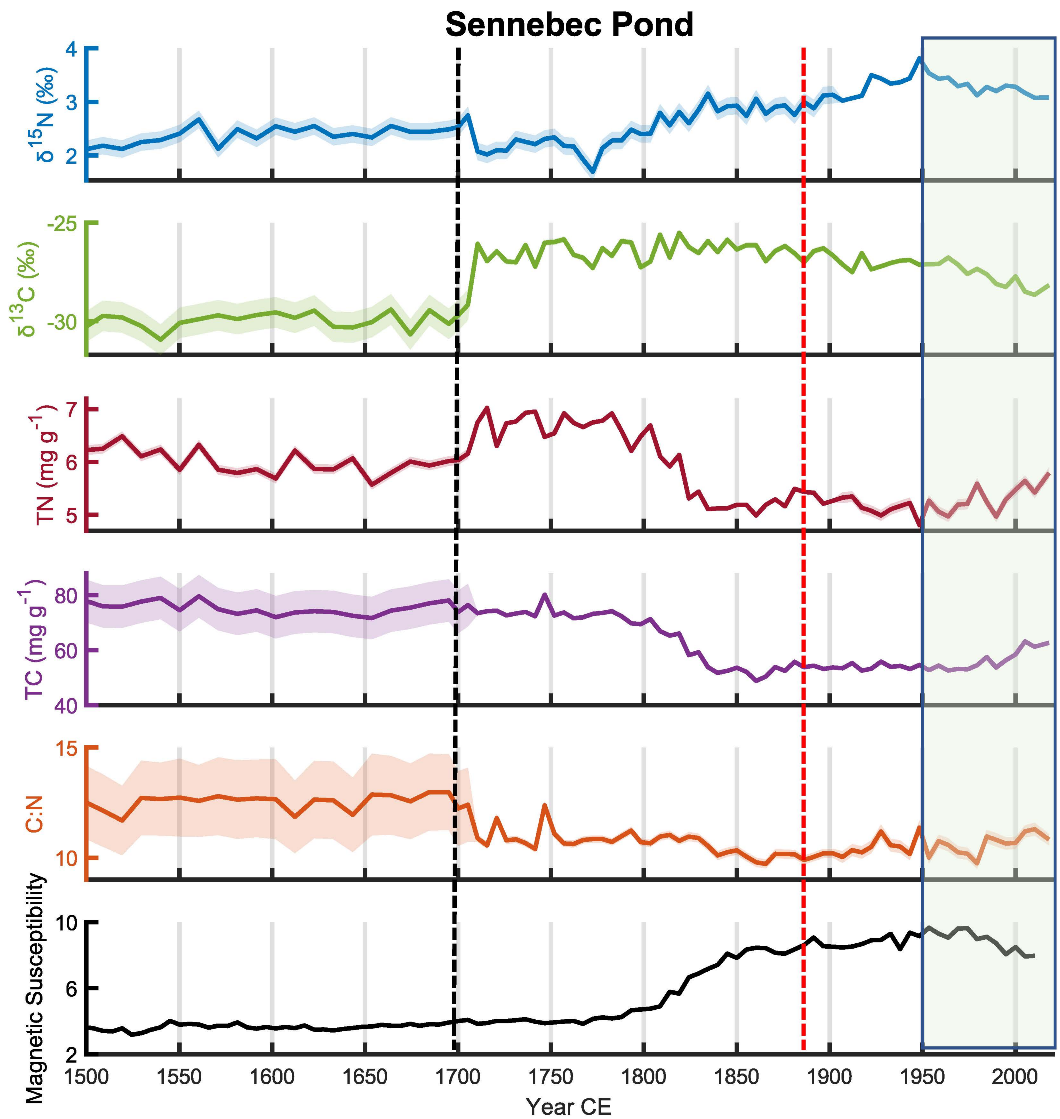

Figure 13- Sennebec Pond $\delta^{15}N$, $\delta^{13}C$, total nitrogen, total carbon, C:N, and magnetic susceptibility from 1500 to 2020. Black dashed line represents estimated date of Euro-American incursion; red dashed line represents the peak of land conversion in the area; green shaded region represents the trend of reforestation noted by Rich (2021).

most recent 30 years of the record, when it climbs slowly, ending up just below the average value. It is worth noting that the period before 1700 CE experiences the largest uncertainty of all the geochemical records in the study. This can be attributed to a decrease in precision of the instrument while these samples were being run. This uncertainty is propagated to C:N values as well.

For C:N, values range from 9.71 to 12.98, with an average of 11.12, and an average error of 0.62. Marked by elevated error in the first 200 years of the record, the signal begins just above the average value and remains relatively stable until 1700 CE, at which point it falls sharply. After 1750 CE, the values remain consistently low. A minimum is reached around 1860 CE. After 1930, there is a slight increase in the variability of the signal, though it still remains below pre-1700 CE values.

