



Effects of a century of mining and industrial production on metal contamination of a model saline ecosystem, Great Salt Lake, Utah

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ABSTRACT

Effects of mining and metals production have been reported in freshwater lake sediments from around the world but are rarely quantified in saline lake sediments, despite the importance of these lake ecosystems. Here we used dated sediment cores from Great Salt Lake, Utah, USA, a large saline lake adjacent to one of the world's largest copper mines, to measure historical changes in the deposition of 22 metals. Metal concentrations were low prior to the onset of mining in the catchment in 1860 CE. Concentrations of copper, lead, zinc, cadmium, mercury, and other metals began increasing in the late 1800s, with peaks in the 1950s, concomitant with enhanced mining and smelting activities. Sedimentary metal concentrations in the 1950s were 20–40-fold above background levels for copper, lead, silver, and molybdenum. Concentrations of most metals in surficial sediments have decreased 2–5-fold, reflecting: 1) storage and mineralization of sedimenting materials in a deep brine layer, thereby reducing metal transport to the sediments; 2) improved pollution control technologies, and; 3) reduction in mining activity beginning in the 1970s and 1980s. Despite reductions, concentrations of many metals in surficial sediments remain above acceptable contamination thresholds for aquatic ecosystems with migratory birds, and consumption advisories for mercury have been placed on three waterfowl species. The research also highlights that metal deposition in saline lakes is complicated by effects of hypersaline brines and deep-water anoxia in regulating sediment redox and release of metals to surface waters. Given the importance of saline lakes to migratory birds, metals contamination from mining and metals production should be a focus of saline lake remediation.

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1. Introduction

Mining and industrial activities frequently release toxic metals to lakes directly through waterways (Moore et al., 1991) and indirectly via atmospheric deposition (Nriagu et al., 1982). Some metals biomagnify through food webs, causing harm to the biota and humans who consume aquatic organisms (Suedel et al., 1994). Historical patterns and effects of metal contamination have been recorded globally in the sediments of many aquatic systems (e.g. Child et al., 2018; Ek and Renberg, 2001; Jin et al., 2013; Schuh et al., 2019). While the chronology and magnitude of metals contamination has been recorded for freshwater lakes and estuaries (e.g.

Gawel et al., 2014; Leppänen et al., 2017; Thienpont et al., 2016), sedimentary studies are seldom done in saline lakes (e.g. Naftz et al., 2000).

Metal contamination in saline lakes is of concern because these lakes are important habitats for migratory waterfowl (Ashbaugh et al., 2018; Conover and Vest, 2009; Wurtsbaugh et al., 2011). In some cases, metal contamination can be reduced by volatilization (Diaz et al., 2009b) or by deflation of aerosols and dust particles from exposed lake beds or shorelines (Erdringer et al., 2004; Goudie, 2014); however, lack of surface outflow in hydrologically-closed (endorheic) basins may result in greatly elevated levels of metals in water and sediments (Javor, 1989). Further, many saline lakes have high solute concentrations (e.g., sulfate) which influences metal solubility, chelation, sedimentation and permanent sequestration (Blomqvist et al., 2004; Hamilton-Taylor and Davison, 1995). Hypersaline lakes may also lack burrowing invertebrates that

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otherwise can remobilize metals and oxidize adjacent sediments, thus complicating interpretation of historical pollution patterns (e.g. Chaillou et al., 2003).

Saline lakes may be particularly influenced by industrial and mining activities, as terminal basins are often the main recipient of mining wastewater in arid regions. For example, the Great Salt Lake (GSL), Utah USA, is adjacent to major mining and smelting activities, and a large industrial metropolitan area (Hughes, 1990). Mining for silver, lead and gold began in the nearby Wasatch Mountains ca. 1863 (Ege, 2005; McPhee, 1977; Varley et al., 1921), while the Bingham Canyon Mine of the Oquirrh Mountains (Kennecott Mine in Fig. 1), which is historically one of the world's largest producers of copper and other metals, opened in 1905. The smelter for this mine is now located on the south shore of GSL, while additional smelters were located in the Salt Lake Valley (Hughes, 1990). To the north in Idaho and Utah, large strip mines extract phosphate within the Bear River watershed (Mars and Crowley, 2003), the largest source of water for GSL (Mars and Crowley, 2003). These mines release selenium (Hamilton and Buhl, 2005). Metallurgical plants, railroad maintenance, petroleum refining and other urban manufacturing also release metals into the GSL and airshed (EPA, 2003), leading to potentially toxic levels of metal pollution in the largest lake in the western USA.

Sediment cores from mountain lakes east of GSL have documented regional metal contamination from smelting and industry and its effects (Kada et al., 1994; Moser et al., 2010; Reynolds et al., 2010), whereas the absence of fish in GSL has limited concerns about biological accumulation in its food webs. However, in recent years managers have found mercury, selenium and other metal contamination in the GSL aquatic food chain, potentially, affecting large populations of migratory waterfowl that use the lake as a staging habitat (Aldrich and Paul, 2002). Adams et al. (2015) documented elevated selenium and other metals in the brine shrimp and brine flies that are important food sources for aquatic birds using the lake (Aldrich and Paul, 2002; Roberts, 2013). Additionally, a survey of metals and toxins in the surficial sediments of GSL and its wetlands (Waddell et al., 2009) revealed metals concentrations frequently above Threshold Effects Concentrations (TEC) detrimental to freshwater organisms (MacDonald et al., 2000).

To help understand the history and magnitude of metal contamination in the Great Salt Lake, we compared sedimentary profiles of metals in cores from the south arm of the lake (Gilbert Bay, Fig. 1) to records of regional mining activity. Sedimentary analysis over a 200-year interval provided baseline estimates of metal deposition in the ecosystem, as well as evidence of the timing, magnitude, and recovery from mining activities. Additionally, surface sediments were analyzed from six other sites in the lake to assess the spatial gradient of current contamination. This research on the GSL is a model for defining the fate of metal contaminants in other saline lakes to help protect their aquatic food webs.

2. Study site and methods

2.1. Study site

The Great Salt Lake (Fig. 1), at 5200 km², is the largest lake in the western United States and the largest saline lake in North America. GSL is a hydrologically-closed basin in Utah, USA (41.04 N, -112.28 W), bordered on its eastern and southeastern shores by the Salt Lake City metropolitan area with over two million residents. The lake is impacted by industrial and municipal discharges, as well as by transportation causeways that divide the system into four large bays (Fig. 1).

Regional human population and accompanying industrial development has increased progressively from 8000 inhabitants in 1850 to 2,380,000 by 2020. In 1922, the Northwest Oil Drain was connected to a sewer canal to discharge wastes from refineries and other industries in Salt Lake City into Farmington Bay (Fig. 1), turning the canal into an EPA Superfund Cleanup Site (EPA, 2003).

