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# Historical gold mining increased metal(loid) concentrations in lake sediments from Nova Scotia, Canada

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## Abstract

Historical gold mining operations between the 1860s and 1940s have left substantial quantities of arsenic- and mercury-rich tailings near abandoned mines in remote and urban areas of Nova Scotia, Canada. Large amounts of materials from the tailings have entered the surface waters of downstream aquatic ecosystems at concentrations that present a risk to benthos. We used paleolimnological approaches to examine long-term trends in sedimentary metal(loid) concentrations, assess potential sediment toxicity, and determine if geochemical recovery has occurred at four lakes located downstream of three productive gold-mining districts. During the historical mining era, sedimentary total arsenic and mercury concentrations and enrichment factors increased substantially at all downstream lakes that received inputs from tailings. Similarly, chromium, lead, and zinc concentrations increased in the sediments after mining activities began and the urbanization that followed. The calculated probable effects of concentration quotients (PEC-Qs) for sediments exceeded the probable biological effects threshold (PEC-Q > 2) during the mining era. Although sedimentary metal(loid) concentrations have decreased for most elements in recent sediments, relatively higher PEC-Q and continued exceedance of Canadian Interim Sediment Quality Guidelines suggest that complete geochemical recovery has not occurred. It is likely that surface runoff from tailing fields, urbanization, and climate-mediated changes are impacting geochemical recovery trajectories.

**Key words:** legacy pollution, tailings, mercury, arsenic, geochemical recovery

## Introduction

Historical gold-mining operations have increased metal(loid) concentrations in freshwater sediments from Canada (Azcue et al. 1995; Galloway et al. 2018; Tenkouano et al. 2019; Cheney et al. 2020) and around the world (Odumo et al. 2014; Pinedo-Hernández et al. 2015; Moreno-Brush et al. 2016; Hillman et al. 2017; Yaraghi et al. 2020). The mineralization of gold-bearing ores and various ore-processing approaches (e.g., roasting, crushing, amalgamation) used to extract gold produce finely-ground tailings elevated in toxic metal(loid)s, which can be transported to waterbodies via surface runoff or aerial deposition (Azcue et al. 1995; Pelletier et al. 2020; Perrett et al. 2021). Materials released from gold-mine tailings and mineral-processing by-products can accumulate in aquatic sediments, thus polluting benthic habitats. Direct exposure to mining wastes elevated in metal(loid)s is greatest for sediment-dwelling organisms, and thus aquatic ecosystems can be altered over time (Nasser et al. 2016; Thienpont et al. 2016; Stewart et al. 2018; Little et al. 2020).

Some of the earliest and largest gold mining operations in Canada began in the province of Nova Scotia around the mid-19th century (Cranstone 2002). Historical mining operations played a key role in the economic development of Nova Scotia, which spanned eight decades, with three major gold mining rushes occurring between the 1860s and 1940s (Bates 1987). Historical mining activities were decentralized and occurred in 64 historical gold districts across Nova Scotia. Gold-bearing ores in Nova Scotia are often associated with arsenopyrite, and gold was extracted predominantly by the crushing of ores followed by mercury amalgamation. This approach produced massive quantities of tailings elevated in arsenic (As) and mercury (Hg), among other mining-associated contaminants (Parsons et al. 2012). In some Nova Scotia gold districts, cyanidation was also used after the 1890s to enhance gold recovery (Parsons et al. 2012). Collectively, the 64 gold districts produced more than 3 million tonnes of tailings that were disposed of without treatment into the local environment (Drage 2015).

Both surface water run-off and wind-blown dust from historical mining activities and associated tailing fields throughout Nova Scotia have led to the dispersal of contaminants into the environment for over a century (Wong et al. 1999, 2002; Corriveau et al. 2011; LeBlanc et al. 2020). An assessment of tailings-impacted soil and sediments in 14 historical gold districts showed that As and Hg levels ranged from 10 to 312 000 μg/g (median: 2550 μg/g) and 5 to 350 000 ng/g (median: 1640 ng/g), respectively (Parsons et al. 2012). Arsenic and Hg concentrations in 99% and 71% of 482 samples, respectively, exceeded the Canadian Council of Ministers of the Environment (CCME) Interim Sediment Quality Guidelines (ISQGs) (Parsons et al. 2012). Similarly, aquatic ecosystems downstream of the historical mines showed high levels of Hg and As in contemporary water and sediment samples (Mudroch and Clair 1986; Wong et al. 1999, 2002; Parsons et al. 2012; Clark et al. 2021). Specifically, elevated concentrations of metal(loid)s in streams between tailing fields and downstream lakes suggest that streams are important transport pathways for moving materials (Mudroch and Clair 1986; Wong et al. 1999; Clark et al. 2021). Mercury and As in sediments and soils from historical gold-mine tailing sites have been shown to be toxic and bioaccumulate in invertebrates and plants (Brooks et al. 1982; Dale and Freedman 1982; Chapman et al. 2019, 2020; Leblanc 2019). Previous research from Nova Scotia has provided important insights into recent levels of contaminants in aquatic sediments; however, long-term trends in metal(loid)s associated with historical gold mining and related sediment toxicity are not widely available. Yet, these temporal data are necessary to establish background levels of contaminants prior to historical mining, understand the health of contemporary aquatic ecosystems after the end of mining, and monitor recovery trajectories from decades of metal(loid) inputs.

Paleolimnological studies often rely on measurements of metal(loid)s in lake sediments to track pollution history and quantify the degree of contamination from mining activities across timescales spanning decades to millennia (Cooke and Bindler 2015; Hillman et al. 2017; Jasiak et al. 2021). Here, we investigated metal(loid) concentrations in sediment cores from five lakes within three gold districts in Nova Scotia. Our goals were to (1) examine the magnitude of increases in sedimentary concentrations of total As and Hg relative to pre-mining background levels, (2) compare concentrations against established sediment quality guidelines to assess potential toxicity of the sediments, and (3) determine trends in geochemical recovery in lake sediments following the end of gold mining around the 1940s. The CCME ISQGs for the protection of aquatic life are based on samples from diverse regions across Canada, and include mercury, arsenic, cadmium, chromium, copper, lead, and zinc (CCME 2001). The CCME ISQGs consider the sedimentary toxicity of each metal(loid) independently; however, more often, mine tailings contain complex mixtures of potentially toxic contaminants. Therefore, approaches that integrate multiple contaminants, such as probable effects concentration quotients (PEC-Qs), are also used to assess sediment toxicity (MacDonald et al. 2000; Ingersoll et al. 2001). In this study, we used the paleotoxicity approach described by Rose et al. (2018), which is an application of the PEC-Q framework to dated sediment cores. Since enrichment of contaminants in sediment threatens the integrity of benthic habitats, alters food webs and water quality, and impacts biodiversity, the paleotoxicity approach offers a means to predict the potential toxicity of lake sediments to invertebrates (Cheney et al. 2020). Recently, Cheney et al. (2022) calculated PEC-Q for dated sediment intervals from severely metal(loid)-polluted lakes and then exposed cultured *Daphnia* sp. (common zooplankton in lakes) to the dated sediments. Cheney et al. (2022) observed increased mortality in *Daphnia* sp. that were exposed to dated sediments with high predicted toxicity (i.e., high PEC-Qs). Here, we use the paleotoxicity approach to predict the potential toxicity of sediments from lakes in Nova Scotia that were impacted by historical gold mining activities.