4.4 Statistical Tests

The resultant *p*-values generated from both the two-sample test and the two-sample f test for all five geochemical indicators can be found below in table 3. For the means, a $p < 0.05$ indicates that the hypothesis can be rejected, and that the pre- and post-Euro-American settlement samples have significantly different means. For standard deviations, a $p < 0.05$ also indicates that the hypothesis can be rejected, and that the samples have significantly different variances.

Variables		Sen	LKL	LSBP
$\delta^{15}N$	mean	0.0002	0.0003	0.0011
	SD	2.07E-06	5.22E-06	5.69E-07
$\delta^{13}C$	mean	2.46E-26	0.0009	0.0010
	SD	0.0012	0.0002	0.2703
Total N	mean	0.1269	1.00E-10	0.8350
	SD	3.35E-06	0.0001	0.1154
Total C	mean	4.16E-09	2.33E-10	0.0306
	SD	2.27E-08	0.0130	0.3648
C:N	mean	5.51E-23	0.0129	1.10E-07
	SD	0.0284	0.0005	0.1435

Table 3- The resulting p-values from both the two-sample t test for means and the two-sample f test for variance. p-values < 0.05 reject the hypothesis and are highlighted in red.

5.0 Discussion

5.1 Comparison of Three Study Sites

In comparing the geochemical and sedimentary records of all three study sites, there are several differences that provide more detail around the timing, mechanism, and effects of land-use change on the three lakes. Little Kennebago Lake displays the largest variation in magnetic susceptibility, a proxy for clastic sediment input (Figure 12) when comparing all three study sites. This may indicate that the magnitude of land-use change within the basin, a likely mechanism for increased clastic input, was more impactful relative to Lower South Branch Pond and Sennebec Pond. This is significant in that all three sites experienced some level of land-clearance, and the difference in variation could indicate the relative difference in the scale of change. Differences with historical timelines, as well as discrepancies between the accuracy of historical record-keeping, have previously made comparison between the sites difficult to quantify. In comparing proxies of clastic inputs, like magnetic susceptibility, the relative contributions of activities like timber clearance and harvest between the lakes can become clearer.

Looking at the entirety of the records of magnetic susceptibility, both Little Kennebago Lake (Figure 12) and Lower South Branch Pond (Figure 11) experience periods of increase and decrease over time. The most substantial variation in the record of Little Kennebago Lake occurs after the estimated incursion of humans to the area, whereas the magnitude of change within the record of Lower South Branch Pond is relatively consistent throughout. Both of these records differ from Sennebec Pond (Figure 13), which shows an increase in magnetic susceptibility between approximately 1800 and 1850, which separates periods of otherwise stable values.

The geochemical records display similar trends. Once again, Little Kennebago Lake displays the greatest range in $\delta^{13}C$, TN, TC, and C:N (Figure 12). Like magnetic susceptibility, the most substantial variation in the geochemical record of Little Kennebago Lake occurs after the proposed incursion of humans to the area, in the most recent ~ 120 years of the record. This is supported by the two-sample t test and to-sample f test (Table 3), which had p-values < 0.05 for all five geochemical variables, indicating statistically-significant differences in mean and variance before and after Euro-American incursion. Unlike magnetic susceptibility, this in in rough agreement with the record of Lower South Branch Pond (Figure 11). This may indicate that while the differences between the two basins were substantial enough to cause disparity between the physical signature of land-use change, the geochemical signatures support the hypothesis that subtle changes in the style and magnitude of land-use change are detectable in the geochemical record. All three of the sites show roughly the same baseline $\delta^{15}N$, between 2-3 ‰, and $\delta^{13}C$, between -32 and -25 ‰, which is expected based on similarities in local ecology, geology, climate, and environmental policy.

The biggest difference in the geochemical records of the three sites is the timing of the periods of considerable variation. With the exception of $\delta^{15}N$, the magnitude of change within the record of Lower South Branch Pond is relatively consistent throughout. Similar to magnetic susceptibility, this may support the idea that the anthropogenic impacts in the basin were of a similar magnitude to natural processes. However, the decline of $\delta^{15}N$ is limited to the upper portion of the record, and the disagreement in timing indicates some other factor is independently impacting the signal of nitrogen.

Conversely, Little Kennebago Lake again shows a period of considerable variation after the estimated incursion of humans to the area (Table 3), and Sennebec Pond shows longer periods of relative stability throughout the record. Both illustrate fundamental differences in the style of, and response to, land-use change in the respective basins.