The main body of GSL was bisected by a railway causeway constructed in 1959. Culverts and a breach allow exchange of water and salts between the two sub-basins; however, salinity of northern Gunnison Bay (1500 km²) is normally at saturation (ca. 330 g L⁻¹), whereas the southern arm, Gilbert Bay (2090 km²), varies from 60 to ~330 g L⁻¹ depending on meteorology, lake level and river diversions (Wurtsbaugh et al., 2017). At a lake elevation of 1280 m, the mean and maximum depths of Gilbert Bay are 4.4 and 10.1 m, respectively (Baskin, 2005). Gilbert Bay now receives an underflow of saturated brine from Gunnison Bay, so that, since 1959, half of the southern bay has been underlain by a deep brine layer (monimolimnion) with high concentrations of hydrogen sulfide (Johnson et al., 2019; Jones and Wurtsbaugh, 2014).

The two bays on the eastern side of GSL receive freshwater from the Wasatch and Uinta mountains, resulting in salinities that vary spatially and seasonally from freshwater to hypersaline conditions. Farmington Bay (~310 km²) in the southeast is shallow (mean depth 1.1 m at 1280 m above sea level) and is partially isolated from Gilbert Bay by automobile causeways constructed in 1952 and 1969. Northeastern Bear River Bay (212 km²) has a mean depth of 0.6 m at mean lake elevation and because of inflows from the Bear River, is a freshwater habitat for much of the year (Johnson et al., 2019).

Water quality conditions differ markedly among the bays in GSL (Wurtsbaugh et al., 2012). Farmington Bay is hypereutrophic (mean chlorophyll *a* 141 µg L⁻¹), whereas hypersaline Gilbert Bay (100–160 g TDS L⁻¹) is less productive (winter Chl. *a* ~70 µg L⁻¹), particularly during summer when grazing by abundant brine shrimp, *Artemia franciscana*, decreases chlorophyll concentrations to ca. 1 µg L⁻¹ (Wurtsbaugh, 1992). Brine fly larvae (*Ephydra* spp.), which primarily live on biostromes, are the only benthic metazoan (Wurtsbaugh et al., 2011). GSL brine is composed of 55% Cl, 28% Na, 8% SO₄, 4% Mg and 5% other salts (Sturm, 1980). Oxygen concentrations in Gilbert Bay are usually near saturation in the mixed layer, but the deep brine layer is anoxic below ~6.5 m. Surface water temperatures reach 24 °C in summer and can decline to 0 °C or below in winter. The pH is relatively stable around 8.1 in the mixed layer (Wurtsbaugh and Marcarelli, 2006). Total mercury and selenium concentrations in the mixed layer of Gilbert Bay are near 5 ng L⁻¹ and 0.5 µg L⁻¹, respectively, but mercury concentrations can reach >60 ng L⁻¹ in the deep brine layer (Jones et al., 2009; Jones and Wurtsbaugh, 2014). Water quality is better in Bear River Bay with mean Chl. *a* ~22 µg L⁻¹ (Wurtsbaugh, 2012) and total selenium levels of ~0.5 µg L⁻¹ (Naftz et al., 2009). Less is known about the water quality of Gunnison Bay, other than that salinities are usually near saturation and metazoans are rare (Baxter and Butler, 2020).

2.2. Sediment collection and chronology

Nine sites (Fig. 1, Table S1) were sampled successfully in 2009 and short (<75 cm) cores were collected with a Kajak-Brinkman gravity corer (Glew et al., 2001); however, metals were not analyzed from the core collected at Site 8 in Farmington Bay. Coring failed in Gunnison Bay due to an impenetrable, precipitated halite layer on the bottom. Cores were sectioned into 5-mm increments in the field using a Glew extruder (Glew et al., 2001). We provide dated stratigraphies from two sites (Gilbert Bay 3 and 4), and comparison of metal content from the upper 0–1 cm from eight

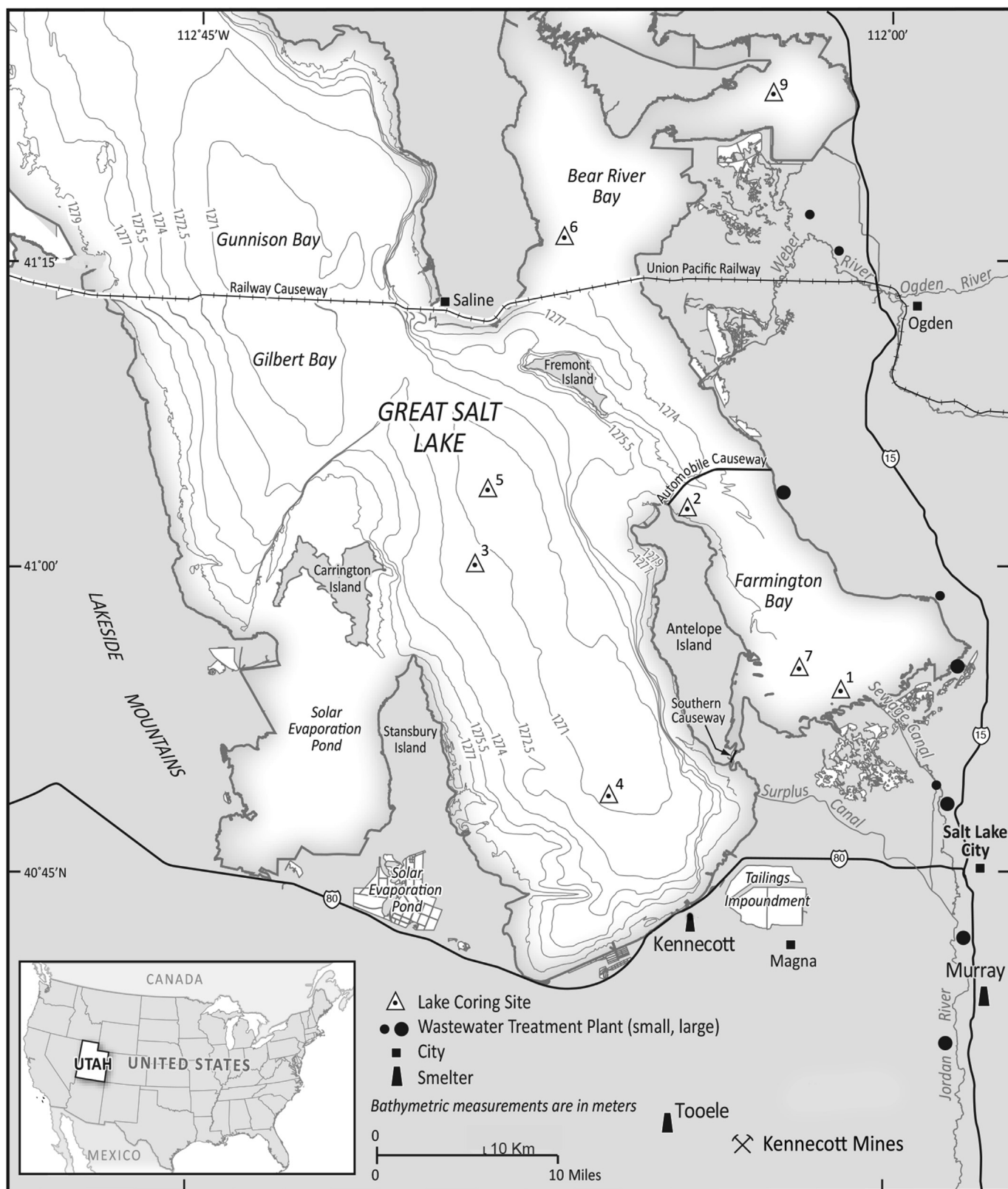


Fig. 1. Map of the Great Salt Lake showing the coring sites, causeways, and the locations of the Bingham Canyon porphyry copper mine (Kennecott Mines) and smelters.

sites to estimate spatial variation across GSL. The two cores from Gilbert Bay were collected from water depths >7 m where an anoxic deep brine layer was present. These regions were known to

have a high rate of sediment deposition (Oliver et al., 2009).

