Environ Monit Assess (2017) 189:257
DOI 10.1007/s10661-017-5952-0

Spatiotemporal assessment (quarter century) of pulp mill metal(loid) contaminated sediment to inform remediation decisions

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Received: 21 February 2017 / Accepted: 17 April 2017

Abstract A bleached kraft pulp mill in Nova Scotia has discharged effluent wastewater into Boat Harbour, a former tidal estuary within Pictou Landing First Nation since 1967. Fifty years of effluent discharge into Boat Harbour has created >170,000 m^3 of unconsolidated sediment, impacted by inorganic and organic contaminants, including metal[loid]s, polycyclic aromatic hydrocarbons (PAHs), dioxins, and furans. This study aimed to characterize metal(loid)-impacted sediments to inform decisions for a $89 million CAD sediment remediation program. The remediation goals are to return this impacted aquatic site to pre-mill tidal conditions. To understand historical sediment characteristics, spatiotemporal variation covering ~quarter century, of metal(loid) sediment concentrations across 103 Boat Harbour samples from 81 stations and four reference locations, were assessed by reviewing secondary data from 1992 to 2015. Metal(loid) sediment concentrations were compared to current Canadian freshwater and marine sediment quality guidelines (SQGs). Seven metal(loid)s, As, Cd, Cr, Cu, Pb, Hg, and Zn, exceeded low effect freshwater and marine SQGs; six, As, Cd, Cr, Pb, Hg, and Zn, exceeded severe effect freshwater SQGs; and four, Cd, Cu, Hg, and Zn, exceeded severe effect marine SQGs. Metal(loid) concentrations varied widely across three distinct temporal periods. Significantly higher Cd, Cu, Pb, Hg, and Zn concentrations were measured between 1998 and 2000, compared to earlier, 1992–1996 and more recent 2003–2015 data. Most samples, 69%, were shallow (0–15 cm), leaving deeper horizons under-characterized. Geographic information system (GIS) techniques also revealed inadequate spatial coverage, presenting challenges for remedy decisions regarding vertical and horizontal delineation of contaminants. Review of historical monitoring data revealed that gaps still exist in our understanding of sediment characteristics in Boat Harbour, including spatial, vertical and horizontal, and temporal variation of sediment contamination. To help return Boat Harbour to a tidal estuary, more detailed sampling is required to better characterize these sediments and to establish appropriate reference (background) concentrations to help develop cost-effective remediation approaches for this decades-old problem.

Keywords Bleached kraft pulp mill effluent · Secondary data · Sediment metal contaminants · Future monitoring · Remediation · Boat Harbour

Electronic supplementary material The online version of this article (doi:10.1007/s10661-017-5952-0) contains supplementary material, which is available to authorized users.

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Published online: 06 May 2017

Introduction

Metals in sediments occur naturally via geological weathering and sediment deposition (Nriagu 1989; Loring et al. 1996). However, most putative metal contamination originates from anthropogenic industrial activities (e.g., metal mining, coal combustion, paper processing, and chlor-alkali plants) (Walker et al. 2003; Parsons and Cranston 2006; Adoli et al. 2011; Tchounwou et al. 2012; Walker and Grant 2015; Walker 2016). Organic-rich sediments impacted by industrial activity can sorb and accumulate high concentrations of metals, which complex with organic matter posing ecological risks to aquatic biota (Hope 2006; Suresh et al. 2012; Walker et al. 2015a). Aquatic sediments impacted by long-term metal contamination from industrial activities often require costly remediation, dredging, or disposal when concentrations exceed sediment quality guidelines (SQGs) (Peng et al. 2009; Walker et al. 2013a, 2015b).

For decades, the pulp and paper industry in Canada has been responsible for generating significant atmospheric and effluent wastewater emissions, sometimes resulting in widespread sediment pollution from organic and inorganic contaminants (Pokhrel and Viraraghavan 2004; Hewitt et al. 2006; Soskolne and Sieswerda 2010; Munkittrick et al. 2013; Hoffman et al. 2015). Pulp mill wastewater effluents can be acutely toxic and associated with deleterious effects in aquatic ecosystems (Sunito et al. 1988; Colodey and Wells 1992; Ali and Sreekrishnan 2001). In Canada, pulp mill effluents are regulated for water quantity and quality under the Pulp and Paper Effluent Regulations (PPER) under the *Fisheries Act* (1985) (PPER 1992; Environment Canada 2003; Roach and Walker 2017). A bleached kraft pulp and paper mill, *the mill*, has operated in Pictou County, Nova Scotia since 1967, producing ~280,000 t of pulp annually (Ogden 1972). Wastewater effluent from the mill is discharged into the Boat Harbour Treatment Facility (BHTF), built and operated by the provincial government, adjacent to Boat Harbour, a former tidal lagoon within the Mi'kmaq Pictou Landing First Nation (PLFN) community (Fig. 1).

The BHTF was upgraded in 1972 consisting of twin *settling ponds* and an "aerated stabilization basin" (ASB) (567,750 m^3 volume). Effluent from the mill (>87,000 m^3/day) is piped beneath East River and discharged into one of two 50,000 m^3 settling ponds (A/B) to allow for sedimentation (Fig. 2). Effluent flows to the ASB (C) for treatment (i.e., oxidation of wastewater) for 5–6 days prior to discharge (D) to the 3,400,000 m^3 *stabilization lagoon* (i.e., Boat Harbour) (E). The compliance monitoring point for federal environmental regulations is the discharge (D) point. Effluent remains in Boat Harbour for 20–30 days before final discharge to the Northumberland Strait via a dammed estuary mouth (F) (JWEL and Beak Consultants 1992). Discharge to Boat Harbour has resulted in the deposition of >170,000 m^3 of unconsolidated contaminated sediments impacted by inorganic and organic contaminants (i.e., metal[loid]s, polycyclic aromatic hydrocarbons [PAHs], dioxins, and furans) (Hoffman et al. 2015).

The chlorine bleaching process generates a suite of carcinogenic, lipophilic chlorinated organic compounds, such as chloroform, dioxins, furans, and phenols, that bioaccumulate in aquatic biota (McLeay 1987; Sunito et al. 1988; Wong et al. 1994). Although most research has focused on chlorinated organic compounds (e.g., dioxins and furans), metal(loids) are also contained in pulp mill effluents (Lewinsky 2007). This study focused on the spatiotemporal assessment of metal(loids) in Boat Harbour impacted sediments. Organic contaminants (e.g., PAHs, dioxins, and furans) were assessed in a separate study. Metal(loid)s in Boat Harbour are presumably derived from pulp mill effluent combined with atmospheric deposition from local point source (PS) emitters, including the mill's smokestacks, a coal-fired thermal generating plant with fly ash emissions, a tire manufacturing plant, and effluent (between 1971 and 1992) from a former chlor-alkali plant (Northern Pulp Nova Scotia Corporation 2015; Fig. 1). Since 1967, cumulative adverse environmental and human health impacts, including poor air and water quality, sediment and soil contamination, and negative impacts to recreational activities, have been a major concern for the local community (Hoffman et al. 2015, 2017; Pictou Landing Native Women's Group et al. 2016). Although the PLFN community have long advocated for closure and remediation of Boat Harbour, a recent effluent leak in 2014 triggered protests, resulting in the passing of the Boat Harbour Act (2015). The magnitude and extent of the mill's effluent leak was deemed deleterious to fish under the federal *Fisheries Act* (1985), resulting in a \$225,000 CAD fine (Pictou Landing Native Women's Group et al. 2016). The Boat Harbour Act legislated cessation of Boat Harbour being used as an effluent treatment facility by January 30, 2020.