5.2 Lower South Branch Pond

The geochemical and sedimentary record of Lower South Branch Pond is primarily influenced by humans through three events and one, more general, trend: the onset of Euro-American influence, in the form of timber harvest, beginning in the area in the latter half of the 19th century; a fire tearing through the region in 1903; the watershed's inclusion in the creation of Baxter State Park in 1938; and, more generally, an increase in the magnitude and rate of nitrogen and carbon emissions of humans in the last 150 years. While the low resolution of some of the data, due in part to the low sedimentation rate within the basin, makes it difficult to ascertain, the approximate timing and appearance of certain trends suggests possible correlation between changes in the record and these anthropogenic influences.

Over the course of the last 500 years, the sedimentary record shows considerable variability pre- and post-Euro-American incursion (Figure 9). The magnitude of clastic input from the period concurrent with land use change is comparable to those associated with natural events. This is shown by the similar values found in the relative maxima of the peaks occurring both pre- and post-Euro-American incursion. This may suggest that the extent of change within the basin was of a similar order of magnitude of a natural event, though the mechanism of change may have been different, which could affect the recovery of the signal to pre-perturbation levels. The $\delta^{15}N$, $\delta^{13}C$, and C:N signals in the geochemical record (Figure 11) remain relatively stable until the late 18th century, which supports the hypothesis that the watershed was subject to limited perturbation in the preceding period.

At the onset of what is generally accepted as the period of more substantial incursion, the latter half of the 19th century, after sustained periods of steady increase, the cores show a decline in both total nitrogen and total carbon. The magnitude of change in total carbon is slightly greater, leading to a decreased C:N ratio over the same period of time. As this trend is reversed later in the 20th century after noteworthy changes in land management, I infer that this initial decline may be a result of land use change within the basin, as regional trends such as atmospheric deposition have persisted throughout. As these changes coincide with a period of increased magnetic susceptibility, the decrease in total carbon and nitrogen could be ascribed to a larger proportion of minerogenic sediment being deposited. Interestingly, however, there is little change in C:N ratio, which would indicate minimal change in the source of organic matter.

I do observe an increase and relative peak in clastic input, as recorded by % LOI, magnetic susceptibility, and dry-bulk density, shortly after 1900, which may be associated with the fire of 1903 (Figure 9). This would be in line with other studies that have found increased erosion rates after fire events (Ahlgren & Ahlgren, 1960; Chen, 2006). While the effects of forest fires on timber harvest be vary depending on the several factors such as burn severity and the biophysical setting of the forest (Peterson et al., 2009), there is little evidence of continued harvest around Lower South Branch Pond after the fires of 1903. For the three aforementioned clastic proxies, relative peaks occur shortly thereafter, around 1927, which could be the delayed signal of increased erosion post-fire.

The period between 1900 and 1938 shows a small spike in both total nitrogen and carbon, which can be interpreted several ways. As previously stated, the fire of 1903 and the subsequent post-fire period severely limited extent of timber harvest, and could have increased rates of erosion, within the basin (Ahlgren & Ahlgren, 1960; Chen, 2006). Additionally, delivery of carbon and nitrogen from burned vegetation to the forest floor can take decades (Peterson et al., 2009), which may explain the delayed peak in total nitrogen and carbon, which occurs around 1927. However, due to a relatively static C:N ratio, the changes in both total nitrogen and carbon could most likely be attributed to a change in flux of clastic sediments.

Interestingly, while other signals experience relative peaks, $\delta^{15}N$ experiences a sudden drop, which is inconsistent with similar studies (Moreno et al., 2016; Stephan et al., 2015). However, the subsequent rise in $\delta^{15}N$, which would be consistent with others, may illustrate the response time of terrestrial events as recorded by $\delta^{15}N$ in the sediment record. This offset from total nitrogen and carbon could indicate subtle differences in the geochemical response of the watershed after fire events, and could merit further investigation.

From the data point most closely associated with the establishment of Baxter State Park in 1938 until present, during which the land has been managed to minimize human impact, all of the clastic proxies show decline, which is in agreement with similar studies (Cook et al., 2020; McLauchlan et al., 2007; Rich, 2021). Geochemically, total nitrogen and carbon increased to at or beyond their respective levels pre-Euro-American influence, while $\delta^{15}N$ and $\delta^{13}C$ decline until the most recent data point, which is in strong agreement with McLauchlin et al. (2007).

5.3 Little Kennebago Lake

From previous research (Cook et al., 2020), it is suggested that the landscape around Little Kennebago Lake, while resilient to natural hydrologic disturbance, has potential of large natural variability in sediment yield due to rare events driving increased delivery of clastic sediment to the lake. Further, the findings demonstrated that human activities can influence suspended sediment yield in a moderately impacted watershed with low background erosion rates. Finally, the recent sedimentary record suggests that forestry best management practices may have effectively reduced the occurrence of the most severe erosional events, despite increased rates of timber harvest.