Estimates of radioisotope activity were performed on 12–14 samples per core at University of Regina following Bunting et al.

(2016). Sediment dating was based on ^{210}Pb activity measured by gamma spectrometry (Appleby and Oldfield, 1983; Schelske et al., 1994) using an Ortec High-Purity Germanium (HPGe) Coaxial Well Photon Detector System. ^{137}Cs activity was measured at 661.7 keV to identify the period of maximum fallout from atmospheric nuclear weapons testing and validate ^{210}Pb dates. Sediment age-depth relations were calculated using the CRS (constant rate of supply) model (Appleby and Oldfield, 1983).

2.3. Analytical methods for metals

Sediment subsamples from each core slice were oven-dried at 70 °C until a constant weight. Metal concentrations in the sediment samples were measured at the Utah Veterinary Diagnostic Laboratory (<http://www.usu.edu/uvdl/>) in Logan, Utah. The nitric acid leachable mineral concentrations in each slice were quantified using inductively coupled plasma mass spectroscopy (ICP-MS). Briefly, weighed sediment samples (~0.5 g) were digested at 90 °C in 10-ml trace mineral grade nitric acid for 4 h to estimate environmentally available metals (EPA, 1996). Digests were diluted 1:20 with 18.2 mOhm ultrapure water to provide a 5% nitric acid matrix and analyzed along with multiple standards. A quality control sample was analyzed with every five sediment samples and values that were not within 5% of targets were re-analyzed. A small number of samples for silver yielded values below detection limits. For these we assumed that concentrations were 80% of the detection limit because adjoining samples in the cores had values just slightly over the detection limit.

Sedimentary metal concentrations were expressed as mass g^{-1} dry matter, following correction for salt content in interstitial waters. The salt correction was particularly important in the uppermost 3–6 cm of the Gilbert Bay cores because unconsolidated sediments were rich in salts. Sodium (Na; % dry sediment) concentrations in the different sections of Gilbert Bay cores were highly correlated with water content (x; %):

$$[\text{Na}] = 0.3018x + 0.0142; R^2 = 0.946, n = 57$$

This equation, and the relationship between Na and total salt in Gilbert Bay water (0.30 g Na: 1.0 g salt; Sturm, 1980), was used to estimate the interstitial salt content. After correction, metal concentrations prior to 1860 were assumed to represent background levels and were used to calculate enrichment factors for metals in more recent strata. Here we focus on 11 metals that were either deposited in high concentrations or which are known to have high toxicity to biota.

2.4. Metal enrichment factors and criteria

We calculated metal enrichment indices to easily compare contamination histories among metals following Hokanson (1980) and Belzile et al. (2004). Maximum Enrichment Factors were calculated as the maximum sedimentary metal concentration relative to mean values recorded prior to industrialization (pre-1860). Current Enrichment Factors were estimated as the ratio of the mean concentration in the uppermost 1 cm relative to the same background values. All indices were estimated from ratios of each metal to that of aluminum, a conservative tracer of watershed erosion (Engstrom and Wright, 1984). This approach assumes most metals are derived from atmospheric, fluvial, or direct industrial sources, rather than eroded materials. Unlike other metals, sedimentary concentrations of erosion-indicators, aluminum and iron, did not change markedly in either core (Tables S2a, b). To provide some context for understanding the potential risks associated with concentrations of metals in GSL sediments, we have used

freshwater sediment Threshold Effects Concentrations (TEC) where some impacts on biota may be possible, and Probable Effects Concentrations (PEC) where impacts are likely (MacDonald et al., 2000).

2.5. Historical production of metals in Utah

Records of the annual production of commercial metals within the State of Utah were obtained from the Utah Geological Survey (2011). Most minerals were processed in Salt Lake Valley near the lake or at the Tintic/Mercur mining district 40–90 km south of GSL (Fig. 1; Ege, 2005). Although small amounts of mercury have been mined in Utah, residues in the lake likely originated from imported mercury used to extract gold and silver with the mercury amalgamation process, Hg released during smelting of deposits containing lead and zinc (Plumlee et al., 1996), and local industrial sources. Consequently, although not ideal, we have used the annual extraction of lead as a metric for mercury use and production in Utah. Although metal smelting is frequently associated with atmospheric deposition of acids, GSL's watershed is underlain primarily by limestone and is therefore well buffered (Ellis, 1986).

3. Results

3.1. Sediment chronology

At Gilbert Bay's core site 4, ^{210}Pb declined in a monotonic fashion to background levels, whereas at site 3, an intermediate peak was noted at ~6 cm depth, possibly representing a change in sedimentation rate (Fig. S1). Distinct peaks of ^{137}Cs were recorded in both cores but maximal specific activities of ^{137}Cs in the early 1960s were recorded only at site 4, consistent with high deposition of this isotope from open-air tests of atomic weapons (~1964). In contrast, maximal ^{137}Cs activity at site 3 appears to precede expected dates by 10–20 yr, either due to isotope migration, low sampling resolution (1 sample per ~15 years), or difficulty fitting ^{210}Pb regressions due to a mid-core peak of ^{210}Pb . At both sites 3 and 4, ^{137}Cs activities declined to near baseline values in surface sediments, suggesting relatively low levels of sediment mixing and post-depositional ^{137}Cs migration. Taken together, the radioisotopes suggest a more reliable chronology for site 4.

3.2. Metal contamination

3.2.1. Temporal variation of sediment contamination

Analyses of Gilbert Bay cores indicated that many metals (e.g., Cu, Hg, Pb, Zn) began increasing in the sediment record in the late 1800s and early 1900s concomitant with mining and smelting activities, before peaking in the mid-1950s and declining in more recently-deposited sediments (Fig. 2, Table S2a, S2b). For example, at both core locations Cu concentrations rose steadily from background levels of 11 $\mu\text{g g}^{-1}$ dry mass (hereafter $\mu\text{g g}^{-1}$) before the onset of commercial mining, to concentrations of 500 $\mu\text{g g}^{-1}$ when copper extraction reached 250,000 metric tons per year in the 1950s. Since the 1950–1960s, concentrations of copper in the sediment record have decreased markedly, although values in surficial sediments are still 10–20 times higher than background levels. Note that sedimentary Cu declines continue despite an overall increase in copper smelting in the past two decades.