## Methods and materials

### Study region and lakes

We selected lakes near three of the most productive historical gold districts in Nova Scotia: Goldenville, Waverley, and Montague (Fig. 1). The Goldenville mining district produced the largest amount of gold (estimated 210 000 troy ounces) in Nova Scotia (Bates 1987). Waverley and Montague historical gold districts placed fourth and fifth, producing 73 000 and 68 000 troy ounces of gold, respectively (Drage 2015). Mining began in all three districts around the 1860s and ended during the 1940s (Bates 1987). Presently, Waverley and Montague gold districts are part of the Halifax Regional Municipality, and the study lakes exist within an urban environment. In contrast, Goldenville is a rural community near the village of Sherbrooke in the eastern coastal area of Nova Scotia. The study region is located south of the Minas Fault Zone and is underlain by the Halifax (meta-siltstone and slate) and Goldenville (meta-sandstone) groups of the Meguma supergroup (Stea et al. 1992). The surficial geology around the study lakes near Montague and Waverley gold mining districts is predominantly till veneer, drumlins, hummocky till, lacustrine, and exposed bedrock (Stea et al. 1992). The surficial geology near Goldenville mining district is mostly stony till plain and drumlins, along with some exposed bedrock (Stea et al. 1992). All three mining districts are located within the south-central Nova Scotia Uplands Ecoregion, where summers are warm and winters are cool, as the climate is moderated by proximity to the Atlantic Ocean (Webb and Marshall 1999).

Gegogan Lake is a small (surface area: 0.21 $km^2$), shallow (maximum depth: 2.2 m) polymictic lake located in the historical Goldenville gold district (Fig. 1; Table 1). It receives surface water inflow from Gegogan Brook, which covers a distance of ~6 km and drains the upstream mining area. Materials in the mine tailings are transported to Gegogan Lake via Gegogan Brook, and previous investigations demonstrate that the lake serves as an important sink for metal(loid) contaminants transported from the upstream tailings field (Wong et al. 1999). Nova Scotia Highway 7 runs along the eastern side of this lake. There is only one seasonal residence on the shores of this lake, and the watershed is entirely forested with mostly conifers.

**Fig. 1.** Map showing the study region. The black diamonds show the location of the three gold mining districts in Nova Scotia. The star in the map of Canada (inset) identifies the location of Nova Scotia. The black circles identify the locations of the five study lakes. The map was made in ArcGIS Online using the Provinces and Territories of Canada basemap.

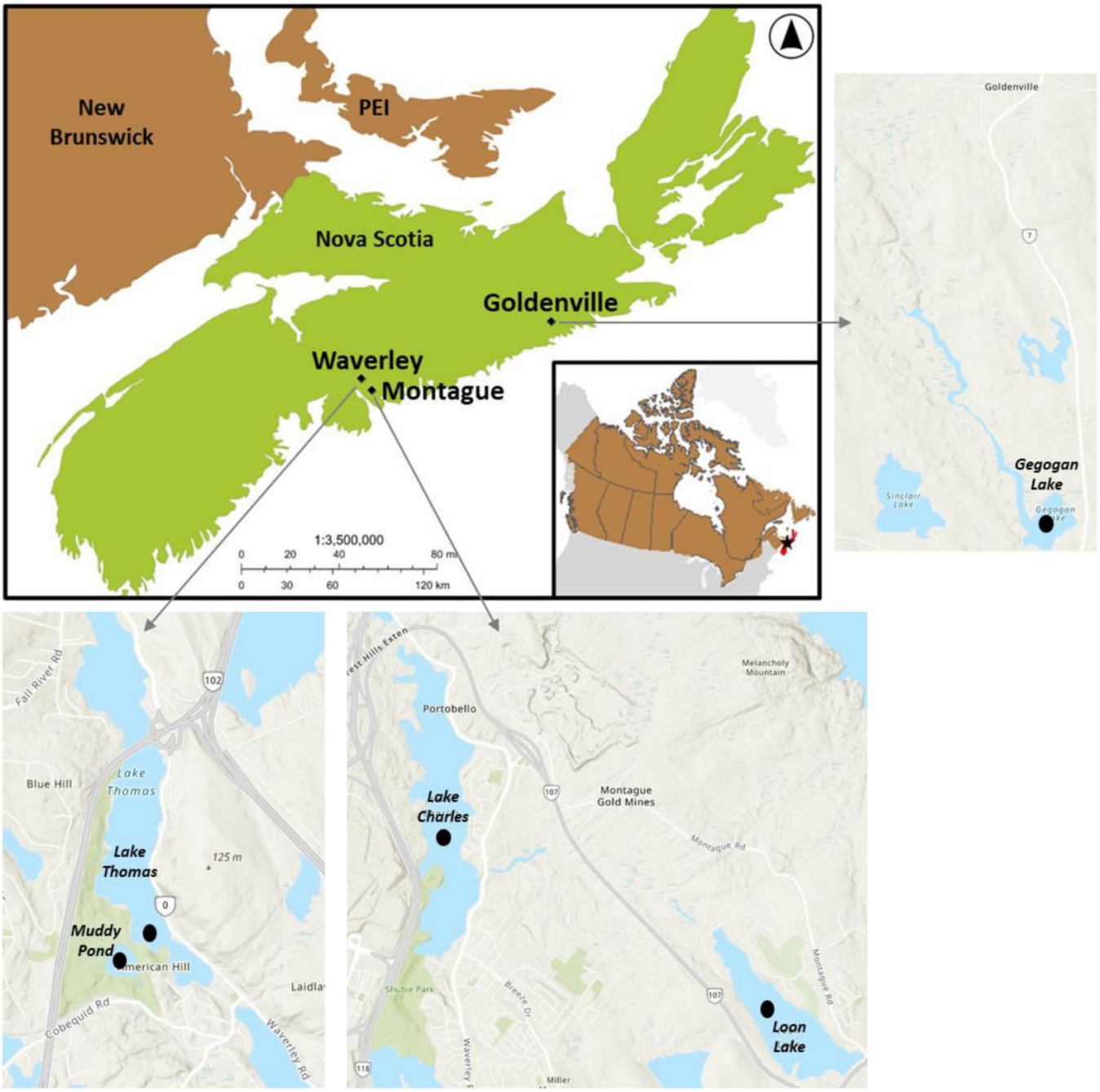



**Table 1.** Select limnological characteristics of the five study lakes from Nova Scotia, Canada.

| Lake | Gegogan | Thomas | Muddy | Charles | Loon |
|---|---|---|---|---|---|
| Latitude (decimal degrees) | 45.0783 | 44.792 | 44.788 | 44.715 | 44.703 |
| Longitude (decimal degrees) | −62.0045 | −63.607 | −63.611 | −63.548 | −63.507 |
| Maximum depth (m) | 2.2 | 17 | 3.5 | 28.1 | 6.6 |
| Surface area ($km^2$) | 0.21 | 1.12 | 0.1 | 1.35 | 0.75 |
| Sampling date | April 2022 | November 2021 | November 2021 | May 2019 | May 2019 |
| Sediment coring depth (m) | 2.0 | 16.7 | 3.3 | 28.1 | 6.6 |

Lake Thomas and Muddy Pond are located in an urbanized watershed flowing through the historical Waverley gold district (Fig. 1; Table 1). Similar to Gegogan, Muddy Pond is also a small (surface area: 0.1 $km^2$), shallow (maximum depth: 3.5 m) polymictic lake that flows into downstream Lake Thomas via a 100 m-long stream. Tailings from Waverley gold mines are located in the southern part of Muddy Pond. Lake Thomas is a larger (surface area: 1.12 $km^2$), deeper (maximum depth: 17 m) dimictic lake with several major roadways around it, including a causeway that was built in the 1960s near its north end to accommodate Nova Scotia Highway 102. Lake Thomas is also part of the Shubenacadie Canal system, which was constructed between 1826 and 1861 to connect Halifax Harbour to the Bay of Fundy.

