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Have natural lake expansion and landscape inundation resulted in mercury increases in flooded lakes of the Great Slave Lowlands (Northwest Territories, Canada)?

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Abstract The inundation of terrestrial vegetation following landscape flooding is an important potential source of mercury to aquatic ecosystems, and may modify mercury cycling, such as through increased methylation. In the Great Slave Lowlands of Canada's Northwest Territories, remarkable landscape flooding has occurred over the recent past, which is the most notable in at least the last several centuries. The potential for this flooding to increase inorganic mercury flux to the lakes of the region has not yet been explored. In this study we used sediment cores from five lakes experiencing a range of recently documented lake expansion to test whether inundation of terrestrial areas has increased the total mercury concentrations in sediments, and resulted in increased total mercury flux. Increases in sedimentary mercury concentrations and fluxes in sediment cores from the expanding lakes were relatively small and within the range of non-expanded systems, suggesting that, to date, flooding has not resulted in major total mercury enrichment, unlike in experimental and natural reservoir impoundments. The potential for increased methylation of existing inorganic mercury following expansion was not explored in this paper because methylmercury is dynamic in sediments and does not preserve well, but is an important consideration for future work.

Keywords Climate change · Contaminants · Flooding · Lake sediments · Paleolimnology · Mercury

Introduction

Increased contaminant exposure of ecosystems through anthropogenic inputs represents a major stressor globally. Mercury (Hg), is a naturally occurring element, released in large quantities by human activities beyond the historical, pre-industrial range. Because of the capacity for long-range transport of mercury in the atmosphere, this contaminant is of particular interest in remote northern regions, which may lack direct sources of anthropogenic pollution. Landscape changes have the potential to alter the movement and mobility of mercury, with concomitant impacts to ecosystems and foodwebs. As mercury

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stored in soils is primarily divalent Hg(II), changing redox conditions, and microbial activity in response to saturation of soils is an important control on total mercury mobility to surface waters, the methylation to bioaccumulating MeHg, and the formation of gaseous Hg^0, and thus its potential flux to the atmosphere (Poulin et al. 2016). Hydrological changes resulting in the inundation of terrestrial terrain, for example during the creation of reservoirs in boreal environments, have been shown to result in an influx of Hg to the newly created aquatic ecosystem in laboratory (Morrison and Thérien 1991), mesocosm (Hall and St. Louis 2004) and whole-lake settings (Hall et al. 2005; Bodaly et al. 2007). The role of large-scale inundation events, such as those associated with impoundment for hydroelectric generation, are well documented sources of Hg to aquatic ecosystems (St. Louis et al. 2004), which have been shown to be present in aquatic foodwebs at multiple trophic levels (Bodaly et al. 1984; Hall et al. 1998). Natural aquatic ecosystem expansion, for example due to beaver dam impoundment, has also been shown to result in enhanced mercury mobility (Roy et al. 2009). These findings have important implications for future mercury dynamics in northern landscapes, where climate change is resulting in shifts in lake hydrological regimes, including lake expansion and shoreline flooding in some instances (Carroll et al. 2011; Parsekian et al. 2011).

A dramatic example of northern lake expansion and landscape inundation linked to recent climate change can be found on the northwest shore of Canada's Great Slave Lake, in the Great Slave Lowlands and Plains ecoregions of the Northwest Territories (Fig. 1). This landscape exhibits little relief, and has a high proportion of water cover, being dominated by wetlands, small ponds, and many large, shallow lake ecosystems (Ecosystem Classification Group 2009). The region also contains the Mackenzie Bison Sanctuary, established in 1963 as habitat for an ecologically important population of wood bison (*Bison bison athabascae*), a distinct sub-population, and North America's largest land mammal (Larter et al. 2000). A recent investigation of changes in the Great Slave lowlands region based on Landsat satellite imagery showed that the proportion of the landscape occupied by water in a 10,000 km^2 area (including the majority of the bison sanctuary) nearly doubled between 1986 and 2010, and that this increase was correlated with climatic variables (Korosi et al. 2017). While the whole of the Great Slave Lowlands and Plains has become wetter recently, the response is quite heterogeneous, with some lakes exhibiting extensive (> 800%) expansion, while others showed a more muted response (Korosi et al. 2017). Lake sediment records were used to extend the record of lake area changes beyond the observational record, and showed no other periods of lake expansion as large and persistent as those occurring in the last ~ 25 years have occurred in at least two centuries (Korosi et al. 2017).