Unlike most metals, concentrations of selenium and arsenic have not declined in recent years (Fig. 3). Background concentrations of Se were ~1 $\mu\text{g g}^{-1}$ at both sites and did not increase substantially until the 1930s–1950s. Since then, sedimentary Se has remained at a relatively constant concentration of 4–6 $\mu\text{g g}^{-1}$. Background levels of arsenic were ~13 $\mu\text{g g}^{-1}$ and began increasing

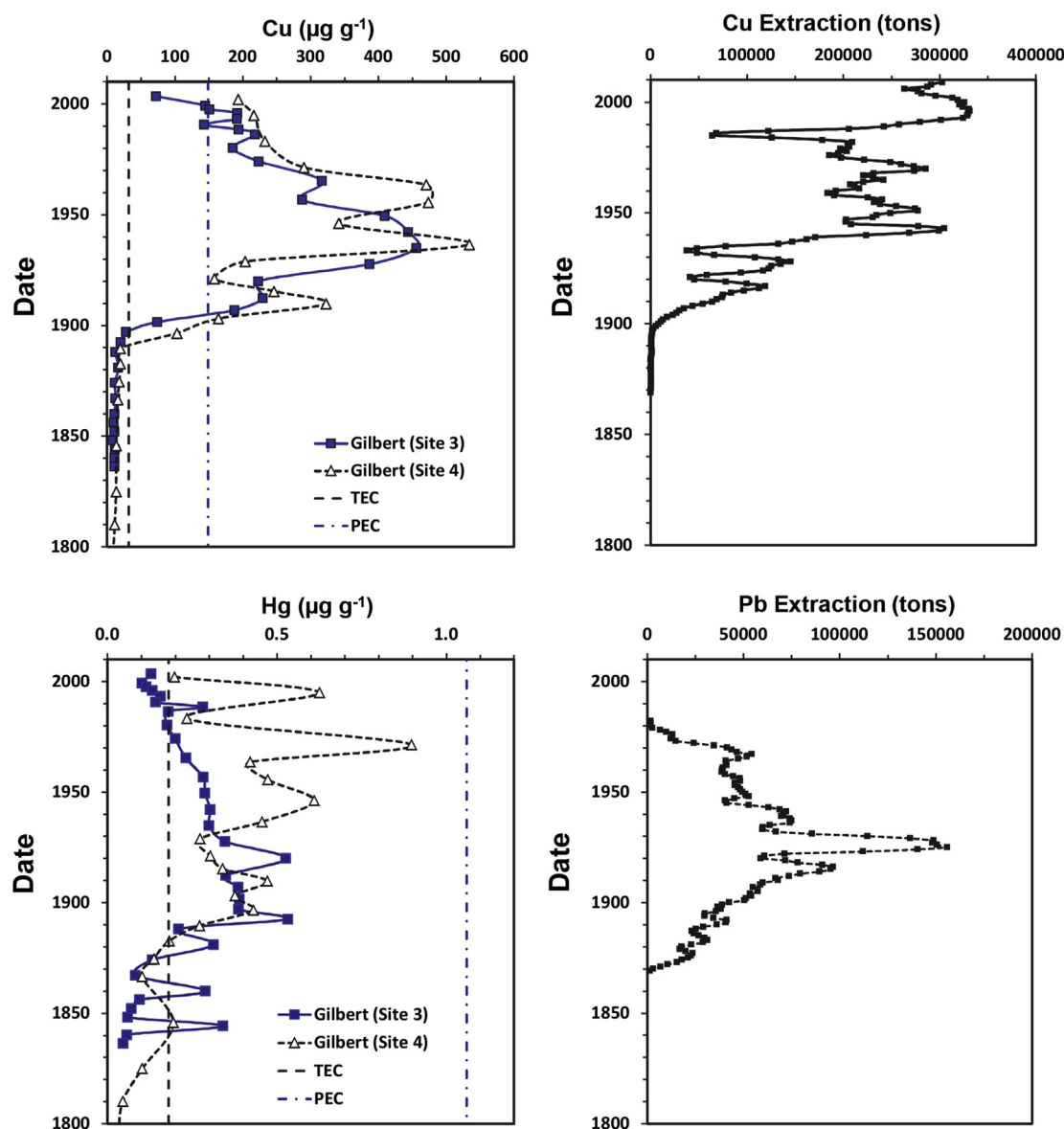


Fig. 2. Left frames: Concentrations of Cu, Hg, Pb and Zn in sediments of different ages from the two coring sites in Gilbert Bay. Concentrations are salt-corrected. Dotted vertical lines show Threshold Effects Concentrations (TEC; - -) and Probable Effects Concentrations (PEC; - · -) of sediments in freshwater ecosystems. Right frames: Metal production in Utah derived from [Utah Geological Survey \(2011\)](#) Utah Geological Survey. Note that although zinc was mined along with lead, it was not valued as a commodity until the early 1900s (K. Krahulec, Utah Geological Survey, personal communication).

during the late 1800s to $30\text{--}50\ \mu\text{g g}^{-1}$.

Enrichment factors for all metals increased from pre-industrial levels to peaks during the mid-20th century, and most declined in recent decades (Fig. S2). Silver and copper in the cores showed the highest enrichment factors, with values > 40 -fold above background, while those of Pb, Mo and Cd ranged from 15 to 35 and Hg was 12-fold above background (Fig. 4). In contrast, Se, Zn and As all had maximum factors less than 6. Current enrichment factors for six (As, Zn, Se, Hg, Sn, Cu) of the 11 metals are less than 50% of the historical maximum enrichment factors but all metals still have 3–18-fold higher concentrations than those recorded in the background era. Concentrations of Zn, As and Se have all decreased marginally, or not at all, compared to the historical maximum enrichment factors.

3.2.2. Spatial variation of sediment contamination

From an ecosystem health perspective, the most relevant metal concentrations are those at the sediment–water interface, as they are the ones that potentially diffuse into the water column and are subject to feeding upon by macroinvertebrates and birds. Spatial analysis of the surface sediments indicates that many metals in Gilbert and Farmington Bays remain above TEC established for freshwaters (MacDonald et al., 2000), whereas Bear River Bay sediments were less contaminated than other GSL sites (Table 1). Sites 1, 4 and 7, closest to smelter locations and other pollution sources (Figs. 1 and 5), had the highest concentrations of metals, values which usually exceeded TEC levels and often PEC levels (Table 1; Fig. 5). Selenium exceeded the freshwater TECs and PECs most frequently (Table 1), with high concentrations at all three sites