Lake Charles is a large (surface area: 1.35 $km^2$), deep (max depth: 28.1 m) dimictic lake located near the historical Montague gold district (Fig. 1; Table 1). It receives mine tailings from the Montague gold district via Barry's Run and Mitchell's Brook. Similar to Lake Thomas, Lake Charles is also part of the Shubenacadie Canal system and now supports extensive commercial and residential development in its catchment. Lake Charles thermally stratifies in the summer. Unlike the other four study lakes, Loon Lake does not receive mine tailings via surface runoff because it is located upstream of the Montague tailings fields (Fig. 1; Clark et al. 2021). Loon Lake is shallow (maximum depth: 6.6 m) and has a relatively well-developed catchment, including a golf course located on the eastern part of the lake. In this study, Loon Lake serves as a reference site to compare with lakes that received metal(loid) pollution from the surface run-off of mine tailings.

## Field work

The study lakes were sampled in May 2019 (Charles and Loon), November 2021 (Thomas and Muddy), and April 2022 (Gegogan). A Glew (1989) gravity corer was used to retrieve sediment cores from the deepest basin of each lake and sectioned onsite at 0.5-cm intervals using a Glew (1988) extruder. The deepest basins for urban lakes were identified using bathymetric maps, and for remote Gegogan Lake, we measured water depths along a few transects to determine the deepest area of the lake. The sediments were kept in a cooler and transported to Mount Allison University, where they were stored refrigerated or frozen until further processing.

## Radiometric dating

Sediment core chronologies were developed by analyzing sediments from select intervals for $^{210}Pb$ and $^{214}Pb$ activities at the Paleoecological Environmental Assessment and Research Laboratory at Queen's University in Kingston, Ontario, Canada. Briefly, freeze-dried sediments were placed in plastic tubes, sealed with epoxy, and then analyzed using gamma spectroscopy (EG&G Ortec germanium detector) following the methods outlined in Schelske et al. (1994). Chronologies were developed using the unsupported concentrations of $^{210}Pb$ and the constant rate of supply (CRS) model described by Appleby (2001). Usually, dates beyond background $^{210}Pb$ activities are extrapolated using established $^{210}Pb$ chronology; however, the extrapolated dates are not anchored to specific sedimentary markers. For Lake Thomas and Muddy Pond, dates beyond background $^{210}Pb$ activities (early-1900s) to the 1860s were extrapolated using downcore trends in Hg levels. Once deposited in lake sediments, Hg undergoes limited post-depositional mobility and can be used as a supplementary stratigraphic marker of pollution inputs (Baskaran et al. 2014; Outridge and Wang 2015). We used the initial rise in sedimentary Hg concentrations to identify the approximate start of mining activities and the use of Hg amalgamation. Dates between the last reliable $^{210}Pb$ date and the 1860s were determined using a linear equation, and these dates should be interpreted with caution. For this study, we extrapolated dates until the 1860s because we were interested in examining the temporal trends in metal(loid)s before and after gold mining activities that began around the mid-1800s.

## Metal(loid) analysis

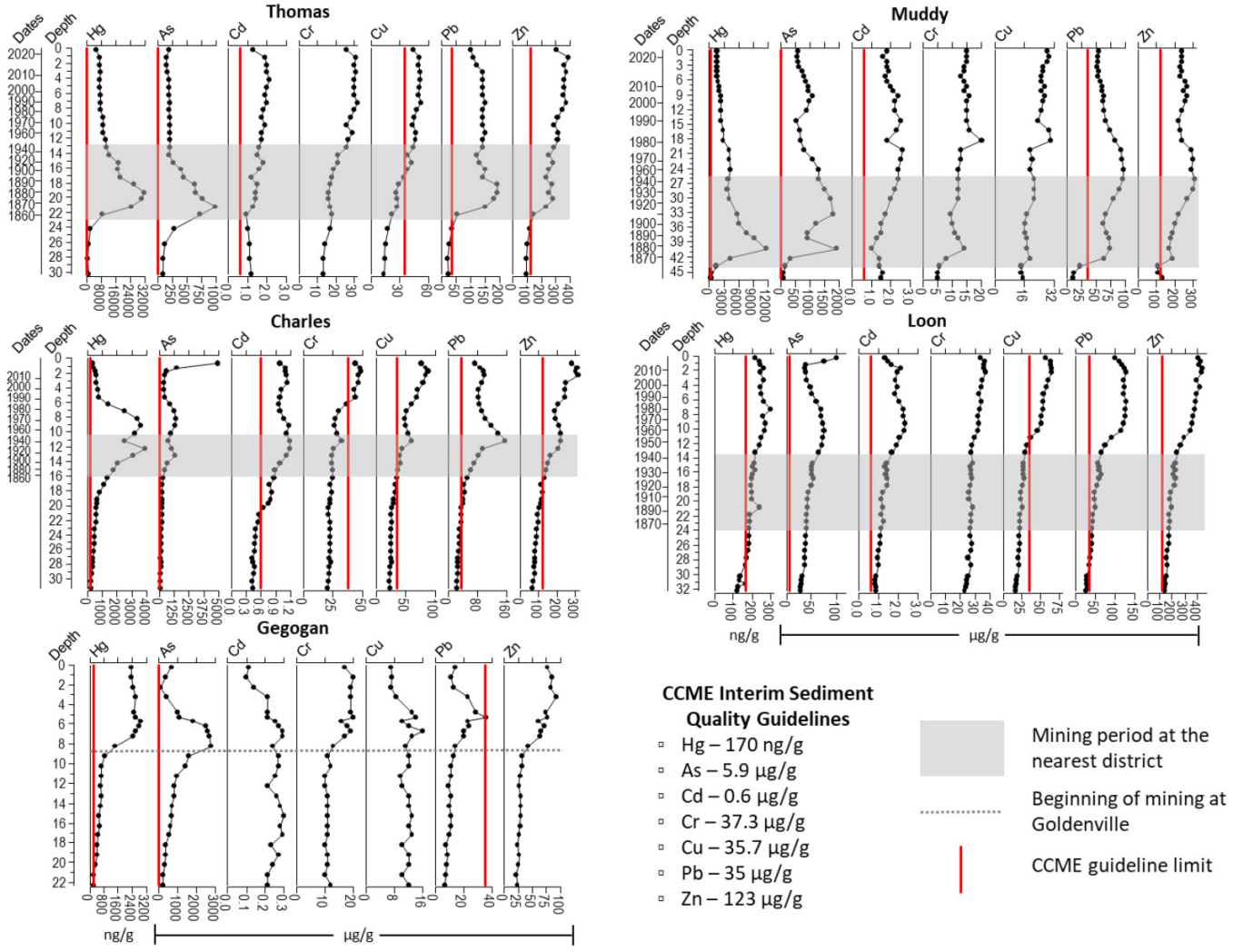
Sedimentary concentrations of aluminum, arsenic, barium, cadmium, calcium, chromium, cobalt, copper, iron, lead, magnesium, manganese, molybdenum, nickel, potassium, selenium, silver, sodium, strontium, thallium, titanium, uranium, vanadium, and zinc were measured in freeze-dried sediments using inductively coupled plasma mass spectrometry (ICP-MS). Sediment samples from Lake Charles and Loon Lake were analyzed at Bureau Veritas Mineral Laboratories Ltd. (BVML, Vancouver, BC, Canada) and published in Clark et al. (2021). Sediment samples from Lake Thomas, Muddy Pond, and Gegogan Lake were analyzed at the Analytical Services Unit (ASU, Kingston, ON, Canada). Both laboratories are Canadian Association for Laboratory Accreditation-certified facilities. At the ASU, sample processing was derived from USEPA Method 200.7, where samples were digested using an aqua regia digestion and diluted for ICP-MS analysis. Metal(loid) concentrations are reported as μg/g dry weight. Mercury samples at the ASU were analyzed using a direct mercury analyzer (DMA-80) following USEPA Method 7473, and concentrations are reported as ng/g dry weight (except in Fig. 3, where units were converted to μg/g). Additionally, information regarding laboratory approaches is included in the Supplementary Materials. To assess quality assurance and quality control, blanks, replicates, controls, and certified reference material were analyzed at the laboratories. Results of the laboratory blanks, replicates, controls, and certified reference materials for the elements are provided in the Supplementary Materials.