The geochemical record (Figure 12) shows varied responses to timber harvest in the watershed. Despite a record of only 500 years, $\delta^{15}N$ displays relative stability in the period preceding Euro-American influence (~1891). The same can be said for C:N; though total nitrogen and carbon do vary somewhat during this period, they do so proportional to each other. This may suggest that while the magnitude of nitrogen and carbon input to the basin may have fluctuated, the source did not vary meaningfully. $\delta^{13}C$, however, does experience substantial variability before 1891. This could indicate that the lake underwent periods of higher and lower primary productivity, independent of anthropogenic influence. It could also suggest that $\delta^{13}C$ may be the most sensitive of the studied geochemical indicators to changes in sediment yield.

Changes in the geochemical record support the sedimentary record in constraining the specific timing of noteworthy land use change in the basin as being around 1900 CE, when the record shows a marked spike in $\delta^{15}N$ and drops in total nitrogen and carbon. These signal shifts are in agreement with those found in McLauchlin et al. (2007), which were associated with timber harvest. The most substantial deviation in $\delta^{15}N$, total nitrogen, and total carbon occurs at this onset of timber harvest, suggesting that this initial phase of land use change had the greatest impact on the geochemistry within the basin. This would support the findings of Cook et al. (2020). These findings are further supported by the two-sample t test and two-sample f-test (Table 3), which indicated that the all five geochemical indicators had both significantly different means and different variances pre- and post-Euro-American settlement. $\delta^{15}N$ wavers, experiencing a minimum shortly before 1950 before, generally, increasing over the most recent 70-year period. Likewise, both total nitrogen and carbon also experience minimums during this time, though they precede the $\delta^{15}N$ minimum by a decade, occurring around 1936

These minimums do correspond with the most noteworthy event in the 500-year C:N record, which also occurs between 1932 and 1936. This timing corresponds to the creation of the Upper Dam, which occurred in 1932. While it could be proposed that these values are in response to the construction of the dam, these geochemical signatures appear to return to pre-1932 levels within a few years, suggesting these impacts to be short-lived.

Shortly after 1950, the primary method of log transport shifted from river-based drives to trucking, though this shift is difficult to detect in the geochemical record. Aside from two major signals, at the onset of harvest and the construction of the dam, there remains increased variability in the record after timber harvest began. This would suggest the increased variability is a direct result of timber harvest.

Furthermore, it continues to modern-day, indicating that while changes in forestry best management practices may have effectively reduced the occurrence of the most severe erosional events, it has done little to minimize geochemical disturbances. This is further supported by the concurrent work of Seal (2022), who has found greater variability in strontium isotopes after onset, and suggests that timber harvest in the Little Kennebago Lake watershed has fundamentally altered sediment provenance.

5.4 Sennebec Pond

The work of Rich (2021) suggests accelerated patterns of sediment delivery to Sennebec Pond corresponds to periods of accelerated land use change. Additionally, GIS analysis showed recent trends of reforestation within the watershed in the last 70 years. In examining the geochemical record, corroborating these trends is more complicated, perhaps due to the more complicated land use history of the area.

Settlement and anthropogenic perturbation in the basin, historically, is not as well-defined as that in the Little Kennebago Lake and Lower South Branch Pond basins. While there appears to be some discontinuity in the geochemical record (Figure 13) around 1700, this shift is not well defined. Furthermore, the error present in $\delta^{13}C$ and total carbon values during this period makes it difficult to discern the magnitude of change that occurs. Still, the agreement in shifts, particular of $\delta^{13}C$ and total nitrogen, shortly after 1700 could suggest this was the onset of detectable land use change as recorded by lake sediments. The initial drop in $\delta^{15}N$ at this time suggests that timber harvest may not have been the primary mechanism of change initially. The enrichment of $\delta^{13}C$ suggests that the primary production with the lake increased at this time (Brenner et al., 1999; Torres et al., 2012). The shift in C:N suggests lake sediments received a higher proportion of algae.

Similar to Little Kennebago Lake, there does appear to be increased relative variability in geochemical signals after the onset of anthropogenic influence at this time, though the absolute values remain fairly stable with some exceptions. The results of the two-sample t-test (Table 3) indicate that $\delta^{15}N$, $\delta^{13}C$, total C, and C:N all have statistically-significant differences in mean values pre- and post-Euro-American incursion. Based on the record from the sediment core (Figure 13), $\delta^{15}N$ experiences a minimum around 1772, then rises steadily until about 1950, after which it slowly declines to the present. This 1950 peak corresponds with the findings of Rich (2021) and the general shape of the trend is in agreement with McLauchlan et al. (2007); land clearance occurred throughout the late 18th, entire 19th, and early 20th centuries, until 1950 when a general trend of reforestation began.