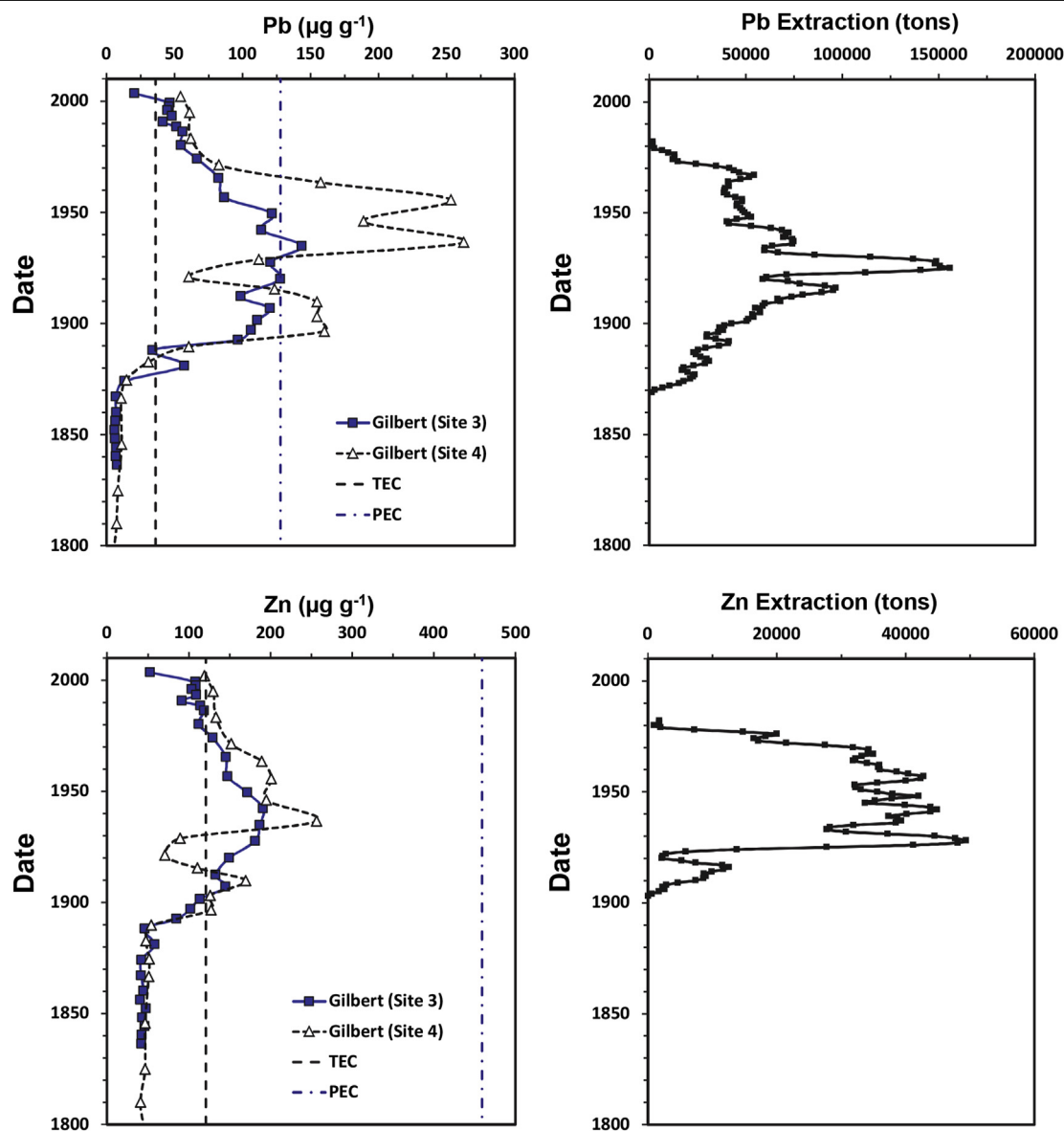


Fig. 2. (continued).

in Gilbert Bay. Selenium also exceeded the TEC, but not the PEC, at all three sites in Farmington Bay, whereas Bear River Bay sediments exceeded TEC at only one site. Concentrations of Cu and As exceeded TECs throughout Gilbert and Farmington bays but not in Bear River Bay. Surficial mercury concentrations exceeded the TEC at five of the six sites in Gilbert and Farmington Bays, but not in Bear River Bay deposits. Cadmium, Pb and Zn concentrations were above TECs at one or more sites in Gilbert and Farmington bays.

4. Discussion

4.1. Historical trends of metals

Paleolimnological analyses showed that the onset of metal contamination was concomitant with mining and metals production in the lake's watershed. Further, patterns of spatial distribution of metals showed that sites proximal to mines and smelters had greater contamination than those distant to the sources. The chronology of metal contamination in GSL sediments is consistent

with those described for the Uinta Mountain lakes, 90–155 km east of GSL (Kada et al., 1994; Moser et al., 2010; Reynolds et al., 2010), suggesting the same source. Isotopic analyses of Pb in Uinta Mountain lake sediments showed that the contamination there was due to smelting in Midvale located 15 km south of Salt Lake City and the Bingham Canyon Kennecott mine (Reynolds et al., 2010). Increases in most metal concentrations in GSL sediments were usually 2–3-fold greater than those observed in the Uinta Lakes, while Cu deposition was 15-fold greater. Contaminants of Uinta Mountain lakes arrive atmospherically (Reynolds et al., 2010), so differences with GSL likely reflect a combination of greater transportation distance and local pollution via runoff at GSL. While we cannot separate these mechanisms in this paper, Couillard et al. (2008) noted a similar difference in contamination of lakes located adjacent to the source of metal pollution compared to those 30 km distant that received only atmospheric influx.

Periodic releases of material from a Kennecott/Rio Tinto tailings pond (Fig. 1) 500 m from the south shore of GSL may have contributed irregularly to the metals loading to proximal Site 4.

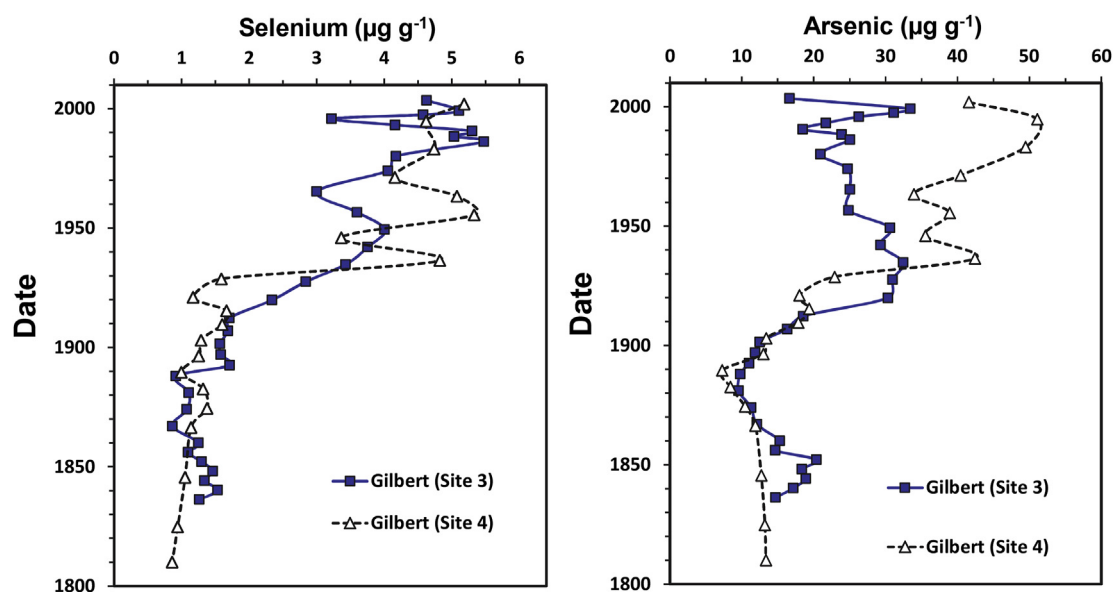


Fig. 3. Concentrations of selenium (left) and arsenic (right) from two sediment cores in Gilbert Bay of the Great Salt Lake.