## Data analysis

The Canadian Council of Ministers of the Environment ISQGs (CCME 2001) for freshwater sediments are available for total mercury (170 ng/g), arsenic (5.9 μg/g), cadmium (0.6 μg/g), chromium (37.3 μg/g), copper (35.7 μg/g), lead (35 μg/g), and zinc (123 μg/g); therefore, these seven metal(loid)s are the primary focus here, and are presented in Fig. 2. Stratigraphic profiles for all 25 elements are provided in Supplementary Figs. 2*a*–2*e*.

In addition to examining long-term trends in concentrations of the seven metal(loid)s, we also used enrichment fac-

**Fig. 2.** Metal(loid) concentrations in the sediments from the five study lakes are scaled by core depth (cm) with $^{210}$Pb-estimated years plotted. The mining periods are identified with a grey box for Lake Thomas, Muddy Pond, Lake Charles, and Loon Lake. The beginning of the mining period for Gegogan Lake is indicated with a grey dashed line. The CCME Interim Sediment Quality Guidelines for the seven metal(loid)s are marked with a solid red line. The red lines are only included when at least one sediment interval exceeds the guideline threshold.






tors (EFs) to assess the magnitude of increases in sedimentary As and Hg levels, the two most important potentially toxic elements associated with historical mining in Nova Scotia. Several approaches were considered to assess metal(loid) enrichment in lake sediments (Cheney et al. 2020; Jasiak et al. 2021; Jacques and Pienitz 2022). For instance, lithogenic elements (e.g., aluminum, titanium) are often used for EF calculations in studies, while in other studies, EF calculations were based on pre-mining concentrations. In this study, we calculated EFs based on the sedimentary metal(loid) concentrations from the pre-mining period using the following equation:

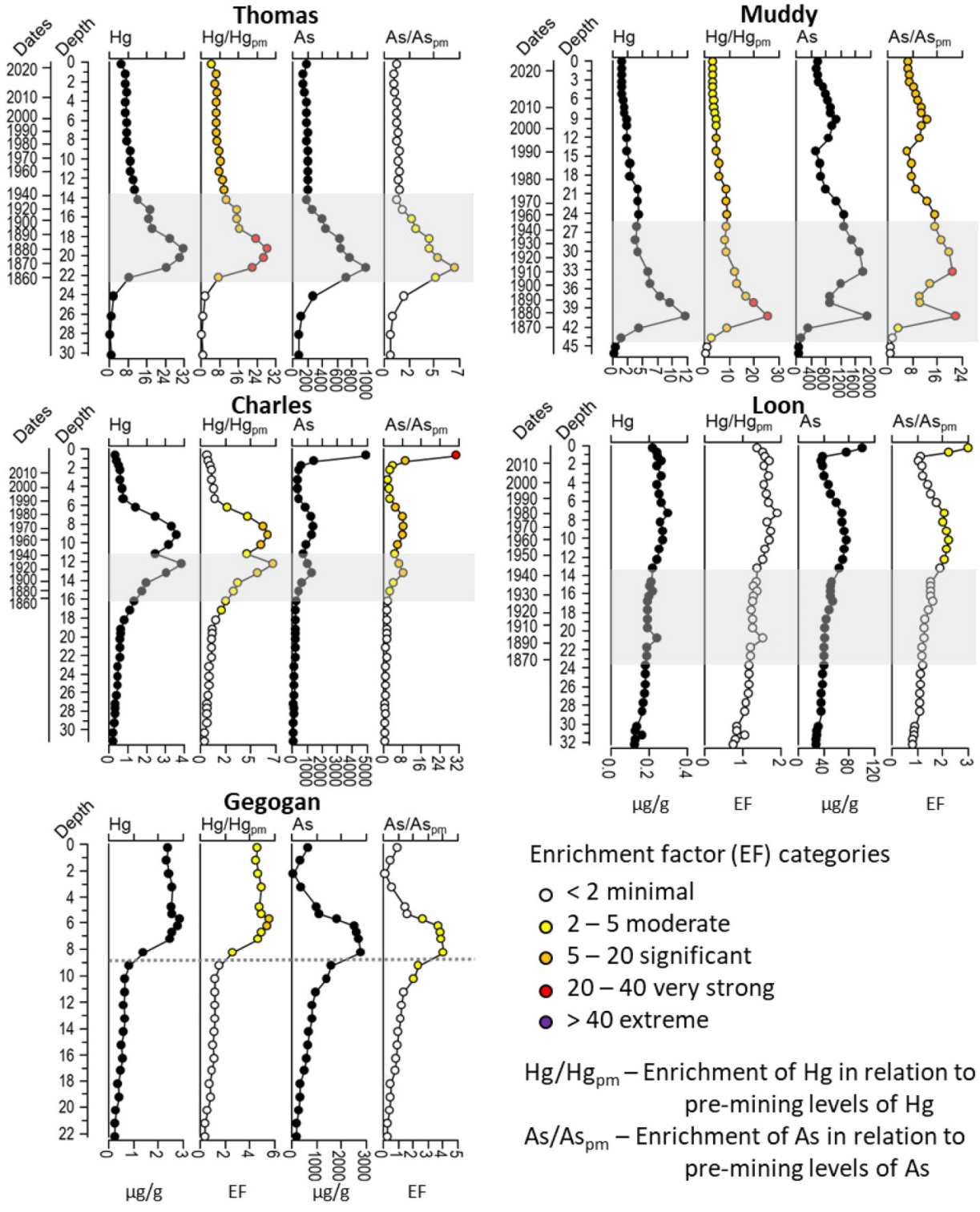
$$\mathrm{EF_{pm}} = A_{\mathrm{sample}}/A_{\mathrm{reference}}$$

where $A$ is the element of interest, and its concentration in a sample is divided by the mean concentration in the pre-mining sample(s) based on the chronology of the dated sediments.