In the early 19th century, the records show a decline in total nitrogen and total carbon, which could reflect changing land use patterns in response to the Industrial Revolution and an increase in terrigenous sediment, which serves to dilute both C and N in the signal. Shortly after these shifts, we see a slow decline of $\delta^{13}C$, which could indicate a reduction on the primary productivity of the lake, though relatively static signals in both $\delta^{15}N$ and C:N make it difficult to discern the exact cause.

Ultimately, the multiple mechanisms of land use change as well as the uncertainty in timing of Euro-American settlement and development make analysis of the geochemical record of Sennebec Pond difficult. While the geochemical indicators appear stable over long periods of time, the multiple inputs may be convoluting the signals and additional interpretation remains difficult without further analysis.

5.5 Geochemical Comparison

5.5.1 McLauchlan et al. (2007)

When the results of this study are compared to those of McLauchlan et al. (2007), there are some similarities in geochemical trends (Figure 14). Though the $\delta^{15}N$ values found in the records of the three study sites are generally higher than that of Mirror Lake, the focus of McLauchlan et al. (2007), shapes of the peaks in the 20th

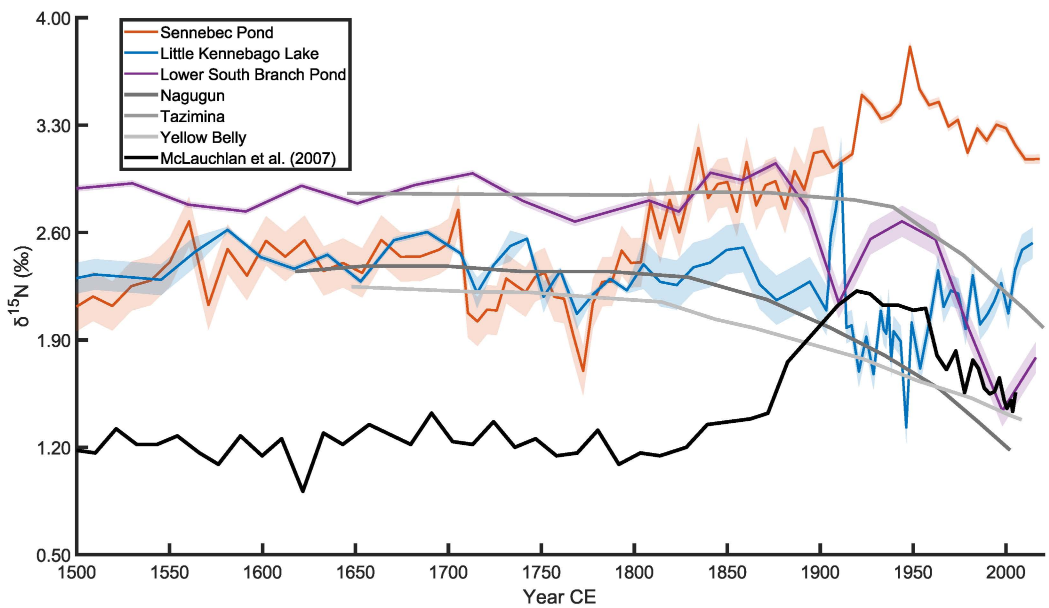

Figure 14- $\delta^{15}N$ for all three study lakes plotted with the record from similar lakes from Holtgrieve et al. (2011) and McLauchlan et al. (2007).

century do bear some resemblance, specifically that of Sennebec Pond, where the land-use timeline is most similar during that period. McLauchlan et al. (2007) ascribed the shape of this peak as the impact of, and subsequent recover from, timber harvest. If true, the similarity in signal of Sennebec Pond during its time of reforestation could support the decline of $\delta^{15}N$ as an indicator of recovery in an area affected by timber harvest, though the inclusion of the work of Holtgrieve et al. (2011), addressed below, does at some complication to this interpretation.