Landscape flooding in the Mackenzie Bison Sanctuary is inundating terrestrial vegetation on the margins of lakes and ponds, drowning the sedges and grasses utilized as the preferred forage by bison, and potentially driving them out of the sanctuary in search of other food sources (Korosi et al. 2017). This widespread flooding of terrestrial material may also be resulting in an influx of mercury to the lake ecosystems, similar to the flooding that occurs with natural impoundments, such as in beaver ponds (Roy et al. 2009), and analogous to hydroelectric impoundments (Teisserenc et al. 2014). Satellite and field-based AQ1 observations have shown that the recent flooding is primarily refilling old lake basins, though the lakes have not been as large as their current area in at least the last 200–300 years (Korosi et al. 2017). It is conceivable that, in this hydrologically dynamic landscape where seasonal increases in water levels may have always occurred, but the recent, persistent lake expansion is likely due to climate-related changes, flooded lakes may record a less clear response in mercury change compared to beaver impoundments and reservoirs. We explore this scenario using a paleolimnological approach in a strategically selected series of lakes in this rapidly changing region (Fig. 1). As lake sediments represent a faithful record of changes in total mercury over time (Lockhart et al. 2000), we reconstructed the recent history of mercury accumulation in order to test the hypothesis that landscape flooding in the Mackenzie Bison Sanctuary has resulted in increased mercury concentrations in recently expanded lakes.

Study site

The study region is located within the Great Slave Plains High Boreal, and Great Slave Lowlands Mid Boreal (Level IV) ecoregions (Ecosystem Classification Group 2009). The region has limited relief, is

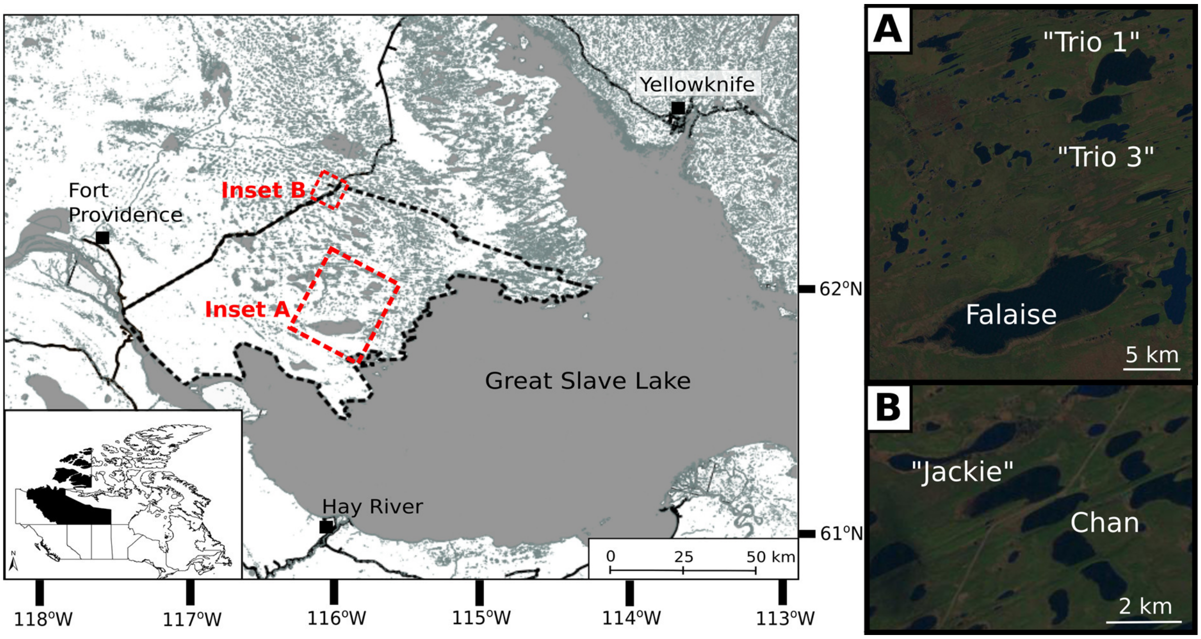


Fig. 1 (a) Map of the study area in the Great Slave Lowlands and Plains region of Canada's Northwest Territories. The area designated as the Mackenzie Bison Sanctuary is delineated with a black dashed line. Landsat images of the study lakes from 2010 are presented in (a, b)