Previous paleolimnological studies have used different approaches to determine the severity of sediment contamination based on EFs (e.g., Chassiot et al. 2019; Cheney et al. 2020; Jasiak et al. 2021; Jacques and Pienitz 2022). In this study, we used the categories recommended by Sutherland (2000), whereby EF < 2 suggests minimal enrichment, EF 2–5 suggests moderate enrichment, EF 5–20 suggests significant enrichment, EF 20–40 suggests very strong enrichment, and EF > 40 suggests extreme enrichment.

The probable effects concentrations (PECs) for sediment, above which there is a higher likelihood of adverse impacts to exposed biota, were developed as part of the consensus-based sediment quality guidelines (MacDonald et al. 2000). Published sediment PEC values are available for arsenic (33 µg/g), cadmium (4.98 µg/g), chromium (111 µg/g), cop-

**Fig. 3.** Sedimentary concentrations of total mercury (Hg) and arsenic (As), along with calculated enrichment factors, are scaled by core depth (cm) with $^{210}$Pb-estimated years plotted. The mining periods are identified with a grey box for Lake Thomas, Muddy Pond, Lake Charles, and Loon Lake. The beginning of the mining period for Gegogan Lake is indicated with a grey dashed line.




per (149 µg/g), lead (128 µg/g), mercury (1.06 µg/g), nickel (48.6 µg/g), and zinc (459 µg/g; MacDonald et al. 2000). Recognizing that aquatic sediments frequently have mixtures of potentially toxic metal(loid)s, probable effects concentrations quotient (PEC-Q) is a promising approach to assessing the potential toxicity of aquatic sediments polluted by multiple, co-occurring contaminants (MacDonald et al. 2000). The PEC-Q approach to assessing potential sediment toxicity to biota has

been successfully applied to sites across North America, including aquatic sediments from Nova Scotia (Ingersoll et al. 2001). Recently, Rose et al. (2018) applied the PEC-Q approach to samples from lake sediment cores to estimate past changes in sediment toxicity. Since we had the measurements for all eight metal(loid)s that have PEC guidelines, all eight were included in the calculations. The PEC-Q for each sample was calculated using the following equation:

$$\text{PEC} - \text{Q} = \Sigma \left( \frac{A_{\text{sample}}}{\text{PEC}_A} \right) / n$$

where $A$ is the element of interest, and its concentration in a sample is divided by the PEC guideline, and $n$ is the number of contaminants that have the PEC guideline included in the calculation of the PEC-Q for a sample. Previous investigations have noted that when PEC-Q is below 0.5, biological effects are unlikely; when PEC-Q is greater than 2, biological effects are probable; and biological effects are possible when PEC-Q is between 0.5 and 2 (Ingersoll et al. 2001; Rose et al. 2018; Cheney et al. 2020).

# Results

## Radiometric dating

Reliable sediment core chronologies were developed using the unsupported activities of $^{210}$Pb and the CRS model for Lake Thomas, Muddy Pond, Lake Charles, and Loon Lake (Supplementary Fig. 1). The dating profiles of Lake Charles and Loon Lake were published in Clark et al. (2021). Generally, the $^{210}$Pb activities declined with depth. A reliable sediment core chronology could not be developed for Gegogan Lake as the $^{210}$Pb activities were very low (<100 Bq/kg) in the sediments. Although very low $^{210}$ Pb activities in lake sediments are uncommon in temperate regions, a similar observation was made at another shallow lake in northeastern Nova Scotia (Middle Lake; Dunnington et al. 2020). In the absence of $^{210}$Pb-based sediment core chronologies, other geochronological markers can be used to identify time periods. As we described earlier, increases in sedimentary Hg concentrations can be a reliable marker to identify the beginning (~1860) of Hg amalgamation, which started with historical gold mining practices in this region due to its inputs from upstream tailings fields (Wong et al. 1999; Clark et al. 2021). Specifically, we used the initial rise in Hg levels in the Gegogan Lake sediment core around 8 cm to approximate the onset of mining and ore amalgamation activities at Goldenville around the 1860s. However, interpretations of sedimentary metal(loid) trends, PEC-Qs, and EFs for Gegogan Lake should be viewed with caution as a reliable chronology for this core could not be developed.

## Trends in sedimentary metal(loid) concentrations

Pre-mining sedimentary metal(loid) concentrations were relatively stable in the lake sediment profiles prior to the 1860s (Fig. 2 and Supplementary Figs. 2*a*–2*e*). However, pre-mining concentrations varied among the five sites, and background As and Hg concentrations were often greater than the CCME ISQG. Sedimentary As and Hg concentrations increased around the 1860s at Thomas, Muddy, Charles, and Gegogan lakes when regional gold mining activities began (Fig. 2 and Supplementary Figs. 2*a*–2*e*). The highest sedimentary Hg concentration (31 400 ng/g) was recorded at Lake Thomas around the 1880s. Sedimentary concentrations of Cr, Pb, and Zn also increased at Thomas, Muddy, Charles, and Gegogan after mining activities started (Fig. 2). Increases in sedimentary Cd concentrations during the mining period were observed at Thomas, Muddy, and Charles, while at Gegogan, [Cd] decreased after the onset of gold mining. Unlike the four lakes that are located downstream of gold mines, the sedimentary metal(loid) levels remained stable during the mining period at Loon Lake, which, as an upstream lake, did not receive inputs of surface waters from the tailings fields of the nearby Montague gold district (Fig. 2).

Generally, As, Hg, and Pb concentrations decreased after regional gold mining activities ended around the 1940s at the four lakes downstream of the historical mines. There were two exceptions to this pattern: (1) Lake Charles, where As levels were highest in the most recent sediments; and (2) Gegogan Lake, where Hg concentrations continue to remain at levels similar to the peak during historical mining (Fig. 2). Both Cr and Zn levels remain elevated in the post-mining sediments at all the sites, including the reference site, Loon Lake. Sedimentary Cu levels have been increasing gradually at Thomas, Muddy, and Charles lakes during the last ~160 years, except at Gegogan, where they have decreased. The notable increases in sedimentary Cd, Cu, Pb, and Zn concentrations at Loon Lake began after the 1950s when urbanization of the watershed occurred. An abrupt increase in As concentrations was observed in the post-2010 sediments at Loon Lake, while other elements showed a minor decreasing trend post-2010 (Hg, Cd, and Pb; Fig. 2).

