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Baseline

Mercury concentrations in marine sediments near a former mercury cell chlor-alkali plant in eastern Canada

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ABSTRACT

Concentrations of total mercury (THg) were measured in coastal marine sediments near a former chlor-alkali plant in Chaleur Bay, New Brunswick. The chlor-alkali plant has been a local point source of THg since operation began in 1963. Historical THg contamination of marine sediments and biota has been widely reported. No baseline assessment has been conducted following plant closure in 2008. Surface (0–2 cm) oxidized marine sediments were sampled along a single 5.2 km transect radiating from the former plant and analysed for THg. THg concentrations ranged from 0.04–0.28 μg g^{-1}. Some localised THg concentrations exceeded Canadian marine sediment quality guidelines (n = 4), but all samples (n = 14) were significantly lower than previous studies conducted during plant operation. Plant closure (source control) and natural sediment recovery likely responsible for attenuating THg concentrations, but burial in deeper anoxic sediments may increase bioavailability of Hg that could pose ecological risks to marine biota.

Global mercury (Hg) concentrations have been increasing since the industrial revolution (Dietz et al., 1996; Campbell et al., 2005). Industries contributing to global Hg accumulation include coal combustion, gold mining and chlor-alkali plants (Seritti et al., 1987; UNEP, 2002). Anthropogenic Hg releases from industrial activities have caused widespread pollution in the marine environment by accumulating in coastal sediments and bioaccumulating in marine biota (Campbell et al., 2005; Walker et al., 2013a, 2013b, 2013c, 2013d; Walker and MacAskill, 2014; Walker et al., 2015; Walker and Grant, 2015).

Chaleur Bay in northeastern New Brunswick, Canada has received Hg inputs from a variety of anthropogenic sources. Former industries in the area included a chlor-alkali plant, pulp and paper mill (both closed in 2008) and a thermal generating station (closed in 2011), all known for Hg releases in the environment. The largest industrial use of Hg during the 20th century was the chlor-alkali process which used electrolysis (Hg being the anode) for separating chlorine (for bleaching in the pulp and paper industry) and sodium (to make caustic soda) from brine (Leopold, 2002). The chlor-alkali plant located in Dalhousie, New Brunswick used Hg cell technology to produce chlorine and caustic soda (Trip et al., 2000). Production began in 1963 with treated effluents discharged into Chaleur Bay (Fig. 1). Contamination sources from the chlor-alkali process were effluents and atmospheric emissions, reportedly releasing 1.5 and 45.6 kg of Hg, respectively in 2002 (EC, 2014), making it one of the highest local point sources of Hg in the region during operations (Wilson and Travers, 1976; Sensen and Richardson, 2002; Garron et al., 2005; Fraser et al., 2011). Effluents from Hg chlor-alkali production are regulated under the *Chlor-Alkali Mercury Liquid Effluent Regulations* of the *Fisheries Act*, limiting daily discharges to <2.50 g tonne^{-1} of chlorine produced. During operation the plant released 4.95 g THg d^{-1}, through two effluent discharge points (Garron et al., 2005). Surface water and groundwater from the site have been treated since plant closure. To date, ongoing quarterly acute lethality testing at two effluent discharge points using rainbow trout based on Environment Canada (1990) methods have been negative (unpublished data).

The chlor-alkali plant (and other industrial activities) has been a local point source of THg in Chaleur Bay since operation began in 1963, leading to contamination of marine sediments (Cranston et al., 1974; Wilson and Travers, 1976; Matheson and Bradshaw, 1985; Cranston, 2000; Garron et al., 2005; Parsons and Cranston, 2006) and biota (Garron et al., 2005; Fraser et al., 2011). The objective of this study was designed to better understand baseline conditions of THg concentrations in Chaleur Bay by collecting surface sediments along a single transect in August 2011 (post-closure) for THg.