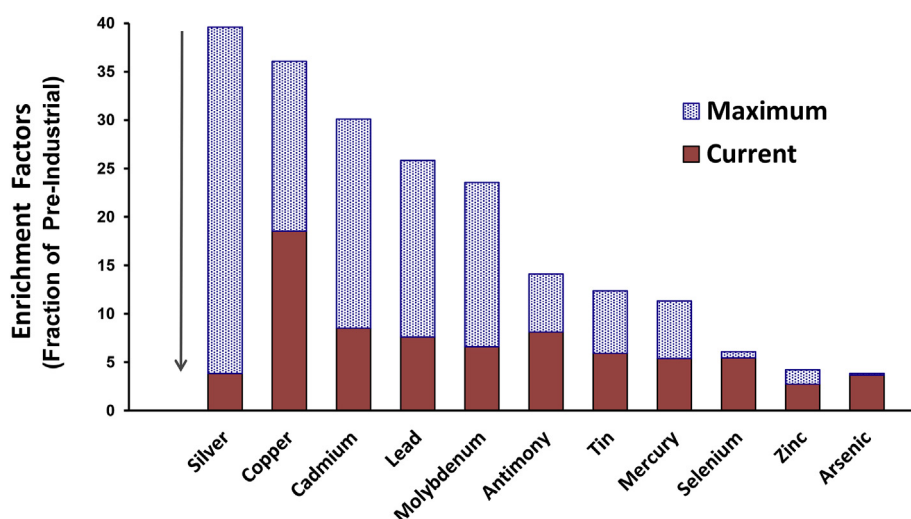


Fig. 4. Maximum Enrichment Factor (maximum/background) in the sediment core from Gilbert Bay (Site 4) (stippled, blue) and the Current Enrichment Factors (surface concentration/background; solid, red) for 11 metals and semi-metals. These values are based on the actual maxima (not the 3-pt running means that are shown in Fig. S2). The down-arrow depicts how concentrations of many metals in Gilbert Bay are now considerably lower than during the highest contamination period in the 1950s and 1960s. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Historical records indicate that tailings pond dikes failed in 1941–42, 1944, 1951, 1964 and 1969, although it is unclear how much material entered GSL (EPA, 2002). Additionally, in 1965, an experimental tailings capture system failed when a south-shore dike containing 56,000 m³ of Kennecott tailings material was destroyed by wave action (EPA, 2002). Typical tailings material at that site contains ~30 µg g⁻¹ As, ~240 µg g⁻¹ Pb and ~6 µg g⁻¹ Se. If this material was spread over just 25% of the southern area of Gilbert Bay, it would have deposited a layer about 0.8 mm thick elevating metal concentrations in the sediments for several years.

Mercury concentrations in the lake's sediments are particularly alarming, given that methylmercury concentrations in the lake's deep brine layer are among the highest recorded in the USA (Naftz et al., 2008). Like many metals, Hg concentrations in Gilbert Bay sediments declined from the mid-20th century maxima. Current atmospheric deposition of Hg to GSL is not abnormally high

(Peterson and Gustin, 2008); consequently, we infer that more recent reductions in sedimentary Hg largely reflect diminished legacy effects of early mining activities and, possibly, formation of the deep brine layer.

Improved smelting techniques and, in some cases, lower metal production, likely contributed to declines in sedimentary metal concentrations since the mid-20th century. Lead and Zn production in Utah ceased ca. 1980, consistent with modern sediment concentrations of ca. 22–33% and ca. 45–53% of the peaks in the 1940s and 50s, respectively. Similarly, the use of lead in gasoline peaked in the early 1970s and has since declined to near zero (Laws, 1993). In contrast, Cu production in Utah has increased to record levels, so the decreases in sedimentary Cu to 33–42% of historical maximum (Figs. 2 and 4) likely reflects improved industrial practices. Atmospheric pollution declined markedly with the Clean Air Act of 1963 and subsequent amendments in the 1970s, while an improved

Table 1
Average concentrations ($\mu\text{g g}^{-1}$ dry) of metals in the top 1-cm of cores taken in three bays of the Great Salt Lake. Threshold effects concentrations (TEC) and the probable effects concentrations (PEC) (MacDonald et al., 2000) are also shown. For silver, the Washington State Standard (2013) is shown as a TEC. Lake sediment concentrations exceeding the TECs are shown in bold numerals, and those exceeding the PECs are underlined.

Metal	Toxicity Criteria		Gilbert Bay			Farmington Bay			Bear River Bay	
	TEC	PEC	3	4	5	1	2	7	6	9
Metals exceeding threshold effect concentrations (TEC) at one or more sites										
Selenium	1.0	4.0	<u>4.9</u>	<u>4.9</u>	<u>4.3</u>	2.2	3.3	2.0	1.1	0.7
Copper	32	149	109	205	91	133	42	126	16	16
Zinc	121	459	80	124	47	242	68	195	43	35
Lead	36	128	34	58	21	119	25	79	18	15
Arsenic	10	33	25	46	18	17	12	20	7	5
Cadmium	1.0	5.0	1.2	2.4	0.7	2.5	0.8	2.4	0.4	0.4
Mercury	0.18	1.06	0.12	0.41	0.30	0.81	0.18	0.63	0.04	0.05
Silver	0.6		0.02	0.07	<0.03	1.71	0.10	1.89	0.08	0.13
Other Metals										
Cobalt			3.3	4.3	1.6	5.0	1.5	4.0	4.0	4.0
Chromium	43.4	111.0	9.1	11.4	6	32.3	6.7	22.0	8.8	7.2
Molybdenum			34.7	16.7	21.6	1.1	18.7	1.0	0.4	0.1
Nickel	22.7	48.6	10.0	14.1	6.5	11.6	7.2	13.1	12.5	9.6
Antimony			0.43	0.64	1	0.04	0.43	0.05	0.03	0.02
Tin			0.83	1.64	0.62	2.53	0.43	1.67	0.03	0.02
Titanium			0.42	0.83	1.40	0.40	1.37	0.34	0.46	0.32
Vanadium			14.7	19.5	9.7	19.9	10.2	19.6	9.1	8.2

