Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Spatial patterns of sediment contamination and their influence on benthic infaunal communities in a highly tidal and industrial estuary in Atlantic Canada

Andrew J. Guerin^a, Karen A. Kidd^{a,b,*}, Marie-Josée Maltais^c, Angella Mercer^c, Heather L. Hunt^c

^a Department of Biology, McMaster University, Hamilton, ON, Canada

^b School of Earth, Environment and Society, McMaster University, Hamilton, ON, Canada

^c Department of Biological Sciences, University of New Brunswick, Saint John, NB, Canada

ARTICLE INFO

Keywords: Marine sediments Metal pollution Tidal harbours Infauna

ABSTRACT

Sediment contamination can be elevated in ports, harbours, and estuaries with legacies of exploitation, negatively impacting infaunal invertebrate communities. Saint John Harbour (45.25° N, 66.05° W), New Brunswick, Canada, is an active harbour with strong tides and a long history of human activity. To examine spatial patterns of sediment contamination, samples were collected between 2011 and 2021 from subtidal sites near potential contaminant sources. Invertebrate data from the same samples were used to investigate potential effects on biological communities. Contaminant concentrations in the inner parts of the harbour were elevated compared to reference sites, but generally did not reach levels comparable to other highly contaminated harbours in the region. Effects on invertebrates were detectable, particularly at sites with higher contamination, although physical factors (depth, sediment grain size) were more important. Dynamic tidal conditions in the harbour may reduce the accumulation of contaminants in subtidal sediments and their impacts on infaunal communities.

1. Introduction

Estuaries and natural harbours can be focal points for contamination from human activities (Mayer-Pinto et al., 2015), including local discharges of industrial effluents and municipal wastewaters (Gallagher and Keay, 1998; Solis-Weiss et al., 2004), intense shipping activity, modification of hydrodynamic regimes (Cutroneo et al., 2017), and upstream activities such as logging and mining (Schäfer et al., 2022). Sediments are a sink for many contaminants and, as such, sediment quality reflects these legacies of activity (Wetzel et al., 2013), with elevated contaminant levels in industrialized areas (e.g. Gibert et al., 2009; Walker et al., 2015). Contaminants of concern include metals (e. g., lead, mercury, zinc), which can have concentrations several orders of magnitude higher than natural background levels, and organic compounds such as polycyclic aromatic hydrocarbons (PAHs), some of which are only found in marine sediments as a result of anthropogenic activity (Eça et al., 2021). These contaminants can have negative impacts on the health and diversity of benthic organisms (Fukunaga et al., 2010; Schäfer et al., 2022), even at relatively low concentrations (Ellis et al., 2017; Nelson et al., 1988) and can be detrimental to human health, for example via ingestion of contaminated seafood (Okpala et al., 2018). Effects are often most severe close to pollution sources (Bubb and Lester, 1994), although contaminants can also be transported long distances from sources by currents (Gallagher and Keay, 1998).

Marine infaunal invertebrate communities are directly impacted by the quality of the sediments that they inhabit. Increasing sediment contamination generally leads to reductions in species richness and diversity, as well as changes in community composition; pollutionsensitive species decline in abundance with increasing contamination, while numbers of so-called 'opportunistic' species can increase (Grassle and Grassle, 1974; Mayer-Pinto et al., 2015; Olsgard and Hasle, 1993). The latter may also decline at higher impact levels (Dhainaut-Courtois et al., 2000; Ellis et al., 2017), and consequently overall abundances of sediment-dwelling invertebrates can be highest at intermediate levels of anthropogenic impact (Gray et al., 1990). Infaunal communities can respond relatively quickly to changes in sediment quality, and their limited range of movement means that spatial patterns in community composition and diversity can reflect spatial patterns in anthropogenic

https://doi.org/10.1016/j.marpolbul.2023.115872

Received 31 August 2023; Received in revised form 28 November 2023; Accepted 29 November 2023 Available online 14 December 2023





^{*} Corresponding author at: Department of Biology, McMaster University, Hamilton, ON, Canada. *E-mail address:* karenkidd@mcmaster.ca (K.A. Kidd).

⁰⁰²⁵⁻³²⁶X/© 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

impact (Hartley, 1982). Infaunal community sampling is therefore a common method for monitoring of marine environmental status, and for detection of contaminant impacts (e.g. Gray et al., 1990; Walker et al., 2015; Schäfer et al., 2022). While there is a large body of literature on sediment contamination and infaunal communities in active ports and harbours, few of these studies have examined sites with both considerable histories of anthropogenic exploitation and intense hydrodynamic regimes, which can in some cases be associated with lower sediment contamination (Callaway et al., 2020; Foulquier et al., 2020).

1.1. Saint John Harbour

Saint John Harbour, New Brunswick, Canada, is situated at the mouth of the Wolastoq (Saint John River), which has a drainage area of 55,400 km² (Marsh, 2015). The harbour has a tidal range of 8 m and is subject to strong tidal currents (Delpeche, 2006; Toodesh, 2012), making it an extremely dynamic environment. It also has a long history of anthropogenic impact, including historical mining in the river catchment (Ray and Macknight, 1984); discharge of municipal wastewater from Saint John (population 120,000); the presence of multiple industries including pulp and paper mills, a brewery, oil and gas import and export terminals, and an oil refinery; and the presence of Port Saint John, the largest port in Atlantic Canada, with navigable channels maintained by dredging. Despite these activities, past surveys have generally categorized the harbour as a whole as relatively unpolluted (Ray and Macknight, 1984; Wildish and Thomas, 1985), with severe sediment contamination restricted to areas close to point sources (Diesbourg et al., 2023).

The history of anthropogenic activity in Saint John Harbour makes it difficult to determine whether contaminant levels at particular sites indicate acute impacts, or are representative of background conditions, resulting from local geology combined with regional anthropogenic inputs. It is therefore necessary to establish a baseline for what can be considered 'normal' sediment conditions in the absence of genuinely pristine sites. One approach is to select a series of reference sites which are suitably distant from impact sources to be representative of minimally impacted conditions in the area, and to repeatedly sample these sites. The resulting data can be used to generate 'ranges of normal', which account for temporal variability within individual sites and background spatial variability among sites (Arciszewski and Munkit-trick, 2015). Measurements outside of these ranges are indicative of acute localised impacts, which could be the result of nearby anthropogenic activity, and can therefore be considered 'red flags', prompting further monitoring or possible management interventions.

1.2. Aims of current study

The aims of this study were to use data from samples collected from multiple sites close to potential sources of industrial contamination in Saint John Harbour to 1) characterise the spatial variability in sediment quality (metals, PAHs, grain size and organic carbon), 2) compare these measures to baseline ranges generated using data from reference sites in the Harbour (Guerin et al., 2023), and 3) examine the extent to which contaminants affect infaunal invertebrate communities in highly tidal coastal environments.

2. Methods

2.1. Field sampling

Samples were collected in Fall (Oct/Nov) 2011–2013 and 2017–2021 from sites in Saint John Harbour, New Brunswick (NB), Canada (Fig. 1, Table 1). Six reference sites were sampled on all sampling dates to generate baseline data for minimally contaminated sites and evaluate potential indicator species (Guerin et al., 2023; Pippy et al., 2016). An additional 15 sites were selected based on their proximity to potential sources of contamination (Table 1). Sites 15, 16, 24 and 25 were close to the Black Point dredge spoil disposal site. Sites 20–22 were within the Courtenay Bay channel north of the breakwater, close to the Irving Oil terminal and other industrial sites. Marsh Creek also flows into this section of the harbour; untreated municipal wastewater was discharged into this system prior to the commissioning of treatment



Fig. 1. Location and sampling frequency of sites in Saint John Harbour, New Brunswick, Canada. Sites 1–6 and 13 are reference sites (open squares), all other sites are potentially contaminated sites (filled circles; three sites with gray-filled symbols were sampled 5 or more times). Letters show some potential contaminant sources: A - outflow of Marsh Creek into harbour; B - outflow of Little River into harbour; C - pulp and paper mill; D - oil refinery; E metal recycling facility; F - crude oil and LNG terminal. Dotted polygon: boundary of the Black Point dredge disposal site. Right panel indicates years in which sites were sampled; reference sites were sampled in all years.

Table 1

Site information. Latitude and Longitude varied among individual samples; representative values are given. Depths are adjusted for tidal height at time of sampling.