These contaminated sediments are now poised for remediation, commencing in 2020, by the province of Nova Scotia, estimated at \$89 million CAD. The goal of remediation will be to return Boat Harbour to its pre-1967 tidal state. Despite a plethora of monitoring

Fig. 1 Location of Boat Harbour in Pictou County, Nova Scotia relative to communities (e.g., Pictou, and Pictou Landing First Nation [PLFN]), Fergusons Pond (a previously studied reference site), and local point source emitters (e.g., pulp and paper mill [PS.1], tire manufacturing facility [PS.2], coal-fired thermal electrical generating station [PS.3], former chlor-alkali facility [PS.4]) (Google Earth©)

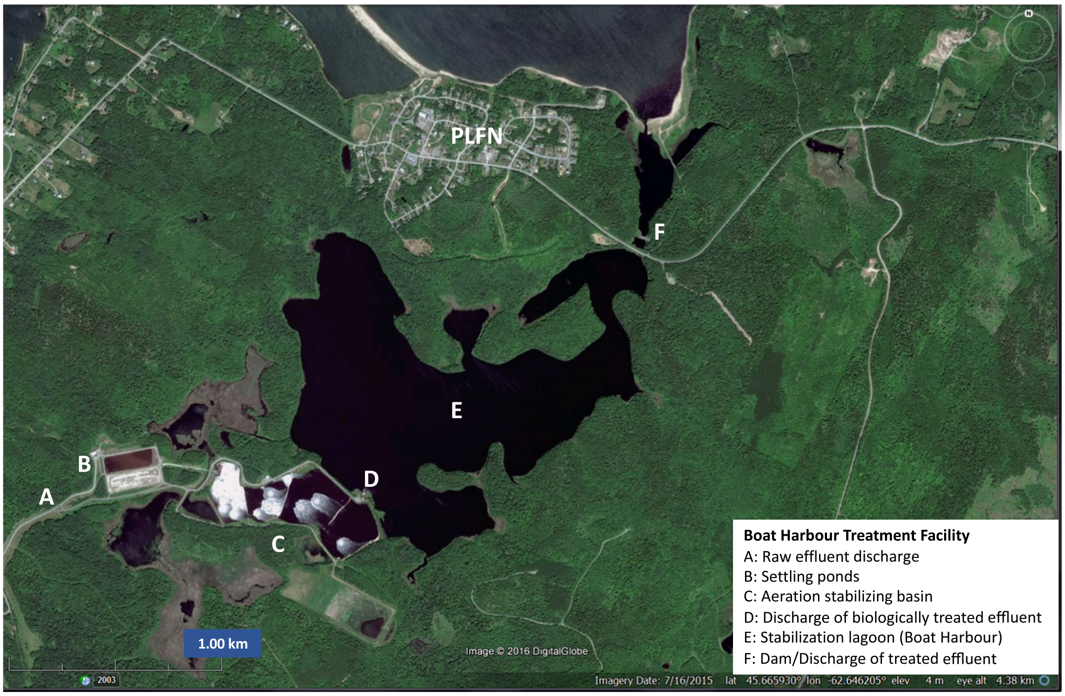

Fig. 2 Components of the Boat Harbour Treatment Facility (BHTF) relative to neighboring Pictou Landing First Nation (PLFN) community (Google Earth©)

studies, gaps still exist in our understanding of sediment characteristics (e.g., depth of impacted sediment, including spatiotemporal extent and magnitude of impacts). Therefore, a comprehensive sediment characterization examining metal(loid) contamination was conducted by reviewing secondary data to examine spatiotemporal extent and magnitude of impacts to help inform remediation management decisions. The aims of this study were to (i) assess spatiotemporal coverage of long-term monitoring data (1992–2015) related to metal(loid) sediment concentrations across Boat Harbour and reference locations using geographic information system (GIS) techniques, (ii) determine vertical and horizontal delineation based on collection methods, (iii) compare sediment metal(loid) concentrations to current Canadian SQGs, and (iv) identify potential gaps in long-term monitoring data to help inform future remediation management decisions.

Materials and methods

Historical document review

Secondary data consisting of >200 documents (including government reports and peer-reviewed literature) were reviewed for relevant sediment metal(loid) chemistry data (Supplementary material Table S1). Relevant data included (i) metal(loid) sediment concentrations, (ii) georeferenced locations, (iii) sediment depth and collection methods, and (iv) consistent laboratory analytical methods. Only eight reports contained relevant information on sediment metal(loid) chemistry data collected from Boat Harbour and reference locations (Jacques Whitford Environment Limited [JWEL] 1997, 1999, 2001, 2005; JWEL and Beak Consultants 1992, 1993; Stantec 2013, 2016). Sediment samples reported in this study were collected between 1992 and 2015.

Sediment sampling and analysis

Boat Harbour currently operates as a freshwater treatment lagoon but will be returned to tidal influences following sediment remediation. Therefore, reported metal(loid) parameters were compared to current Canadian Council of Ministers of the Environment (CCME) freshwater and marine SQGs (CCME 2016). CCME interim sediment quality guidelines (ISQGs) represent lowest effect levels, below which contaminants have little chronic or acute effect on biota; CCME probable effect levels (PELs) are equivalent to severe effect levels, above which biota are highly likely to be negatively impaired by contaminants (Walker et al. 2015a, b). Sediment concentrations <ISQGs were considered uncontaminated, between ISQGs and PELs were considered moderately contaminated, and >PELs were considered severely contaminated (Walker et al. 2015a).

For GIS data entry, criteria were identified for each report: metal(loid) parameters, parameter concentrations, sample location (x, y coordinates in decimal degrees), site identification, date of sample, sample depth (cm), and environmental media (i.e., unconsolidated sediment/deposits or marine sediment). To attain a unified coordinate system, sample location coordinates were generated by overlaying report maps in Google Earth©. Coordinates of centroids were identified for horizontal composite samples to represent average sample locations.

Based on study sampling methods, *sample type* was either categorized as *grab* (e.g., Ekman grab) or *core* (e.g., Piston corer). Additionally, samples were categorized as *composite* or *discrete*. Vertical composites represent horizontal layer mixing, and horizontal composites represent aggregates of sub-samples from an area (e.g., like incremental sampling [see Hadley and Bruce 2014]). Discrete samples were collected from specific horizontal locations or vertical intervals. Assumptions were made for sample depth, when necessary. Grab samples were assumed to represent 0–15 cm, unless otherwise stated. Centroid (mid-range) sample depths were used to assess vertical variation of metal(loid) concentrations. A contoured map displaying mean sediment thickness for Boat Harbour (stabilization lagoon) were referenced to determine approximate sample depths for JWEL (2001, 2005) and Stantec (2013), assuming sediment depth remained consistent (Supplementary material Fig. S1).

Box and scatter plots were used to display temporal (1992–2015) and spatial (vertical) variation, respectively, of metal(loid) sediment concentrations. Significant temporal differences ($p < 0.05$ level) were determined by one-way analysis of variance (ANOVA) followed by a Tukey's test using Minitab. Spatiotemporal variation of discrete locations and relative metal(loid) concentrations across Boat Harbour were assessed using GIS mapping techniques for this long-term monitoring dataset over three distinct temporal periods: 1992–1996, 1998–2003, and 2004–2015. Time periods were chosen due to break points or shifts in concentrations. Despite limitations of GIS data, spatial analysis using these three time periods provided qualitative trends to help provide direction for future surveys to collect appropriate spatial data for quantitative interpolation.