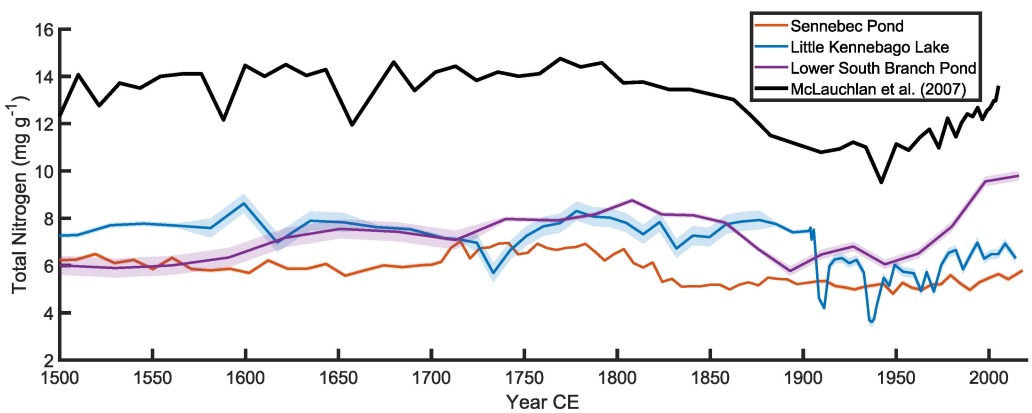

Figure 15- Total nitrogen for all three study lakes plotted with the record from McLauchlin et al. (2007).

There is, however, one caveat when comparing these results: the samples from Mirror Lake were dried in an oven at 65° C, which could have led to some fractionation (Bosley & Wainright, 1999; Kaehler & Pakhomov, 2001) and uniformly altered the $\delta^{15}N$. While this may have less impact on the general shape of the record, it could affect the absolute values, in which case the McLauchlin et al. (2007) curve could be in greater agreement with those of the study areas.

The records of total nitrogen (Figure 15) also have similar trends, though, again, the absolute values are notably different. The Mirror Lake record shows relative stability until the onset of timber harvest, after which it dips, experiencing a minimum around 1950, and rises in the most recent 70 years. This shape aligns well with the total nitrogen record of Lower South Branch Pond, indicating some possible similarities in nitrogen processes during this period. The records of total carbon (Figure 16) are less similar, showing little agreement in both value and trends. The

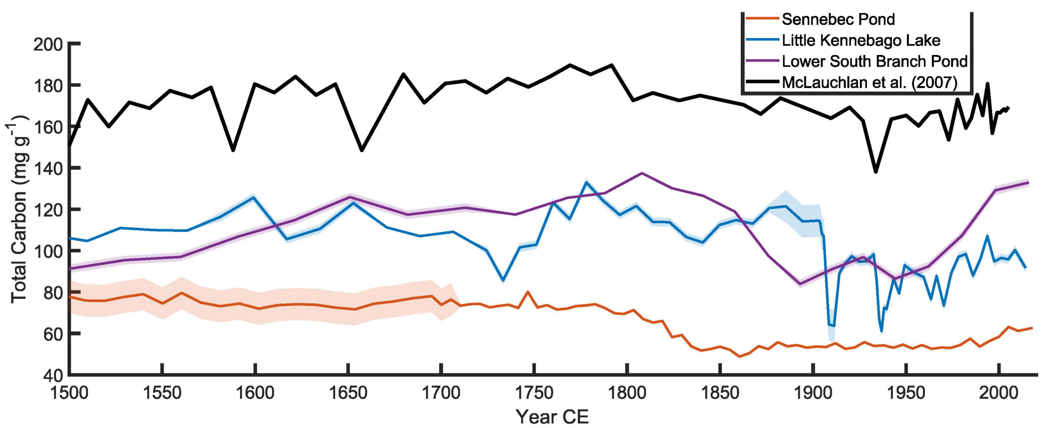

Figure 16- Total carbon for all three study lakes plotted with the record from McLauchlin et al. (2007).

Mirror Lake record, again, has elevated total carbon values when compared to the three study sites, and shows little variation throughout the record. It is most similar to the Sennebec Pond record, which also displayed very little variation other than a slight in the early 19th century, a signal which is present, albeit weakly, in the Mirror Lake record. The records of C:N (Figure 17) show some agreement in shape with Little

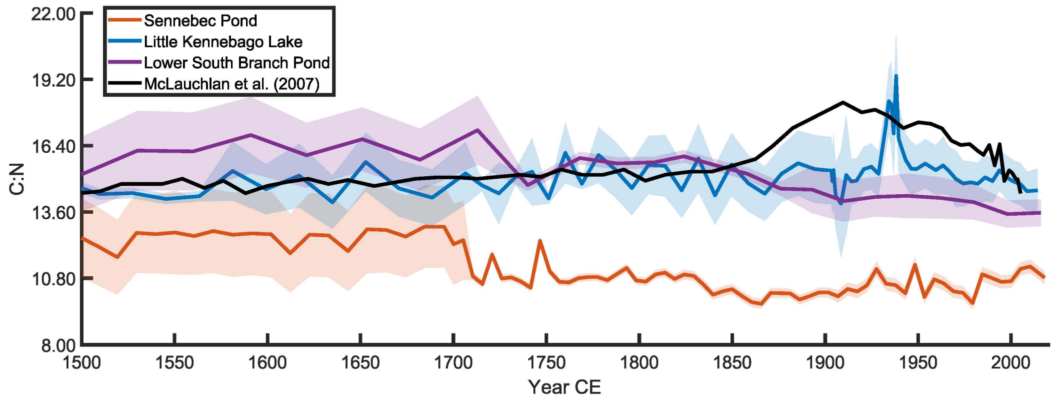

Figure 17- C:N for all three study lakes plotted with the record from McLauchlin et al. (2007).