This study followed an approach used in an earlier study in October 2001 (pre-closure) by Garron et al. (2005) for temporal comparison (10 yr). A single transect was established close to the shoreline in front of the former plant to represent potential near-field impacts and ran northeast for 5.2 km across the mouth of the Restigouche River to 1.5 km east of Pointe de Fleurant, Québec (Fig. 1). Seven sampling stations were selected (distributed 50 m, 100 m, 300 m, 600 m, 1.2 km, 3.0 km and 5.2 km) along the transect and were based on stations used by Garron et al. (2005) in one of their transects (transect C). A reference station was chosen along the southeast shore near Eel

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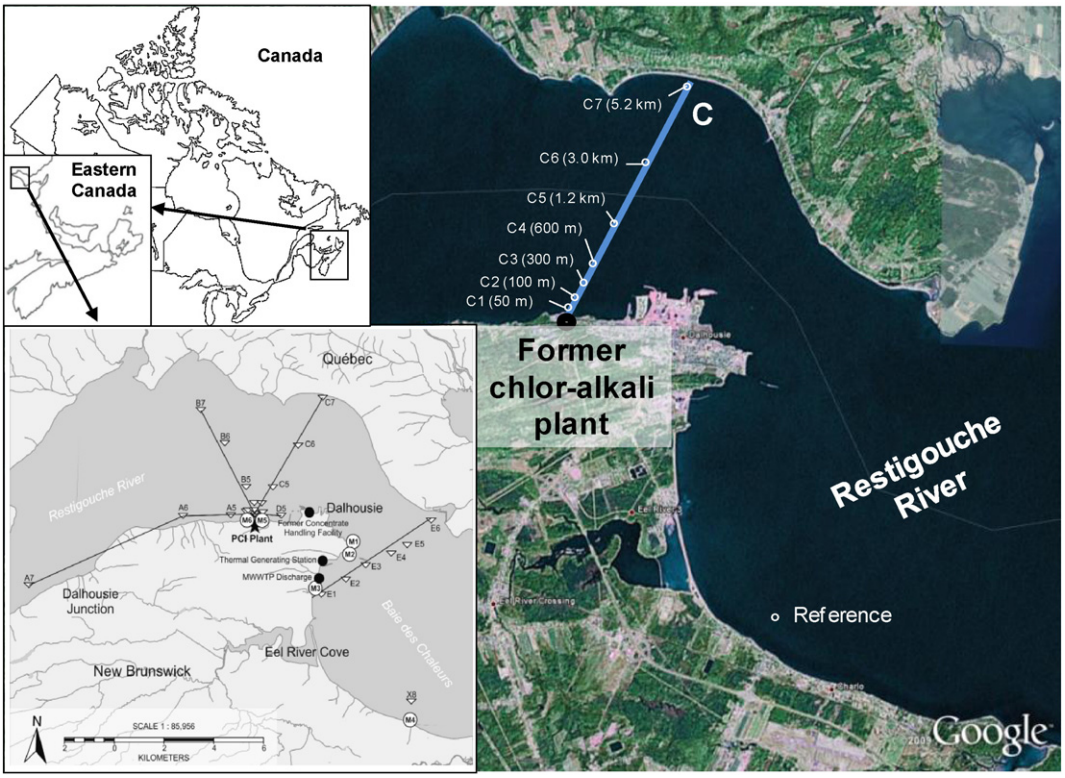

Fig. 1. Sediment sampling stations in Chaleur Bay, New Brunswick (2011). Top inset indicates where this study was conducted within Canada. Bottom inset indicates sediment sampling stations used in 2001 (adapted from Garron et al., 2005) (Google Maps 2015).

River Cove (11 km downstream), which was previously reported as being uncontaminated with THg (Wilson and Travers, 1976; Garron et al., 2005).

Duplicate surface (0–2 cm) sediment samples were collected at each station using an Ekman grab, taking care to minimize disturbance of sediment before sub-sampling (Walker and Grant, 2009; Grant et al., 2013). Samples including one blind field duplicate were analysed by Maxxam Analytics Inc. (Standards Council of Canada accredited) for analysis of total Hg based on US-EPA245.5 (US-EPA, 2005). Analyses were reported on a dry weight (dw) basis. Blind field duplicates were collected for every ten samples. Method blanks, spike blanks, matrix spikes and duplicate samples were analysed with samples. Spiked blank results were control charted and met specific acceptance criteria. Detection limits (0.01 μg g^{-1}) are shown in Table 1. Significant differences of Hg concentrations were determined with Student's t-Test (temporal differences) and Spearman rank correlation analysis (spatial differences) using Minitab. Sediments were compared to previous studies in the area and against guidelines developed by Canadian Council of Ministers of the Environment (CCME, 2014; Table 1). CCME interim sediment quality guidelines (ISQGs) are equivalent to lowest effect level, below which contaminants have little chronic or acute effect on biota, and CCME probable effect levels (PELs) are equivalent to severe effect level, above which biota are very likely to be negatively affected by contaminants (Walker et al., 2015).