Quality control, data analysis, and limitations

Samples (including blind field duplicates) were analyzed by commercial laboratories (Standards Council of Canada accredited) for analysis of total metal(loid)s (dw) basis, unless otherwise indicated. Method blanks, spike blanks, matrix spikes, duplicate samples, and quality control protocols varied, so individual reports are cited for more detailed information. Detection limits (DL) varied between 0.01 and 10 mg/kg (i.e., DLs for Cr = 2–5, Cd = 0.02–0.50, Cu = 2–5, Hg = 0.01–2, Pb = 0.10–10, Zn = 2–5 mg/kg) across reports.

Most historical sediment sampling was conducted a priori of remediation, but metal(loid) concentrations from different reports using different sampling techniques (grabs vs. cores, composite vs. discrete) were reported without modification (except for data reported <DLs). Censored data with metal(loid) concentrations <DL were substituted using 0.05 DL values (de Solla et al. 2012; MacAskill et al. 2016). For this study, data independence between years was assumed due to variation of abiotic (e.g., sampling techniques and deposition of fresh sediment) and biotic (e.g., bioturbation) conditions in Boat Harbour. Therefore, statistical analysis using ANOVA was used to analyze temporal differences in metal(loid) concentrations (i.e., concentrations at time T + 1 were independent of concentrations at time T + 0). Interpolation of geo-spatial data for metal(loids) was challenging, due to limitations of the data (i.e., too few data points temporally and spatially distributed for the entire survey period). Consequently, hotspot mapping of metal(loid) concentrations was not conducted.

Results and discussion

Spatiotemporal assessment

Review of secondary data revealed wide variation in sampling techniques (e.g., using grabs, cores, discrete, and composite sampling) and sample depth (e.g., shallow vs. deep). Approximately 38% of samples were grabs, while 62% were cores, making temporal comparisons between reports, using different techniques, extremely challenging. Sediments across Boat Harbour were highly organic with mean total organic carbon values ranging from 4 to 27% (JWEL and Beak Consultants 1993; JWEL 2001). Subsurface conditions consist of anthropogenic black freshwater organic sediment, between 12- and 26-cm thick, underlain by a distinct layer of organic silt, glacial till, and sedimentary bedrock (Spooner and Dunnington 2016; Stantec 2016).

Spatiotemporal analysis of a quarter century of sediment metal(loid) concentrations revealed coverage of sampling sites was lacking (Fig. 3). The western half of Boat Harbour had limited coverage (JWEL and Beak Consultants 1992; JWEL 2005) compared to the eastern tidal channel (JWEL 1999, 2001). However, JWEL and Beak Consulting (1993), JWEL (1997), and Stantec (2013) sampled from fewer locations. A recent study by Stantec (2016) sampled from isolated coves with few comparable historic locations. Most (69%) Boat Harbour samples from 103 samples and 81 locations were from shallow horizons, between 0 and 15 cm, leaving deeper horizons under-characterized. These data gaps provide challenges for accurate horizontal and vertical delineation of impacted sediment for future remedy decisions (Fig. 4; Supplementary material Table S2, Fig. S1).

Despite wide variation in sampling techniques, seven metal(loid)s, As, Cd, Cr, Cu, Pb, Hg, and Zn, frequently exceeded low effect freshwater ISQGs (56.3–88.4%; n = 58–91) and low effect marine ISQGs (23.0–91.3%; n = 23–94). Six, As, Cd, Cr, Pb, Hg, and Zn, exceeded severe effect freshwater PELs (1.0–63.1%; n = 1–53) and four, Cd, Cu, Hg, and Zn, exceeded severe effect marine PELs (20.4–54.4%; n = 21–56) (Table 1; Figs. 4 and 5). SQG exceedances were predominantly found in surface horizons (Fig. 4). Results were also mirrored in geo-spatial data (Figs. 6, 7, and 8; Supplementary material Figs. S2, S3, and S4). Significantly higher concentrations of Cd, Cu, Pb, Hg, and Zn were measured in 1998 and 2000 compared to those measured before and/or after, and often exceeding both freshwater and marine CCME PEL guidelines (Fig. 5). Elevated concentrations of Cd, Cu, Pb, Hg, and Zn measured between 1998 and 2000 were presumably because ASB improvements in 1996 increased oxygen concentrations in Boat Harbour. Enhanced decomposition of organic material by aeration may have also led to sediment volume reduction and simultaneous increases in relative metal[loid] concentrations. Furthermore, changes in operational control during 1996, potential metal(loid) inputs from other local PS

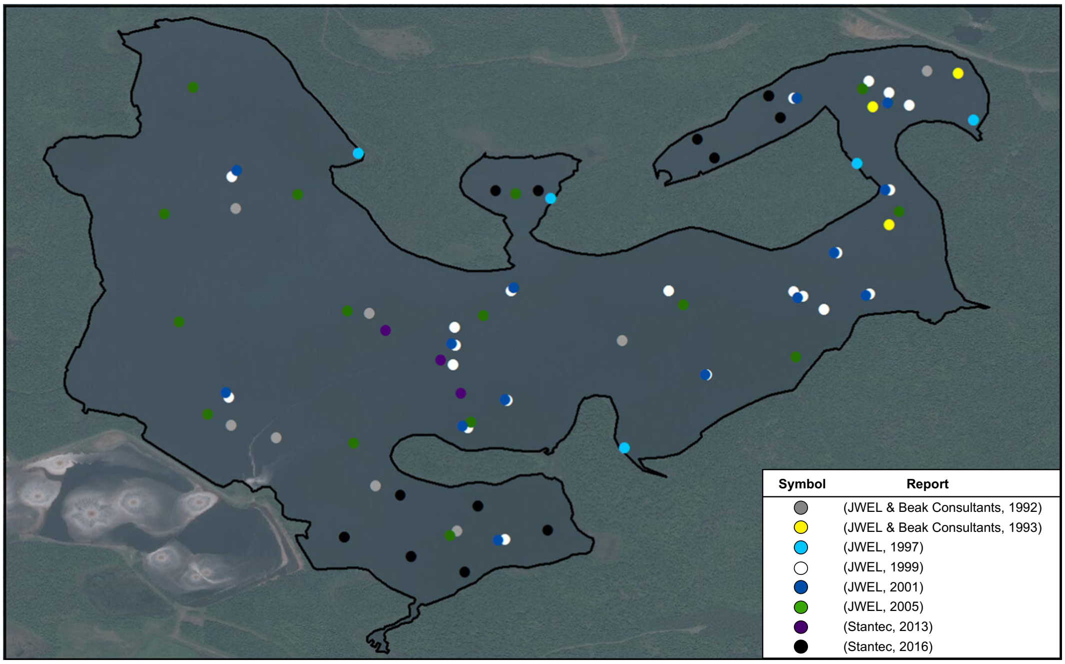

Fig. 3 Spatiotemporal coverage (1992–2015) of sediment sampling sites in Boat Harbour. *Colored circles* indicate when samples were collected/analyzed

emission sources, or dredging of ASB sediments could partly explain increases in metal(loid) concentrations during this period (JWEL 1999). However, since peak metal(loid) concentrations in 1998, there appears to have been a steady decline, except for Hg (Fig. 5).

