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Asbestos mining waste impacts on the sedimentological evolution of the Bécancour chain of lakes, southern Quebec (Canada)

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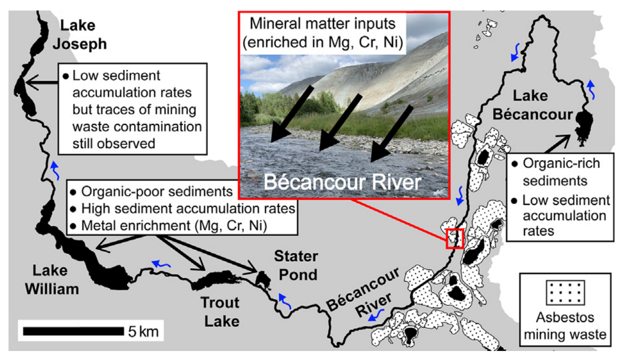
HIGHLIGHTS

- Impacts of asbestos mining wastes on aquatic ecosystems are poorly known.
- Paleolimnology shows that massive asbestos waste erosion occurs in Thetford Mines.
- Asbestos mining wastes are transported over at least 25 km by the Bécancour River.
- Asbestos mining wastes induce high sediment accumulation rates in lake ecosystems.
- Asbestos mining wastes cause heavy metal enrichment (Cr, Ni) of lake sediments.

GRAPHICAL ABSTRACT



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ABSTRACT

More than a century (1877–2011 CE) of asbestos mining activities in the Thetford Mines region have resulted in the accumulation of gigantic mineral waste piles on the banks of the Bécancour River (southern Quebec, Canada). This river widens downstream from the mining sites to form a chain of lakes, successively: Stater Pond, Trout Lake, Lake William and Lake Joseph. A previous paleolimnological investigation revealed that waste erosion and transport strongly modified and polluted Trout Lake. However, questions remain about the extent of the mining contamination within the Bécancour River Basin and its impacts at other spatial scales. Here, we aimed to address this lack of knowledge by analyzing the sedimentological evolution of Stater Pond and lakes William, Joseph and Bécancour (upstream reference site). Radiometric dating (^{210}Pb, ^{137}Cs, ^{14}C) and analyses of geochemical composition (ICP-AES/ICP-MS), computed tomography, magnetic susceptibility, loss-on-ignition and grain size were performed on sediment cores retrieved at these sites. In contrast to Lake Bécancour, yet similar to Trout Lake, we found that Stater Pond and Lake William have received high mineral matter loads since the creation of the Lake Asbestos Mine during the 1950s. Recent lake sediments at these downstream sites were highly enriched in magnesium, chromium and nickel. Comparison of their geochemical signature with that of sedimentary source materials from within the drainage basin demonstrated that they predominantly originate from mining waste erosion. Because of this issue, Stater Pond and lakes Trout and William are nowadays exposed to very high sediment accumulation rates (up to 1.4 cm yr^{-1}; 0.6 g cm^{-2} yr^{-1}) and heavy metal enrichment. Evidence for contamination was also found in Lake Joseph, indicating that wastes are transported and deposited over ≥25 km downstream from the mining sites. Our study highlights the high risks and dangers associated with asbestos pollutants in aquatic ecosystems.

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1. Introduction

Over recent decades, worldwide asbestos production has abruptly declined following increasing awareness of its negative impacts on human pulmonary health. The exploitation of this resource and its use are now banned in many countries. However, wastes resulting from decades of mining are still heaped on the ground in many regions of the planet. Their impacts on surrounding aquatic ecosystems are generally unknown and assumed to be minimal, yet only very few studies have been completed to verify this aspect. It is known that leachate waters originating from asbestos mining waste sites are alkaline (Meck et al., 2006; Wu, 2011) and can induce strong pH changes in streams draining across old mining sites (Masi and Bourget, 2007; GROBEC, 2015a; Donati-Daoust and Dubois, 2020). Asbestos contaminants from mining activities have also been shown to cause metal pollution and asbestos fiber enrichment in surface waters and sediments (e.g., Monaro et al., 1983; Meck et al., 2006; Koumantakis et al., 2009; Kumar and Maiti, 2015). Nevertheless, assessments of their impacts on aquatic ecosystem components and functioning over long time scales and at broad landscape levels remain scarce. In Canada, the flagrant lack of knowledge regarding this important environmental issue has recently been acknowledged by a public inquiry ('Bureau d'audiences publiques sur l'environnement', BAPE) mandated by the Government of Quebec to examine the situation of asbestos mining wastes within the province (BAPE, 2020).

For more than a century (1877–2011 CE), the Thetford Mines region in southern Quebec has been among the most important chrysotile asbestos production centers in the world. This has resulted in the accumulation of 489,000,000 tons of mine tailings (milling waste) and 539,000,000 tons of waste rock spread over more than 15 km^2 within the Bécancour River drainage basin (Beaudoin et al., 2008; GROBEC, 2015b). This river originates at the outflow of Lake Bécancour and flows downstream through the city of Thetford Mines while cutting across several gigantic mining waste piles located on its banks (Fig. 1). It then widens to form a chain of successive ponds and lakes (hereafter collectively referred to as lakes), namely from upstream to downstream: Stater Pond (' Étang Stater'), Trout Lake ('Lac à la Truite'), Lake William and Lake Joseph (at 25 km distance from the last mining site). These fluvial water bodies are of great importance for the region as they are surrounded by many residences and extensively used for water sports and leisure. They are also important biodiversity sites, notably Stater Pond which hosts vulnerable bird and turtle species. Unfortunately, poor water quality and strong siltation due to large sand deposits that enter these systems are posing threats to their survival in recent years, as documented by in situ/on-the-ground surveys and time series aerial photographs (Le regroupement des 4 lacs, 2015; Miquelon, 2018; Mercier, 2019).

In a previous paleolimnological investigation of a sediment core from Trout Lake (Jacques and Pienitz, 2021), we documented sediment accumulation rates that dramatically increased following the drainage of Black Lake ('Lac Noir'), a former upstream lake which was completely drained and excavated between 1955 and 1959 CE for the creation of the Lake Asbestos Mine. We provided evidence for mining waste erosion triggering the extremely high sediment inputs into Trout Lake since this event. Indeed, its modern sediments were organic-poor and highly enriched in metals associated with asbestos contaminants (i.e. magnesium, chromium and nickel). Questions remained about the travelling distance of mining wastes within the Bécancour River Basin (BRB) and their influence on metal pollution and lake sedimentation rates at other spatial scales. In the present study, our objective was to assess the impacts of past asbestos mining activities and waste erosion on the other three downstream components of the BRB chain of lakes: Stater Pond (1st downstream site), Lake William (3rd downstr. site) and Lake Joseph (4th downstr. site; Fig. 1). We present the results obtained from the examination of sediment cores retrieved from these aquatic systems and from Lake Bécancour (upstream headwater site), which we used as reference. We analyse their sedimentological evolution over the last decades/centuries and compare it to that of Trout Lake (2nd downstr. site) to provide a complete portrait of historical changes and modern conditions within the BRB basin. Our main hypothesis was that clear evidence of asbestos mining contamination and impacts would be found in all lakes located downstream from the mining sites, but not in Lake Bécancour.

The paleolimnological approach used in this study relies on the combined use of multiple physical and geochemical proxies. Radiometric dating, as well as analyses of geochemical composition, computed tomography, magnetic susceptibility, loss-on-ignition and grain size were performed on the sediment cores. In addition, we collected samples from various sources of sedimentary material in the upper part of the BRB to compare their geochemical signature with that of our sediment cores. Our work provides much needed knowledge of the environmental impacts of asbestos mining wastes to ensure the preservation of BRB lakes. It should also serve as an important reference study for other world regions with similar geographical and historical contexts.

2. Study site

Stater Pond and lakes Bécancour, Trout, William and Joseph are located in the upper part of the BRB at elevations between 190 and 390 m above sea level. Thetford Mines, which is comprised of several former mining villages/towns that have been merged in 2001 CE, was the most populated center of the region with 25,403 inhabitants in 2016 CE (Statistics Canada, 2019). Our fluvial study lakes have variable morphometric and limnological characteristics. Their surface area ranges between 0.4 and 4.9 km^2, and maximum depth between 2.5 and 30.1 m (Table 1). During fieldwork in 2017, water pH ranged between 7.9 and 8.9, specific conductivity between 73 and 337 μS cm^{-1}, and total phosphorus between 11 and 122 μg L^{-1} (mesotrophic to hypertrophic states). All lakes had low transparency waters (Secchi disk: 0.3–2.4 m) and hypolimnia were depleted in oxygen (O_2), especially at Stater Pond and Lake Joseph (1.8–4.6% O_2). Stater Pond and lakes Trout, William and Joseph receive municipal wastewater from a few discharge points in their catchment. In addition, agricultural lands are abundant in the region, yet nutrient enrichment may also originate from riverine residences.

Modern settlement in the region began around 1810 CE with the arrival of many American and European families near Trout Lake and Lake Joseph (Barry, 1999). Colonization intensified around 1830 CE and led to the foundation of what would later become 'Saint-Ferdinand' on the borders of Lake William in 1834 CE (Marcoux-Dubois and Fréchette-Laframboise, 1984). It was further accelerated following the discovery of asbestos deposits in Thetford Mines in 1876 CE with the initiation of mining activities the following year (Fortier, 1983). Several mines were rapidly created, mainly upstream from Black Lake (ca. 10 km from Stater Pond and Trout Lake). Regional asbestos production increased markedly after 1945 CE and peaked following the drainage of the lake. Between 1955 and 1959 CE, sediments extracted from Black Lake were disposed of in sedimentation basins and storage areas along the Bécancour River. Previously in 1954 CE, a dam and dike were built downstream from Stater Pond, resulting in a major increase in its surface area and its connection to the river. The purpose was to replace the hydrological role of Black Lake and also to trap sediments and mining wastes transported by the Bécancour River during the drainage and following years (Piette, 1953; Cloutier, 1965). Regional asbestos production dropped in 1980 CE and kept declining until all mining activities ceased in 2011 CE. In the past, a few chromite mines were also sporadically active on each side of Black Lake between 1894 and 1944 CE (Fortier, 1983; Gaudard, 1993).