dominated by wetlands, with a multitude of small marl ponds, and a number of large, shallow lake ecosystems. Bedrock is primarily Cambrian to Devonian aged limestone, sandstone, and dolomite. Surficial geology is composed of a thick mantle of glacial and postglacial deposits greater than 80 m in depth (Craig 1965), composed of tills and glaciolacustrine deposits from glacial Lake McConnell. Soils are variable throughout the region, including Organic soils in wetland areas, coarse-textured Brunisolic soils especially near beach ridges, and Gleysols adjacent to ponds. Permafrost in the region is sporadic discontinuous, with organic Cryosols associated with peat plateaus (Ecosystem Classification Group 2009).

Five study lakes were selected in the Great Slave Plains and Lowlands ecoregions for analyses (Fig. 1, Table 1). Four of the lakes are located in the core of the Mackenzie Bison Sanctuary (MBS) and to the east of NWT HWY 3, which forms the western edge of the MBS. The fifth lake, "Jackie Lake" (unofficial name) is located on the periphery of the MBS to the west of NWT HWY 3. In order to assess the potential for heterogeneous responses in mercury change associated with varying lake size, the lakes range in area from the largest lake in the MBS, Falaise Lake, to Chan Lake, a small lake located along NWT HWY 3 (Table 1). In order to test our prediction that landscape flooding has resulted in increased mercury accumulation, lakes that have exhibited recent expansion are compared to sites that have not expanded in the recent past (Table 1; Korosi et al. 2017).

Materials and methods

Sediment cores were collected from the deepest location in each of the five lakes through the late-winter ice in March of 2012, using a Glew-type gravity corer (Glew 1989). Sediment cores were extruded into 0.5 cm intervals using a Glew-type vertical extruder (Glew 1988), and kept < 10 °C during transport and prior to analyses. The sediment cores were the same as utilized for the analyses presented in Korosi et al. (2017), which included determination of total organic carbon, nitrogen content, C:N elemental ratio, and ^{13}C and ^{15}N stable isotope analyses. Selected intervals were prepared for ^{210}Pb and ^{137}Cs-based radioisotopic dating using gamma spectroscopy, with details presented in Korosi et al. (2017), with the constant rate of supply (CRS) model used for sediment age

Table 1 Study lake locations, recent surface area and Landsat-derived area change (%) between 1986 and 2010 (from Korosi et al. 2017)

Lake	Latitude (°N)	Longitude (°W)	2010 surface area (ha)	Percent change 1986–2010 (%)
Falaise	61.47642	116.15280	5637.6	+ 824
“Trio 1”	61.64026	116.05184	1036.4	+ 462
“Trio 3”	61.59762	116.07063	306.4	+ 20
“Jackie”	61.89678	116.55987	151.0	+ 313
Chan	61.89079	116.54170	66.2	+ 51

Lake names in quotation marks are unofficial

determination (Fig. 2). Modelled errors associated with the dates corresponding to the observed recent lake expansion were low (less than 5 years for dates since 1950). Freeze-dried and homogenized samples were analyzed for total mercury by thermal decomposition with gold trap amalgamation and cold vapour atomic absorption spectrometry (CV-AAS) using a Nippon Instruments SP-3D mercury analyzer with a detection limit of 0.01 ng per sample size. Required sample masses ranged from 17 to 30 mg (dry weight). Measurement accuracy was estimated by running blanks and calibrated with MESS-3 (91 ± 9 ng g^{-1}, Natural Resources Canada) as reference material, every 10 samples. Results of mercury concentration analyses were determined per unit mass dry sediment weight, as well as per unit mass of organic carbon. As the concentration of organic carbon throughout the sediment cores did not change significantly (Korosi et al. 2017), the profiles of total mercury per unit organic carbon did not differ from per unit dry weight, and as such only the latter are discussed below. Mercury fluxes to lakes were estimated based on the modelled sedimentation rate determined via the CRS model utilized for sediment chronology development for the sediment cores for all five lakes (Fig. 2).