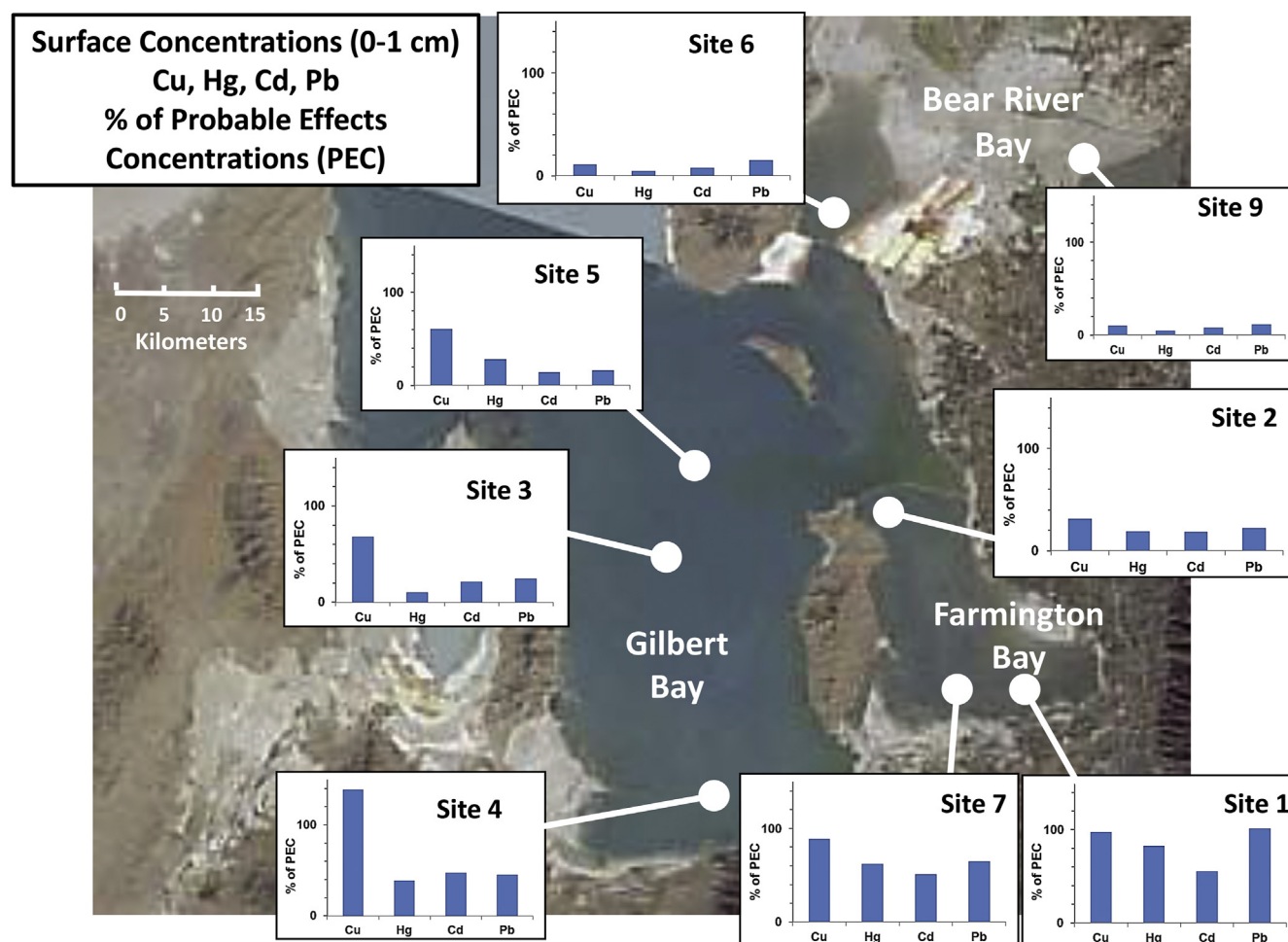


Fig. 5. Mean surface (0–1 cm) concentrations of copper (Cu), mercury (Hg), cadmium (Cd) and lead (Pb) at eight sites in the Great Salt Lake expressed as a percentage of the Probable Effects Concentrations (PEC) developed for freshwater organisms (Table 1; MacDonald et al., 2000).

economic market for recovered sulfur also reduced industrial discharges. As a result of emission controls and economic factors,

Kennecott Copper (now Rio Tinto), has reduced emissions of sulfuric acid from $\sim 22 \text{ MT h}^{-1}$ in 1974 to $\sim 0.03 \text{ MT h}^{-1}$ by 1998

(Newman et al., 1999).

The 370-m smokestack built at the Kennecott smelter in 1974 also helps disperse metals away from GSL. Consistent with enhanced atmospheric dilution, sediment Cu concentrations in GSL have decreased ~50% relative to peak values (Fig. 4), whereas those in Uinta Mountain lakes 85–100 km downwind have exhibited negligible decreases (c.f. Fig. 2 and Reynolds et al., 2010). Pelletier et al. (2020) noted that Canadian gold mines with stacks of 46–116 m had metal deposition radii of 27–40 km. Consequently, the 370-m Kennecott stack may now disperse metals beyond even the Uinta Mountain lakes.

Although improved smelting techniques and, in some cases, lower metal production likely contributed to declines in sedimentary metal concentrations, most metals in the sediments of GSL began to decline ca. 1950 before the advent of these changes (Site 4; Fig. 2, S1). Instead, declines are coeval with construction of the Southern Pacific railway causeway in 1959 which bisected GSL and led to the formation of the deep brine layer in Gilbert Bay (Domagalski and Eugster, 1990; Johnson et al., 2019). This dense, anoxic brine layer is usually present below a depth of ~6.5 m, and has a density (up to 1.2 g cm^{-3}) greater than that of many phytoplankton and detrital particles (Reynolds, 2006). As many metals reach the lake bottom via sedimentation of phytoplankton and detritus (Maszczyk and Wurtsbaugh, 2017), this deep brine layer can suspend material above the sediments, and allow mineralization and formation of dissolved phases in the deep water column. This hypothesized mechanism is supported by Jones and Wurtsbaugh (2014) who reported that particulate carbon was 120 to 200-fold higher in the deep brine layer than in the mixed layer of GSL, and that dissolved mercury was 9-fold higher in deep waters. Approximately 40% of metals in the deep brine water may be entrained into the upper mixed layer each year (Jones and Wurtsbaugh, 2014), potentially reducing metals accumulation in profundal deposits but exposing brine shrimp and other biota to Hg in the upper water column. Deep brine layers have been reported in other saline lakes (Anderson, 1958; Miller et al., 1993; Tiffany et al., 2007), but their effects on metal sedimentation and recycling have not previously been investigated.