Extraction of asbestos fibers was done without chemical reagents (Fortier, 1983; Villeneuve, 2013). The mines only discharged very low amounts of liquids into the environment, with the exception of waters

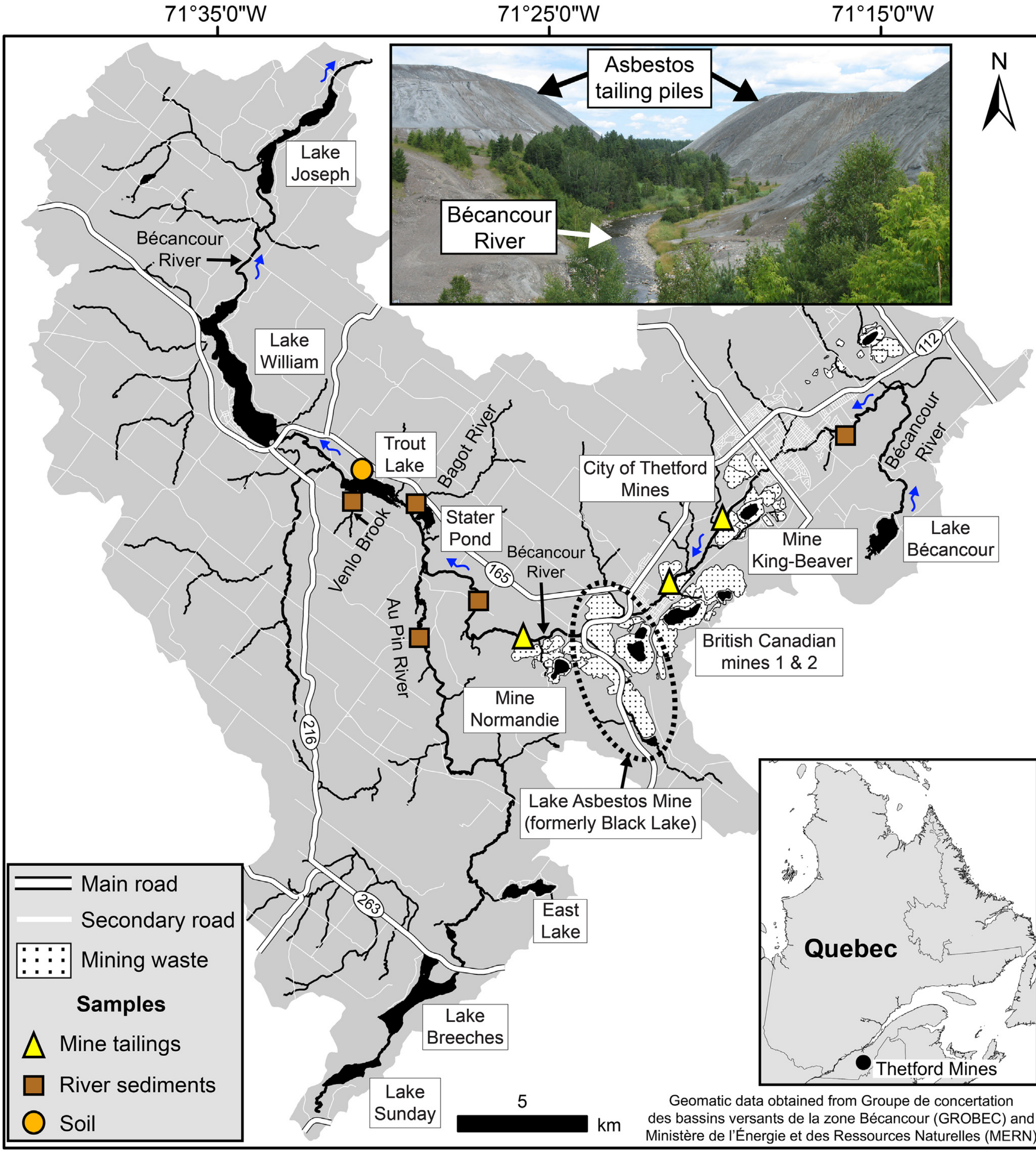

Fig. 1. Map showing the upper part of the Bécancour River drainage basin in southern Quebec, with the location of the samples collected from the sediment sources. The inserted photograph illustrates asbestos mining waste piles on the banks of the river in Thetford Mines (tailings from British Canadian mines 1 and 2).

that naturally accumulated at the bottom of pits and that were pumped out to maintain dry conditions (MDDEFP, 2013). However, they produced very high amounts of solid wastes. Mine tailings are composed of residues from physically processed (milled) mineral matter used to extract chrysotile fibers, whereas waste rock corresponds to bulk material excavated to reach the asbestos ores. Compared to tailing piles, waste rock piles have a more heterogenous grain size composition (meter-size blocks to sub-millimeter grains) and are often more flat and partially covered by vegetation. Therefore, they are likely more stable and assumed to be a less important source of sedimentary material to the Bécancour River.

The upper part of the BRB is located in the Appalachian Mountains and is composed of different geological groups (i.e. lithology is locally variable). It mainly includes peridotite and serpentinite rocks from the Thetford Mines Ophiolite Complex (from which asbestos was extracted), and different kinds of schists, phyllites, sandstones and quartzites (MERN, 2021). The bedrock is generally overlain by a thin layer of till and unconsolidated alluvial, fluvioglacial and glaciolacustrine

Table 1
General characteristics of the study lakes in 2017 CE.

	Lake Bécancour (upstr. site)	Stater Pond (1st downstr. site)	Trout Lake (2nd downstr. site)	Lake William (3rd downstr. site)	Lake Joseph (4th downstr. site)
Location	46°04′08″ N 71°14′40″ W	46°04′28″ N 71°28′26″ W	46°05′05″ N 71°30′16″ W	46°07′28″ N 71°34′22″ W	46°11′25″ N 71°33′24″W
Surface area (km^2)	0.97	0.36	1.24	4.90	2.53
Max depth (m)	3.4	3.9	2.5	30.1	12.0
Secchi (m)	0.9	0.3–0.5	0.8–1.4	1.8–2.4	1.4–1.8
pH	7.9	8.5–8.9	8.3–8.4	8.2–8.3	8.3
Cond (μS cm^{-1})	73–74	243–298	297–337	178–209	169–172
TP (μg L^{-1})	34	122	72	16	11
Hypolimnetic oxygen (%)[a]	N/A	4.6	40.6	<54.8	1.8

Note: Cond, specific conductivity; TP, total phosphorus; Secchi, Secchi disk transparency. Lake metrics are based on pre-existing bathymetric maps (Figs. S1–S4), whereas water chemistry and Secchi disk transparency data are based on 1–2 measurements acquired during fieldwork.
[a] Measurements at coring sites from the water-sediment interface, except for Lake William (14.5 m).

deposits of variable grain size (clay to boulders; Chauvin, 1979; Godbout, 2013). Late Wisconsinan deglaciation and the final retreat of the Bois-Francs Residual Ice Cap occurred around 10,900 yr BP in the upper reaches of the BRB (Parent and Occhietti, 1999).

Climate normals for the 1981–2010 CE period indicate mean annual temperatures of 4.4 and 4.6 °C, and total precipitation of 1228 (rain: 940 mm; snow: 289 cm) and 1310 mm (rain: 946 mm; snow: 364 cm) at Saint-Ferdinand and Thetford Mines meteorological stations, respectively (ECCC, 2019a). However, most recent data (2011–2017 CE), only available for the Thetford Mines station, indicate higher means for temperature (5.1 °C) and total precipitation (1337 mm; ECCC, 2019b). Our study lakes are usually covered by ice from late November to mid-April. Catchment vegetation in the upper part of the BRB is characterized by mixed deciduous and coniferous tree assemblages that mainly include sugar maple (*Acer saccharum*), yellow birch (*Betula alleghaniensis*) and various boreal species (e.g., *Abies* spp., *Larix* spp.; MRNFP, 2003).

3. Materials and methods

3.1. Core collection and sediment subsampling

Sediment cores (one per lake) 1.1–1.3 m in length were retrieved between June and August 2017 in the deepest central zones of Stater Pond (STA) and lakes Bécancour (BEC), William (WIL) and Joseph (JOS; Figs. S1–S4). An Aquatic Research percussion gravity corer equipped with 6.5 cm internal diameter plastic tubes was used. Water-sediment interfaces of the cores were stabilized using sodium polyacrylate (Zorbitrol) prior to transport to the laboratory (Tomkins et al., 2008).

The coring tubes were cut along their longitudinal axis using a Dremel saw and sediments were split in two halves with a fishing line. The surface of each half core was then cleaned and smoothed by running a spatula parallel to the sediment layers. One core half was transversely subsampled at 0.5 cm intervals, whereas the other was kept intact for nondestructive computed tomography (CT), magnetic susceptibility (MS) and X-ray microfluorescence (μ-XRF) analyses. The 0.5 cm subsamples were freeze-dried ca. 48 h before being used for the other analyses.

3.2. Sampling of sediment sources

Different types of sedimentary material (tailings, river sediments and soil) that accumulate/deposit in BRB lakes from various sources were sampled between July and September 2020. For logistical reasons, sampling efforts focused only on the catchments of Stater Pond and Trout Lake. Tailing samples were collected on the bottom slopes of King-Beaver, British Canadian 1 and Normandie mining waste piles where there was evidence of erosion towards the Bécancour River (Fig. 1). An additional tailing sample was taken on the river banks in front of Normandie Mine. In addition, sediments were sampled in the littoral zones of rivers Bécancour (upstream and downstream from the mining sites), Au Pin, Bagot and Venlo that feed either directly or indirectly into Trout Lake. The selection of river sampling sites was determined mainly by ease of access (e.g., near bridges) and presence of important sediment deposits. Finally, a soil sample was collected at the high-water mark on the slopes bordering Trout Lake.

All ten samples were placed into plastic bags for transport to the laboratory where they were transferred to aluminium trays and dried at room temperature for 1 week under a fume hood. The samples were then sieved through a 125 μm mesh to extract fine material suitable for metal analysis and comparable to our lake sediments in terms of particle size (>85% of core grains are below 125 μm).

3.3. Dating and chronology of sediment cores

To establish the chronology of the most recent portion of the sediment cores, the total lead 210 ($^{210}Pb_{tot}$), radium 226 (^{226}Ra) and cesium 137 (^{137}Cs) activities of a large series of dry bulk sediments (0.3–1.0 g) were measured by gamma spectrometry (Table S1). These analyses were performed by the Laboratoire de Radiochronologie of the Centre d'études nordiques (CEN) at Université Laval (Québec City, Canada) using an ORTEC's GWL series High-Purity Germanium (HPGe) Coaxial Well photon detector system equipped with a High-Performance, Low-Background lead Shield (HPLBS). Samples were previously stored for ≥3 weeks in sealed containers to allow radioactive equilibration. Corrections were made for the effect of self-absorption of low-energy gamma rays within the sediments. Detection limits for $^{210}Pb_{tot}$, ^{226}Ra and ^{137}Cs were respectively 0.015, 0.044 and 0.004 Bq g^{-1}, based on measurements from a blank sample that was subtracted from the final results. Unsupported lead 210 ($^{210}Pb_{uns}$) concentrations were calculated by subtracting the $^{210}Pb_{tot}$ activity from that of ^{226}Ra, which is equivalent to supported lead 210 activity ($^{210}Pb_{sup}$; Appleby and Oldfield, 1978). In sediment core BEC, ^{226}Ra concentrations were very low and even undetected in most samples (Table S1). We corrected its values by adding the $^{210}Pb_{tot}$ mean background concentration (corresponding to $^{210}Pb_{sup}$ activity).