Site	Latitude	Longitude	Depth (m)	Sampling Justification
1	45.24928	-66.0251	4.4-6.1	Reference site
2	45.23315	-66.06933	7.4–9.8	Reference site
3	45.22913	-66.02783	7.8–9.6	Reference site
4	45.20687	-66.06358	12.3-19.8	Reference site
6	45.20145	-65.96908	32.0-38.1	Reference site
13	45.20428	-66.1	16.3-21.1	Reference site
14	45.26635	-66.02947	0–4.9	Courtenay Bay, Little River, paper mill, oil refinery
15	45.21243	-66.0306	6.5–14.6	Proximity to Black Point dredge disposal site
16	45.21077	-66.04167	7.1–8.4	Proximity to Black Point dredge disposal site
19	45.24723	-66.05658	0.2–1.1	Digby Ferry Terminal, metal
20	45.27367	-66.04275	0–1.2	Oil terminal, industry, Marsh Creek
21	45.27088	-66.04248	4.6–5.2	Oil terminal, industry, Marsh Creek
22	45.26967	-66.04348	0–6.8	Oil terminal, industry, Marsh Creek
23	45.25052	-66.05677	0.2–14.8	Digby Ferry Terminal, metal
24	45.21975	-66.01765	10.2–11.2	Proximity to Black Point dredge
25	45.21617	-66.0255	12.1–12.6	Proximity to Black Point dredge
27	45.2541	-66.05977	2.4–3.2	Digby Ferry Terminal, metal
30	45.2736	-66.06768	0-5.5	Long Wharf. Port Saint John
31	45.26637	-66.06917	5.9-9.1	Port Saint John cargo terminal
32	45.26797	-66.07073	4.7–10.7	Port Saint John cargo terminal
33	45.25785	-66.06037	8.8–10.3	Digby Ferry Terminal, metal recycling plant

facilities from 2011 to 2014, and sediments in the creek and in the Harbour near shore are known to have very high concentrations of PAHs, mostly because of historical contamination with creosote (Diesbourg et al., 2023). Site 14 was in Courtenay Bay, near a paper mill, oil refinery and the mouth of Little River, another system known to have elevated PAH concentrations (Diesbourg et al., 2023; Loughery et al., 2014). Sites 30–32 were in the innermost portion of the harbour next to Port Saint John terminals, and near to a pulp and paper mill and the inflow from the mouth of the Wolastoq (Saint John River). Sites 19, 23, 27 and 33 were north of Partridge Island, close to the Digby ferry terminal and a metal recycling facility. Each site was sampled in one or more years, and reference sites were sampled in all years (Fig. 1). On each occasion five or more replicate sediment samples from each site were collected using a $\sim 0.1 \text{ m}^2$ Smith-McIntyre grab sampler; GPS positions and water depths of each grab were recorded.

2.2. Sample processing and analysis

Each sample was divided into two portions, one for physical / chemical analysis and one for enumeration of invertebrate infauna. Subsamples from the physical / chemical portion of each grab were collected using pre-cleaned 5 cm depth, 6.4 cm diameter (~160 ml volume) cores, and frozen at -20 °C for storage. After removal of macroinvertebrates and debris, 20 g aliquots were taken for loss-onignition (LOI) analyses; these were dried at 105 °C for 16 h, weighed, heated to 550 °C for 3.5 h, and reweighed. Percent loss after heating to 550 °C (LOI₅₅₀) was converted to percent total organic carbon (TOC) using an empirically-derived relationship based on 20 samples analysed directly for TOC by Research Productivity Council (RPC; Fredericton, NB) using a LECO combustion/infrared method. The remaining material was freeze-dried for a minimum of three days prior to further analysis.

For grain size determination, aliquots of 10-50 g dried sediment were shaken in a sequential sieve stack; material retained on each sieve, and below the finest sieve, was weighed and converted to percentages in four categories: Silt and Clay (< 0.125 mm), Fine and Medium Sand (0.125–0.5 mm), Coarse Sand (0.5–1 mm), and Granules and Pebbles (> 1 mm). PAHs were extracted from 10 g aliquots of homogenised sediment and quantified using a gas chromatograph-mass spectrometer (Agilent 6890/5975B GC-MS). Total mercury concentrations (herein 'mercury' or 'Hg') in 0.03 g aliquots of homogenised sediment were measured using a direct mercury analyser (Milestone DMA-80). Concentrations of 14 other potential metal/metalloid contaminants were also measured: As, Cd, Co, Cr, Cu, La, Mg, Mn, Ni, Pb, Rb, Sr, V, and Zn. Aliquots (0.5 g) of dried and homogenised samples were microwave digested in 10 ml of metal-grade nitric acid and diluted via addition of 40 ml of Milli-Q water. A known quantity of Yttrium was also added as an internal standard. Sample metal concentrations were then obtained using an inductively coupled plasma-optical emissions spectrophotometer (ICP-OES, iCAP 6500 Duo, Thermo Fisher Scientific). Element analysis for samples collected from 2011 to 2018 was conducted at UNB Saint John; from 2019 to 2021, samples were sent to RPC (Fredericton, NB, Canada); precision and detection limits are given in Supplementary Table S1. 2018 samples were analysed by both laboratories to allow comparison and possible correction. For additional details of field sampling and sample analysis protocols (including QA/QC procedures) refer to Van Geest et al. (2015) and Guerin et al. (2023). Data are reported on a dry weight basis.

The infaunal analysis portion (surface area 0.0321 m^2) was sieved through a 0.5 mm mesh under running seawater and retained organisms were preserved in 95 % ethanol. These were subsequently identified to the highest possible taxonomic resolution (usually species), although some species were merged to higher taxonomic levels when identification was uncertain or potentially inconsistent over time. Taxonomic names were verified using the World Register of Marine Species (WoRMS, 2022).

2.3. Data analysis outline

All data handling and analysis were done using R version 4.1.0 (R Development Core Team, 2021) excluding multivariate analyses which were done using PRIMER 7.0.21 with PERMANOVA+ (Anderson et al., 2008; Clarke and Gorley, 2015). As a result of the complexity of the combined physical, chemical and biological data, analysis was performed in several stages, following the necessary preprocessing (see following sections and Supplementary Table S2). A subset of data were first analysed to examine temporal variation at individual sites, since prior analysis of reference site data detected significant interannual variation (Guerin et al., 2023). Spatial variation among sites in physical and chemical variables was then examined, comparing measurements at individual sites to baseline data generated using the reference site samples, and to relevant sediment quality guidelines where possible, including the Threshold Effects Level (TEL) and Interim Sediment Quality Guideline (ISQG) levels (Buchman, 2008; CCME, 1999). Invertebrate data were compared among sites using a similar approach for univariate diversity measures, and using multivariate analysis. Finally, relationships between physical / chemical and biological data were examined using univariate and multivariate modelling approaches.

2.4. Data preparation

 $2.4.1. \ \ \text{Data preparation} - \text{sediment grain size, organic carbon and PAHs}$

Grain size, TOC, and PAH data required minimal preprocessing. PAH data were only available for 2012, 2013 and 2018, and not for all sites.

2.4.2. Data preparation – metals

Since mercury determinations were performed independently from other metals and in the same lab over time, no preprocessing was required. For other metals, previous comparison of concentrations measured by UNB and RPC for the same set of samples from 2018 showed that for most elements, data from the two laboratories were not comparable, and could not be combined (Guerin et al., 2023). However, data for two elements (copper, nickel) from the two labs were directly comparable without adjustment, and for two other elements (lead, zinc) data were amenable to simple mathematical correction; RPC-measured lead concentrations were therefore adjusted by +3.32 mg/kg, and RPC-measured zinc concentrations were adjusted by +3.34 mg/kg (Supplementary Table S2).

2.4.3. Data preparation – invertebrates

Invertebrate abundances were used to calculate three diversity indices for each sample; taxonomic richness, Shannon diversity, and AZTI Marine Biotic Index (AMBI; Borja et al., 2000). AMBI uses information on the ecology of sampled species to generate an index of pollution status, and was calculated using AMBI v6.0 (species list updated December 2020). For species absent from the AMBI database, the score for a suitable congener was used (3.6 % of species) or the species was assigned the score for a higher taxonomic level (9.1 % of species); if this was not possible, or if congeners had a range of different ecological group (EG) scores, the species was included in the data but marked as 'not assigned' (5.6 % of species, < 2 % of individuals for all samples, except one with 14.3 % 'not assigned'). Modified EG scores for North American marine environments (Gillett et al., 2015) were used for some species where appropriate (6.1 % of species). AMBI scores were not calculated for samples with fewer than 4 taxa or individuals, as AMBI can suffer from reduced robustness in such cases (Borja and Muxika, 2005); this included samples from Site 14 (one from 2013, one from 2018 and all samples from 2019), Site 22 (one sample from 2018, one from 2019 and two from 2020) and Site 30 (two samples from 2021).

For multivariate analysis, invertebrate abundance data were first fourth-root transformed; this down-weights numerically abundant taxa, preventing them from dominating subsequent analysis. Between-sample similarity matrices were calculated using the Bray-Curtis similarity measure, and centroids were calculated for each site on each sampling date, enabling the calculation of a matrix of similarities between sitedate pairs.