Station C1 was characterized by coarse sandy sediments containing some gravel and shell fragments with shallow (<1 cm) light brown oxic surface horizon and abundant eel grass (*Zostera marina*). Stations C2–C5 (100 m to 1.2 km) were characterized by soft fine grained silty sediments with 1–3 cm oxic surface horizons containing abundant infauna. Stations C6 and C7 (3–5.2 km) along the transect and reference station were characterized by coarse sandy sediments with deep (>3 cm) light brown oxic surface horizons. All samples were collected from shallow coastal sediments (1.5 to 9 m deep). Mean reference sediment Hg concentrations (0.045 μg g^{-1}) were lower than background Hg levels reported in Chaleur Bay by Wilson and Travers (1976) (0.1 μg g^{-1}), Garron et al. (2005) (0.075 μg g^{-1}), and lower than background coastal sediments in Nova Scotia reported by Loring et al. (1996) (0.1 μg g^{-1}) (Table 1).

THg concentrations in surface sediments ranged from 0.04–0.28 μg g^{-1}. Fig. 2 compares results from this study (2011, circles) against Garron et al. (2005) (2001, triangles). There has been a significant decrease ($p \leq 0.05$) in sediment THg concentrations at all stations (including reference), since sediments were last sampled in 2001 during operation. Spearman rank correlation analysis indicated that THg concentrations along the transect were negatively correlated with distance from the plant ($p < 0.005$). No samples exceeded PEL concentrations (0.70 μg g^{-1}). Only stations C1 (50 m away) and C2 (100 m away) exceeded ISQG concentrations (0.13 μg g^{-1}). In contrast, Garron et al. (2005) reported exceedances up to 1.3 km away. All other stations were <ISQG. Stations C5, C6 and C7 were comparable to the mean reference THg concentration (0.045 μg g^{-1}).

Previous studies reported that THg concentrations in sediments near the Dalhousie plant ranged from <0.10 to 8.10 μg g^{-1} dw, (higher near the plant, 5.40–8.10 μg g^{-1}), with contamination occurring up to 2.2 km along the shore from the plant and 1 km out from the shore off the plant (Wilson and Travers, 1976) (Table 1). A decade after operation began, Cranston et al. (1974) reported levels ranging from 0.38 to 0.71 μg g^{-1} near the plant. Results from the Garron et al. (2005) study reported an order of magnitude less THg in sediments compared to Wilson and Travers (1976), and the results of this study were lower still than those reported by Garron et al. (2005) (0.05–1.96 μg g^{-1}). These earlier studies support the hypothesis of decreasing THg sediment concentrations over time via natural remediation of the Restigouche River sediments from burial deposition of recent sediment. Matheson and Bradshaw (1985) found concentrations of THg in Chaleur Bay from <0.01 to 0.32 μg g^{-1}, with the highest concentrations being at those sites directly influenced by the Restigouche River (>0.2 μg g^{-1}). Concentrations along the Quebec and New Brunswick shores were

Table 1
Comparison of THg sediment concentrations with historical regional studies and near other chlor-alkali plants (μg g^{-1} dw).