However, decreasing metal(loid) concentrations may not indicate improvements in overall conditions, as metal(loid)s would not degrade over the quarter century of data examined herein. Overall apparent decreases in concentrations may perhaps be attributable to bioturbation, burial from less contaminated sediment, isolated or under-sampling, improved federal regulatory effluent guidelines (Environment Canada 2013, 2014a) or more stringent provincial industrial approvals (Nova Scotia Environment 2015), improvements to effluent wastewater treatment, or bleached kraft mill effluent currents changing sediment depths and deposition rates, all of which may have effectively attenuated mean concentrations over time. Boxes with diagonal lines show wide variation for all six metal(loid) concentrations over 25 years: Cd, 6.55 ± 5.64; Cr, 47.04 ± 22.98; Cu, 60.27 ± 40.98; Pb, 54.20 ± 26.36; Hg, 0.67 ± 0.42; and Zn, 501 ± 413 mg/kg (n = 80–103) (Fig. 5). This illustrates a quarter century of composite concentrations presumably reflecting variation in sampling depths and techniques. This also represents the spatiotemporal inventory of metal(loid)s in Boat Harbour (i.e., the entire contaminant inventory poised for remedy treatment).

Metal(loid) concentrations measured in 2015 were all below freshwater and marine PELs, and many were ≤ISQGs. Furthermore, 2015 concentrations were not significantly different than concentrations measured in 1992 (~25 years earlier). Significant decrease of metal(loid) concentrations in 2015 compared to 1998, and to a lesser extent 2000, were likely attributable to dilution by vertical composite sampling in 50-cm increments, whereby unconsolidated sludge and sediment were mixed with less contaminated marine sediment in deeper horizons (Fig. 4). Metal(loid) concentrations in sediment using different techniques (e.g., cores vs. grabs) and subsequent subsampling techniques (e.g., surface vs. deep sediment or composite vs. discrete) showed wide spatiotemporal variation between 1992 and 2015. Such inconsistent sampling methods can yield "bad data." For example, depending on coring methods and quality assurance protocols used, convex upward deformation (*edge smearing*) could have biased horizontally sectioned data. Core deformation can limit the ability to resolve high concentration layers, decrease peak concentration, and increase depths where high contaminant concentrations are observed (Dunnington and

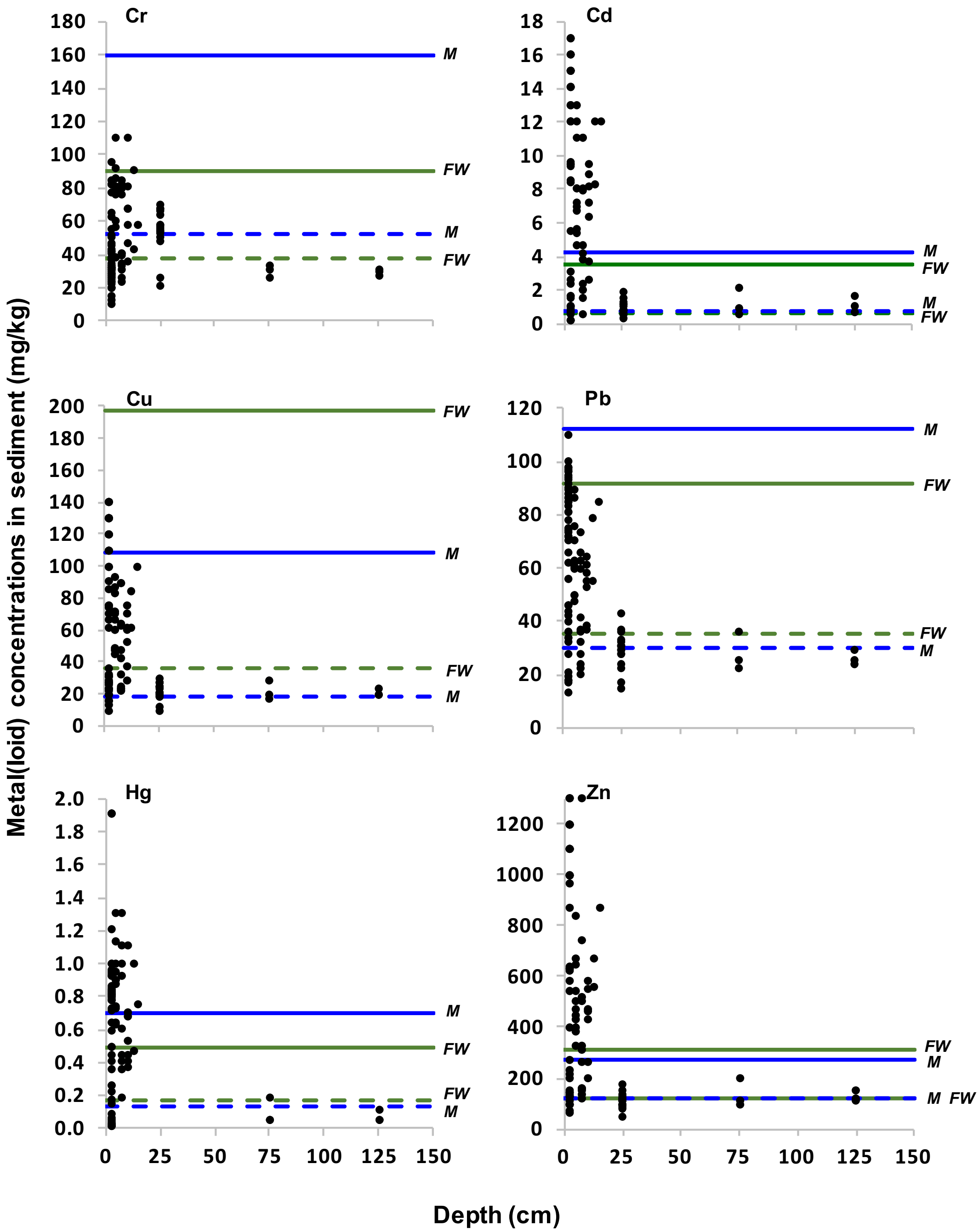

Fig. 4 Vertical variation of metal(loid) concentrations with sediment depth (cm). CCME freshwater (*FW*; *dark green*) and marine (*M*; *blue*) sediment quality guidelines are indicated using *solid horizontal lines* for PEL and *dashed lines* for ISQG values (CCME 2016)

Spooner 2017), resulting in gross over estimates of volumes of contaminated sediments to treat (Stantec 2016).

A recent study by Spooner and Dunnington (2016) reported wide variation of metal(loid) concentrations in organic unconsolidated sedimentation in Boat Harbour, using X-ray fluorescence (XRF) techniques. Unconsolidated sedimentation was distinguished from marine sediment visually, and by characteristically high values for Cu and Pb, likely due to cation capture by

Table 1 Canadian Council of Ministers of the Environment (CCME) sediment quality guideline (SQG) exceedances for metal(loid)s in the stabilization lagoon (i.e., Boat Harbour) between 1992 and 2015. Total sediment samples (n) are indicated under each element parameter

Parameter	Freshwater				Marine			
	ISQG		PEL		ISQG		PEL	
	SQG limit	Number of exceedances (%)	SQG limit	Number of exceedances (%)	SQG limit	Number of exceedances (%)	SQG limit	Number of exceedances (%)
As (n = 100)	5.9	59 (59.0)	17	1 (1.0)	7.24	23 (23.0)	41.6	0 (0.0)
Cd (n = 103)	0.6	91 (88.4)	3.5	58 (56.3)	0.7	88 (85.4)	4.2	55 (53.4)
Cr (n = 103)	37.3	58 (56.3)	90	4 (3.9)	52.3	39 (37.9)	160	0 (0.0)
Cu (n = 103)	35.7	58 (56.3)	197	0 (0.0)	18.7	94 (91.3)	108	21 (20.4)
Pb (n = 103)	35	70 (68.0)	91.3	11 (10.7)	30.2	79 (76.7)	112	0 (0.0)
Hg (n = 84)	0.17	68 (81.0)	0.486	53 (63.1)	0.13	70 (83.3)	0.7	43 (51.2)
Zn (n = 103)	123	81 (78.6)	315	55 (53.4)	124	80 (77.7)	271	56 (54.4)