2.4.4. Data preparation – analyses linking biological with physical / chemical data

Only samples with matched physical / chemical and biological data were included for these analyses. Depth was also included as a potential explanatory variable. Preliminary data exploration revealed that Site 14 samples were very different in physical, chemical, and biological measures from those from other sites, and were therefore not included in the dataset for these analyses. PAH concentrations were not included as these were not consistently measured in all years. Remaining variables were log-transformed to reduce right skew, apart from Rb, which did not benefit from transformation, and percent Silt/Clay, which was inverselog transformed: SiltClay_{transformed} = 5 - log(101 - % SiltClay). Where transformed physical or chemical variables were highly correlated (Pearson's $\rho \ge$ 0.95), one was arbitrarily selected as representative (Supplementary Table S2). As metals data from the two laboratories were not comparable, two datasets were prepared for separate analysis; one including data from 2011 to 2013 and 2018 (using metal concentrations measured by UNB) and one including data from 2018 to 2021 (using metal concentrations measured by RPC).

2.4.5. Data preparation – 'normal' ranges for Saint John Harbour

Following the approach described in Arciszewski and Munkittrick (2015), data from the six reference sites were used to calculate 'normal' ranges for selected variables; these were defined as ± 2 standard deviations around the global mean for a site or group of sites. Selected variables were those metals (As, Cd, Cr, Cu, Hg, Ni, Pb, Zn) which had

available TEL / ISOG levels (Buchman, 2008; CCME, 1999), TOC, total PAH, AMBI, species richness, and Shannon diversity. Normal ranges were calculated for the Inner Harbour using data from reference Sites 1, 2 and 3, and for the Outer Harbour using data from reference Sites 4, 6 and 13, using the mean values of each site on each sampling date as replicates. For physical and chemical variables only, additional data were available from August 2011, April and June 2012, June 2013 and June 2014. These data were included in calculations of normal ranges (Guerin et al., 2023) since prior analysis established that their seasonal variation was minimal (Van Geest et al., 2015). For As and Cr, separate normal ranges were calculated using UNB data (2011-2014) and RPC data (2018-2021) since these data could not be combined. For Cd, normal ranges were only generated for 2018-2021, using RPC data, since all UNB Cd measurements were below detection limits. For Pb, separate normal ranges were calculated for 2012, and for 2013 onwards, since Pb concentrations appeared higher from 2013 onwards at the reference sites (Guerin et al., 2023).

2.5. Data analysis

2.5.1. Examination of within-site temporal variation

Most of the potentially contaminated sites were only sampled on one or two occasions. However, three sites (14, 22 and 23) were sampled 5 or more times (Fig. 1). These were examined to determine whether there was significant interannual variation. For each of these sites, the following variables were compared among years using ANOVA with *post-hoc* pairwise comparisons: percent Silt / Clay (Sites 22 and 23), percent Fine and Medium Sand (Site 14), TOC, mercury, copper, nickel, lead, zinc (the metals which could be analysed across all years), invertebrate species richness, Shannon diversity and AMBI. Multivariate species composition data were compared among years for each site using PERMANOVA with post-hoc comparisons. For each site, *p*-values for the ANOVA / PERMANOVA tests for the 12 variables tested were adjusted for multiple comparisons using the Benjamini and Hochberg (1995) method, as were *p*-values for pairwise comparisons for each variable.

2.5.2. Spatial patterns in physical and chemical variables

Variation among sites in grain size, TOC, PAH, and metal concentrations was examined by plotting the mean values for each site in each year, along with associated variability (\pm SE). For TOC, PAH, and metal concentrations, these were compared with the normal ranges generated using the reference site data, and relevant sediment quality guidelines.

2.5.3. Spatial patterns in biological data

Univariate biological indices (taxonomic richness, Shannon diversity, AMBI) were also examined by plotting them alongside the relevant normal ranges generated using the reference site data. For the multivariate data, variation in invertebrate community composition among calculated centroids for each site on each date was visualised using non-metric Multi-Dimensional Scaling (nMDS) ordinations. Twoway PERMANOVA analysis, with year and site as fixed factors, was also used to test for differences among sites. BVSTEP and SIMPER routines in PRIMER were used to identify a list of species which were most important in defining differences among sites and years.

2.5.4. Relationships between physical / chemical variables and infauna

To account for high collinearity among multiple explanatory variables, relationships between physical / chemical variables and species richness, Shannon diversity and AMBI were modelled by Partial Least Squares Regression (PLSR) using the pls package (Mevik and Wehrens, 2007). Data from all sites, including the reference sites, were used, apart from Site 14; data from this site differed substantially from all others in multiple variables. The numbers of PLSR components used in the final models were chosen to minimise Root Mean Square prediction error during leave-one-out cross-validation. Relative contributions of each variable to each of the PLSR models were examined using Variable Influence in Projection (VIP) scores (Wold et al., 2001) obtained using the *mixOmics* package (Rohart et al., 2017). To assess the overall influence of all metal contaminants, a second set of models were also run, using data from the same set of samples, but without metal concentrations.

DistLM models were used to explore the influence of metals on multivariate species composition, using the same physical and chemical data described above, and Bray-Curtis among-sample similarity matrices based on invertebrate data. To evaluate how the potential importance of metals concentrations varied at potentially contaminated sites compared to reference sites, models were run with and without metals data, for all sites together, for reference sites only, and for potentially contaminated Inner Harbour sites only.

Both the PLSR and DistLM analyses were first conducted for data from 2011 to 2013 and 2018, using metal concentrations measured at UNB. Subsequently, the analyses were repeated using the second dataset, which included samples collected from 2018 to 2021, using metal concentrations measured by RPC.

3. Results

3.1. Within-site temporal variation

There was significant interannual variation in physical/chemical and biological data at all three Inner Harbour sites (14, 22, 23) which were sampled five or more times, but there was little sign of consistent change over the period sampled (Table 2, Supplementary Fig. S1). At Site 14, mercury was the only one of the five metals tested that varied significantly (Table 2). Site 22 showed more evidence of statistically significant variability, with most differences involving samples collected in 2013, which had the highest silt and clay content and Cu concentrations, and relatively high richness and diversity. At Site 23, samples from 2013 again tended to differ from those collected in later years, with little other significant variation (Table 2, Supplementary Figs. S1, S2). However, there may have been a spatial component to this 'temporal' variation, since the sampled positions differed slightly in some years (Supplementary Fig. S3).

3.2. Spatial patterns in grain size and TOC

Sediments in the harbour were dominated by the finest fractions (Silt and Clay, grain size <0.125 mm), apart from at two sites (Sites 33 and 14; Fig. 2). Samples from Site 33 had typically <50 % Silt and Clay, with elevated proportions of Fine and Medium Sand (0.125–0.5 mm) and Granules and Pebbles (> 1 mm); at Site 14 sediments were

Table 2

Summary of statistical analyses of temporal variation at Sites 14, 22, and 23, for 7 physical / chemical variables, 4 biological indices, and multivariate species composition data for sediments from the Saint John Harbour, New Brunswick, Canada. Analyses were ANOVA for all univariate data, and PERMANOVA for community composition data. For full details (*F*-statistics, *p*-values) see Supplementary Table S3.

Site	Variables with significant temporal variation	Variables with no significant variation
14	% Fine & Medium Sand, TOC	
	Hg	Cu, Ni, Pb, Zn
	Species richness, AMBI	Shannon diversity
	Community composition	
22	% Silt & Clay, TOC	
	Cu, Ni, Pb, Zn	Hg
	Species richness, Shannon diversity	AMBI
	Community composition	
23	% Silt & Clay	TOC
	Pb	Hg, Cu, Ni, Zn
	Species richness, AMBI	Shannon diversity
	Community composition	

predominantly Fine and Medium Sand (Supplementary Fig. S4). Total organic carbon content was generally low (< 1 %) across most sites and dates, with mean values always <2 %. There was little evidence of organic enrichment at potentially impacted sites; mean TOC was generally within the range of normal values expected for the harbour, although it was slightly higher in some years at Sites 22, 30, and 32 (Fig. 2). At Site 14, TOC was very low, and below the expected normal range for the Inner Harbour.

3.3. Spatial patterns in metal and PAH concentrations

Sediment metal concentrations were highest in the inner reaches of the Harbour, declining at sites further out in the Inner and Outer Harbours (Fig. 3 and Supplementary Fig. S5). Apart from sites within inner Port Saint John (30-31) and Courtenay Bay (20-22,14) and Site 33, mean metal levels were always within the expected normal ranges. The highest measured concentrations for most metals were normally in inner Courtenay Bay (20–22); metal concentrations at these sites were usually above normal ranges (Figs. 3, 4, and Supplementary Fig. S5). However, concentrations were generally not extremely high and were usually below TEL, although Cu concentrations at Sites 20 and 22 in 2013 were slightly above TEL, and As and Ni were almost always above TEL at these sites. Concentrations at Sites 30-33 tended to be slightly lower, often falling within normal ranges. Site 14 differed markedly from the other sites, with elevated copper, lead and zinc concentrations; these were often higher than the relevant TELs, and even above the Probable Effects Level (PEL) threshold for zinc (271 mg/kg dw) in some individual samples. However, concentrations of some other metals (e.g. mercury, nickel) were similar to other Inner Harbour sites (20-22,30-32), and concentrations of chromium and arsenic were lower than at all other potentially contaminated sites, and below normal ranges on some dates. PAHs were measured on fewer dates (2012, 2013 and 2018) and mean values were always below the TEL of 1.684 mg/kg dw, although at Sites 20-22 they were consistently higher than the normal range for Inner Harbour sites (Fig. 4).