Clean ^{137}Cs peaks associated with the 1963 CE global fallout maximum from nuclear weapons (Appleby, 2001) were noticed in sediment cores STA, WIL and JOS (Fig. S5). Unsupported ^{210}Pb activities below these peaks were very low and variable, hence could not be used to establish chronologies (Fig. S5). Ages for sediment layers younger than 1963 CE were established using Appleby's (2001) equations for composite (piecewise) models. The surface of the cores (t_1: 0 yr) and the ^{137}Cs peaks (t_2: 54 yrs) were used as chronological anchor points. For each core, the mean $^{210}Pb_{uns}$ flux during the period spanning/covering the 1963–2017 CE section was calculated using the following formula (Appleby, 2001):

$$P = \frac{\lambda \Delta A}{e^{-\lambda t_1} - e^{-\lambda t_2}} \quad (1)$$

where λ is the ^{210}Pb decay constant (0.03114 yr^{-1}), ΔA is the $^{210}Pb_{uns}$ inventory difference between stratigraphic levels x_1 and x_2 of known ages t_1 and t_2. Assuming a constant flux, the date associated with each analyzed level (x) of this section was calculated using the principles of the Constant Rate of Supply (CRS) model by resolving the following equation (Appleby, 2001):

$$\frac{P}{\lambda} e^{-\lambda t} = \frac{P}{\lambda} e^{-\lambda t_1} + \Delta A(x_1, x) \quad (2)$$

where ΔA (x_1, x) is the $^{210}Pb_{uns}$ inventory difference between stratigraphic levels x_1 and x of ages t_1 and t. The most recent part (<150 yrs) of sediment core BEC was dated in a standard way by using all $^{210}Pb_{uns}$ concentrations and applying the CRS model (Appleby, 2001). For this core, no ^{137}Cs peak could be used to provide chronological information or validation (Fig. S5).

Supplemental chronological points for all sediment cores were obtained from stable lead (Pb) concentrations, which were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS; see Section 3.8). Stable Pb content in North American lake sediments has significantly increased since 1850 CE as a result of pollution driven by global industrialization (e.g., Dunnington et al., 2020). Using the CRS model, Blais et al. (1995) identified that this rise originated near 1886 CE in southern Quebec lakes (standard deviation: 30 yrs; 95% confidence interval: ±15 yrs). Consistent with Blais et al. (1995), we attributed this date to the first stratigraphic level (from bottom to top) where the stable lead concentration exceeded twice the background concentrations in each sediment core (Fig. S6).

Variations in *Ambrosia* (ragweed) pollen concentrations were also analyzed in the sediment cores. Modern colonization and agriculture in North America have caused an important and rapid increase of this pollen type which can be used as a cultural and chronological marker in sediment archives (McAndrews, 1988). The pollen analyses were performed by the Laboratoire de paléoécologie at Université de Montréal (Montréal, Canada). For each core, a series of 0.2–1.0 g subsamples of dry bulk sediment of selected stratigraphic levels were mixed with a solution of exotic (*Eucalyptus*) pollen grains of known concentration and treated using standard techniques described by Faegri and Iversen (1989). Microscope slides were prepared and *Ambrosia* pollen grains in the samples were enumerated at 400× magnification along random transects until at least 50 exotic pollen grains were recorded. Consistent with Jacques and Pienitz, 2021, we estimated that the first points of *Ambrosia* pollen increase above (>2×) mean background concentration values in sediment cores STA, WIL and JOS corresponded to the year 1825 CE ± 15 yrs (Fig. S7), as modern settlement within these lake's catchments (and within a radius of ca. 50 km) mainly occurred between 1810 and 1840 CE (Marcoux-Dubois and Fréchette-Laframboise, 1984; Barry, 1999). The *Ambrosia* rise in sediment core BEC was not used as chronological marker because of greater uncertainty regarding its timing.

To determine the chronology of older core sediments, a large series of dry bulk sediments (0.3–1.0 g), plant debris and wood fragments from various stratigraphic levels were submitted to CEN's Laboratoire de Radiochronologie for carbon 14 (^{14}C) dating (Table S2). When quantity was sufficient, the samples were first pretreated using hydrochloric acid (HCl) and sodium hydroxide (NaOH). They were then transformed to carbon dioxide (CO_2) before being transferred to the Keck Carbon Cycle AMS Facility at the University of California (Irvine, U.S.) to be dated by accelerator mass spectrometry (AMS). The radiocarbon ages were corrected for total isotopic fractionation.

All chronological points identified from ^{210}Pb, ^{137}Cs and ^{14}C dating, and the stable Pb and *Ambrosia* rises were used to construct Bayesian age-depth models using the R package Bacon 2.5.6 (Blaauw and Christen, 2011; Blaauw et al., 2021). Radiocarbon ages were calibrated according to the IntCal20 dataset (Reimer et al., 2020). For cores WIL and JOS, dates derived from bulk sediment material were not considered in the models as they were older than those obtained from vegetal debris and wood fragments, which usually provide more reliable ages (Björck and Wohlfarth, 2001; Table S2). Furthermore, dates from stratigraphic levels above the *Ambrosia* and stable Pb rises, and older than those provided by these chronological markers, were discarded as they were not considered indicative of the true deposition ages. An error of ±1 yr was attributed to ages at the core surfaces (2017 CE) and the ^{137}Cs peaks (1963 CE). Error distributions with long tails were assigned to ^{14}C dates by using the default parameters of the student-t distribution (t.a = 3, t.b = 4). Error distributions with short tails were used for non-radiocarbon dates by setting t.a = 33 and t.b = 34. Age-depth models were built from 1 cm thick sections. Boundaries resetting the memory of the models were set at certain levels where important and abrupt stratigraphic changes occurred (STA: 93 cm; WIL: 40 cm, 72 cm; JOS: 10 cm). Prior settings for sedimentation times (yr cm^{-1}) were based on the examination of preliminary chronological data (for acc.mean) and on values calculated by Goring et al. (2012; for acc. shape, with minimum values fixed at 1.1). The memory priors were set to allow highly variable sedimentation times based on the parameters used by Blaauw and Christen (2011; mem.strength = 20, mem. mean = 0.1).

Weighted mean ages were extracted from the models and used for the presentation of results and discussion. Historical sediment linear accumulation rates (LAR; cm yr^{-1}) were determined from the sedimentation times calculated *a posteriori* by Bacon for each 1 cm section. Sediment mass accumulation rates (MAR; g cm^{-2} yr^{-1}) were obtained by dividing the dry mass (g cm^{-2}) of each 1 cm section by the corresponding number of years.

3.4. Computed tomography

Computed tomography (CT) scans of the sediment cores were acquired using a Siemens SOMATOM Definition AS device at Centre Eau-Terre-Environnement of Institut national de la recherche scientifique (INRS-ETE). CT scans illustrate changes in sediment density and allow a better visualization of certain sedimentary structures (Fortin et al., 2013). Lighter and darker grey tones respectively represent higher and lower densities. Tones expressed on a grayscale indicative of light intensity (black: lower; white: higher) were extracted along a central longitudinal line (resolution: 0.6 mm). Data treatment was done with the Fiji distribution of the ImageJ software (Schindelin et al., 2012).

3.5. Magnetic susceptibility

Variations in magnetic susceptibility (MS) at the surface of the intact half sediment cores were measured at room temperature with a Bartington MS3 device equipped with a hand-held MS2E probe. Data were acquired at 1 cm intervals with Bartsoft software version 4.2.1.1. For each depth, 4 measurements were performed and a mean value was calculated.

3.6. Loss-on-ignition

Loss-on-ignition (LOI) analyses were performed on 0.2–0.3 g subsamples of dry sediments from all 0.5 cm core slices. They were placed into a laboratory oven for 24 h at 105 °C to eliminate residual humidity, and then weighed. Following recommendations outlined by Heiri et al. (2001), they were afterwards transferred into a muffle furnace at 550 °C for 4 h and the organic matter (OM) content of the sediments (lost due to combustion) was estimated by weighing the samples again.

3.7. Grain size

Loss-on-ignition residues were retrieved at 1 cm intervals and analyzed with a Horiba laser diffraction particle size analyzer at the Laboratoire de géomorphologie et de sédimentologie of Université Laval. Sodium hexametaphosphate (5.5%) was added to the samples at least 24 h before the analyses to help deflocculate the mineral matter. An ultrasound treatment (time up to 200 s) was also applied to several samples with aggregated grains. The mean grain size (MGS) of each sample was calculated according to the Folk and Ward method (Blott and Pye, 2001) using GRADISTAT version 8.0 (Blott, 2010).

3.8. Geochemistry

ICP-AES and ICP-MS analyses were performed to examine the metal content of the sediment cores and samples collected from sediment sources within the BRB. Subsamples of 0.3–1.2 g of dry material were submitted to INRS-ETE laboratories where they were analyzed using Agilent 5110 SVDV and Thermo Scientific X Series II spectrometers. Subsamples of the sediment cores were generally taken at 4 cm intervals. However, stable Pb concentrations were analyzed in a few supplemental selected levels. A total digestion method involving nitric acid (HNO_3), perchloric acid ($HClO_4$), hydrofluoric acid (HF) and HCl was used to fully solubilize the samples. Concentration data were obtained for the following chemical elements: aluminium (Al), arsenic (As), barium (Ba), calcium (Ca), cadmium (Ca), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), potassium (K), lithium (Li), magnesium (Mg), manganese (Mn), sodium (Na), nickel (Ni), phosphorus (P), lead (Pb), sulfur (S), scandium (Sc), strontium (Sr), titanium (Ti) and zinc (Zn). The accuracy and precision of the analyses were ensured by several control procedures, including laboratory tests on duplicates and certified reference materials #8704 (Buffalo River Sediment) from the National Institute of Standards and Technology (NIST; U.S. department of commerce) and LKSD-2 and LKSD-4 (lake sediments) from Natural Resources Canada (NRCan). Measured concentrations for the standards (4 sets for each of them; $n = 12$) all differed by <15% from the expected values.

For each sediment core, complementary high-resolution semi-quantitative data for a wide range of chemical elements were also acquired by μ-XRF. Although variations in elemental concentrations obtained through this method generally followed the same trends as those provided by ICP-AES and ICP-MS analyses, μ-XRF results appeared to be influenced by changes in sediment physical characteristics despite normalization. Therefore, they were considered less reliable and are only presented here as supplementary material.

To evaluate the increased or decreased levels of elements analyzed by ICP-AES and ICP-MS in recent lake sediments (1750–2017 CE), enrichment factor (EF) values were calculated from the following equation (Chassiot et al., 2019):

$$EF = \frac{(x/Ti)sample}{(x/Ti)reference} \tag{3}$$

where x and Ti are respectively the concentrations of a given element and of Ti at a given stratigraphic level (sample) and in precolonial (reference) sediments of each core. Expressing concentrations as a ratio to Ti allows to account for changes in sediment organic matter content and grain size (Chassiot et al., 2019). Reference ratios were obtained by averaging values from the entire sequences of precolonial sediments, except for sediment core BEC where only ratios from 12 to 24 cm were averaged as its precolonial sediments covered a much longer time sequence and captured more important chemical changes. For comparison purposes, EF values were also calculated for Trout Lake's sediment core (TRU) using data from Jacques and Pienitz, 2021. An EF value < 1 indicates an impoverishment; an EF value of 1 no enrichment; and an EF value > 1 an enrichment.

Finally, a principal component analysis (PCA) was performed to visualize the evolution of the geochemical signature of lake sediments through time and to compare it with samples collected from sediment sources within the BRB. Computation was done with Canoco version 5.0 (ter Braak and Šmilauer, 2012). Only data obtained by ICP-AES and ICP-MS analyses were considered in the PCA. Elements As, Fe, Mn, P and S were excluded because their concentrations in lake sediments are often strongly influenced by local redox conditions (Engstrom and Wright, 1984; Boyle, 2001). Similarly, Pb concentrations were omitted because they are highly dependent on atmospheric depositions. Concentration data for Ba, Cd, Co, Cr, K, Li, Mg, Na, Ni, Sr and Ti were log-transformed for analyses following recommendations by Canoco Adviser. All variables were also centered and standardized. Trout Lake's sediment core from Jacques and Pienitz, 2021 was also included in the PCA.