Results

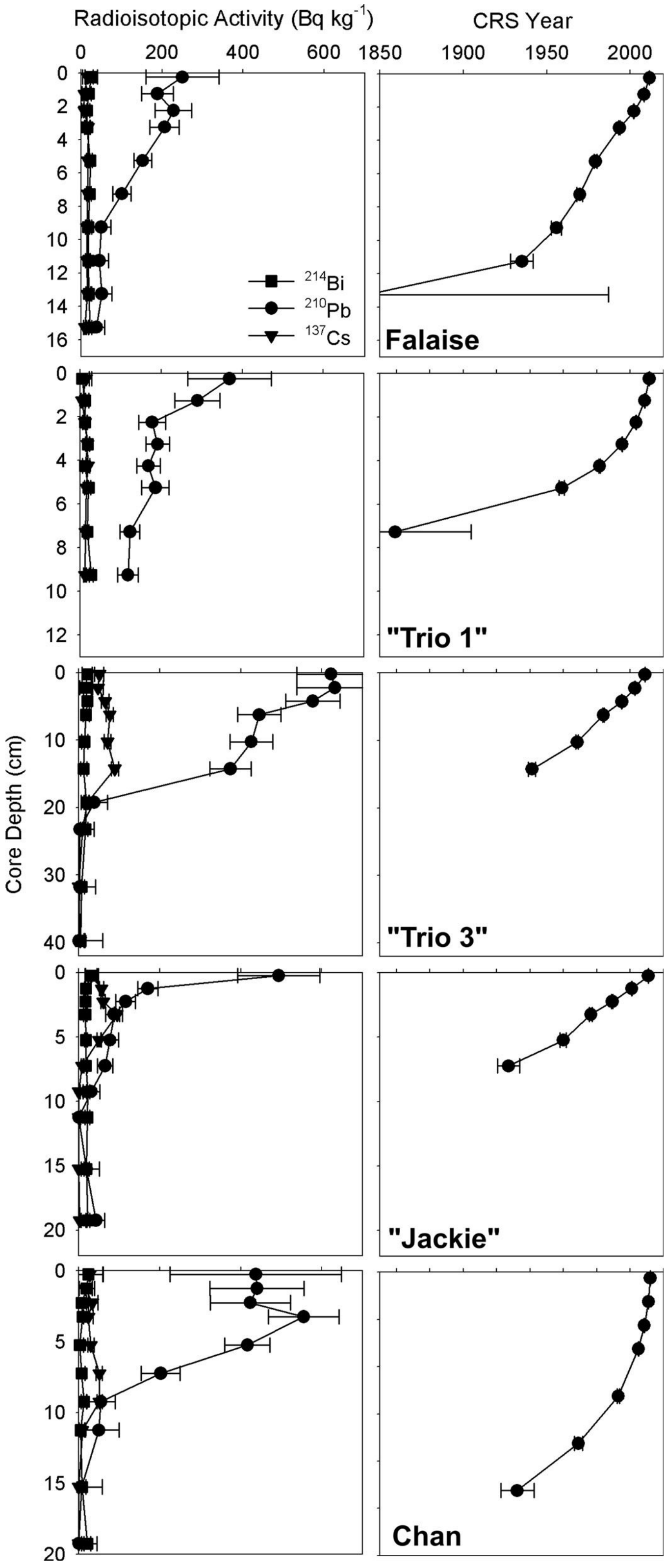
Changes in sedimentary mercury concentrations