Location	Range Hg (DL)	Mean Hg	Date sampled	Dominant industry	Reference
Chaleur Bay, Dalhousie, New Brunswick (NB)					
Chaleur Bay, Dalhousie, NB (plant)	0.38–0.71 (0.10)	ND	1974	Hg cell chlor-alkali plant, thermal power plant	Cranston et al. (1974)
Chaleur Bay, Dalhousie, NB (bay)	0.27–0.62 (0.10)	ND	1974	Hg cell chlor-alkali plant, thermal power plant	Cranston et al. (1974)
Chaleur Bay, Dalhousie, NB (upstream)	<0.10–0.50 (0.10)	0.30	1976	Hg cell chlor-alkali plant, thermal power plant	Wilson and Travers (1976)
Chaleur Bay, Dalhousie, NB (plant)	5.40–8.10 (0.10)	6.80	1976	Hg cell chlor-alkali plant, thermal power plant	Wilson and Travers (1976)
Chaleur Bay, Dalhousie, NB (downstream)	0.10–4.60 (0.10)	1.80	1976	Hg cell chlor-alkali plant, thermal power plant	Wilson and Travers (1976)
Chaleur Bay, Dalhousie, NB	<0.01–0.32 (0.01)	ND	1985	Hg cell chlor-alkali plant, thermal power plant	Matheson and Bradshaw (1985)
Chaleur Bay, Dalhousie, NB (bay)	0.14–0.17 (0.01)	ND	2000	Hg cell chlor-alkali plant, thermal power plant	Cranston (2000)
Chaleur Bay, Dalhousie, NB (plant)	0.05–1.96 (0.02)	0.43	2001	Hg cell chlor-alkali plant, thermal power plant	Garron et al. (2005)
Chaleur Bay, Dalhousie, NB (bay)	<0.01–2.4 (0.01)	ND	2006	Hg cell chlor -alkali plant, thermal power plant	Parsons and Cranston (2006)
Plant closure			2008	Plant closure	Plant closure
Chaleur Bay, Dalhousie, NB (plant)	0.04–0.28 (0.01)	0.10	2011	Hg cell chlor-alkali plant (closed), thermal power plant (closed)	Present study
Nova Scotia (NS)					
Wine Harbour, NS	<0.05–74.3 (0.05)	ND	2004	Historical gold mining (1860–1940)	Little et al. (2015)
Isaacs Harbour, NS	<0.05–16.0 (0.05)	ND	2004	Historical gold mining (1860–1940)	Little et al. (2015)
Seal Harbour, NS	<0.05–1.30 (0.05)	ND	2004	Historical gold mining (1860–1940)	Little et al. (2015)
Isaacs and Country Harbours, NS	<0.05–0.16 (0.05)	ND	2008	Historical gold mining (1860–1940)	Walker and Grant (2015)
Sydney Harbour, NS	0.10–0.49 (0.05)	ND	2009/12	Steel coking industry, coal combustion (1901–1988)	Walker et al. (2013b)
Background coastal sediments, NS	0.10	ND	1995	Perceived as un-impacted by industry	Loring et al. (1996)
Other chlor-alkali plants					
Laurentian Trough, St. Lawrence, Canada	0.10–0.52 (0.05)	ND	ND	Hg cell chlor-alkali plant	Gobeil and Cossa (1993)
Saguenay Fjord, St. Lawrence, Canada	<0.02–10.0 (0.02)	0.50	1991/92	Hg cell chlor-alkali plant	Gagnon et al. (1997)
Lake Balkyldak, Pavlodar, Kazakhstan	0.36–617 (0.10)	151.5	2001	Hg cell chlor-alkali plant	Ullrich et al. (2007)
Thane Creek–Mumbai Harbour, India	6.23–8.21 (0.10)	ND	1978	Hg cell chlor-alkali plant	Zingde and Desai (1981)
Thane Creek–Mumbai Harbour, India	0.21–1.19 (0.01)	ND	2000	Hg cell chlor-alkali plant	Ram et al. (2009)
CCME ISQG	0.13	–	–	–	CCME (2014)
CCME PEL	0.70	–	–	–	CCME (2014)

DL — detection limits presented in parentheses; ND — not determined; CCME ISQG and PEL — Canadian Council of Ministers of the Environment SQGs, Interim Sediment Quality Guidelines and Probable Effects Levels (CCME, 2014).

found to be generally <0.05 μg g^{-1}. Those concentrations are consistent with the reference concentrations found in this study (0.045 μg g^{-1}). Surface sediments collected by Cranston (2000) had elevated concentrations of THg in samples offshore of Dalhousie (0.14–0.17 μg g^{-1}). Other known point sources of THg in the Restigouche River and the Chaleur Bay area include former Bowater pulp and paper mill at Dalhousie and thermal generating power plant (Cranston, 2000; Parsons and Cranston, 2006; EC, 2014).