4. Results

4.1. Chronology

Age modelling revealed that sediment core BEC (length: 113 cm) covered the last 9300 yrs of regional history, with only the upper 12.25 cm being deposited since the onset of modern settlement (ca. 1810 CE; Fig. 2). In contrast, sediment core STA (113.5 cm) captured the last 1210 yrs, with 97.75 cm of sediments deposited since 1810 CE. Basal layers of WIL (126.5 cm) and JOS (105 cm) sediment cores were dated to 510 and −320 CE, while the onset of modern settlement corresponded to levels 72.75 cm and 21.25 cm, respectively. Maximum error associated to the weighted mean ages extracted from the models ranged between 2 and 26 yrs for 1940–2017 CE, 13 and 56 yrs for 1800–1940 CE, and 31 and 617 yrs for <1800 CE.

4.2. Sediment accumulation rates

All study lakes had low sediment accumulation rates prior to 1785 CE (<0.07 cm yr^{-1} and 0.02 g cm^{-2} yr^{-1}; Fig. 3). LAR and MAR slightly increased in all study lakes between 1785 and 1860 CE, coinciding with the modern settlement period (ca. 1810–1880 CE). The most important increases occurred in Lake William, where rates stabilized around 0.26 cm yr^{-1} and 0.12 g cm^{-2} yr^{-1} during this epoch. LAR and MAR then remained fairly stable until they increased again between 1905 and 1925 CE in Lake William, reaching mean values of 0.33 cm yr^{-1} and 0.14 g cm^{-2} yr^{-1} between 1925 and 1935 CE. Sediment accumulation rates surged in all downstream sites after 1935 CE and peaked between 1940 and 1965 CE near the beginning of the intensive mining period (1945–1980 CE), following similar trends previously observed for Trout Lake (Jacques and Pienitz, 2021). A maximum MAR of 1.11 g cm^{-2} yr^{-1} was reached in Stater Pond in 1958 CE. The accumulation rates in all downstream lakes then remained high but generally followed decreasing trends from 1965 to 1990 CE. MAR displayed small peaks near 1970 CE and were especially lower after 1975 CE. Between 1990 and 2017 CE, during the mining decline period (ca. 1980–2017 CE), LAR (1.3–1.4 cm yr^{-1}) did not vary much at Stater Pond but MAR (0.56–0.85 g cm^{-2} yr^{-1}) continued to decrease while remaining much higher than at any other sites (including Trout Lake). Meanwhile, rates in Lake William (means: 0.50 cm yr^{-1}, 0.22 g cm^{-2} yr^{-1}) and Lake Joseph (means: 0.10 cm yr^{-1}, 0.05 g cm^{-2} yr^{-1}) were lower than during the previous period and fairly stable. At Lake Bécancour, LAR (0.05–0.14 cm yr^{-1}) and MAR (0.01–0.03 g cm^{-2} yr^{-1}) progressively increased between 1940 and 2017 CE but remained low.

4.3. General stratigraphy and physical characteristics

Sediment core BEC was composed of dark brown watery and organic-rich sediments (Fig. 4). Only the upper 4 cm were more lightly

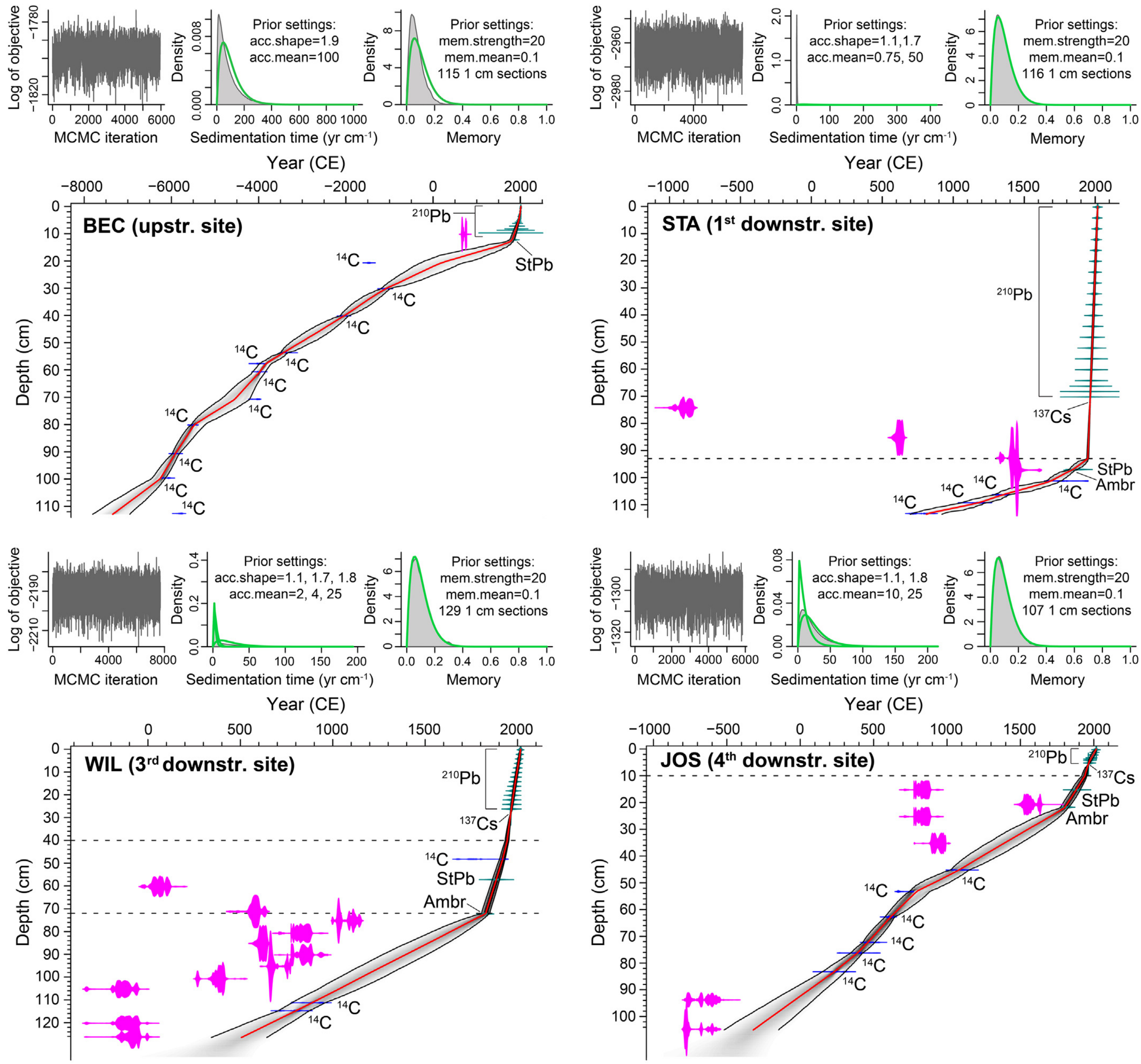

Fig. 2. Age-depth models established using Bacon for sediment cores retrieved in Lake Bécancour (BEC), Stater Pond (STA), Lake William (WIL) and Lake Joseph (JOS). Note: the red lines correspond to weighted mean ages, whereas the black lines delimit 95% confidence intervals. Darker shaded areas represent higher age probabilities. The horizontal dashed lines mark the boundaries set for each model. Calibrated ^{14}C dates included and discarded from the models are respectively represented by the blue and purple markers, whereas the green markers are non-radiocarbon dates (^{210}Pb, ^{137}Cs, StPb [stable Pb rise], Ambr [*Ambrosia* rise]). For each model, the upper left panel depicts the Markov Chain Monte Carlo (MCMC) iterations performed to estimate the sedimentation times (upper middle panel) and their memory (i.e. variability; upper right panel). Prior and posterior distributions for the sedimentation times and memories are respectively represented by the green curves and grey histograms.

coloured and visibly distinct. Sediment density was negatively correlated with OM values throughout the entire stratigraphic sequence. In precolonial core sediments, OM content followed a decreasing trend from 113 to 104.25 cm and reached a low plateau (28.0–29.0%) between 104.25 and 97.75 cm (−6610 to −6170 CE; 8560–8120 cal yr BP). It increased again and attained maximum levels (38.2–43.2%) between 88.75 and 84.25 cm (−5830 to −5640 CE; 7780–7590 cal yr BP). Values then decreased abruptly but remained high (34.8–40.6%), forming a mostly flat plateau from 83.75 to 54.75 cm (−5620 to −3530 CE; 7570–5480 cal yr BP). OM content declined and remained lower in the rest of precolonial sediments, especially between 53 and 27 cm (−3350 to −650 CE; 5300–2600 cal yr BP) where values of 28.7–33.1% were measured. MGS was highly variable throughout the whole stratigraphic sequence, but values were generally lower (17.4–22.2 μm) from 113 to 97.75 cm (−7350 to −6170 CE; 9300–8120 cal yr BP) and slightly higher (19.0–25.5 μm) from 88.75 to 54.75 cm (−5830 to −3530 CE; 7780–5480 cal yr BP). In most recent sediments, a marked increase in sediment magnetic susceptibility accompanied by a growing density and a decreasing OM content (23.3–27.2%) were observed from 6 to 0 cm core depth (1951–2017 CE).

Sediment core STA displayed major stratigraphic changes (Fig. 4). Levels 113.5 to 97 cm (810–1844 CE) consisted of organic-rich sediments. Important trends towards lower sediment density, higher OM content (29.8–51.6%) and coarser grains (19.1–24.3 μm) were observed