The concentration of total mercury (THg) in the sediment core from Falaise Lake showed only small variations over time (Fig. 3). Over the recent past, a relatively small magnitude increase in THg was observed since the mid-1980s, which was strongly correlated to the marked (> 800%) increase in lake area (Spearman rank correlation, $n = 7$, $r_s = 0.94$, $p < 0.001$) (Fig. 3). The highest concentration recorded in Falaise Lake, $\sim$ 33 ng g^{-1} in the surface sediment interval, was well below the sediment quality guidelines for the protection of aquatic life set at 170 ng g^{-1} by the Canadian Council of Ministers for the Environment (CCME 1999) The THg concentration in “Trio 1” Lake increased slightly from the earliest part of the record ($\sim$ 1850) until $\sim$ 1950, after which it decreased, despite the increase in lake surface area observed for this system (Fig. 4a). “Trio 3” Lake recorded the highest magnitude THg concentration in the sediment cores in this study (Fig. 4b). The concentrations of THg in “Trio 3” decreased after $\sim$ 1975, during which time the lake area exhibited some fluctuation, but no directional change (Fig. 4b). In “Jackie” Lake, Hg concentration exhibited a rapid increase after $\sim$ 1990 from $\sim$ 25 to $\sim$ 45 ng g^{-1}, which tracked closely the timing and direction of lake area increase inferred from Landsat imagery (Fig. 5a). In Chan Lake, Hg concentration showed several small magnitude changes throughout the period represented by this sediment core, which were not related to lake area, which increased slightly ($\sim$ 51%) over the period of record (Fig. 5b). Correlations were not conducted for the latter four lakes due to a low number of samples that had overlapping area estimates and mercury determinations. All sedimentary total mercury values were below the CCME sediment quality guidelines.

Changes in mercury flux

The total mercury flux to the sediment in Falaise Lake decreased after $\sim$ 1940, until the onset of lake

Fig. 2 Radioisotopic activity and constant rate of supply modelled sediment age, along with associated error, for the five sediment cores from the Great Slave Lowlands and Plains region



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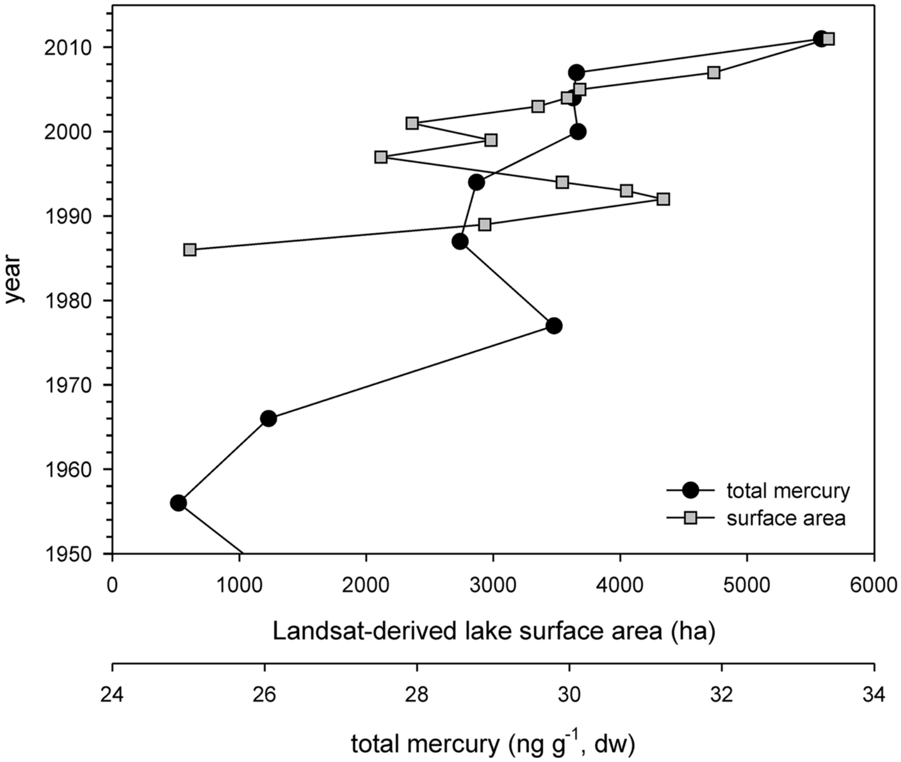

Fig. 3 Sedimentary record of total mercury concentration (per gram dry sediment weight) and Landsat-derived lake surface area for Falaise Lake, Northwest Territories, Canada