Kennebago Lake before timber harvest began, but display little similarity after. The C:N values do show some agreement in value with both Sennebec Pond and Lower South Branch Pond in this same period, which could indicate similar sources of organic matter before changes in land use.

The $\delta^{13}C$ record of the three study sites and McLauchlan et al. (2007) (Figure 18) is perhaps the most interesting comparison. With the exception of similar absolute values to Sennebec Pond in the period before 1700, they show little similarity pre-1900. However, the all exhibit the same decline, both in rate and magnitude, after this

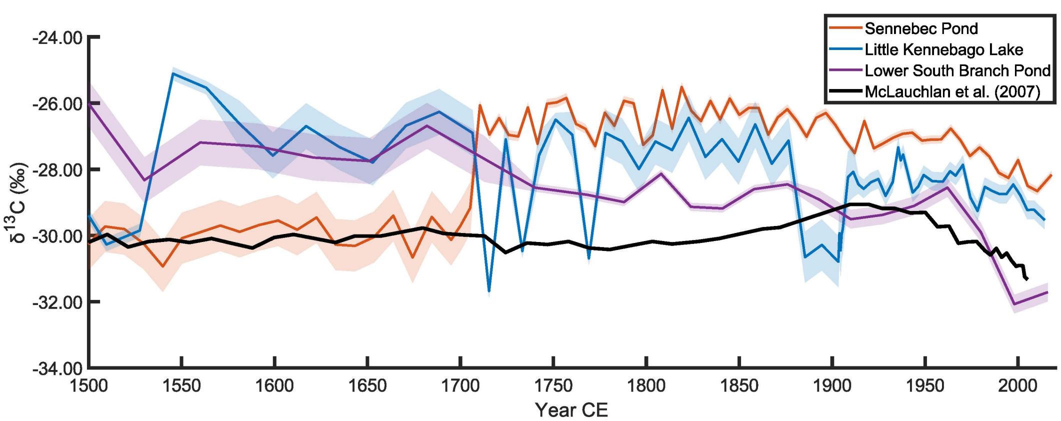

Figure 18- $\delta^{13}C$ for all three study lakes plotted with the record from McLauchlin et al. (2007).

time. Due to the varying histories of land use change during this period, it may be suggested that they were all subject to the same regional forcing that has, independent of local conditions, affected their $\delta^{13}C$ records in concert.

5.5.2 Holtgrieve et al. (2011)

The work of Holtgrieve et al. (2011) suggests a ubiquitous signal of anthropogenic emissions in $\delta^{15}N$ records of near-pristine, remote lakes in the northern hemisphere, characterized by a decline in $\delta^{15}N$ that begins in the 19th century, and accelerates in to the 20th century (Figure 14). While virtually every watershed in the northeast United States has been impacted in some way by humans, geographically, both Lower South Branch Pond and Little Kennebago Lake are a considerable distance from large population centers and could be considered remote by New England standards. Likewise, Lower South Branch Pond has been managed to minimize human impacts for over 70 years and may be as close to pristine as is possible in the northeastern United States.

To compare the signal found in Holtgrieve et al. (2011) to my study sites, three lakes from Holtgrieve et al. (2011) with comparable $\delta^{15}N$ values before the onset of anthropogenic signal were used: Nagugun and Tazima Lakes, both found in Alaska, and Yellow Belly Lake in Idaho (Figure 14). In those lakes, the magnitude in depletion in $\delta^{15}N$ in the last 70 years is similar to that found in Lower South Branch Pond and Sennebec Pond, which could suggest anthropogenic input of nitrogen as a contributor to the more recent $\delta^{15}N$ signal. The $\delta^{15}N$ record of Little Kennebago Lake displays little similarity to those of Holtgrieve et al. (2011). This is most likely due to the extent of land use change within the basin in the past century. A lack of evidence for a regional scale, anthropogenic, atmospheric input could suggest the local, watershed-scale processes are of sufficient magnitude as to dictate $\delta^{15}N$ values up to present-day.

Lower South Branch Pond, bears the most similarity, both historically and currently, to the sites examined in Holtgrieve et al. (2011). Timber harvest, the primary mechanism of land use change in the watershed, was short-lived. An enrichment of $\delta^{15}N$, the indicator of timber harvest found in McLauchlan et al. (2007), is not evident in the record. If the depletion event found around 1910 is attributed to the forest fire that occurred a few years before, suddenly the $\delta^{15}N$ record from Lower South Branch Pond bears a striking resemblance to those found in Holtgrieve et al. (2011). This is noteworthy in that it could suggest the magnitude of land use change in the basin was not substantial enough overpower anthropogenic atmospheric inputs.