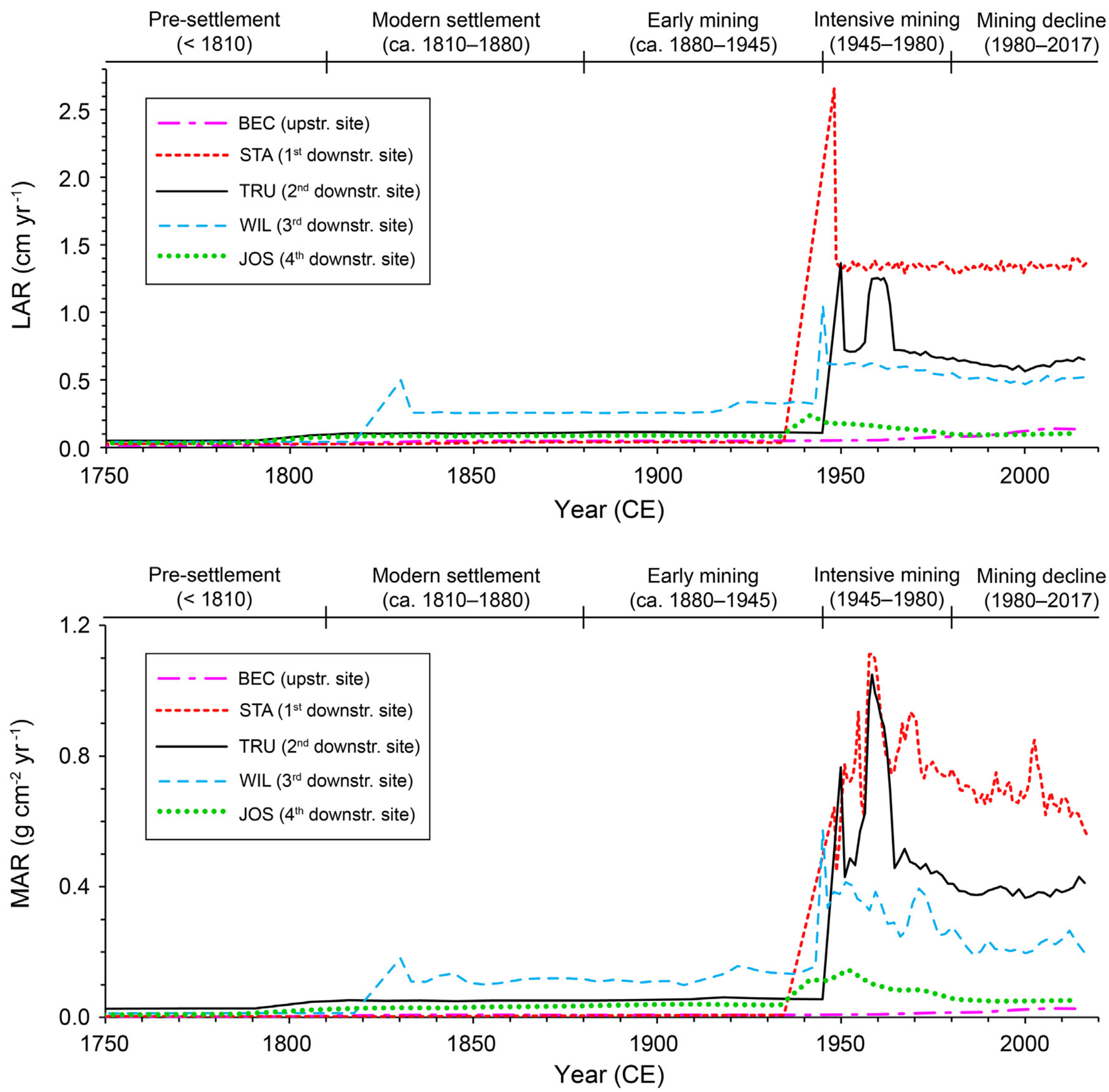

Fig. 3. Temporal evolution (1750–2017 CE) of linear accumulation rates (LAR; upper panel) and mass accumulation rates (MAR; lower panel) of sediments at the coring sites of Lake Bécancour (BEC), Stater Pond (STA), Trout Lake (TRU; Jacques and Pienitz, 2021), Lake William (WIL) and Lake Joseph (JOS).

between 108.25 and 97 cm (1240–1844 CE). From 97 to 85.5 cm (1844–1953 CE), decreases in MGS (15.6–22.5 μm) and OM (8.8–48.7%) values occurred. These changes were accompanied by pronounced increases in density and magnetic susceptibility above 93 cm (1948 CE), roughly corresponding with the intensification of mining activities in Thetford Mines (1945 CE). From 85.5 to 75 cm (1953–1961 CE), sediments comprised several laminae and distinct layers, with variable MGS (6.6–16.5 μm; up to 18% of clay) and OM values (4.1–14.7%). Sediments from 75 to 0 cm (1961–2017 CE) were fairly uniform, with a light brown colour (greyish at daylight). The density and magnetic susceptibility were high, whereas the OM content was low (5.5–8.8%). MGS increased (12.4–18.0 μm) from 75 to 56 cm (1961–1975 CE) and stabilized around 17.2 μm from 56 to 0 cm (1975–2017 CE).

Sediment core WIL also displayed a dynamic stratigraphy (Fig. 4). From 126.5 to 72.25 cm (510–1820 CE), sediments had a dark brown colour with an OM content of 15.0–20.9% (mean: 18.7%) and MGS of 13.7–19.5 μm (mean: 15.9 μm). Within this interval, notable variations in OM content included a growing trend from 122.25 to 109.75 cm (610–920 CE), a return to lower values from 99.25 to 80.75 cm (1170–1620 CE), and a renewed increase from 80.75 to 72.25 cm (1620–1823 CE) coinciding with a peak in MGS values. Sediment density and magnetic susceptibility both followed inverse trends at these depths. From 72.25 to 40.25 cm (1823–1944 CE), modern settlement and land development corresponded with abrupt increases in density and magnetic susceptibility, as well as lower average OM content (12.5%). MGS was also generally lower (13.9 μm) between 1820 and 1925 CE, but peaked (13.1–19.2 μm) between 1925 and 1940 CE. Peaks in density and magnetic susceptibility, and low OM content (5.7–11.3%) from 40.25 to 18.75 cm (1944–1980 CE) coincided with the period of intensive mining activities. Within this depth interval, a greyish sediment layer with fine grains (7.4–12.8 μm; up to 15% of clay) was present from 40.25 to 31.5 cm (1944–1958 CE). From 18.75 to 0 cm (1980–2017 CE), average magnetic susceptibility and OM content (9.6%) remained respectively higher and lower than pre-1944 CE means, whereas the density was similar. MGS oscillated between 12.1 and 20.4 μm.

Sediment core JOS was more homogenous than cores STA and WIL (Fig. 4). In precolonial sediments, density and MGS (17.4–20.8 μm) reached highest values between 105 and 90.25 cm (−320–50 CE), whereas OM content was concurrently at its lowest (11.7–13.8%). OM values gradually rose (11.7–17.6%) from 90.25 to 52.75 cm (50–810 CE), were slightly lower (15.0–16.5%) from 46.75 to 31.25 cm (1030–1510 CE) and increased again (15.4–17.7%) between 31.25 and

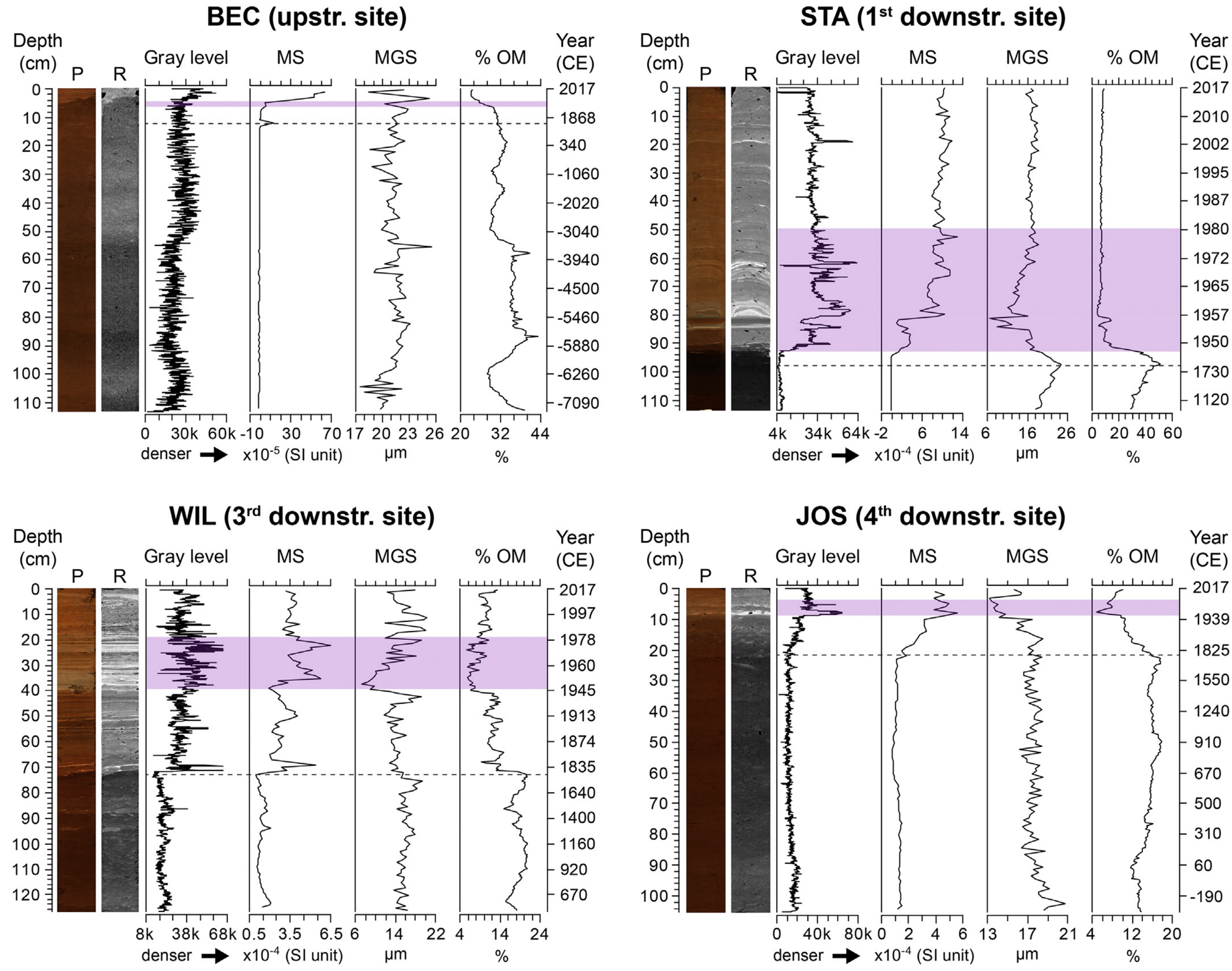

Fig. 4. Photographic (P) and radiographic (R) images of the sediment cores collected in Lake Bécancour (BEC), Stater Pond (STA), Lake William (WIL) and Lake Joseph (JOS), with stratigraphic profiles of their physical characteristics. Note: the horizontal dashed line marks the onset of modern settlement within the Bécancour River basin (ca. 1810 CE), whereas the purple shaded area represents the period of intensive mining activities (1945–1980 CE) in the Thetford Mines region. MS, magnetic susceptibility; MGS, mean grain size; % OM, organic matter percentage.

21.75 cm (1510–1800 CE). Lower magnetic susceptibility values were also measured from 62 to 21.75 cm (630–1800 CE). From 21.75 to 8.75 cm (1800–1945 CE), modern settlement and land development in the catchment coincided with gradual increases in sediment density and magnetic susceptibility, as well as decreasing MGS (14.2–18.6 μm) and OM (9.7–16.0%) values. From 8.75 to 3.5 cm (1945–1981 CE), the period of intensive mining activities then corresponded with peaks in sediment density and magnetic susceptibility, and low MGS (13.4–14.8 μm) and OM values (4.9–9.7%). From 3.5 to 0 cm (1981–2017 CE), density and magnetic susceptibility remained high, whereas MGS (13.2–16.4 μm) and OM content (8.4–10.3%) increased.

4.4. Element concentrations and enrichment factors

In precolonial core sediments, chemical element concentrations varied in accordance with changes in OM content. In Lake Bécancour, sediments deposited between −7350 and −3530 CE (9300–5480 cal yr BP) generally showed higher concentrations of As, Ca, Cd, Cu, Ni, S and Sr, although their abundance was significantly lower within the −6610 to −6170 CE interval (8560–8120 cal yr BP; Fig. S8). These elements displayed lower concentrations between −3530 and −750 CE (5480–2700 cal yr BP), whereas Al, Ba, Co, Cr, K, Li, Mg, Na, Ti and Zn were more abundant. Concentrations of most elements dropped between −750 and 1810 CE (2700–140 cal yr BP). In other lakes, sediment geochemical changes preceding modern settlement were less important (Figs. S9–S11). The absolute concentration of most elements progressively increased within the anthropogenic horizon of the sediments in all study sites.