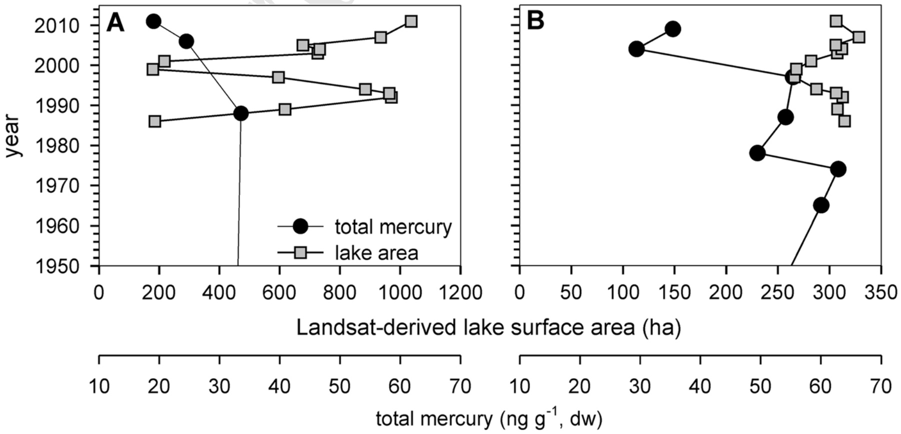

Fig. 4 Sedimentary record of total mercury concentration (per gram dry sediment weight) and Landsat-derived lake surface area for **a** Trio 1 and **b** Trio 3 lakes, Northwest Territories, Canada. The "Trio Lakes" are located close together, but have drastically different histories of recent expansion (Fig. 1, Table 1)

expansion in the late 1980s, after which it increased through to the top of the core (2010) (Fig. 6a). The THg flux increased from ~ 1800 ng m^{-2} year^{-1} in ~ 1980 to ~ 3000 ng m^{-2} year^{-1} in the surface sediments (Fig. 6a). Mercury flux in "Trio 1" Lake remained low throughout the record, and increased from ~ 1930 until ~ 1990, after which it was constant until the uppermost (surface) sediment interval, where it decreased (Fig. 6b). The sedimentation rate in Lake "Trio 3" was the lowest of the sediment cores modelled in the study, and similarly THg flux was also low throughout the period for which sedimentation rate was estimated (since ~ 1960) (Fig. 6c). During this period the flux of mercury did not change markedly (Fig. 6c). Mercury flux and sedimentation rate in "Jackie" Lake increased rapidly from the bottom of the core, which continued over the recent period of rapid lake expansion (Fig. 6d). The THg flux in "Jackie" Lake was of the highest magnitude of the sediment cores from the five study

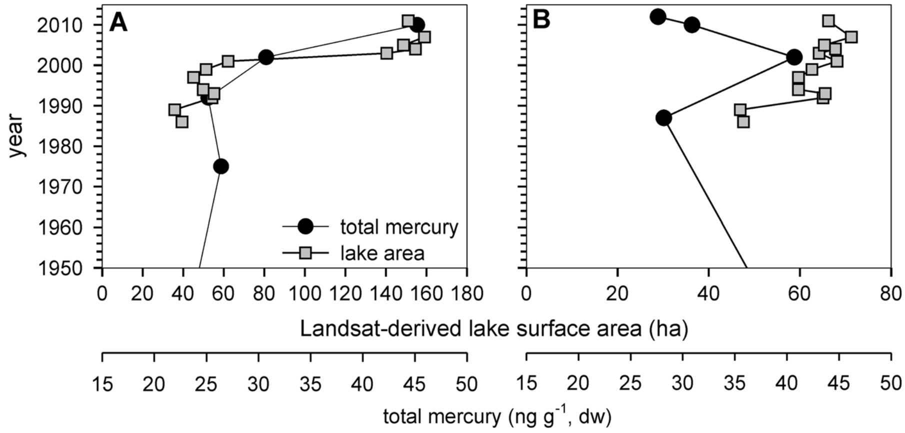

Fig. 5 Sedimentary record of total mercury concentration (per gram dry sediment weight) and Landsat-derived lake surface area for **a** "Jackie Lake" (unofficial name) and **b** Chan Lake, Northwest Territories, Canada