3.4. Spatial patterns in invertebrate infauna

Shannon diversity was lower in the Inner Harbour compared to the Outer Harbour (Fig. 5), with the lowest values being recorded at Sites 14, 21, and 22. Mean diversity was sometimes lower than the expected normal ranges derived from reference site data. Species richness was also lowest in the Inner Harbour, often below the expected normal range, especially at Sites 14 and 22 (Supplementary Fig. S6). Similarly, AMBI was higher in the Inner Harbour (implying greater contamination) although values were generally within relevant normal ranges, and AMBI values were remarkably low at Site 14 (Supplementary Fig. S6). Site 33 was unusual compared to adjacent sites (19, 23, 27), having higher levels of some contaminants but also higher richness and diversity, and lower AMBI than expected based on normal ranges for the Inner Harbour.

Invertebrate community composition varied among sites (Fig. 6); all sites differed significantly from all other sites in all years, with very few exceptions (PERMANOVA, Table 3). Outer Harbour sites (15, 16, 24, 25) formed a relatively tight cluster on the nMDS despite not all being sampled in the same year. These sites had similar species compositions to Outer Harbour reference sites (Supplementary Fig. S7); abundances of potentially pollution-sensitive species (e.g., *Nucula proxima* and *Nephtys incisa*) were comparatively high, some opportunistic polychaetes (*Cossura longocirrata, Tharyx* spp. and *Chaetozone* spp.) were present but not dominant, and opportunistic oligochaete species (*Clitellio arenarius* and *Tubificoides benedii*) were generally absent. In the Inner Harbour, Sites 19, 23, 27, 33 formed a group with relatively similar community composition. Opportunistic species (e.g., *C. arenarius, T. benedii*) were present (though not in very high abundances), while abundances of the pollution-sensitive bivalve *N. proxima* were low. These sites were also



Fig. 2. Percent Silt and Clay (grain size <0.125 mm; for other grain size fractions see Supplementary Fig. S4) and total organic carbon (TOC; $\% \pm$ SE), collected in the fall at potentially contaminated sites in the Saint John Harbour, New Brunswick, Canada. For TOC, dashed and dotted reference lines indicate upper and lower bounds of normal ranges for the Inner and Outer Harbour areas respectively, based on reference site data. Sites arranged based on approximate locations within the Harbour, moving further offshore left to right: inner Port of Saint John (30–32); inner Courtenay Bay (20–22); Courtenay Bay (14); Outer Port (33,19,23,27); Outer Harbour (24,25,15,16); see Fig. 1.

distinguished by moderate abundances of another polychaete species, *Micronephtys neotena*; this species was present in smaller numbers at a few other Inner Harbour sites but was very rare in the Outer Harbour. Of this group of sites, Site 27 was the least similar to the others, due to the presence of larger abundances of the opportunist *T. benedii*, but much lower abundances of *Cossura longocirrata*, *Tharyx* spp. and *Chaetozone* spp.

The six sites within the inner reaches of the Harbour (20-22, and 30-32) were distinct from the reference sites and tended to be more spatially and temporally variable (Fig. 6). Sites appeared to form two groups: those within Courtenay Bay (21,22, excluding 20) and those closest to the mouth of the Saint John River (30-32). Site 20, despite being in Courtenay Bay, had a more similar community composition to the latter group. All six sites had very low abundances of pollutionsensitive taxa, and higher abundances of several pollution-tolerant and opportunistic taxa (e.g. Cossura longocirrata, Polydora cornuta, Tharyx / Chaetozone spp., Clitellio arenarius and Tubificoides benedii; Supplementary Fig. S7). Sites 30-32 tended to have a somewhat higher overall species richness (Supplementary Fig. S6), but also higher abundances of C. arenarius and T. benedii. Finally, Site 14 was very distinct from all other sites in all years. Abundances of individual species (including opportunistic species) were very low (Supplementary Fig. S7). However, one reportedly pollution-sensitive species (Bathyporeia quoddyensis) was more abundant at this site than any of the others.

3.5. Relationships between physico-chemical and biological data

For the analysis using the UNB metals data (samples collected from 2011 to 2013, and 2018), the most important predictor of species richness, Shannon diversity and AMBI was depth; richness and diversity increased with depth, while AMBI declined (Fig. 7). Sediment grain

variables (percent silt / clay, percent granules / pebbles) were important predictors of diversity and AMBI but not richness; diversity was higher (and AMBI lower) in samples with less silt / clay and more coarse sediment. While most metals were clearly not important predictors, certain were, particularly mercury (which was one of the most influential variables for richness, diversity and AMBI), copper (somewhat important for diversity and AMBI), and lead (diversity). Analyses using the RPC-analysed metals data (samples from 2018 to 2021) showed similar patterns (Supplementary Fig. S8), with depth and sediment properties among the more important predictors, along with concentrations of some metals.

For all variables (richness, diversity, AMBI, species composition), using both UNB and RPC data, models including metals explained more variance than models excluding metals (Table 4). This difference was particularly marked for DistLM models including only samples from potentially contaminated sites in the Inner Harbour: the full model explained almost 80 % of the variance ($r^2 = 78$ % for Inner Harbour contaminated sites model including UNB metals data), compared to 56 % for the equivalent model without metals.

4. Discussion

4.1. Sediment contamination in Saint John Harbour

In general, Saint John Harbour sediments do not appear to be severely contaminated, even relatively close to potential pollutant sources. Metal concentrations in Outer Harbour sediments were generally within baseline ranges generated using data from reference sites. While concentrations in Inner Harbour sediments were often above these ranges, even at these sites contaminant concentrations were generally not much outside the normal ranges, and were mostly below



Fig. 3. Mean copper, nickel and zinc concentrations \pm SE (mg/kg dw) for each date sampled at potentially contaminated sites. Zinc RPC data (2019–2021) were adjusted by +3.34 mg/kg (see Methods for details). Dashed and dotted reference lines indicate upper and lower bounds of normal ranges for the Inner and Outer Harbour areas, respectively, based on reference site data (see Methods for details). Solid orange line indicates TEL / ISQG (Cu: 18.7 mg/kg; Ni: 15.9 mg/kg; Zn: 174 mg/kg), solid red line indicates PEL for Zn (271 mg/kg). PEL for Cu: 108 mg/kg and Ni: 42.8 mg/kg not shown. For additional metals (Pb, As, Cr, Cd), see Supplementary Fig. S5. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

TEL / ISQG thresholds. Concentrations of two metals, As and Ni, were often higher than their ISQGs. This likely reflects naturally-occurring background concentrations, since As concentrations, for example, are known to be high in much of New Brunswick (Klassen et al., 2009), and naturally elevated As concentrations are found in many marine sediments (Hartwell et al., 2018; Mirlean et al., 2012). Comparison with other harbours in Atlantic Canada suggests that sediment metal concentrations in much of Saint John Harbour (including As and Ni) are also similar to harbours considered minimally impacted by anthropogenic contamination (Table 5). Concentrations of most metals were lower than those found in sediments in Lunenberg and Sydney harbours, two Nova Scotia harbours with long histories of municipal and industrial contamination (Loring et al., 1996; Walker et al., 2015), and were up to two orders of magnitude lower than those in a heavily contaminated European port (Gibert et al., 2009). PAH levels in Saint John Harbour were also low compared to other harbours; mean TPAH at Inner Harbour sites was 0.51 mg/kg (range 0.03-2.05), which is similar to Sydney Harbour reference sites (< 0.5 mg/kg), and much lower than Sydney Harbour contaminated sites (up to 97 mg/kg; Walker et al., 2015). Comparisons with other studies should be viewed with caution, however, since differences can reflect methodological variation as well as actual differences in contamination. Nevertheless, these results corroborate previous findings that the subtidal areas of Saint John Harbour are not excessively contaminated (e.g. Ray and Macknight, 1984); it is likely

that this is driven by the relatively open nature of the harbour and the highly dynamic tidal regime, which ensure a high degree of water exchange and therefore rapid dispersal of contaminants. There have been few studies conducted in similarly energetic coastal sites over similar timescales. A study of Swansea Bay, UK, an area subject to strong hydrodynamic disturbance, found that anthropogenic influences on sediment communities were not substantial (Callaway et al., 2020). In this case the dynamic conditions were driven by an intense wave climate, rather than by tides, but the authors conclude that "ecosystems driven by a strong hydrodynamic regime can be relatively resistant to human activities"; this likely applies to Saint John Harbour.