Limited CCME SQG values available for other metal(loid)s (CCME 2016). ISQG indicates CCME interim sediment quality guidelines and PEL indicates CCME probable effect levels

organic matter and effluent discharge and loss on drying. Effluent impacted sediment (evidenced visually and by metal concentrations and metal ratios) were <30 cm at all stations (Spooner and Dunnington 2016). Underlying low-porosity marine sediments may serve as an indication of baseline conditions of sediment chemistry and biological status pre-mill operations and warrant further chemical characterization to help determine depth of impacted sediment prior to remediation. Ferguson's Pond (located 2.5 km NE of Boat Harbour on the Northumberland Strait) was previously used as a reference site (i.e., JWEL 1997, 1999, 2001) and considered a priori un-impacted by industrial activities (Fig. 1). Over the past 25 years, Boat Harbour sediment metal(loid) concentrations were up to 20 times higher than samples collected from Fergusons Pond (n = 4), which were all <ISQGs. Fergusons Pond similarly requires further investigation to better understand local baseline conditions and to help guide remedial objectives.

Sediments in many coastal regions adjacent to industrial areas in Nova Scotia are large sinks for, and potential sources of, metallic contaminants (Loring et al. 1996). However, limited data of contaminants in Nova Scotian coastal sediments exist (e.g., Walker et al. 2013b, 2015a; Walker and Grant 2015). This is particularly true for the Northumberland Strait marine environment near Boat Harbour (Dalziel et al. 1993). For comparison, mean background metal(loid) concentrations based on natural variation in pristine marine sediments around Nova Scotia were Cd = 0.3, Cu = 40, Pb = 40, Hg = 0.1, and Zn = 150 mg/kg (Loring et al. 1996). Metal(loid) concentrations in Boat Harbour greatly exceeded these regional background concentrations, suggesting that metal inventories were largely attributed to anthropogenic activities. A pre-remediation baseline study by Walker et al. (2013b) assessed contaminants in Sydney Harbour, near a former steel and coking facility (subject to historical effluent and atmospheric metal inputs), and found Cu, Hg, Pb, and Zn concentrations exceeded background levels reported by Loring et al. (1996). However, overall metal(loid) concentrations were considerably lower in Sydney Harbour compared to Boat Harbour (Walker et al. 2013b).

The mill's effluent is likely the primary source of metal(loid) loadings, but other local sources may have contributed to metal burdens in Boat Harbour. Coal combustion and chlor-alkali plants are global contributors to Hg (UNEP 2008). For decades, Canadian chlor-alkali plants have been sources of Hg in surficial sediments (Wilson and

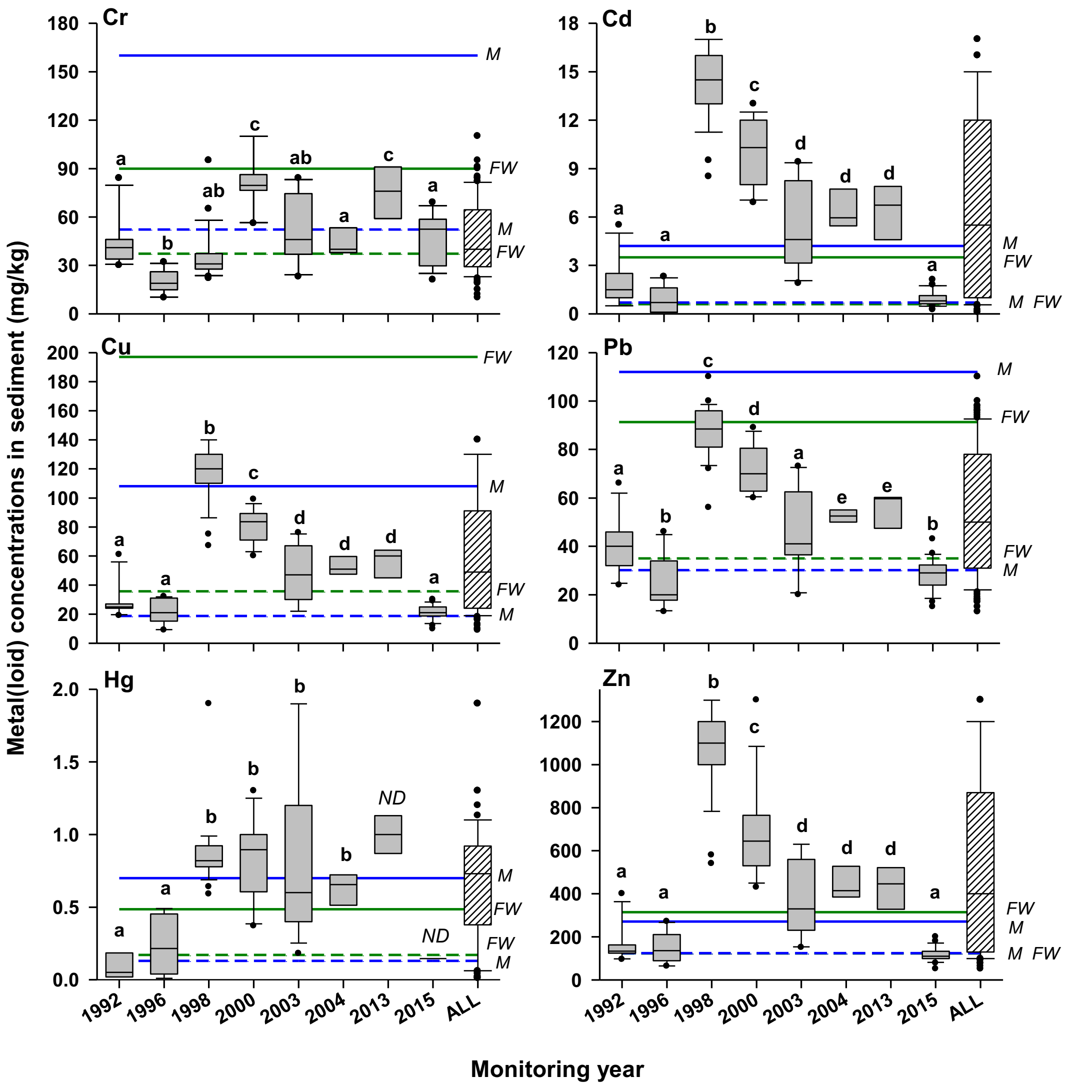

Fig. 5 Temporal variation (~25 years) of metal(loid) concentrations in surface sediment measured by ICP-MS. CCME freshwater (*FW*; *dark green*) and marine (*M*; *blue*) sediment quality guidelines are indicated using *solid horizontal lines* for PEL and *dashed lines* for ISQG values (CCME 2016). Significant temporal differences were determined by one-way ANOVA followed by Tukey's test; *years attributed with same letters* were not significant and *those with different letters* were significantly different ($p < 0.05$ level). Number of sediment samples analyzed in any given year varied widely (1992 = 11, 1996 = 10, 1998 = 26, 2000 = 14, 2003 = 12, 2004 = 5, 2013 = 3, 2015 = 22). *ND* not determined (too few samples for one-way ANOVA). *Boxes with diagonal lines* indicate variations in metal(loid) concentrations across all years

Travers 1976; Gobeil and Cossa 1993; Gagnon et al. 1997; Garron et al. 2005; Parsons and Cranston 2006; Walker 2016). A former nearby chlor-alkali plant reported a Hg release in combined effluent from the mill, which may explain elevated Hg concentrations in Boat Harbour (Taylor 2015). Mean total Hg (THg) concentrations measured ~25 years were higher than CCME freshwater and marine PELs. Furthermore, the THg contaminant inventory remains intact within the sediment profile, with potential for future contaminant disturbance or resuspension.