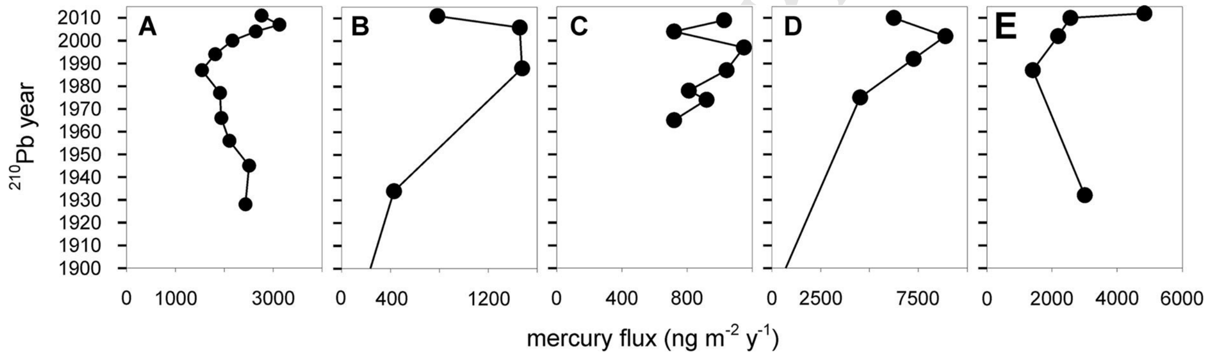

Fig. 6 Total mercury flux to the sediments (per gram dry weight, per square meter, per year) for **a** Falaise, **b** "Trio 1", **c** "Trio 3", **d** "Jackie", and **e** Chan lakes, in the Mackenzie Bison Sanctuary, Northwest Territories, Canada

lakes, with the surface sediments recording a mercury flux of $\sim$ 7500 ng m^{-2} year^{-1} (Fig. 6d). In Chan Lake THg flux decreased slightly between $\sim$ 1930 and the late 1980s, after which it increased through to the surface sediment intervals, consistent with the small magnitude lake area increase (Fig. 6e). The mercury flux estimates for the smaller lakes were of a higher magnitude than the larger study lakes in the study (Fig. 6).

Discussion

Is landscape flooding releasing mercury to lakes?

The potential for mercury stored in vegetation and soils surrounding the lakes to be released following flooding has been well documented (St. Louis et al. 2001, 2004; Roy et al. 2009; Teisserenc et al. 2014). However, most studies on mercury release in larger-scale lake ecosystems have been due to anthropogenic impoundment for hydroelectric generation, and not due to natural landscape flooding. Natural impoundment, such as through the bio-engineering activities of beavers, are also a source of mercury to aquatic ecosystems (Roy et al. 2009), though the magnitude of impoundment from these actions is smaller. The recent lake area changes of Falaise Lake are correlated with increased total mercury, though the magnitude of the increase is small (Fig. 3). Similarly, total mercury in "Jackie" Lake, a much smaller ecosystem, has increased coincident with recent lake expansion (Fig. 5a). In "Trio 1" Lake, which has undergone the second greatest increase in surface area of the study lakes, total mercury has decreased over the recent past (Fig. 4a). The highest overall concentration of total mercury in the sediments of the five study lakes was recorded in Lake “Trio 3” which has not expanded recently (Fig. 4b). In Chan Lake, which has increased in surface area moderately compared to the other lakes (51%), there was no concurrent, directional increase in total mercury concentration in the sediments (Fig. 5b). Sedimentary mercury concentrations found throughout the profiles of all five sediment cores analyzed in this study are well below the sediment quality guidelines for the protection of aquatic life set at 170 ng g^{-1} by the Canadian Council of Ministers for the Environment (CCME 1999), and the range of values are similar to those found in other studies of subarctic lakes in Canada (Lockhart et al. 1995, 1998; Muir et al. 2009; Brazeau et al. 2013).

Increases in fluxes of mercury to the sediment correspond relatively closely to the timing of the recent lake expansion in Falaise and “Jackie” lakes, as well as in Chan Lake, which increased marginally in area. Despite these increasing mercury flux trends, the absolute values recorded are low, at least an order of magnitude less than lakes in northern Scotland, for example (Yang and Rose 2003). The highest flux values (Chan and “Jackie” lakes) were similar to the lowest values documented for undisturbed sites from AQ2 northern Canada (Lockhart et al. 1995), western Greenland (Bindler et al. 2001) and Scandinavia (Renberg 1986; Verta et al. 1989). Both the mercury concentration and flux data suggest that landscape flooding in the Great Slave Lowlands region has not resulted in the release of large amounts of total mercury to the sediments of impacted lake ecosystems.