## CCME Interim Sediment Quality Guidelines (ISQGs)

All sediment samples, including intervals from the pre-mining period, had As concentrations that exceeded the CCME ISQG of 5.9 μg/g (Fig. 2). Similarly, the total Hg ISQG of 170 ng/g was exceeded in all sediment samples except for six pre-mining samples from Loon Lake (Fig. 2). After regional mining activities began, however, the concentrations of both As and Hg greatly exceeded the ISQGs at Thomas, Muddy, Charles, and Gegogan lakes (Fig. 2 and Table S1). Chromium concentrations increased at most sites, but the levels remained below the ISQG of 37.3 μg/g in almost all samples, except for sediments deposited after ~1990 at Lake Charles. Generally, Pb and Zn concentrations at Thomas, Muddy, and Charles exceeded ISQGs after mining activities began (Fig. 2). Both Pb and Zn also increased at Gegogan Lake after mining activities began, but both elements remained below ISQGs, with the exception of one sample (Fig. 2). Cadmium concentrations at Thomas, Muddy, and Loon exceeded the ISQG (0.6 μg/g) throughout the entire record, including pre-mining periods (Fig. 2). Sedimentary Cu concentrations at Gegogan Lake and Muddy Pond remained below the ISQG (35.7 μg/g).

The Cu ISQG threshold was exceeded at Charles and Thomas after mining activities began and after the 1950s at Loon Lake.

### Sedimentary arsenic and mercury enrichment factors

Overall, the post-1860s sediments from Thomas, Muddy, Charles, and Gegogan lakes were enriched in both As and Hg (Fig. 3). The magnitude of enrichment differed among the four lakes downstream from tailings fields. During the mining period, sediments at Muddy Pond reached As and Hg EFs >20, and the highest EF (42) for Hg was observed around the late-1870s (Fig. 3). Similarly, very high enrichments of As and Hg were also observed at Lake Thomas between the 1860s and 1940s (Fig. 3). At Gegogan Lake and Lake Charles, sedimentary As and Hg levels reached moderate levels of enrichment after regional mining activities began. The Hg EFs at Lake Charles, Lake Thomas, and Muddy Pond in the recent sediments decreased, while at Gegogan Lake, Hg EFs continued to be elevated at values near those of the peak in the core coincident with early mining activities. The As EFs show signs of recovery at Lake Thomas, Muddy Pond, and Gegogan Lake. At Charles, the highest As EF (31) was observed in the most recent sediments. The lowest levels of As and Hg enrichment were observed at reference Loon Lake, where Hg EFs always remained below the minimal enrichment threshold.

### Probable effects concentration quotients (PEC-Q)

The PEC-Q values in our study lakes were strongly influenced by sedimentary As and Hg concentrations, as these two mining-associated elements were present at high levels following the establishment of historical mining in the watersheds. Before gold mining operations began, the PEC-Qs at Lake Charles, Lake Thomas, Muddy Pond, and Loon Lake were below the probable biological effects thresholds (<2; Fig. 4). At Gegogan Lake, however, several pre-mining intervals had PEC-Qs greater than 2. When mining activities began around the 1860s, the PEC-Q increased by several-fold at Gegogan, Charles, Thomas, and Muddy, relative to the pre-mining period. PEC-Qs exceeded the probable biological effects threshold at Charles, Thomas, and Muddy after mining began. The PEC-Q at Gegogan, Thomas, and Muddy has decreased in recently deposited sediments. Although Lake Charles displayed a similar decreasing trend after mining activities ended, recent sediments recorded the highest PEC-Q because of high As levels. The PEC-Q levels at Loon Lake have always been below 1, but they have increased gradually since the mid-20th century.

## Discussion

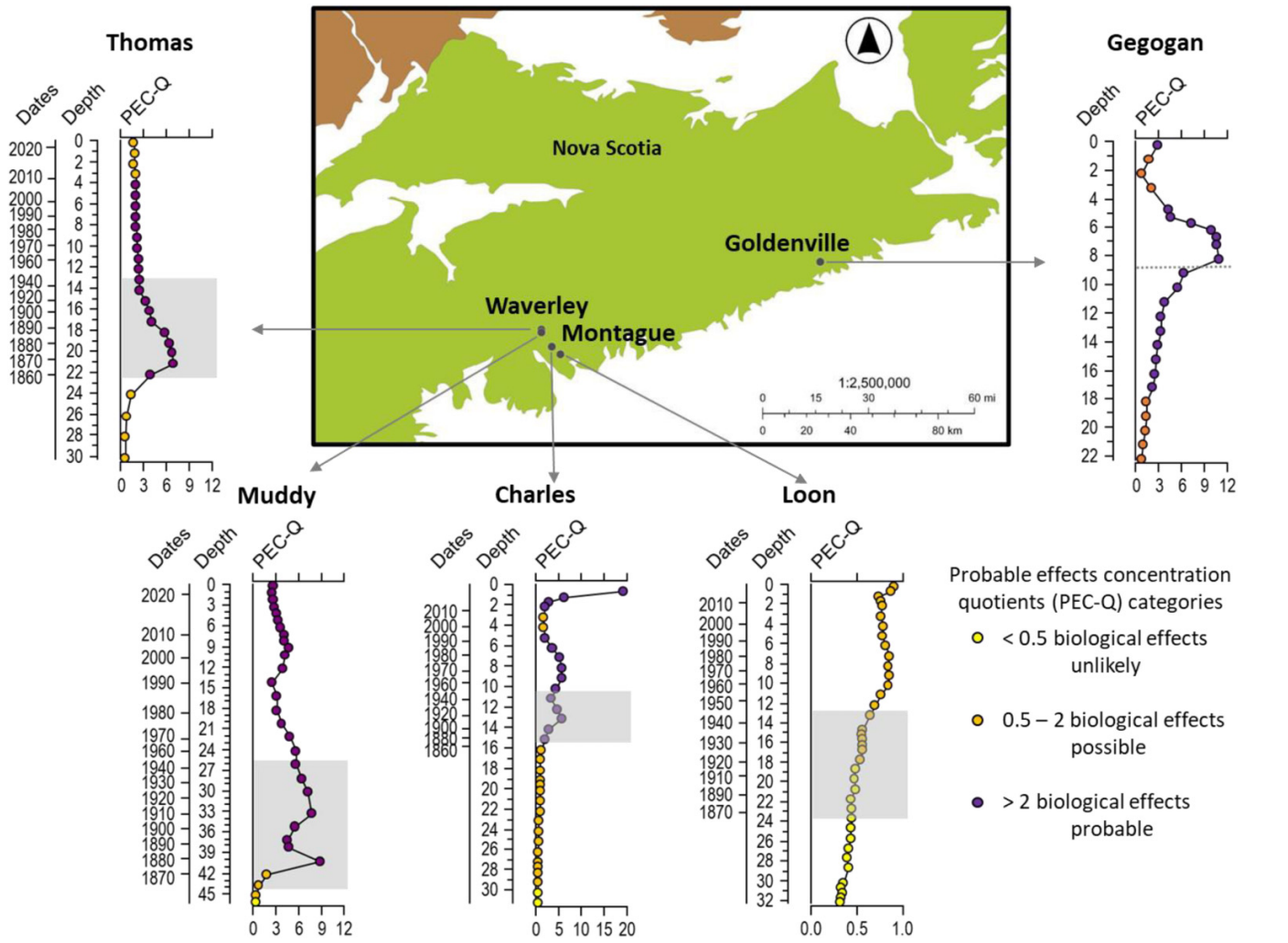
### Sedimentary As and Hg contamination due to historical gold mining and mineral processing

Despite differences in background or pre-mining total As and Hg concentrations in the sediments of each study lake, concentrations of mining-associated elements have increased from background levels at all study lakes. Increases in sedimentary As and Hg occurred earlier and were much larger at the four study lakes downstream of historical-mining tailing fields relative to Loon Lake, which represents an urban reference lake located upstream of the Montague gold district. Greater sedimentary As and Hg levels at four downstream lakes coincided with the beginning of gold mining activities in the 1860s. Meanwhile, Hg and As concentrations at Loon Lake remained much lower during the mining period and only increased during the period of urbanization (Clark et al. 2021). This shows that tailings-contaminated material transported downstream via surface waters and wind was an important source of As and Hg pollution to downstream lake ecosystems, which is consistent with earlier investigations (Mudroch and Clair 1986; Wong et al. 1999, Wong et al. 2002; Clark et al. 2021).