However, at more contaminated sites, particularly Sites 20, 21 and 22, infaunal communities showed signs of being impacted by contamination; pollution-sensitive species were generally absent, and opportunistic species were present in higher abundances. These sites are sheltered behind a breakwater and subject to higher sediment deposition; this area of the port is also regularly dredged. There are also multiple potential contaminant sources nearby: an oil terminal, a pulp mill, and the outflow from Marsh Creek. As a result of historical contamination, PAH concentrations in Marsh Creek sediments are very high, ranging from 2.6 to 168 mg/kg (Diesbourg et al., 2023). It is therefore unsurprising that sediments at Sites 20–22 had higher PAH concentrations than other sites, although these were still below the ISQG threshold. PAH data were available for fewer sites in the harbour, and



Fig. 4. Mean total mercury concentrations \pm SE (µg/kg dw) and Total PAH \pm SE (mg/kg dw) for each date sampled at potentially contaminated sites. Dashed and dotted reference lines indicate upper and lower bounds of normal ranges for the Inner and Outer Harbour areas, respectively, based on reference site data (see Methods for details). Solid orange line indicates TEL / ISQG for Total PAH (1.684 mg/kg, PEL = 16.77 mg/kg). TEL/ISQG and PEL for mercury not shown (130 µg/kg and 700 µg/kg, respectively).



Fig. 5. Mean Shannon diversity (± SE) of infaunal invertebrates in Fall samples collected over several years at potentially contaminated sites in the Saint John Harbour, New Brunswick, Canada. Reference lines show upper and lower bounds for expected 'normal' ranges based on reference site data (see Methods for details): long dashes - Inner Harbour; short dashes - Outer Harbour (Sites 4 and 13); dots - Site 6.

only for some years; it may be beneficial to collect more data on PAH concentrations for a wider range of sites.

Site 14 was physically and chemically distinct from all other sites, having very low organic carbon content and low concentrations of PAHs and some metals (e.g. As, Cd, Cr). This is likely related to the physical properties of the sediment; sediments at Site 14 were predominantly Fine and Medium Sand, with very low Silt and Clay content. Sediment contaminant concentrations and organic carbon content are often strongly related to the proportion of finer sediments (Loring, 1979; Schintu et al., 2016), so lower contaminant concentrations at this site are unsurprising. However, concentrations of Cu, Pb, and Zn, three elements which are often highly correlated in contaminated systems

(Gibert et al., 2009), were considerably higher than at the other sites. This suggests that these metals were preferentially bound to other fractions, such as fine and medium sand, or that the limited amounts of finer sediments present were contaminated with extremely high levels of these metals. Measurement of metal concentrations for specific grain size fractions would be required to resolve this question. Ray and Macknight (1984) also found the highest concentrations of Cu and Zn in this area of the harbour, close to the mouth of Little River, attributing this to industrial pollution from the nearby oil refinery and paper mill. Compared to other parts of the harbour, the area is comparatively shallow and not subject to the same intense tidal stresses (Parrot et al., 2002), making it a possible site for accumulation of contaminants. The



Fig. 6. Spatial relationships for infaunal invertebrate data collected from the Saint John Harbour, New Brunswick, Canada in fall of several years: nMDS ordination based on distances among centroids for each year at each of the potentially contaminated sites. Centroids calculated from 5 samples at each site on each date, using Bray-Curtis similarities calculated from fourth root-transformed abundance data.

Table 3

2-way PERMANOVA table for comparison of invertebrate community composition among sites and years in Saint John Harbour, New Brunswick, Canada. Almost all pairwise comparisons were significant (p < 0.05); as such, only nonsignificant pairwise comparisons among sites (p > 0.05, after adjustment using the Benjamini and Hochberg (1995) method) are listed, excluding those comparing only reference sites.

Global PERMANOVA

GIODAI PERIMANOVA									
	df	SS	MS	Pseudo-F	р				
SITE	20	570,520	28,526	31.8	< 0.001				
YEAR	7	38,616	5516.6	6.15	< 0.001				
SITE * YEAR	54	152,302	2820.4	3.14	< 0.001				
Residual	338	303,190	897						
Total	419	1,106,100							
Pairwise comparisons among sites with $p.adj > 0.05$ (no significant difference)									
2012	none								
2013	24 vs 3	24 vs 3 ($p = 0.334$), 15 vs 25 ($p = 0.232$), 20 vs 22 ($p = 0.228$)							
2018	none	none							
2019	22 vs 2	22 vs 1 ($p = 0.07$)							
2020	none	none							
2021	none	none							

unique physical and chemical characteristics of this site were also reflected in the infauna. Species richness and diversity were among the lowest sampled - mostly below normal ranges based on Inner Harbour reference sites - which would be expected given the relatively high concentrations of Cu, Pb, and Zn; these are known to have additive effects on benthic infauna (Fukunaga et al., 2011). In contrast, AMBI values for samples from Site 14 included some of the lowest recorded, which would normally be associated with low contamination and good environmental status. This may reflect the fact that several samples from Site 14 were not assigned AMBI scores because too few individuals and species were present. Overall abundances at Site 14 were low, as were abundances of opportunistic species which respond to organic enrichment and are associated with fine sediments. Site 14, with little fine sediment and little organic carbon, may not be suitable habitat for these opportunists, regardless of contamination level, and AMBI may therefore not be a useful indicator of contamination status at this site.

4.2. Temporal changes

A previous analysis of Saint John Harbour reference site data did not identify any clear shifts in contaminant levels or infaunal communities that could be attributed to broader changes in the harbour, such as increases in shipping activity or the cessation of raw sewage discharges (Guerin et al., 2023). Similar changes in sewage management and anthropogenic contamination in other harbours and estuaries have had clear effects (Borja et al., 2006; Gallagher and Keay, 1998), but in these cases the marine sediments were more heavily contaminated to begin with, such that improvements were more easily identified. For Sites 14, 22 and 23, there was some significant interannual variation, but no clear directional change that might indicate environmental improvement or decline. Furthermore, at least some of this variation may be driven by differences in sampling location (Supplementary Fig. S3). Over longer time scales, comparison with published data from the 1970s-1980s (Ray and Macknight, 1984) also suggests that there has not been substantial change in contamination levels, although obviously the numbers and locations of samples collected are not equivalent (Table 5). Organic carbon levels in the harbour were also similar to those recorded in the 1970s (Wildish and Thomas, 1985) indicating little change in the intervening decades.



Fig. 7. Variable Influence in Projection (VIP) scores for physical and chemical variables included in univariate PLSR models using UNB metals data (2011–2013,2018) for (a) species richness, (b) Shannon diversity, and (c) AMBI for samples from the Saint John Harbour, New Brunswick, Canada. Variables with VIP < 1 (dashed line) are not typically considered influential predictors, but this is a guideline and not a definitive cut-off. Symbol fill indicates direction of predictor-response variable relationship: black filled symbols - positive relationship, open symbols - negative relationship, gray symbols - relationship unclear / very weak. GP = percent Granules and Pebbles; SC = percent Silt and Clay, CS = percent Coarse Sand. For VIP plots for RPC data (2018–2021) see Supplementary Fig. S8.

Table 4

Comparison of variance (r^2) explained by models with and without metals data for species richness, Shannon diversity and AMBI (best partial least-squares regression models) and multivariate species composition data (DistLM models) from sediment samples collected from the Saint John Harbour, New Brunswick, Canada. Site 14 data were excluded because all samples were substantially different from all other data in biological and physical / chemical variables.

	Model type	Sites	Data	UNB data	RPC data
				r ²	r ²
Species richness	PLSR	All sites	All variables	48.09	58.33
			No metals	32.48	34.55
Shannon diversity	PLSR	All sites	All variables	48.48	46.24
			No metals	40.73	24.35
AMBI	AMBI PLSR All sites		All variables	62.79	70.17
			No metals	50.86	61.87
Community composition	DistLM	All Sites	All variables	41.57	41.51
-			No metals	26.39	26.64
	DistLM	Reference sites only	All variables	41.61	42.76
			No metals	24.04	30.80
	DistLM	Potentially contaminated sites (Inner Harbour)	All variables	77.97	44.38
			No metals	56.39	24.96

4.3. Factors influencing infaunal communities in anthropogenically impacted coastal habitats

Physical factors (depth and sediment characteristics) were important predictors of infaunal community metrics and multivariate species composition in Saint John Harbour. This is consistent with studies on estuarine systems throughout the world, which have commonly found that infaunal abundance, richness, and diversity have significant relationships with depth and sediment composition (e.g. Currie and Small, 2005; Hartwell et al., 2018; Callaway et al., 2020). It is important to note that depth and sediment grain size may not influence infauna directly, but may be proxies for other, unmeasured variables (Jordà Molina et al., 2019; McArthur et al., 2010; Snelgrove and Butman, 1994). In Saint John Harbour, depth is also directly confounded with other variables, including metal concentrations: contamination and depth both tend to decline with distance from the mouth of the Saint John River. Other physical and environmental factors have also been found to be important for benthic communities, including temperature and oxygenation (Jordà Molina et al., 2019), but these are less likely to be important in Saint John Harbour, since the waters are very well mixed. Hydrodynamic conditions may be an important driver; Wildish and Thomas (1985) identified a large area of the harbour with a relatively impoverished infauna, which they attributed to a strong erosion-deposition cycle driven by tidal stresses. Some sites in the inner parts of the harbour can be considered intertidal; this includes Sites 19, 20, 22 and 30 (Table 1) which were sampled at high tide but are exposed at low tides. This would explain the presence of species such as Polydora cornuta which, in addition to being pollution-tolerant (Pearson and Rosenberg, 1978), is also found in intertidal environments (Levin, 1984).