Most elements displayed EF values near or below 1 in colonial core sediments of lakes Bécancour, William and Joseph, whereas values were generally a little higher (1.0–1.8; 3.4 for Pb) at Stater Pond (Fig. 5). Enrichment levels of most elements then significantly increased in sediments deposited during the early mining period (ca. 1880–1945 CE) in lakes Bécancour and William. Manganese (13.0), As (2.5) and S (2.4) enrichment levels reached maximum values in the sediments from this epoch within core WIL. In all downstream sites, EF values of Mg, Cr, Ni, K, Al, Ba, Li and Sc formed pronounced peaks between 1940 and 1960 CE, at the transition between the early and intensive mining periods. Fluctuations were, however, less important in Lake Joseph. Between 1960 and 1975 CE, EF values for Mg, Cr and Ni followed new increasing trends in cores STA and WIL. Within these cores, Mg and

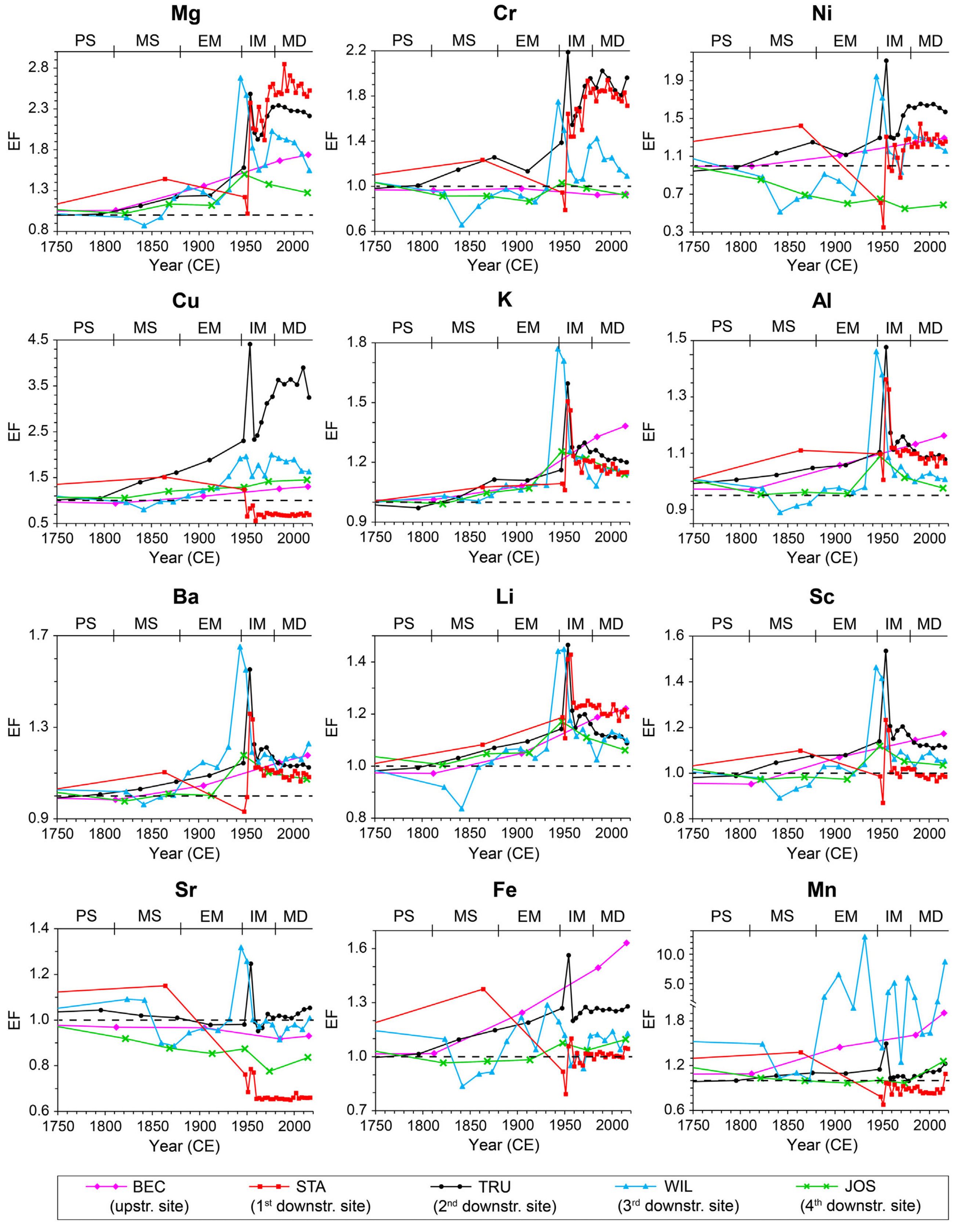

Fig. 5. Enrichment factors (EF) of chemical elements analyzed by ICP-MS and ICP-AES as a function of the year of deposition (1750–2017 CE) in sediment cores from Lake Bécancour (BEC), Stater Pond (STA), Trout Lake (TRU; Jacques and Pienitz, 2021), Lake William (WIL) and Lake Joseph (JOS). Note: stratigraphic levels below or above the horizontal dashed line (EF = 1) are respectively impoverished or enriched in the analyzed elements. PS, pre-settlement period; MS, modern settlement period; EM, early mining period; IM, intensive mining period, MD, mining decline period.

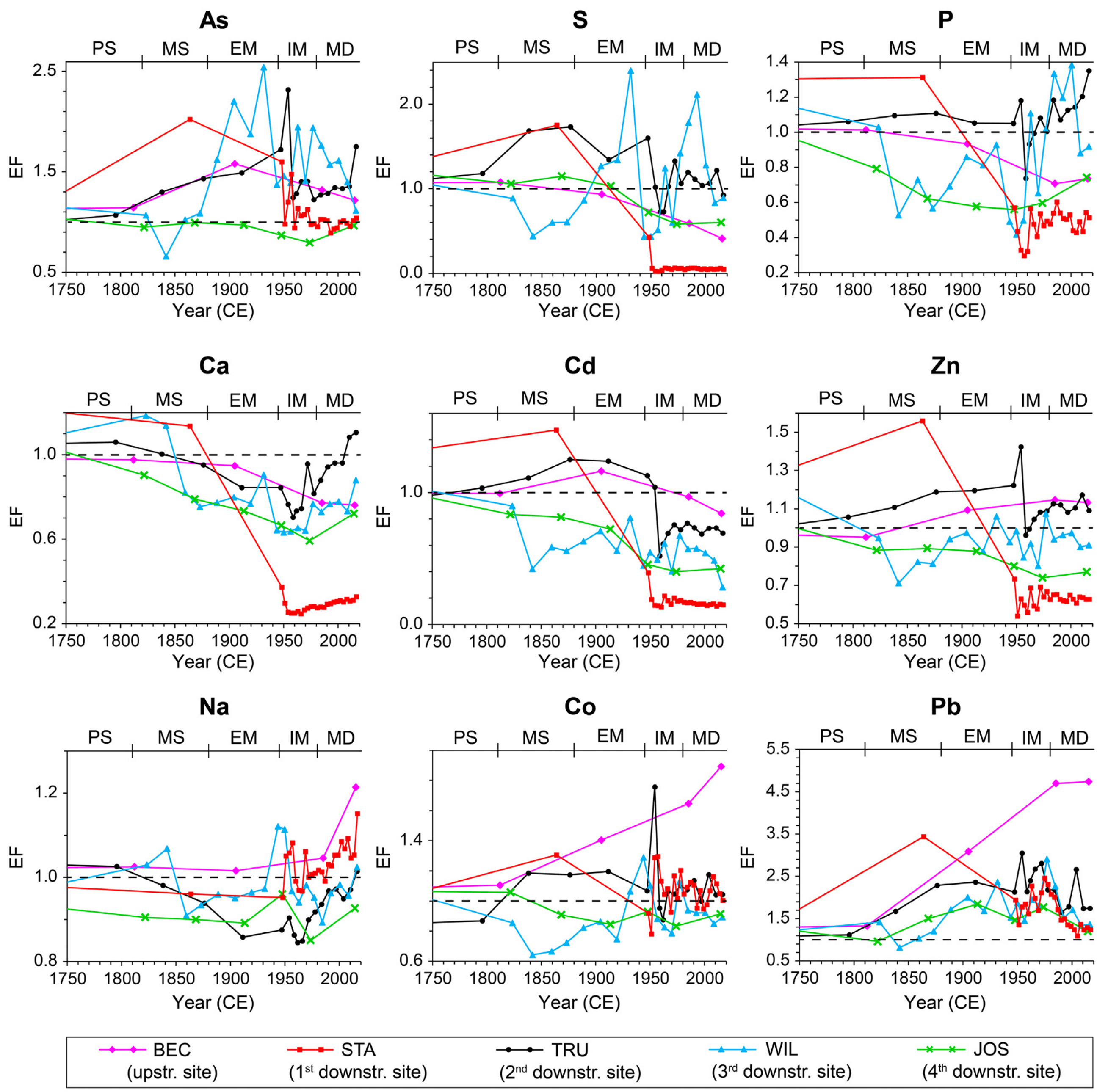

Fig. 5 (*continued*).

Cr enrichment levels remained high between 1975 and 2017 CE, although decreasing trends were observed in WIL. For this period, the highest mean EF values for Mg and Cr were found in core STA (respectively, 2.6 and 1.8), followed by cores WIL (1.8 and 1.3), BEC (1.7 and 0.9) and JOS (1.3 and 0.9). Mean EF values were also high for Cu in core WIL (1.8). In comparison, EF values of 2.3, 1.9 and 3.5 were measured respectively for Mg, Cr and Cu in Trout Lake's sediments deposited between 1975 and 2017 CE. Meanwhile, mean EF values for K, Al, Ba, Li, Sc and Fe in all downstream lakes only varied between 1.0 and 1.3. Relatively high mean EF values were noted for Pb (4.7), Co (1.8), Mn (1.8), Fe (1.6) and K (1.4) in sediments deposited since 1980 CE at Lake Bécancour. All lake sediments deposited since modern settlement were impoverished or only weakly enriched in Sr, P, Ca, Cd, Zn and Na, although notable EF increases during the mining decline period were observed for P in sediment core WIL and for Na in sediment cores BEC and STA.