The high sedimentary As levels observed during the mining period in our study lakes are comparable to those reported from gold mine-impacted sites in other regions of Canada, such as Madoc, Ontario (Tenkouano et al. 2019) and Yellowknife, Northwest Territories (Wagemann et al. 1978; Mudroch et al. 1989; Andrade et al. 2010; Thienpont et al. 2016; Cheney et al. 2020; Jasiak et al. 2021). It is notable that the peak sedimentary Hg concentrations from the late 1800s reported at Lake Thomas (31 400 ng/g) and nearby Muddy Pond (11 600 ng/g) are among the highest published results in Canadian lake sediments. For example, peak Hg levels of ~20 000 ng/g were reported from highly-contaminated Phantom Lake near Flin Flon, Manitoba, smelters (Ma et al. 2013). Similarly, exceptionally high Hg levels (>17 000 ng/g) were reported in St. Lawrence River sediments impacted by effluent from a mercury cell chlor-alkali plant and pulp paper factory near Cornwall, Ontario, where industrial activities led to severe sediment pollution (Razavi et al. 2013; Moir et al. 2021). However, broadscale assessments of sedimentary Hg concentrations from other lakes in Canada typically report much lower values because most sites are primarily affected by long-range atmospheric deposition (Muir et al. 2009; Drevnick et al. 2016; Roberts et al. 2019; Galloway et al. 2023). Although Lake Thomas received tailings from Muddy Pond, it may have also received tailings from other mines within the Waverly district (Geological Survey of Canada 1901) that disposed of tailings near its shoreline. Hence, sedimentary concentrations during the mining era were much higher at Lake Thomas relative to Muddy Pond. Further, Muddy Pond is a shallower pond where metal(loid)s may be distributed evenly across the basin, while Lake Thomas is a deeper lake with a steep depth gradient where metal(loid)s may be focused on the deepest points, as observed in other studies (Blais and Klaff 1995).

The enrichment of sedimentary As and Hg during the past ~150 years varied among the study lakes. Proximity to tailing fields, water residence time, inputs via surface waters, and organic matter availability may have influenced the enrichment of these two mining-associated elements at each site, in addition to input from local bedrock and derived surficial materials. For example, during the mining era, the highest EFs for As and Hg were recorded at Muddy Pond, which was expected because tailings were disposed of directly on the southern shore of this shallow pond. Previous

**Fig. 4.** The predicted toxicity of sediments using the probable effects concentration quotients (PEC-Q) is scaled by core depth (cm) with $^{210}$Pb estimated years plotted for the five study lakes. The mining periods are identified with a grey box for Lake Thomas, Muddy Pond, Lake Charles, and Loon Lake. The beginning of the mining period for Gegogan Lake is indicated with a grey dashed line. The map was made in ArcGIS Online using the Provinces and Territories of Canada basemap.




studies from Nova Scotia have shown that the streams running through tailing fields are a key pathway by which historical mining contaminants are transported (Mudroch and Clair 1986; Parsons et al. 2012). Both Gegogan and Charles lakes receive inflow from streams that run through extensive tailings fields. Although Goldenville was the most productive historical mine in Nova Scotia and generated the most tailings (Drage 2015), As and Hg enrichment at Gegogan was modest relative to Muddy Pond, Lake Thomas, and Lake Charles. Gegogan Lake is located 6 km south of Goldenville tailing fields; hence, it is possible that more tailing-derived metal(loid)s were integrated into Gegogan Brook sediments closer to the tailing fields (Wong et al. 1999), which likely reduced the amount of metal(loid)s input to the lake.

## Sediment quality guidelines and paleotoxicity in mine-impacted lakes

This study and previous research have highlighted the legacy As and Hg pollution of aquatic ecosystems near abandoned gold mines in Nova Scotia and their exceptionally high concentrations relative to other Canadian watersheds where historical gold mining occurred (Parsons et al. 2012; LeBlanc et al. 2020; Clark et al. 2021). Gold mine tailings in Nova Scotia, however, also contain several other metal(loid)s known to harm sediment-dwelling biota (Wong et al. 1999, 2002; Parsons et al. 2012). We also observed increases from pre-mining background levels in sedimentary Cr, Pb, and Zn at all downstream lakes after mining activities began in the 1860s. A growing number of studies emphasize the importance of approaching mine tailings pollution as a mixture of contaminants, even when one or two elements may be present in much higher concentrations relative to others (Leppänen et al. 2017; Little et al. 2020; Perrett et al. 2021; Sivarajah et al. 2021). The presence of co-occurring metal(loid)s, albeit at varying levels, should be considered when assessing sediment quality and potential toxicity because biota are often exposed to multiple contaminants in aquatic ecosystems (Rose et al. 2018).

The CCME ISQGs serve as environmental benchmarks and can be useful tools to assess sediment quality; however, they should be applied with caution and within an appropriate environmental context involving a recognition of "background" or pre-impact levels. This is true in regions where background metal(loid) concentrations may be elevated due to local geology. Both As and Hg concentrations are higher than the ISQGs in almost all samples from our study lakes, including sediments from the pre-mining period. The background As concentration is likely higher in lakes from this region because of the weathering of arsenian pyrite and arsenopyrite veins that occur within the Cambro-Ordovician metapelites of the Meguma Group hosting the gold that was mined in this area (Graves and Zentilli 1982; Kennedy and Drage 2017). In a regional survey of lakes near the former Giant and Con mines (Yellowknife, Northwest Territories), Cheney et al. (2020) also observed As levels in 18 of 20 lakes that exceeded the ISQG in pre-mining sediments due to local geology. Similar observations were also made by Galloway et al. (2015), where As concentrations were higher in surface sediments from a suite of lakes around Yellowknife. Importantly, after historical mining activities began around the 1860s in Nova Scotia, sedimentary concentrations of As and Hg exceeded the ISQGs by several fold at all four study lakes located downstream of tailing fields. For instance, during the mining era, the peak Hg concentration of 31 400 ng/g at Thomas was 185 times higher than the ISQG, and the As concentration of 2800 μg/g at Gegogan was 475 times higher than the ISQG. The magnitude of the increase in Pb and Zn was modest relative to As and Hg, but concentrations of these elements were below ISQG limits pre-mining and crossed the thresholds after mining activities began, and other human activities contributed substantial amounts of Pb to the environment (Dunnington et al. 2020). The exceedance of multiple ISQGs to varying degrees shows that the sediments of lakes that received mine tailings were polluted by a variety of metal(loid)s that likely resulted from differences in ore sources, mineral processing, and transport pathways.