Given the substantial output of freshwater from the Saint John River, variation in salinity may influence the infaunal communities in the harbour. Previous studies have detected reduced surface salinities at sites throughout Saint John Harbour, but as a result of the strong tidal influences the salt wedge penetrates well into the harbour at all states of the tide (Neu, 1960; Toodesh, 2012). Consequently, all sites within the Outer Harbour, along with reference Sites 1, 2, and 3 (Fig. 1), can reasonably be considered fully marine at the seabed under all conditions. However, the remaining sites may experience variable freshwater influence, with the magnitude and duration of dilution depending on their location and the level of freshwater discharge from the river. Under conditions of low discharge, salinity at the seabed may remain fully marine for most of the tidal cycle in almost all areas of the harbour. However, under high-discharge conditions such as the spring freshet, some sites may experience marked reductions in salinity. Freshwater influences on infauna would be strongest at Sites 30-32, which are closest to the mouth of the river, while Sites 20-22, being near to the outflow from Marsh Creek, could also experience reductions in salinity. However, without time series of direct measurements of salinity at the seabed, and ideally measurements of sediment pore water salinity, it is not possible to know what conditions the infauna experienced.

Metal concentrations, though generally low, had a statistically detectable effect on infauna. For every variable examined (richness,

Table 5

Selected mean metal concentrations (mg/kg dw, with ranges in italic font where provided) in sediments of Saint John Harbour and other locations in Atlantic Canada, and an example from a highly impacted European port (Barcelona, Spain). BD = Below Detection; minimum recorded values were less than the method detection limit.

	Years	As	Cd	Cr	Cu	Hg	Ni	Pb	V	Zn
Atlantic Canada, baseline / minimally contaminated harbours										
Bay of Fundy ^a	1970s	8	0.220	57	15	0.030	15	20	70	51
		4–15	.03-0.52	15-352	5–32	.02-0.09	3-46	8-42	27–136	18–104
Annapolis Basin, NS ^b	1988	5	0.040	76	12	0.020	17	16	55	39
A -		4–8	.03-0.08	50-226	9–20	.01-0.03	13–36	14–19	42–91	33-49
Pubnico Harbour, NS ^b	1993	6	0.190	43	9	0.020	18	20	47	42
		4-8	.09-0.34	30–60	7–11	.1-0.3	14-22	15–25	37–58	30-52
Sydney Harbour, NS ^c - reference sites	2013	4–13	0.3	6–12	2–13	0.1		4–13		24–61
Atlantic Canada, contaminated harbours										
Lunenberg Harbour, NS ^b	1002	14	1 310	93	58	0.220	27	42	91	159
Eulenberg Harbour, No	1992	5 20	24 1 71	85	56	0.220	16 73	42	50 100	10 204
Sudney Harbour NS south arm ^c	2013	12 22	.24-1.71	22 22	17 63	01027	10-75	35 120	50-100	49-304 80.250
Sydney Harbour, NS, south ann	2015	12-25	0.3-0.92	23-32	17-05	0.1-0.27		35-120		89-230
Highly contaminated harbour										
Barcelona ^d	2002–2005		0.9–19.8	32–109	55–784	0.9–20	18–47	52–696		106–1165
Saint John Harbour ^e										
Port Area ("Rodney Terminal")	1970s-1980s		0.10		12.6	0.020	16.3	20.9		46.7
,			.01-0.31		10-17	.01-0.04	9-40	14-39		34-59
Courtenay Bay			0.220		11.5	0.030	16.7	30.8		65.9
			.06-0.61		8-22	.01-0.3	7-27	9-67		44-106
Outer Harbour			0.08		15.1	0.03		21.9		69.2
			BD-0.4		8–106	BD-0.09		17–30		29-442
o total main f										
Saint John Harbour	0000		. 0.01	10.0	11		10	10.5	04.0	(5.7
"Inner Herbeur"	2022	4	< 0.01	12.8	11		10.0	12.5	24.2	05.7
Inner Harbour		0.33	0.08	19.8	10		18.2	12	29.8	35.8
Thi Can beach		10.2	0.03	10.2	21.5		17.2	83.2	28.5	119
Saint John Harbour (this study)										
Reference sites ($n = 253$)	2011-2021	6.1	*0.05	21.6	*7.3	*0.01	16.8	11.6	34.4	42.9
		2–13	BD-0.21	10-43	BD-18	BD-0.04	8–31	5–24	15-67	20-89
'Contaminated' sites ($n = 115$)		7.3	*0.07	24.8	13.2	0.020	20.1	16.8	37.6	56
		4–12	BD-0.17	15-49	6–32	.01-0.1	14–30	9–54	24–56	32–79
Site 14 (<i>n</i> = 30)		4	*0.05	16.1	37.1	0.010	22.1	**37.7	26.1	203.7
		3–5	BD-0.16	12–21	19–61	.005-0.02	16–28	**22-51	20–33	118–372

^a Loring (1979).

^b Loring et al. (1996).

^c Walker et al. (2015).

^d Gibert et al. (2009).

^e Ray and Macknight (1984).

^f Diesbourg et al. (2023).

* for calculation of means, values < detection limit have been replaced with random values below the relevant detection limit.

** excluding two samples with very high values: 136, 334 mg/kg.

diversity, AMBI, and community composition), models including metals data explained more of the variance than models including only physical variables and organic carbon content. This difference was particularly marked for models of community composition at potentially contaminated sites in the Inner Harbour, where depth was less likely to be an important variable (since it was more consistent among sites) and where sediment characteristics were mostly uniform. Impacts on the infauna can also be inferred from the higher abundances of opportunistic and pollution-tolerant taxa at the Inner Harbour sites. This adds to evidence from other studies that have shown that even where sediment contaminant concentrations are below thresholds of concern, impacts are still detectable (Ellis et al., 2017; Nelson et al., 1988). It is also important to note that while a range of sites within the harbour were sampled between 2011 and 2021, it is likely that areas of higher contamination exist, particularly within very nearshore, intertidal areas (Diesbourg et al., 2023).

4.4. Conclusions

Despite a long history of locally intense anthropogenic activity, the subtidal marine sediments of Saint John Harbour remain relatively free of serious contamination, particularly compared to similarly active ports and harbours in Atlantic Canada and elsewhere. It is likely that the most important reason for this is the highly dynamic tidal regime, since for much of the harbour's history, industrial effluents and raw domestic sewage have been discharged directly into the harbour and the rivers that drain into it. This is not to imply that there are no potential areas of concern. Fine sediments in the vicinity of Site 14 may be highly contaminated, and this warrants further investigation. High spatial variability in the inner reaches of Port Saint John implies that there could be locally high concentrations of contaminants at sites that were not sampled herein. Although metal and PAH concentrations were mostly below guideline levels, they still had a detectable influence on infaunal communities. Richness and diversity were lower at more contaminated sites, while AMBI was higher, and samples contained greater abundances of opportunistic, pollution-tolerant species.

Monitoring of harbour sediments should continue in order to detect any potential changes resulting from activities in the harbour, which may become apparent over longer timescales.

CRediT authorship contribution statement

Andrew J. Guerin: Data curation, Formal analysis, Writing – original draft. Karen A. Kidd: Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Writing – review & editing. Marie-Josée Maltais: Data curation, Investigation, Methodology, Writing – review & editing, Formal analysis. Angella Mercer: Data curation, Formal analysis, Investigation, Methodology, Writing – review & editing. Heather L. Hunt: Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Writing – review & editing.

Declaration of competing interest

Financial support for this project was received from the Canadian Water Network (WS2011-SJH-3 to HH; WS2011-SJH-2 to KK), Fisheries and Oceans Canada's Coastal Baseline Program (HH and KK), the Canada Foundation for Innovation (201843 to KK), the Jarislowsky Foundation (KK), and Port of Saint John (HH).

Data availability

Data are available from the St. Lawrence Global Observatory public repository: https://doi.org/10.26071/ogsl-1954d7f6-0fc1-42fa.