4.5. Geochemical signature

Clear geochemical distinctions were possible between the different kinds of sedimentary materials collected in this study. Lake and river sediments, along with the soil sample taken at Trout Lake, were associated with higher K, Ba, Ti, Li, Al, Sr, Na and Zn concentrations towards the left end of PCA axis 1 (Table S3; Fig. 6). This was especially the case for sediment cores TRU, WIL and JOS, and for the upper organic-poor section (0–92 cm) of STA, as their sample scores grouped in the part furthest to the left of the PCA plot. Sediments from rivers Bagot, Au Pin and Venlo were also associated with higher Ca concentrations near the bottom end of PCA axis 2. In contrast, K, Ba, Ti, Li, Al, Sr, Na, Zn and Ca were much less abundant in the mine tailing samples which plotted in the upper right corner of the PCA plot with high Mg, Cr and Ni concentrations. Among all sources of sedimentary material sampled, the Bécancour River sediments collected up- and downstream

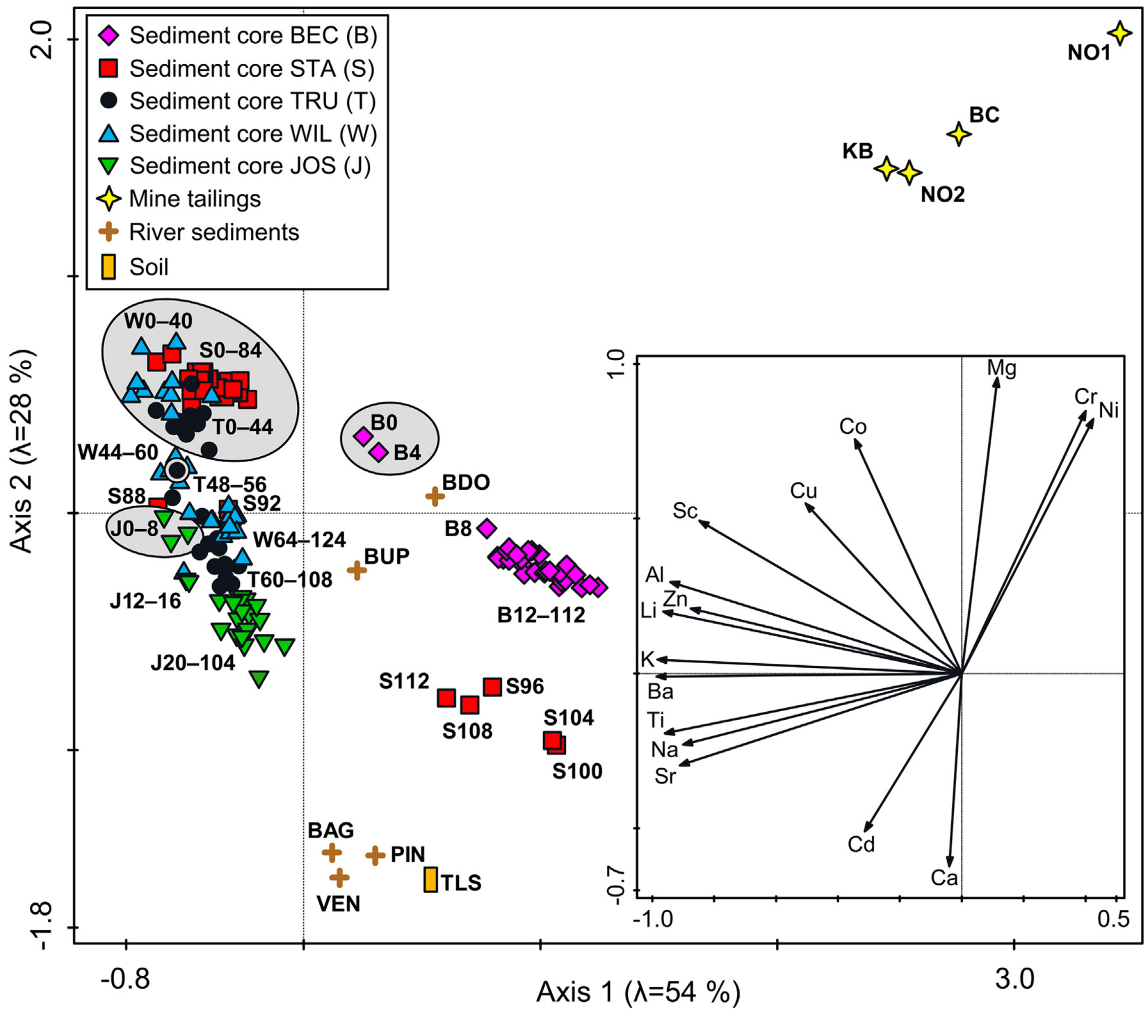

Fig. 6. Principal component analysis (PCA) plots showing the distribution of samples from sediment cores and sediment sources within the Bécancour River drainage basin in terms of metal composition. Note: grey ovals represent the sediment core samples deposited ≥1944 CE, near the intensification of mining activities (1945 CE). Numbers attached to the sediment core samples correspond to rounded depth levels (in centimeters). BAG, Bagot River; BC, British Canadian 1 Mine; BDO, Bécancour River — downstream from mining waste piles; BUP, Bécancour River — upstream from mining waste piles; KB, King-Beaver Mine; NO1, Normandie Mine sample #1; NO2, Normandie Mine sample #2 (border of Bécancour River); PIN, Au Pin River; TSL, Trout Lake soil; VEN, Venlo Brook.

from the mine tailings most resembled the lake sediments based on the position of their sample scores.

The PCA analysis revealed a geochemical signature of lake sediments that evolved over time. For each core, sample scores of sediments deposited before 1870 CE plotted close together, indicating similar geochemical composition. However, sample scores related to years 1870–1944 CE showed more leftward and upward distributions. Sediment composition in Stater Pond changed the most during this time frame. Sample scores of sediments deposited after 1944 CE shifted even more to the left, especially in the case of sediment core BEC. More importantly, they plotted higher along PCA axis 2 towards higher Mg concentrations. Sediments corresponding to this period had almost identical geochemical signatures at Stater Pond, Trout Lake and Lake William, whereas they were more distinct at lakes Bécancour and Joseph. Sediments from downstream Bécancour River were associated with higher Mg, Cr and Ni concentrations as compared to the upstream sample.

5. Discussion

5.1. Pre-settlement period

Sedimentological changes preceding the modern settlement period can be attributed to natural variability. In Lake Bécancour, they closely mirror Holocene climate fluctuations. For example, low OM plateau from −6610 to −6170 CE (8560–8120 cal yr BP; Fig. 4) matches a brief cooling event between −6450 and −6050 CE (8400–8000 cal yr BP) in the Northern Hemisphere (Alley et al., 1997; Shuman and Marsicek, 2016). Maximum OM levels between −5830 and −5640 CE (7780–7590 cal yr BP) can be associated with a peak in air temperatures that was followed by increasing humidity and cooling after −5550 CE (7500 cal yr BP) in southern Quebec and other parts of North America (Bhiry and Filion, 1996; Viau et al., 2006; Shuman and Marsicek, 2016). The decrease in OM after −3530 CE (5480 cal yr BP) coincides with the end of the Holocene thermal maximum, which was accompanied by yet another pronounced cooling event and increase in humidity after −3550 CE (5500 cal yr BP; Viau et al., 2006; Shuman and Marsicek, 2016). These changes in OM content have likely been driven by variations in primary productivity and biomass, as favored by higher air/water temperatures and longer growing seasons. Increases in OM content in Lake William between 1620 and 1820 CE, and in Lake Joseph between 1510 and 1800 CE (Fig. 4) are likely related to reduced summer precipitation after ca. 1550 CE in southern Quebec during the Little Ice Age (ca. 1450–1850 CE; Paquette and Gajewski, 2013). Similar geochemical changes also occurred during the same time interval in Trout Lake (Jacques and Pienitz, 2021). Reduced rain would have resulted in weaker hydric erosion in the drainage basin and hence lower clastic mineral and nutrient inputs into the lakes.

5.2. Effects of early land development

Physical and geochemical proxies revealed that modern settlement after ca. 1810 CE had immediate effects on the sedimentological evolution of Stater Pond and lakes William and Joseph. Decreases in sediment OM content and MGS, accompanied by increasing density (Fig. 4), demonstrate that early human activities (i.e. land clearance and agriculture) resulted in enhanced inputs of fine clastic material into the lakes. Trends towards higher magnetic susceptibility levels denote a greater abundance of magnetic minerals, which is also indicative of enhanced erosion (Sandgren and Snowball, 2001). Consequently, sediment accumulation rates significantly increased (Fig. 3). Sedimentological changes were more abrupt and important in lake William which indicate more rapid and intensive colonization on its banks. Contrary to our other study lakes, no important sedimentological changes were detected in Lake Bécancour's sediments deposited between 1810 and 1880 CE. This is consistent with the probable absence of settlers in that part of the BRB before the onset of mining activities in Thetford Mines.

5.3. Evidence of asbestos mining pollution

Despite a few sedimentological changes, there is no obvious indication of asbestos mining contamination in lakes located downstream from Thetford Mines prior to 1940 CE. Clearly, mining impacts were fairly weak at that time. In our previous study (Jacques and Pienitz, 2021), we found that mining pollution in Trout Lake was greatly enhanced by the drainage of Black Lake (1954–1959 CE; including related preparatory works), which resulted in the deposition of high amounts of fine mineral particles in this water body where it caused sediment accumulation rates to surge. The results that we obtained here indicate that this event also had similar effects on Stater Pond and on lakes William and Joseph following an upstream to downstream decreasing gradient of perturbation. Indeed, the sudden and major stratigraphic changes in these lakes at core depths dated between 1940 and 1960 CE are also undoubtedly attributable to it. However, it is curious that sediment accumulation rates maintained high levels in all downstream lakes in the decades that followed (Fig. 3). Moreover, it is odd that their OM content remained low (Fig. 4), as that they all underwent severe and immediate eutrophication after the drainage event (Jacques and Pienitz, 2021; O. Jacques, unpublished data). Lake eutrophication most commonly causes sediment OM to increase as a result of enhanced biological productivity (e.g., Pienitz et al., 2006; Laperrière et al., 2008). In a 'normal' context, one might have expected to find much higher percentages of organic material than the 5.5–8.8% in a hypereutrophic small and shallow water body such as Stater Pond. Our results indicate that significant amounts of mineral matter, diluting the organic phase of the sediments, have been supplied to the lakes since 1960 CE.

High enrichment levels of Mg and Cr in recent lake sediments in Stater Pond and lakes Trout and William (Fig. 5) provide strong evidence that these high mineral matter inputs were triggered by the erosion of mining waste piles. Preferential increases in these metals over other detrital/minerogenic elements, such as K and Al, indicate that their high abundance cannot be simply attributed to generalized enhanced soil erosion in the Bécancour River drainage basin, for example. The Thetford Mines Ophiolite Complex that underlies the mining sites of the region has a different mineralogical composition than other geological groups that cover the vast majority of the BRB basin (MERN, 2021). As illustrated in this study, asbestos mining wastes therefore also present a distinct geochemical signature. We showed that they contain very high concentrations of Mg and Cr (Fig. 6; Table S3), hence are undoubtedly the main source of these metals to downstream lakes since 1960 CE. Although Mg enrichment levels are low in Lake Joseph's recent sediments, they are still higher than those of other detrital elements (Fig. 5) which indicate that this water body also receives small amounts of mining wastes.