Metal(loid)s (including THg) may become more bioavailable to aquatic biota under certain anoxic conditions. For example, sediment bioavailability is affected by porosity, organic content, grain size, pH, redox, metal oxide and sulfide contents, and bioturbation, all of which can increase anoxic conditions (Nedwell and

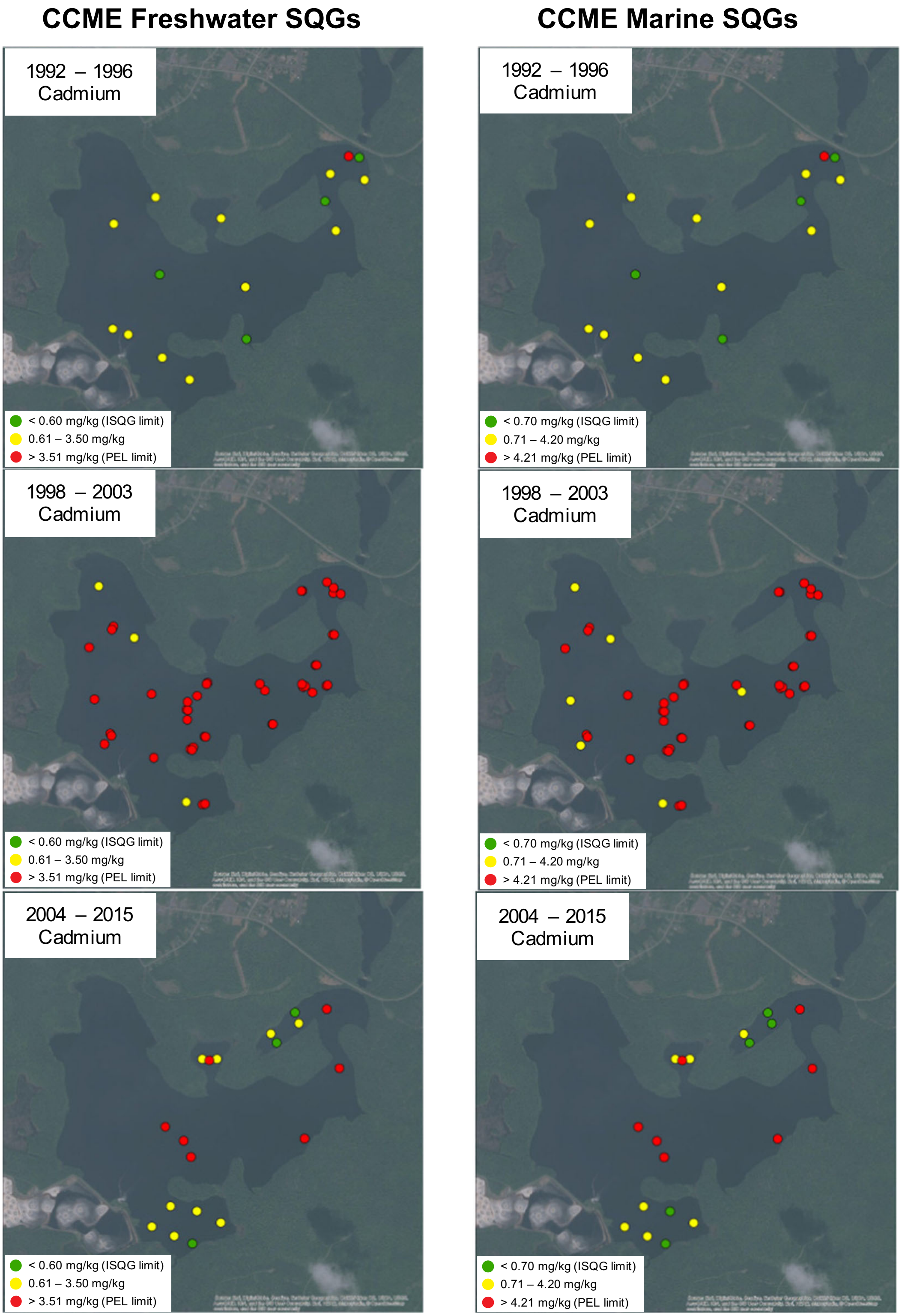

Fig. 6 Spatiotemporal variation of Cd concentrations compared to CCME freshwater (*green*, <0.60 [ISQG]; *yellow*, 0.61–3.50; *red*, >3.51 [PEL] mg/kg) and marine (*green*, <0.70 [ISQG]; *yellow*, 0.71–4.20; *red*, >4.21 [PEL] mg/kg) SQGs in Boat Harbour sediment over three periods: *top* 1992–1996, *middle* 1998–2003, and *bottom* 2004–2015