Acknowledgements

The crew of the Huntsman Marine Science Centre's research vessel Fundy Spray and many field assistants made the field sampling possible. Marie-Josée Abgrall, Lacey Haddon, and Tammy Bo assisted with lab analyses in addition to field work. Members of the Saint John Harbour Environmental Monitoring Partnership and Kelly Munkittrick provided advice on study design and data analysis.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2023.115872.

References

- Anderson, M.J., Gorley, R.N., Clarke, K.R., 2008. PERMANOVA+ for PRIMER: Guide to Software and Statistical Methods. PRIMER-e, Plymouth, UK.
- Arciszewski, T.J., Munkittrick, K.R., 2015. Development of an adaptive monitoring framework for long-term programs: an example using indicators of fish health: defining normal framework development. Integr. Environ. Assess. Manag. 11, 701–718. https://doi.org/10.1002/ieam.1636.
- Benjamini, Y., Hochberg, Y., 1995. Controlling the false discovery date: a practical and powerful approach to multiple testing. J. R. Stat. Soc. B 57, 289–300. https://doi. org/10.1111/j.2517-6161.1995.tb02031.x.
- Borja, A., Muxika, I., 2005. Guidelines for the use of AMBI (AZTI's Marine Biotic Index) in the assessment of the benthic ecological quality. Mar. Pollut. Bull. 50, 787–789. https://doi.org/10.1016/j.marpolbul.2005.04.040.
- Borja, Å., Franco, J., Pérez, V., 2000. A marine biotic index to establish the ecological quality of soft-bottom benthos within European estuarine and coastal environments. Mar. Pollut. Bull. 40, 1100–1114. https://doi.org/10.1016/S0025-326X(00)00061-8.
- Borja, Á., Muxika, I., Franco, J., 2006. Long-term recovery of soft-bottom benthos following urban and industrial sewage treatment in the Nervión estuary (southern Bay of Biscay). Mar. Ecol. Prog. Ser. 313, 43–55. https://doi.org/10.3354/ meps313043.
- Bubb, J.M., Lester, J.N., 1994. Anthropogenic heavy metal inputs to lowland river systems, a case study. The River Stour, U.K. Water Air Soil Pollut. 78, 279–296. https://doi.org/10.1007/BF00483037.
- Buchman, M.F., 2008. Screening Quick Reference Tables (SQuiRTs) (No. NOAA OR&R Report; 08-1). National Oceanic and Atmospheric Administration, United States.

- Callaway, R., Fairley, I., Horrillo-Caraballo, J., 2020. Natural dynamics overshadow anthropogenic impact on marine fauna at an urbanised coastal embayment. Sci. Total Environ. 716, 137009 https://doi.org/10.1016/j.scitotenv.2020.137009.
- CCME, 1999. Canadian sediment quality guidelines for the protection of aquatic life. In: Canadian Environmental Quality Guidelines. Winnipeg, MB.
- Clarke, K.R., Gorley, R.N., 2015. PRIMER v7: User Manual/Tutorial. PRIMER-e, Plymouth, UK.
- Currie, D.R., Small, K.J., 2005. Macrobenthic community responses to long-term environmental change in an east Australian sub-tropical estuary. Estuar. Coast. Shelf Sci. 63, 315–331. https://doi.org/10.1016/j.ecss.2004.11.023.
- Cutroneo, L., Carbone, C., Consani, S., Vagge, G., Canepa, G., Capello, M., 2017. Environmental complexity of a port: evidence from circulation of the water masses, and composition and contamination of bottom sediments. Mar. Pollut. Bull. 119, 184–194. https://doi.org/10.1016/j.marpolbul.2017.03.058.
- Delpeche, N., 2006. Observations of advection and turbulent interfacial mixing in the Saint John River estuary, New Brunswick, Canada. (M.Sc.E.). University of New Brunswick., Fredericton, NB, Canada.
- Dhainaut-Courtois, N., Pruvot, C., Empis, A., Baudet, K., 2000. Les peuplements macrozoobenthiques, indicateurs des qualités physico-chimiques et chimiques des sédiments portuaires - exemple du port de Boulogne-sur-Mer (Manche). Bull. Soc. Zool. Fr. 125, 49–62.
- Diesbourg, E., MacDonald, M., Reid, H.B., MacKinnon, R., Reinhart, B., Mercer, A., Crémazy, A., 2023. State of polycyclic aromatic hydrocarbon (PAH) contamination in the Saint John Harbour, New Brunswick, Canada. Mar. Pollut. Bull. 189, 114760 https://doi.org/10.1016/j.marpolbul.2023.114760.
- Eça, G.F., Albergaria-Barbosa, A.C.R., de Souza, M.M., Costa, P.G., Leite, A.S., Fillmann, G., Hatje, V., 2021. Polycyclic aromatic hydrocarbons in sediments and shellfish from Todos os Santos bay, Brazil. Mar. Pollut. Bull. 173, 112944 https:// doi.org/10.1016/j.marpolbul.2021.112944.
- Ellis, J.I., Clark, D., Atalah, J., Jiang, W., Taiapa, C., Patterson, M., Sinner, J., Hewitt, J., 2017. Multiple stressor effects on marine infauna: responses of estuarine taxa and functional traits to sedimentation, nutrient and metal loading. Sci. Rep. 7, 12013. https://doi.org/10.1038/s41598-017-12323-5.
- Foulquier, C., Baills, J., Blanchet, H., D'Amico, F., Rihouey, D., 2020. Soft-bottom community responses in a marine area influenced by recurrent dumping activities and freshwater discharges. Mar. Pollut. Bull. 156, 111259 https://doi.org/10.1016/ j.marpolbul.2020.111259.
- Fukunaga, A., Anderson, M., Webster-Brown, J., Ford, R., 2010. Individual and combined effects of heavy metals on estuarine infaunal communities. Mar. Ecol. Prog. Ser. 402, 123–136. https://doi.org/10.3354/meps08457.
- Fukunaga, A., Anderson, M.J., Webster-Brown, J.G., 2011. Assessing the nature of the combined effects of copper and zinc on estuarine infaunal communities. Environ. Pollut. 159, 116–124. https://doi.org/10.1016/j.envpol.2010.09.012.
- Gallagher, E., Keay, K., 1998. Organism-sediment-contaminant interactions in Boston Harbor. In: Stolzenbach, K.D., Adams, E.E. (Eds.), Contaminated Sediments in Boston Harbour. MIT Sea Grant Publication 98-1, Cambridge, MA, pp. 89–132.
- Gibert, O., Martínez-Lladó, X., Martí, V., Díez, S., Romo, J., Bayona, J.M., de Pablo, J., 2009. Changes of heavy metal and PCB contents in surficial sediments of the Barcelona harbour after the opening of a new entrance. Water Air Soil Pollut. 204, 271. https://doi.org/10.1007/s11270-009-0044-6.
- Gillett, D.J., Weisberg, S.B., Grayson, T., Hamilton, A., Hansen, V., Leppo, E.W., Pelletier, M.C., Borja, A., Cadien, D., Dauer, D., Diaz, R., Dutch, M., Hyland, J.L., Kellogg, M., Larsen, P.F., Levinton, J.S., Llansó, R., Lovell, L.L., Montagna, P.A., Pasko, D., Phillips, C.A., Rakocinski, C., Ranasinghe, J.A., Sanger, D.M., Teixeira, H., Dolah, R.F.V., Velarde, R.G., Welch, K.I., 2015. Effect of ecological group classification schemes on performance of the AMBI benthic index in US coastal waters. Ecol. Indic. 50, 99–107. https://doi.org/10.1016/j.ecolind.2014.11.005.
- Grassle, J.F., Grassle, J.P., 1974. Opportunistic life histories and genetic systems in marine benthic polychaetes. J. Mar. Res. 32, 253–284.
- Gray, J., Clarke, K., Warwick, R., Hobbs, G., 1990. Detection of initial effects of pollution on marine benthos: an example from the Ekofisk and Eldfisk oilfields, North Sea. Mar. Ecol. Prog. Ser. 66, 285–299. https://doi.org/10.3354/meps066285.
- Guerin, A.J., Kidd, K.A., Maltais, M.-J., Mercer, A., Hunt, H.L., 2023. Temporal and spatial trends in benthic infauna and potential drivers, in a highly tidal estuary in Atlantic Canada. Estuar. Coasts. https://doi.org/10.1007/s12237.023.01222.w
- Atlantic Canada. Estuar. Coasts. https://doi.org/10.1007/s12237-023-01222-w. Hartley, J.P., 1982. Methods for monitoring offshore macrobenthos. Mar. Pollut. Bull. 13, 150–154. https://doi.org/10.1016/0025-326X(82)90084-4.
- Hartwell, S.I., Apeti, A.D., Pait, A.S., Radenbaugh, T., Britton, R., 2018. Benthic habitat contaminant status and sediment toxicity in Bristol Bay, Alaska. Reg. Stud. Mar. Sci. 24, 343–354. https://doi.org/10.1016/j.rsma.2018.09.009.
- Jordà Molina, È., Silberberger, M.J., Kokarev, V., Reiss, H., 2019. Environmental drivers of benthic community structure in a deep sub-arctic fjord system. Estuar. Coast. Shelf Sci. 225, 106239 https://doi.org/10.1016/j.ecss.2019.05.021.
- Klassen, R.A., Douma, S.L., Ford, A., Rencz, A., Grunsky, E., 2009. Geoscience modelling of relative variation in natural arsenic hazard potential in New Brunswick (No. 2009–7). https://doi.org/10.4095/247834.
- Levin, L.A., 1984. Life history and dispersal patterns in a dense infaunal polychaete assemblage: community structure and response to disturbance. Ecology 65, 1185–1200. https://doi.org/10.2307/1938326.
- Loring, D.H., 1979. Baseline levels of transition and heavy metals in the bottom sediments of the Bay of Fundy. Proc N S Inst Sci 29, 335–346.
- Loring, D.H., Rantala, R.T.T., Milligan, T.G., 1996. Metallic contaminants in the sediments of coastal embayments of Nova Scotia. Can. Tech. Rep. Fish. Aquat. Sci. 2111, 268.
- Loughery, J.R., Arciszewski, T.J., Kidd, K.A., Mercer, A., Hewitt, L.M., MacLatchy, D.L., Munkittrick, K.R., 2014. Understanding the chronic impacts of oil refinery