The variable trend of mercury change, as well as the small magnitude of increase in both concentration and flux in those sites that are recording increases, contrasts with the well-documented enrichment of mercury in lake water and sediments following reservoir impoundment (Teisserenc et al. 2014). There are several reasons why this may be the case. Though the persistence of recent expansion is unique over the last several 100 years, these shallow basins are undoubtedly wetted periodically during snowmelt or periods of heavy precipitation. This would result in relatively consistent and regular leaching of mercury into the lake, as opposed to soils in reservoir impoundments that have been building up inorganic mercury over long timescales, which are then incorporated into lakewater and sediment rapidly, and in bulk. In addition, the composition of the soils could result in conversion to gaseous Hg^0, which would be lost to the atmosphere, and not transported to the lake sediments (Poulin et al. 2016), and thus detailed sampling of local soil properties would be helpful for understanding mobilization and transformation properties. A second confounding factor may be the regular occurrence of fire in this region, which burns soil organic matter reserves and depletes soil mercury concentrations, also through release of gaseous mercury (Mailman and Bodaly 2005; Friedli et al. 2003). The burning of land prior to impoundment has been suggested as a mechanism for decreasing methylmercury production, through the loss of inorganic mercury (Mailman et al. 2006). Fire regime is a critically important control on vegetation structure within subarctic boreal regions (Johnson 1979), and its influence may extend to landscape mercury cycling as well. Fire return intervals are projected to decrease in the Great Slave region as a result of climate warming (Boulanger et al. 2014), and historical fire frequency may be higher in the Northwest Territories region than in eastern Canada where most of the studies of impoundment-driven mercury enrichment have occurred (Bergeron et al. 2004).

Total organic content (TOC) in the sediments of all of the lakes is relatively low ∼ 20–30%, and did not change as a result of recent flooding (Korosi et al. 2017). Non-peat soils in the Great Slave Lowlands region, derived from Glacial Lake McConnell sediments (Smith 1994) tend to have a high proportion of clay and are relatively low in organic carbon (Ecosystem Classification Group 2009), in comparison to nearby areas such as the Tathlina and Kakisa lake watersheds to the west and south (TOC averages 40–50%), where strong increases in mercury have been reported (Korosi et al. 2015). The concentrations of mercury in uncontaminated soil samples were measured at 55 ng g^{-1} in both the B and C horizons of brunisolic samples from near Fort Providence, less than 70 km from Falaise Lake (McKeague and Kloosterman 1974). This suggests Great Slave Plains and Lowlands soils derived from Glacial Lake McConnell sediments are likely naturally low in mercury. Importantly, the local distribution of sporadic permafrost in the region is not well documented. Frozen ground may play a role in influencing the movement of mercury in soils, by limiting mobility, and future thaw could result in altered mercury availability, especially during fire events (Turetsky et al. 2006).

It is important to note that throughout this study we have focused on reconstructing the history of total mercury changes in these lake ecosystems through sediment records. We have not reconstructed changes in methylmercury, the most toxic and bioaccumulative form of mercury, as historical trends cannot be reliably reconstructed from lake sediment records. There exists the potential for landscape flooding in the region to alter rates of methylation and demethylation, particularly if peatlands are inundated (Heyes et al. 2000), and increased deposition of mercury is not necessarily required for methylmercury to enter and accumulate in foodwebs (Bodaly and Fudge 1999). As changing methylation activity would not be obvious from sedimentary profiles of total mercury, further study is needed before we can definitively say that expansion has not altered the mercury cycling and availability in these lakes.

Conclusions

The recent, extensive landscape flooding that has occurred in the Great Slave Lowlands, including the Mackenzie Bison Sanctuary, resulted in increases in total mercury and mercury flux in the sediments from two recently flood lakes, though the magnitude of these increases was small ($\sim$ 25–33 ng g^{-1} in Falaise Lake, and $\sim$ 25–45 ng g^{-1} in “Jackie” Lake). In another flooded site, no increase in total mercury was observed. Total mercury concentrations in sediments in all of the lakes studied in the region are well below Canada’s sediment quality guidelines, set at 170 ng g^{-1}. The potential for landscape flooding to have resulted in changes in mercury cycling, including altered rates of methylation, remains an important knowledge gap that requires future study.

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