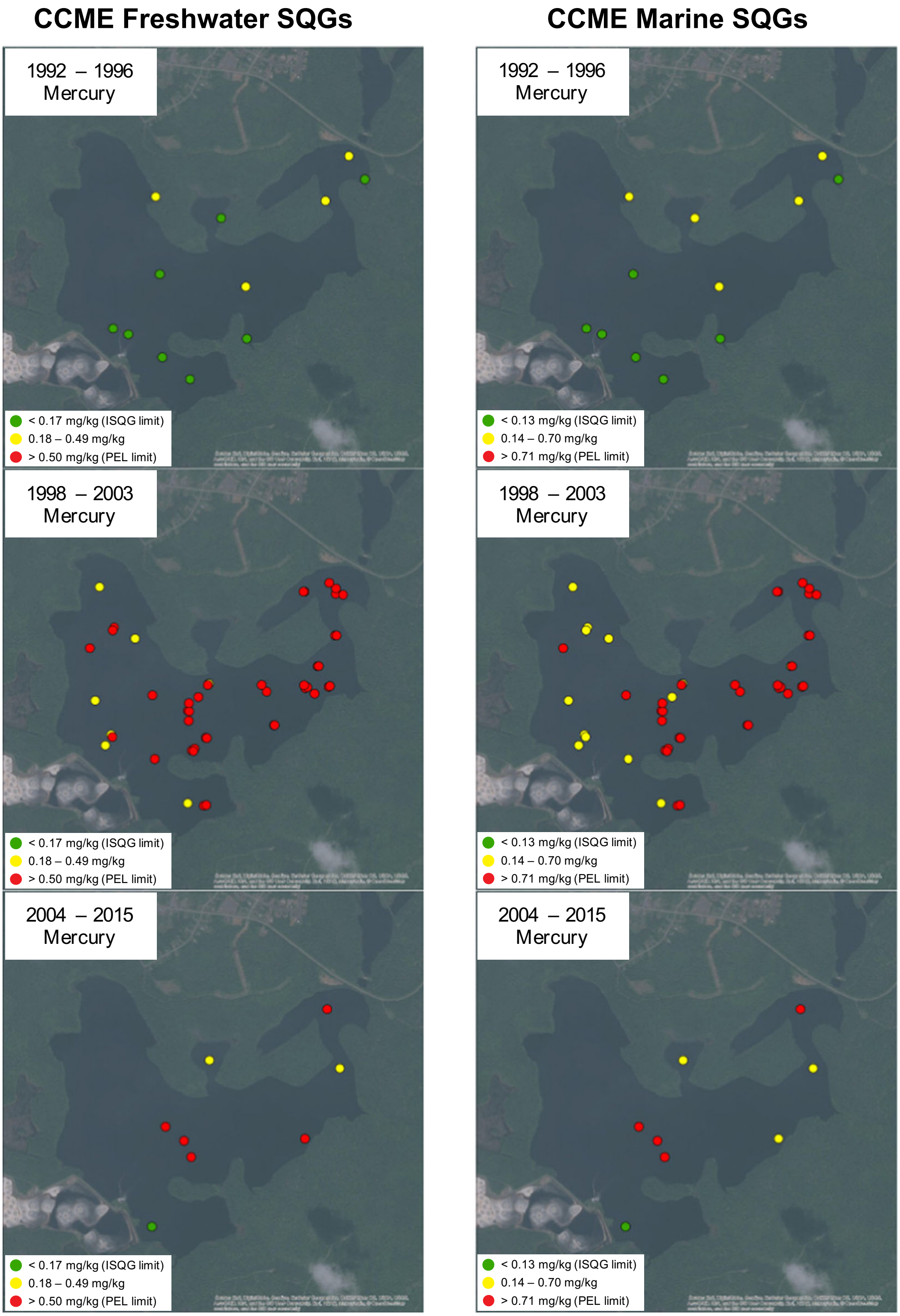

Fig. 7 Spatiotemporal variation of Hg concentrations compared to CCME freshwater (*green*, <0.17 [ISQG]; *yellow*, 0.18–0.49; *red*, >0.50 [PEL] mg/kg) and marine (*green*, <0.13 [ISQG]; *yellow*, 0.14–0.70; *red*, >0.71 [PEL] mg/kg) SQGs in Boat Harbour sediment over three periods: *top* 1992–1996, *middle* 1998–2003, and *bottom* 2004–2015

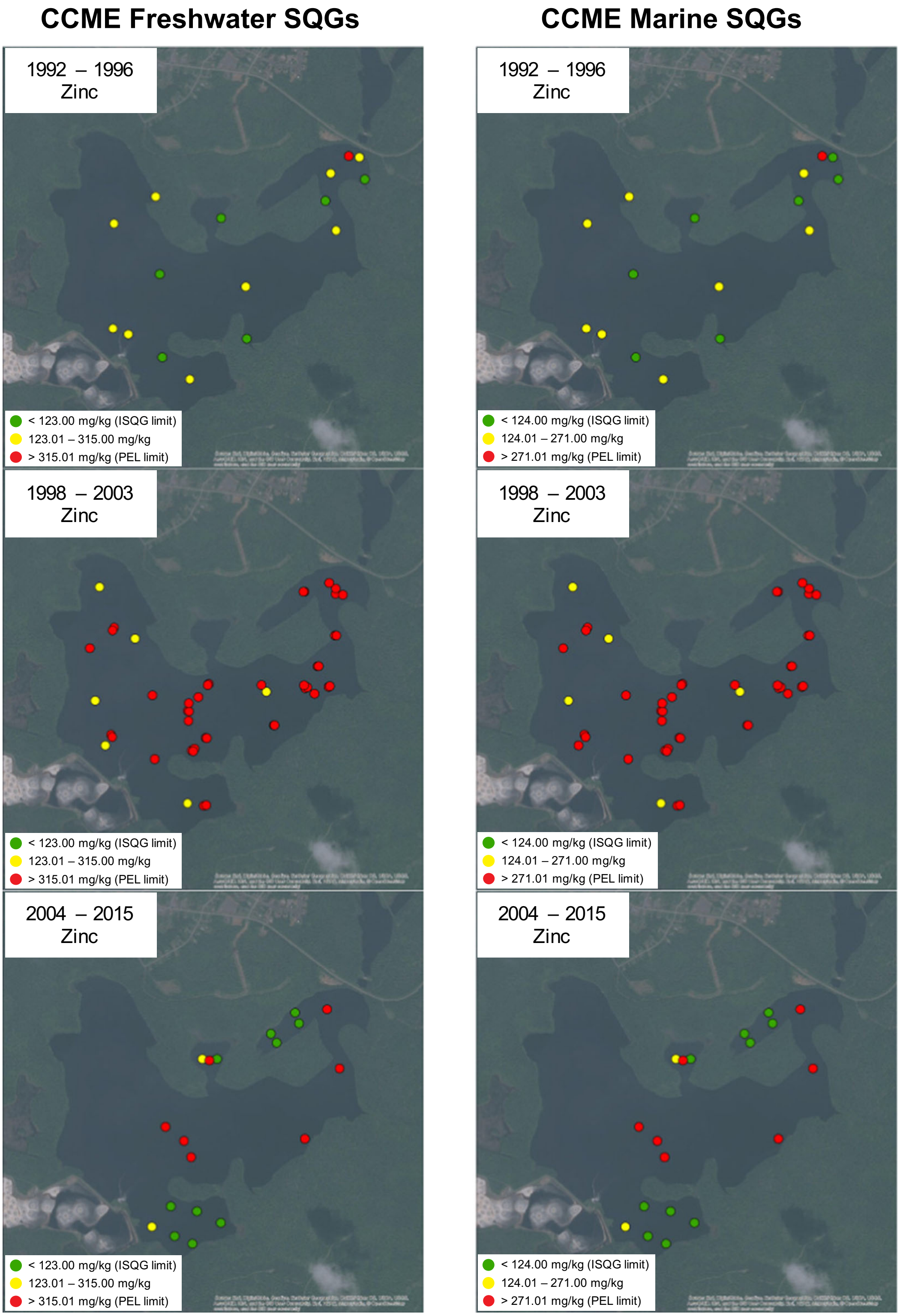

Fig. 8 Spatiotemporal variation of Zn concentrations compared to CCME freshwater (*green*, <123 [ISQG]; *yellow*, 123–315; *red*, >315 [PEL] mg/kg) and marine (*green*, <124 [ISQG]; *yellow*, 124–271; *red*, >271 [PEL] mg/kg) SQGs in Boat Harbour sediment over three periods: *top* 1992–1996, *middle* 1998–2003, and *bottom* 2004–2015

Walker 1995; Walker 2016). Under anoxic conditions, THg may become bioavailable to aquatic biota via the microbial process of methylation which forms methyl Hg (MeHg) (Walker 2016; Roach and Walker 2017). While previous studies analyzed THg, bioavailable MeHg was under-characterized in Boat Harbour sediments (Jackson 2016). Because of the potential for bioaccumulation, MeHg threatens aquatic biota and human health due to the consumption of fish (Zhang et al. 2012). Although a recent fish survey in Boat Harbour by Oakes (2016) found limited numbers of pollution tolerant fish (e.g., Mummichog and Ninespine sticklebacks), historical effluent discharge into Northumberland Strait may have released MeHg to the marine environment, warranting investigation prior to, during, and post remediation.

Additionally, a nearby coal-fired thermal generating station reported releases of atmospheric emissions of all six metal(loid)s identified in this study reported under the National Pollution Release Inventory (NPRI) program (Environment Canada 2014b). The mill's smoke stacks may have also contributed to metal(loid) burdens in Boat Harbour (Hoffman et al. 2015). However, according to a recent substance report submitted to NPRI, no on-site releases of metal(loid)s to air or water were reported by the mill (Environment Canada 2014b). However, Adoli et al. (2011) reported that mean concentrations of Cd, Cu, Cr, and Zn in mosses <1 km radius of a pulp mill were significantly higher than reference stations, suggesting atmospheric emissions from pulp mills may also contribute to local metal loadings. Atmospheric deposition of metals via the mill's smoke stacks may likely continue post remediation, despite installation of a precipitator in 2015 (Hoffman et al. 2015). To properly understand potential remediation management options or ecological risks associated with contaminated sediments in Boat Harbour and downgradient marine environments, more information related to sediment characteristics and potential sources are required.