The low redox potential ($<-200 \text{ mV}$) and abundant sulfides ($>100 \text{ mg l}^{-1}$) in the stagnant brine layer (Jones and Wurtsbaugh, 2014) may also affect metal deposition (Domagalski and Eugster, 1990). Those authors analyzed the metals in the water column and sediments of GSL and concluded that the deep brine layer traps metals, but ultimately facilitates the permanent storage of Cu, Zn, Pb and Co by co-precipitation with pyrite (FeS_2) in the anoxic, reducing sediments. Although, Mo and Cd also precipitate as sulfides, these metals remain mobile if exposed to oxidizing conditions, such as occurred in 2014–15 (Yang et al., 2000) after our cores were taken. Others working in both fresh and marine waters have found that low redox potentials and high sulfide concentrations reduce the mobility of many of the metals (Engstrom and Wright, 1984; Hamilton-Taylor and Davison, 1995; Smrzka et al., 2019). These analyses suggest that the redox changes induced by formation of the anoxic deep brine layer should have increased sedimentary concentrations of these metals, whereas their concentrations have all decreased in recent decades (Fig. 3; Appendix 1). These complex patterns point to the need for an improved understanding of how redox conditions can alter the phase states of metals and influence their movements between sediments, brines, and waters of saline lakes.

In contrast to most metals, concentrations of the metalloids Se and As have increased or remained relatively stable during the past 50 years (Fig. 3). Arsenic occurs in both the Mercur gold ores and lead-zinc ores of western Utah. Selenium occurs at low levels in the Bingham Canyon porphyry copper deposits and is released from

smelting activity (Newman et al., 1999). Although Se is also released into the Bear River watershed from phosphate mining districts in Idaho and the Uinta Mountains (Hamilton and Buhl, 2005; Mars and Crowley, 2003), Diaz et al. (2009a) reported that this river provides only 26% of Se influx compared to riverine inputs from the Salt Lake Valley watershed. Both As and Se are emitted from smelting activities in the Great Salt Lake Valley, as demonstrated locally from tailings wastewater (EPA, 2002). Concentrations of these metalloids, particularly As, were highest at coring site 4 which is closest to the Kennecott smelter (Fig. 1) and discharge creeks, demonstrating a continued local source of metalloids (Naftz et al., 2009).

It is unclear whether the stable or increasing concentrations of selenium and arsenic are due to increased pollution from mining alone, as both Se and As can be mobile in sediments and may respond to changing redox conditions. For example, in oxygenated freshwater sediments, As can bind to Fe- and Mn- oxyhydroxides, whereas these oxides can be dissolved and As released into sediment porewaters, migrate upwards in the sediments, and enter the overlying water under reducing conditions (Martin and Pedersen, 2002; Schuh et al., 2019). For example, Whitmore et al. (2008) attribute recent declines in sedimentary As to eutrophication of a Florida lake and development of hypolimnetic anoxia and reducing sediments. In contrast, marine waters which have high sulfate concentrations like GSL can incorporate both As and Se into highly-insoluble pyrite (FeS_2) when redox potentials approach -200 mV (Rickard and Luther, 2007; Smrzka et al., 2019), a condition seen in the deep brine layer of GSL (Jones and Wurtsbaugh, 2014). In principle, these processes could interact to trap local sources of contamination in the southern basin of GSL. Better understanding of the mechanisms sustaining elevated levels of Se and As is needed because surficial sediments in Great Salt Lake still exceed TEC and PCC values to harm aquatic wildlife (Table 1).

Although concentrations of most metals in the Great Salt Lake's sediments have declined, complete recovery will require many decades. For example, Haas et al. (2019) demonstrated that ~50 years were needed for a polluted lake to recover after Romans abandoned their settlement in what is now Switzerland. Similarly, Jamieson (2014) found that copper concentrations in the surface sediments of Yellowknife Lake (Canada) declined to baseline levels 25 years after control of excessive emissions from nearby Giant Mine, although Pelletier et al. (2020) estimated that 97 years would be needed for full recovery from lead pollution from the same mine. Overall, recovery rates in GSL may be similar, suggesting that centennial timeframes are needed to return managed lakes to baseline metals concentrations. For example, in Utah commercial production of lead and automobile emissions nearly ceased by 1981 but current Pb levels still remain six-to nine-fold higher than background levels.

4.2. Spatial distribution and toxicity of metals

Concentrations of metals in the surface sediments varied widely in the three bays, with highest values at the southern end of Farmington Bay and Gilbert Bay closest to mining and smelting, and lowest levels in northeastern Bear River Bay. High concentrations of metals at Site 1 in Farmington Bay also may result from the proximity of this site to the outflow of the combined Sewer Canal and the NW Oil Drain (Sorensen and others, 1988). Samples from Bear River Bay generally had relatively low levels of contamination (Table 1), with only selenium slightly exceeding TEC guidelines at one site. We infer that the low absolute concentrations in Bear River Bay reflect greater distance to source, hydrological isolation by the train causeway, and higher dilution by riverine materials (Allred and Judd, 2002).

Concentrations of most metals in southern-most Gilbert and Farmington Bay surface sediments exceeded their TECs, while Cu, As, and Hg also exceeded PECs. These bays are critical feeding areas for millions of migratory birds represented by 257 species (Baxter and Butler, 2020). Brine fly larvae and brine shrimp are important vectors that transfer metals to migratory waterfowl, but these invertebrates are resistant to metal pollution (Brix et al., 2004). Although exposure to metals above TEC and PEC values are consequently not a direct concern for these invertebrates, trophic transfer to sensitive avian species remains a concern. Bio-magnification factors for Se and Hg in brine flies living in oxic environments are apparently modest (Hg - 2.2X; Se - 1.2X; Wurtsbaugh et al., 2011). Nevertheless, recent analysis of GSL waterfowl revealed that their eggs have Se and Hg levels exceeding toxicity benchmarks or regulatory standards (Ackerman et al., 2015). Additionally, Hg concentrations in three local duck species exceed human dietary thresholds, leading Utah to issue consumption advisories for these birds (Utah Department of Environmental Quality, 2019). Thus, although concentrations of metals have declined in lacustrine sediments, effects of ongoing and legacy contamination remain a threat to the millions of birds utilizing GSL.

5. Conclusions

Great Salt Lake is located next to major mining, smelting and industrial areas and its sediments exhibited marked metal contamination. Although metal pollution in GSL generally peaked in the mid-1950s and declined thereafter, many metal concentrations remain above critical thresholds decades later. Declines in metal content of lake sediments over the past 75 years likely arise from multiple mechanisms including: (1) decreased mining for some elements such as lead and zinc; (2) cleaner smelting technologies; and (3) formation of a deep brine layer that produces reducing conditions and likely reduces sedimentation of metals. Concern remains for Cu, Hg, Cd, Pb, Zn, Se and As, metals whose concentrations in surface sediments remain both elevated above TEC and accessible to invertebrates supporting the food web of migratory and regional birds. Notwithstanding the declining levels of many metals in the lake, concentrations of As and Se were either stable or increasing in the sediments and we suggest that managers should carefully monitor these metals to ensure safe levels to protect wildlife and people. Importantly, our research shows that even when environmental regulations successfully lower metal pollution of saline lakes, toxic effects of these pollutants may linger for many decades.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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