Similarly, the recent decline in the $\delta^{15}N$ record from Mirror Lake, the focus of McLauchlan et al. (2007), also bears similarity to those found in Holtgrieve at el. (2007). While the depletion in $\delta^{15}N$ was originally attributed to forest recovery, it would be difficult to discern the strength of individual factors, including anthropogenic atmospheric input. A key difference between Mirror Lake and Lower South Branch Pond is that the original land use change in Mirror Lake was significant enough to alter the baseline $\delta^{15}N$ signal, in this case an enrichment, while Lower South Branch Pond saw virtually no enrichment during the period of timber harvest.

6.0 Conclusions

Sennebec Pond, Little Kennebago Lake, and Lower South Branch Pond all preserve a continuous record of the geochemistry within the lake basin across the past >500 years. Consistency across the records of all three study sites as well as similarities with other regional and global studies support my interpretation of lake sediments as accurate records of the geochemical responses to changes in land use with a basin. Thus, this study provides the following answers to the original research questions and goals.

(1) Contribute to the long-term record of nitrogen availability in New England.

Prior to this study, there were few records of nitrogen and carbon composition and content derived from New England lake sediments. This study has expanded that record to include data on nitrogen and carbon isotopes, as well as total nitrogen and carbon content, for the last 500+ years.

(2) Determine the geochemical signature of various changes in land use in northern New England.

Across the three study sites, there are three very different patterns of $\delta^{15}N$ that may be attributable to the different land use histories at each site. While previous studies, such as McLauchlan et al. (2007), were able to suggest distinct geochemical trends in response to a specific land use, the complexity and uniqueness of our study sites made it difficult to make any such interpretations. Rather, this study may suggest the ability of geochemical proxies such as $\delta^{15}N$ to elucidate more subtle changes in the style of land use change provided the main mechanism remains the same, such as timber harvest at Little Kennebago Lake.

(3) Compare variations in nitrogen levels and sources through time in lakes with a variety of histories.

Due to the proximity of the three study sites, as well as similarities in climate and regional policy, this study provided an appropriate comparison of the geochemical records of three lakes with differing land use histories. Through statistical analysis, it was determined that there are significant differences in the means of pre- and post-Euro-American settlement sample populations for most geochemical indicators of both Sennebec Pond and Little Kennebago Lake, as well as for $\delta^{15}N$ in Lower South Branch Pond.

(4) There is increased variability in geochemical indicators after the introduction of Euro-American settlers.

The geochemical records of Sennebec Pond and Little Kennebago Lake show a statistically-significant increase in variability after the onset of land use change (Table 3) precipitated by the arrival of Euro-American settlers. This is most evident in the record of Little Kennebago Lake, which has a more defined land use history, and displays a greater degree of perturbation in the record after the onset of timber harvest in the late 18th century. This supports the hypothesis. Lower South Branch Pond displayed an increase in variability in only $\delta^{15}N$ after Euro-American settlement, though the cutoff date used for Euro-American arrival occurs near the onset of the detectable anthropogenic nitrogen signal as recorded by Holtgrieve et al. (2011), meaning the change in variability cannot be solely attributed to changes in land use within the basin.

(5) The regional signal of anthropogenic nitrogen detected by Holtgrieve et al. (2011) is detectable in northern New England Lakes with more complex land-use histories.

The $\delta^{15}N$ record from Lower South Branch Pond shows a trend of depletion in $\delta^{15}N$ on a similar magnitude, and with similar timing, of the anthropogenic signal found in Holtgrieve et al. (2011). This is further supported by the lack of any signal of enrichment during the period of timber harvest, though the signal is interrupted by what has been interpreted to be the effects of a significant fire. This supports the hypothesis that the signal recorded in Lower South Branch Pond is that of a larger, regional signal of anthropogenic nitrogen remotely deposited in the lake.

(6) Subtle changes in land use management, specifically in the style of timber harvest, are detectible in the geochemical records in lake sediments.

The variability seen in the $\delta^{15}N$ record, as well as the C:N record, of Little Kennebago Lake during the period of sustained timber harvest (~1891 to present) suggests response to some change, especially when compared to the trend found in McLauchlan et al. (2007), which looked to identify the geochemical signature of timber harvest. These results suggest that changes in timber harvest technique and management have affected the geochemistry within the watershed. This supports the hypothesis, though additional study, including ancillary data and a finer sampling interval, would provide more insight.

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