The overall patterns in the PEC-Q values at all lakes tracked trends in bulk sedimentary concentrations of As and Hg because these elements were present at substantially higher levels than other elements included in the PEC-Q calculations. Concomitant with sedimentary metal(loid) increases, the PEC-Qs also increased markedly after mining activities began and reached levels that are likely to cause long-term biological effects (PEC-Q $>$ 2). Our observations are consistent with sedimentary PEC-Q trends reported from Yellowknife, where atmospheric deposition of As and other metal(loid)s from local gold mining led to more than half of the 20 study lakes crossing the biological effects probable threshold (Cheney et al. 2020). At Gegogan Lake, 9 of 14 sediment samples from the pre-mining period also had PEC-Q $>$ 2, consistent with the higher background levels of sedimentary As and Hg. Cheney et al. (2020) also observed PEC-Q $>$ 2 in the pre-mining samples of four lakes near the former Giant Mine in Yellowknife. While PEC-Q values help to predict the toxicity of sediments to exposed biota, the measurements should be compared with past biological and mineralogical changes to assess potential cause-and-effect relationships between legacy pollution and biological recovery.

## Implications for ecosystem recovery and aquatic biota

Nearly eight decades after historical gold mining activities ended in Nova Scotia, the average sedimentary concentrations of As and Hg have decreased from peak mining-era concentrations at most of the downstream lakes. Notable exceptions to geochemical recovery include As in Lake Charles and Hg in Gegogan Lake, which suggests that post-depositional mobility and continued inputs from the watershed or contaminated sediments influence lake-specific recovery patterns. For example, the tailing fields near Goldenville and Montague gold mining districts have been used by off-road vehicles, which has accelerated the erosion of tailings that are moved into aquatic ecosystems downstream (Parsons et al. 2012). ISQG-listed metal(loid)s (Zn, Cu, Pb, Cd, and Cr) are also higher in the recently deposited sediments relative to the pre-mining period, which may also reflect other land-use change due to urbanization combined with regional atmospheric deposition (Dunnington et al. 2020; Clark et al. 2021). These higher levels of metal(loid)s are reflected in the continued exceedance of ISQGs for individual elements and PEC-Q levels that are indicative of probable or possible biological effects. Our results suggest that there are signs of geochemical recovery at these lakes, but elevated sedimentary metal(loid) levels suggest that other factors are impeding recovery. For example, it is possible that Gegogan Lake may be continually receiving mine waste from the Goldenville tailing fields given that current Hg concentrations continue to be as high as they were during the mining era (Wong et al. 1999). Unlike Hg, As levels have decreased in Gegogan sediments, and the difference in the post-mining trends between these two elements could be associated with the heterogenous element distribution of the historical Goldenville gold-mine tailings field (Parsons et al. 2012). It is also possible that certain sections of the tailing fields may have higher levels of Hg relative to As (Parson et al. 2012). This should be an important consideration when developing future site-specific remediation plans for Gegogan Brook and Gegogan Lake.

An increase in As was observed in the most recent sediments of Lake Charles, and a similar trend was also observed in the sediment core from Loon Lake, although not nearly the same magnitude as at Charles (Clark et al. 2021). Arsenic, a redox-sensitive element, is known to be mobile in lake sediments after deposition. Arsenic enrichment in surficial lake sediments has been observed in severely As-polluted lakes around Yellowknife, Northwest Territories (Andrade et al. 2010; Galloway et al. 2018; Schuh et al. 2018; Van Den Berghe et al. 2018). Arsenic mobility and cycling in lake sediments are partly controlled by redox conditions near the sediment–water interface and in shallow pore waters, which can facilitate reductive dissolution of As sorbed to iron and/or manganese oxy(hydr)oxides (Belzile and Tessier 1990; Smedley and Kinniburgh 2002; Couture et al. 2010; Van Den Berghe et al. 2018). Furthermore, labile organic matter can serve as a substrate for microbial growth and mediate authigenic pre-

cipitation of arsenic-sulphides that can be an effective means of sequestering As (Galloway et al. 2018; Van Den Berghe et al. 2018). In the case of Lake Charles, we suspect that hypolimnetic anoxia develops during thermal stratification in late summer. Furthermore, primary production has increased at both Charles and Loon lakes in recent years in response to climate-mediated changes and urbanization (Clark et al. 2021). The decay of algal matter consumes oxygen, which, coupled with a stratified water column, can promote the development of anoxia at the sediment–water interface and in shallow pore waters. In fact, the highest PEC-Q measurement (19) in this study was calculated for the surface sediments of Lake Charles, as they contained the highest As levels. Mobility of As in the sediments and high PEC-Q measurements at these urban lakes have implications for delaying aquatic recovery, particularly the recovery of benthic habitats where sediment-dwelling organisms in the profundal zone are exposed to contaminants.

Ecosystem recovery is an important goal in environmental remediation programs, and paleolimnological studies provide insight into pre-disturbance conditions that can be used to identify potential recovery targets (Smol 2019). However, lake ecosystems and their catchments are also impacted by multiple stressors, which present certain challenges to identifying realistic recovery targets and determining how long it will take to reach a "recovered" state, particularly within the context of present climatic changes, including extreme weather events. In Nova Scotia, historical gold mining occurred for nearly eight decades, and eight more decades have passed since the cessation of major mining activities, yet sedimentary metal(loid) concentrations have not returned to background values. This suggests that complete geochemical recovery may not be a realistic target in severely polluted lakes. As recorded in our urban lakes, changing land use and climatic conditions can independently or synergistically influence geochemical recovery trajectories. Even in the absence of mining pollution, land-use changes related to urbanization, such as residential development and road construction, also have the potential to increase metal(loid) delivery to local lakes (Sivarajah et al. 2020), as observed at Loon Lake (Clark et al. 2021). In Loon Lake, all seven metal(loid) concentrations have increased, and six of them have crossed the ISQG guidelines, while PEC-Q is also moving towards possible biological effects threshold. The cumulative impacts of urbanization and climate-mediated limnological changes (e.g., longer seasonal duration of thermal stratification, extended growing season, and greater primary production) can promote the development of low oxygen conditions in bottom waters, and thus create conditions promoting the mobility of redox-sensitive metal(loid)s (Palmer et al. 2019). Therefore, sediments may become sources of metal(loid)s in the water column rather than sinks in deeper lakes. Additionally, in remote Arctic lakes, changes in organic matter quantity and quality have also shown to influence geochemical cycling of metal(loid)s (Outridge et al. 2019). Extreme precipitation events also have the potential to weather and move significant quantities of metal(loid)s from tailing fields into downstream lakes, which can further impede geochemical recovery. Hence, it is important to implement monitoring programs to track metal(loid) movement from land to water and within lakes, given expectations of climatic changes.



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### Data availability

Data generated or analyzed during this study are available from the corresponding author upon reasonable request.

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The authors declare there are no competing interests.

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# Supplementary material

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