The results that we obtained regarding chemical element concentrations within the tailings are also similar to those of previous sampling campaigns (Dupéré et al., 2007; Wu, 2011; Villeneuve, 2013). It has been pointed out that the chemical/mineral composition of tailings was fairly homogenous within each mining pile and between piles from different mining sites (Dupéré et al., 2007; Thibault, 2011; Wu, 2011; Villeneuve, 2013). As we illustrated, in addition to Mg and Cr, tailings are also enriched in Ni. With Fe, it certainly accounted for most of the increases in magnetic susceptibility levels in recent sediments. However, although relatively high Ni enrichment levels were measured in Trout Lake's recent sediments, it was curiously not the case in Stater Pond's sediments (Fig. 5). Comparison of EF values for this metal is biased by the naturally higher Ni/Ti ratio of Stater Pond's precolonial sediments. In fact, mean ratio values are higher in sediments deposited after 1960 CE in Stater Pond (0.058) than in Trout Lake (0.042). Copper pollution in Trout Lake and Lake William might also originate from mining sources. Although this metal is not reputed to be abundant in asbestos wastes, it was present in notable abundance in Normandie Mine piles (Table S3). However, concentrations of this metal were similar in the sediment samples from the Bécancour River collected up- and downstream from the waste piles (Table S3), which suggests they may not be the dominant or only important source of Cu pollution in the lake's catchments. Modern agricultural practices can be another important source of Cu pollution (Mantovi et al., 2003), which may also have accounted for some of the concentration changes in the sediments. Here again, low Cu enrichment levels in Stater Pond's recent sediments are biased by the naturally high Cu/Ti ratio of its precolonial sediments.

The PCA performed in this study (Fig. 6) highlighted the major influence of sedimentary inputs from the Bécancour River and tailing piles on the geochemical composition of lake sediments downstream from Thetford Mines. At first glance, it also suggests that recent sediment core samples are still very different than asbestos wastes. However, the analysis that we performed is simplistic. First, it did not take into account physical and chemical alterations of tailings before and after they enter the Bécancour River. For example, there is probably an important segregation of the mineral fractions composing the residues during erosion events and due to transport in water. Most chrysotile asbestos fibers likely remain in suspension in water and are never deposited into the sediments, while other denser minerals sink to the lake bottoms (Schreier, 1989). The fibers $[Mg_3(Si_2O_5)(OH)_4]$ can represent 40% of the total volume of bulk tailing samples (Villeneuve, 2013) and therefore account for an important part of their Mg content. Second, even though we removed redox-sensitive elements from our PCA analysis, it did not take into consideration all post-depositional mechanisms that may alter the composition of lake sediments, such as contributions from groundwater.

5.4. Triggers of the asbestos waste pollution

The drainage of Black Lake exerted a huge impact on the sedimentological evolution of BRB lakes, which extended far beyond the 1954–1959 CE period. We believe that subsequent mining waste depositions within lakes located downstream from Thetford Mines are mainly attributable to the disappearance of Black Lake, which facilitated their transport over longer distances. As discussed in Jacques and Pienitz, 2021, we suppose that this former lake previously acted as a sediment trap and played an important buffer (protection) role for Trout Lake, as well as lakes William and Joseph. In this regard, our results demonstrate that Stater Pond with its dam and dike never efficiently replaced Black Lake. In addition, mining contamination was favored by the massive amounts of waste material that were disposed of on the banks of the Bécancour River during the excavation works. The opening of the Lake Asbestos Mine in 1958–1959 CE also coincided with an important mining boom in the region which resulted in important and rapid expansions of mine tailing piles (Figs. S12–S14).

The lower Mg, Cr and Ni enrichment levels noted between 1960 and 1975 CE (Fig. 5) coincided with higher sediment accumulation rates (Fig. 3) and generally lower MGS values (Fig. 4) as compared to the more recent sediments deposited between 1975 and 2017 CE. This likely corresponds to a period of intensive erosion and transport of waste materials freshly excavated from Black Lake (which included fine clays; Piette, 1953) and deposited along the Bécancour River. This is based on the assumption that these materials contained lower proportions of Mg, Cr and Ni as compared to the mine tailings. Moreover, small peaks in MAR near 1970 CE (Fig. 3) correspond well with the diversion of a ca. 1 km section of the Bécancour River during expansion works at King-Beaver Mine in 1971 CE (Fig. S12). The river has also been enlarged, channeled and excavated over more than 10 km near the upstream entry of the city in 1971–1972 CE. Lower sediment accumulation rates after 1975 CE correspond with a reduction of such extreme erosional events, and also likely reflect higher stability of waste materials excavated from Black Lake. Conversely, higher Mg, Cr and Ni enrichment levels during this most recent period are indicative of a higher proportion of mine tailing materials in the sediments.

5.5. The case of Lake Bécancour

In contrast to downstream lakes, our results reveal that Lake Bécancour has not received high loads of mineral matter during recent years. Although the OM content of its sediments decreased after 1950 CE (Fig. 4), it remained high and sediment accumulation rates maintained low levels (Fig. 3). This is consistent with its geographical location upstream from the mining waste piles. Yet, significant Mg enrichment in recent sediments of Lake Bécancour (Fig. 5) suggests that it has also been contaminated by asbestos wastes. This could be partly attributed to pollution from a former municipal landfill (closed during the 1970s) located at 1 km distance within its catchment. Asbestos mine tailings have been used to bury the municipal waste, and residue flows towards the lake have been reported in the past. Pollution due to the landfill could as well explain the particularly high Pb, Co and Fe enrichment levels in recent sediments of Lake Bécancour (Fig. 5). Mine tailings were also used as fill material in areas bordering the lake, which could have further contributed to asbestos waste contamination of the sediments. However, mature soils and vegetation currently cover the former landfill and no outcrops of mine tailings are visible around the lake. Although leachate waters could still be an important source of Mg ions to the lake, we suspect that supplementary waste inputs originate from eolian erosion and transport of asbestos pollutants, as the lake is located east of the old mining sites in the direction of predominant winds.

Decreasing OM content after 1950 CE and increases in detrital element concentrations have also undoubtedly been independently favored by repeated natural and human-induced lake level fluctuations following the construction of a dam at the outflow of Lake Bécancour in the late 1930s (Gaudreau, 2002). In particular, it seems possible that an important water rise occurred in Lake Bécancour during the drainage of Black Lake based on plans drawn by Piette (1953). Changes in water column depth likely favored increased erosion in the littoral zone of the lake. Moreover, residential developments on its shores between 1950 and 1966 CE, and in 2003–2004 CE, have probably also contributed to enhanced inputs of clastic material.

5.6. Impacts of asbestos mining wastes

Our findings contradict the long-existing perception that the impacts of asbestos mining wastes on the Bécancour River and its chain of lakes are minimal, as expressed repeatedly in the past (e.g., Cloutier, 1965; Bérubé, 1991; Arbour, 1994). This view was largely based on the notion that hard and thick carbonate crusts have formed at the surface of the wastes following the slowdown and end of mining activities (Beaudoin et al., 2008, 2017), thus apparently cementing and stabilizing the piles. However, we did not observe such hard crusts at our sampling sites and obvious signs of extensive recent tailing erosion and transport were visible. Villeneuve (2013) also had previously noted that not all pile surfaces were cemented. The fact that Mg and Cr enrichment levels in Stater Pond's sediments remained very high after 1980 CE (Fig. 5) is proof for ongoing active waste erosion. However, their decreasing trends in lakes Trout, William and Joseph, and concurrent non-proportional changes in sediment accumulation rates, imply that asbestos waste deposition has slightly diminished at these sites during recent years.

The most significant and perceptible impact of waste erosion on BRB aquatic ecosystems is the exceptionally high sediment accumulation rates it generates. As a result, Stater Pond, Trout Lake and Lake William are experiencing lake infilling at a fast pace. Their depth and surface areas are rapidly decreasing, with problematic sediment accumulations occurring near river inlets (also noticed at Lake Joseph). The short- and long-term consequences of this precipitated 'lake ageing' will be far more critical in Stater Pond and Trout Lake, considering that they are particularly shallow basins.

Asbestos mining wastes also caused metal enrichment of BRB lake sediments. Enrichment levels of Mg and Cr in recent sediments display a clear gradient of contamination downstream from the mining sites (Fig. 5), with Stater Pond being the most impacted water body, followed by lakes Trout, William and Joseph (only weakly affected). Similar to our study, Kumar and Maiti (2015) found important Cr and Ni concentrations in pond and stream sediments near a chromite-asbestos mine of India that decreased with increasing distance from waste influence. Important Mg, Cr and Ni pollution gradients have also been noted in stream sediments located downstream from naturally exposed serpentine asbestos deposits within the Sumas River Watershed on the Canada-U.S. border (Schreier, 1987; Smith et al., 2007; Schreier and Lavkulich, 2010, 2015).

Chromium and Ni are heavy metals that may have adverse effects on the environment. Their mean background concentrations were naturally high in all our study sites. In lakes Bécancour, Trout and William, they ranged between 112 and 191 mg kg^{-1} for Cr and between 147 and 186 mg kg^{-1} for Ni, hence already exceeding the 'probable effect concentration' (PEC) thresholds of respectively 111 mg kg^{-1} and 48.6 mg kg^{-1} proposed by MacDonald et al. (2000; Table S4). Concentrations above the PEC are likely to be harmful for sediment-dwelling organisms. Chromium and Ni are now 1.4 to 1.9 times more abundant in sediments deposited since the drainage of Black Lake (>1960 CE) in lakes Bécancour, Trout and William (Cr: 168–267 mg kg^{-1}; Ni: 208–356 mg kg^{-1}). Modern concentrations are also respectively 5.1 and 3.7 times higher at Stater Pond as compared to precolonial times (Cr: 221 vs 43 mg kg^{-1}; Ni: 264 vs 72 mg kg^{-1}). Although indigenous aquatic organisms of the region were certainly adapted to the naturally high Cr and Ni levels, the new enhanced concentrations now potentially impair aquatic life and reduce biodiversity. Contrary to Cr and Ni, mean background Cu values were generally lower at our study sites (12–33 mg kg^{-1}). However, excluding Lake Bécancour's samples, the Cu content in sediments deposited after 1960 CE is now 1.8 to 3.4 times higher and at all sites exceeds the 'threshold effect concentration' (TEC) of 31.6 mg kg^{-1} established for this metal, above which harmful effects may be observed in sediment-dwelling organisms (MacDonald et al., 2000).

6. Conclusions

We previously highlighted that important erosional and depositional processes of asbestos mining wastes occurred during many decades at Trout Lake and were still ongoing in 2017 CE (Jacques and Pienitz, 2021), almost four decades after regional mining activities began to decline. Here, we demonstrated that mining pollution in the BRB extends far beyond this lake. Consistent with our initial hypothesis, we found that it is also present in Stater Pond, Lake William and Lake Joseph, following a gradient of contamination spread over at least 25 km downstream from the former mining sites. In addition, we unexpectedly found evidence of waste inputs into Lake Bécancour, located upstream from Thetford Mines. Contamination levels are most pronounced since the Black Lake drainage event (1954–1959 CE) that facilitated the transport of pollutants towards downstream lakes, and which also coincided with the expansion of waste piles on the banks of the Bécancour River.

Erosion and transport of mining wastes profoundly altered the sedimentological trajectory of BRB lakes. They caused sedimentation accumulation rates to explode in downstream lakes, at magnitudes consistent with their respective contamination level, and therefore accelerated their ageing. They also resulted in increased concentrations of harmful metals (Cr, Ni) in their sediments. Our study highlights that erosion control measures must be implemented rapidly to ensure the sustainability of BRB lakes. Given the damaging impacts of asbestos mining wastes that we documented in this study, we recommend that close monitoring should be performed on aquatic systems in other regions of the world that have been or are currently affected by this kind of mining industry.

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CRediT authorship contribution statement

Olivier Jacques: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Reinhard Pienitz:** Conceptualization, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

We received funding from organizations that may or may not benefit from demonstrating that asbestos mining wastes pose a threat to the Bécancour chain of lakes.

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Appendix A. Supplementary data

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