A.J. Guerin et al.

- Marsh, J.H., 2015. Saint John River [WWW Document]. The Canadian Encyclopedia. URL. https://www.thecanadianencyclopedia.ca/en/article/saint-john-river (accessed 7.28.22).
- Mayer-Pinto, M., Johnston, E.L., Hutchings, P.A., Marzinelli, E.M., Ahyong, S.T., Birch, G., Booth, D.J., Creese, R.G., Doblin, M.A., Figueira, W., Gribben, P.E., Pritchard, T., Roughan, M., Steinberg, P.D., Hedge, L.H., 2015. Sydney Harbour: a review of anthropogenic impacts on the biodiversity and ecosystem function of one of the world's largest natural harbours. Mar. Freshw. Res. 66, 1088–1105. https:// doi.org/10.1071/MF15157.
- McArthur, M.A., Brooke, B.P., Przesławski, R., Ryan, D.A., Lucieer, V.L., Nichol, S., McCallum, A.W., Mellin, C., Cresswell, I.D., Radke, L.C., 2010. On the use of abiotic surrogates to describe marine benthic biodiversity. Estuar. Coast. Shelf Sci. 88, 21–32. https://doi.org/10.1016/j.ecss.2010.03.003.
- Mevik, B.-H., Wehrens, R., 2007. The pls package: principal component and partial least squares regression in R. J. Stat. Soft. 18 https://doi.org/10.18637/jss.v018.i02.
- Mirlean, N., Medeanic, S., Garcia, F.A., Travassos, M.P., Baisch, P., 2012. Arsenic enrichment in shelf and coastal sediment of the Brazilian subtropics. Cont. Shelf Res. 35, 129–136. https://doi.org/10.1016/j.csr.2012.01.006.
- Nelson, D.A., Miller, J.E., Calabrese, A., 1988. Effect of heavy metals on bay scallops, surf clams, and blue mussels in acute and long-term exposures. Arch. Environ. Contam. Toxicol. 17, 595–600. https://doi.org/10.1007/BF01055828.
- Neu, H.A., 1960. Hydrographic Survey of Saint John Harbour, N. B., Mechanical Engineering Report. National Research Council of Canada. https://doi.org/10.4224/ 40001823.
- Okpala, C.O.R., Sardo, G., Vitale, S., Bono, G., Arukwe, A., 2018. Hazardous properties and toxicological update of mercury: from fish food to human health safety perspective. Crit. Rev. Food Sci. Nutr. 58, 1986–2001. https://doi.org/10.1080/ 10408398.2017.1291491.
- Olsgard, F., Hasle, J.R., 1993. Impact of waste from titanium mining on benthic fauna. J. Exp. Mar. Biol. Ecol. 172, 185–213. https://doi.org/10.1016/0022-0981(93) 90097-8.
- Parrot, D.R., Cranston, R., Li, M., Parsons, M., Kostolev, V., 2002. Monitoring and Evaluation of Conditions at the Black Point Ocean Disposal Site (Report for Environment Canada). Natural Resources Canada, Dartmouth, NS.
- Pearson, T.H., Rosenberg, R., 1978. Macrobenthic succession in relation to organic enrichment and pollution of the marine environment. Oceanography and Marine Biology - An Annual Review 16, 229–311.
- Pippy, B.A., Kidd, K.A., Munkittrick, K.R., Mercer, A., Hunt, H., 2016. Use of the Atlantic nut clam (*Nucula proxima*) and catworm (*Nephtys incisa*) in a sentinel species approach for monitoring the health of Bay of Fundy estuaries. Mar. Pollut. Bull. 106, 225–235. https://doi.org/10.1016/j.marpolbul.2016.02.065.
 R Development Core Team, 2021. R, a language and environment for statistical
- R Development Core Team, 2021. R, a language and environment for statistical computing.

- Marine Pollution Bulletin 198 (2024) 115872
- Ray, S., Macknight, S.D., 1984. Trace metal distributions in Saint John Harbour sediments. Mar. Pollut. Bull. 15, 12–18. https://doi.org/10.1016/0025-326X(84) 90417-X.
- Rohart, F., Gautier, B., Singh, A., Lê Cao, K.-A., 2017. mixOmics: an R package for 'omics feature selection and multiple data integration. PLoS Comput. Biol. 13, e1005752 https://doi.org/10.1371/journal.pcbi.1005752.
- Schäfer, J., Coynel, A., Blanc, G., 2022. Impact of metallurgy tailings in a major European fluvial-estuarine system: trajectories and resilience over seven decades. Sci. Total Environ. 805, 150195 https://doi.org/10.1016/j.scitotenv.2021.150195.
- Schintu, M., Marrucci, A., Marras, B., Galgani, F., Buosi, C., Ibba, A., Cherchi, A., 2016. Heavy metal accumulation in surface sediments at the port of Cagliari (Sardinia, western Mediterranean): environmental assessment using sequential extractions and benthic foraminifera. Mar. Pollut. Bull. 111, 45–56. https://doi.org/10.1016/j. marpolbul.2016.07.029.

Snelgrove, P., Butman, C.A., 1994. Animal sediment relationships revisited – cause versus effect. Oceanogr. Mar. Biol. 32, 111–177.

- Solis-Weiss, V., Aleffi, F., Bettoso, N., Rossin, P., Orel, G., Fonda-Umani, S., 2004. Effects of industrial and urban pollution on the benthic macrofauna in the Bay of Muggia (industrial port of Trieste, Italy). Sci. Total Environ. 328, 247–263. https://doi.org/ 10.1016/j.scitotenv.2004.01.027.
- Toodesh, R., 2012. The Oceanographic Circulation of the Port of Saint John over Seasonal and Tidal Time Scales (M.Sc.E.). University of New Brunswick, Fredericton, NB, Canada.
- Van Geest, J.L., Kidd, K.A., Hunt, H.L., Abgrall, M.-J., Maltais, M.-J., Mercer, A., 2015. Development of baseline data for long-term monitoring of sediment conditions at reference sites in Saint John Harbour, New Brunswick: benthic infaunal invertebrates and sediment contaminants 2011-2013. Can. Manuscr. Rep. Fish. Aquat. Sci. 3076, 97.
- Walker, T.R., Willis, R., Gray, T., Maclean, B., McMillan, S., Leroy, M., Appleton, R., Wambolt, N., Smith, M., 2015. Ecological risk assessment of sediments in Sydney Harbour, Nova Scotia, Canada. Soil Sediment Contam. 24, 471–493. https://doi.org/ 10.1080/15320383.2015.982244.
- Wetzel, M.A., Wahrendorf, D.-S., von der Ohe, P.C., 2013. Sediment pollution in the Elbe estuary and its potential toxicity at different trophic levels. Sci. Total Environ. 449, 199–207. https://doi.org/10.1016/j.scitotenv.2013.01.016.
- Wildish, D.J., Thomas, M.L.H., 1985. Effects of dredging and dumping on benthos of Saint John Harbour, Canada. Mar. Environ. Res. 15, 45–57. https://doi.org/ 10.1016/0141-1136(85)90037-6.
- Wold, S., Sjöström, M., Eriksson, L., 2001. PLS-regression: a basic tool of chemometrics. Chemom. Intel. Lab. Syst. 58, 109–130. https://doi.org/10.1016/S0169-7439(01) 00155-1.
- WoRMS, 2022. World Register of Marine Species [WWW Document]. URL. https://www. marinespecies.org/ (accessed 9.19.22).