Future monitoring and assessment to inform remedy decisions

Technical complexity at contaminated sediment sites arises from physical, chemical, and biological characteristics, spatial variability, and residual disturbance during and after remedial activities (e.g., change in contaminant bioavailability) (IRTC 2011). Due to inherent complexity of large-scale remediation projects, site characteristics (such as source areas, transport mechanisms, background and upstream areas, and key site features) should be clearly identified prior to evaluating and selecting remedial alternatives. Effective management of contaminated sediment requires systematic approaches to identify potential sources of contamination, understanding of spatial extent and magnitude of impacts and associated ecological risks prior to implementing costly remediation activities (Walker et al. 2013a, b; Walker 2014). Subsequent remediation of contaminated sediments can involve in situ or ex situ treatment, capping or follow-up monitoring programs (Magar and Wenning 2006). In Canada, management of contaminated aquatic sites follows established ecological risk frameworks and follows federal and provincial guidelines (under the Nova Scotia *Environment Act*) to help guide remediation decisions (e.g., Chapman 2011; Contaminated Sites Regulations 2013; CCME 2016), alongside engineering considerations and stakeholder engagement (e.g., the Mi'kmaq PLFN community). Stakeholder engagement, especially with elders in the community, will be crucial for understanding the historical pre-mill condition of Boat Harbour and surrounding environment. Stakeholder engagement combined with collection and measurement of local baseline data will help establish remediation end-point goals. For example, the ultimate remediation goal for Boat Harbour is to return it to pre-mill tidal conditions. This may involve ex situ removal or in situ treatment of sediments to achieve concentrations below low effect levels or comparable to local baseline conditions.

This review of Boat Harbour data revealed inherent gaps in sediment characteristics (vertical and spatial coverage) that suggests more detailed sediment sampling (e.g., piston coring, discrete sampling across vertical horizons and greater spatial coverage, and incremental sampling) is required to accurately determine volumes of unconsolidated sediment prior to remediation. Walker et al. (2015a) argued that characterizing contaminated sediments using discrete sampling, such as those used in this study, can produce high degrees of spatial and temporal variability. Incremental sampling can help overcome potential spatial and temporal variability and lower laboratory analysis costs. Another advantage of incremental sampling is that limited numbers of samples are required to achieve a representative and statistically valid characterization of a targeted area (ITRC 2012; Hadley and Bruce 2014). This would include the entire organic sediment profile deposited since the mill began operations. Discrete sampling is also recommended for accurate delineation of these impacted sediments. Prior to conducting high-resolution analysis of horizontally sectioned samples, it is essential to control for deformation when using cores to mitigate bias (Dunnington and Spooner 2017). Spatial analysis with adequate sample vertical and horizontal coverage to create *relief maps* or *concentration hotspots with depths* is also recommended. For example, accurate delineation of impacted surficial sediments (i.e., those exceeding low level [ISQGs] and high level effect levels [PELs]) would allow for removal or treatment of sediments to expose underlying perceived *cleaner* marine sediments, which presumably pose much less of a threat in terms of metal(loid)s. However, confirmatory sampling of underlying marine sediments in Boat Harbour is required prior to remediation to establish pre-mill *background* conditions and remediation end-point goals.

Background conditions typically refer to locations that are not influenced by releases from an industrial site or usually described as either naturally occurring (e.g., consistently present in the environment but not influenced by human activity) or anthropogenic (e.g., influenced by human activity but not related to specific activities at an industrial site) (USEPA 2002). For example, metals occur naturally in sediments around Nova Scotia due to geologic processes and mineralogy of the parent bedrock material (Loring et al. 1996). Background or reference conditions must be considered in virtually all stages of sediment investigations, remedial technology evaluations, and remedial response actions (USEPA 2002). Baseline monitoring over several years using multiple reference sites is critical for establishing benchmarks for comparing contaminated sediment sites pre- and post-remediation (Walker 2014), including baseline studies of water quality and biota in marine receiving waters prior to remediation (Walker et al. 2013c; Walker and MacAskill 2014; Roach and Walker 2017). Few studies have assessed Fergusons Pond or comparable reference sites in any great detail (JWEL 1997; JWEL 1999) but would warrant further study for comparison. Therefore, more extensive assessments (vertical and spatial coverage) using core sampling would be required to determine measurable changes of sediment chemistry at discrete depth intervals that approach baseline conditions.

To properly understand potential ecological risks associated with contaminated sediments in Boat Harbour and the Northumberland Strait, more information related to sediment characteristics are required. A baseline marine environmental effects monitoring (EEM) program (including fish tissue analysis [using crab or lobster hepatopancreas tissue chemistry and mussel or oyster tissue chemistry], sediment and water quality, etc.) should be established in the Northumberland Strait marine receiving environment prior to remediation or disturbance of Boat Harbour sediments. In addition, the EEM program could include up-gradient terrestrial environments (e.g., wetlands to collect water quality, soils, plant, and invertebrate tissue samples) that need to be properly characterized before remediation. The EEM program would also help inform decision makers on the effectiveness of remedy options (e.g., Walker 2014). An EEM program conducted during remediation of the Sydney Tar Ponds in Nova Scotia recommended that several years of baseline data be conducted prior to initiating a remediation program to reflect natural inter-annual variation (Walker 2014). Follow-up studies are also recommended to assess and compare measurable changes of conditions in Boat Harbour post remediation. Additional sampling and chemical analysis of Boat Harbour sediment will establish: (i) effectiveness of remediation in eliminating or minimizing environmental and human health risks, (ii) changes in physical and biochemical processes returning to a tidal lagoon, and (iii) provide temporal and spatial variability in Boat Harbour.

Conclusions

Decades of pulp mill effluent releases into Boat Harbour have resulted in deposition of large quantities of unconsolidated sludge and sediment that require remediation. Metal(loid) concentrations in sediment using a variety of sampling techniques (e.g., cores and grabs) and subsequent sub-sampling techniques (e.g., surface vs. deep sediment or composite vs. discrete) showed wide temporal variation between 1992 and 2015, with concentrations apparently peaking between 1998 and 2000. Despite variation of sampling techniques, Cd, Cr, Cu, Pb, Hg, and Zn frequently exceeded Canadian SQGs, posing significant risks to aquatic ecosystems. Results suggest the mill's effluent as the primary source of metal(loid) loadings; however, other local PS emitters may have contributed.

A critical review of historical data revealed inherent gaps in sediment characteristics, including vertical and spatial coverage. The Boat Harbour Act and the Mi'kmaq PLFN communities' desire to return Boat Harbour to a pre-mill tidal estuary state will require the ex situ removal or in situ treatment of sediments to achieve concentrations that are below low effect levels or at least comparable to local baselines conditions. To achieve remediation end-point goals, more detailed sediment sampling (e.g., piston coring, discrete sampling across vertical horizons, and greater spatial coverage) are required in Boat Harbour and nearby reference sites prior to costly remediation.

Acknowledgements This study was supported by Nova Scotia Lands. Valuable input and improvements to this paper were provided by representatives of the Boat Harbour Environmental Advisory Committee, Pictou Landing First Nation and two anonymous reviewers. We would also like to thank Farrah Stevens for confirming lack of reference site data.

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