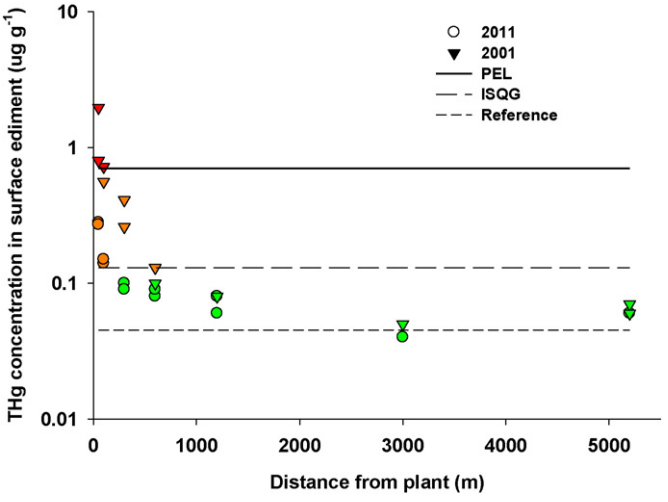

Fig. 2. Spatial and temporal variation of THg concentrations in surface sediment (μg g^{-1} dw) along a single transect (circles, 2011; triangles, 2001 data from Garron et al. (2005)). Horizontal lines represent PEL (solid), ISQG (medium dashes) and mean reference station concentrations (short dashes). Symbols in red indicate PEL exceedances (0.70 μg g^{-1}), orange ISQG exceedances (0.13 μg g^{-1}) and green <ISQG.

Chlor-alkali plants across Canada have been major point sources of THg in surficial sediments for decades (Gobeil and Cossa, 1993; Gagnon et al., 1997). Similar plants globally have also contributed to THg accumulation in aquatic environments (Zingde and Desai, 1981; Ullrich et al., 2007; Ram et al., 2009). However, many have reported decreases in concentrations following plant closure (Table 1). THg concentrations in marine and estuarine sediments around the world vary widely due to influences by both anthropogenic and natural sources (Mitra, 1986), and can range from <0.02 μg g^{-1} to as high as 617 μg g^{-1} near a chlor-alkali plant operating in Lake Balkyldak, Kazakhstan (Ullrich et al., 2007). Background sediment mercury concentrations around Nova Scotia have been estimated at 0.10 μg g^{-1} (Loring et al., 1996), which are consistent with concentrations measured in reference stations in this study (0.045 μg g^{-1}).

Overall, sediment Hg concentrations were lower than previous studies in the area and exceedances of ISQGs (considered lowest effect thresholds) were highly localised (<100 m). Furthermore, current baseline conditions in Chaleur Bay were lower than other former industrial coastal areas in eastern Canada (Table 1). Results suggest there has been some natural recovery of Chaleur Bay sediments from burial by recent sediment deposition. Although rates of sediment deposition were not assessed, deposition rates in shallow coastal estuaries in the region are low (<1 cm $year^{-1}$) (Lee et al., 2002; Walker et al., 2013b). Natural capping has been observed in other contaminated harbours in the region (e.g., Walker et al., 2013a; Walker and Grant, 2015).

This limited quantitative dataset (THg) will serve as baseline information, as envisioned by Baseline Editors of this journal (e.g., Richardson, 2012), against which future monitoring can be compared during decommissioning operations of nearby industrial facilities. THg concentrations in sediments have decreased significantly since closure and have decreased by an order of magnitude since the Wilson and Travers (1976) study. Whilst sediment THg contaminant concentrations are lower than those previously reported, most of the contaminant inventory remains intact below the sediment surface, with the potential risk for

future contaminant disturbance or resuspension. Furthermore, THg may become more bioavailable to marine biota under anoxic conditions. Sediment stability and bioavailability is often affected by many factors such as: porosity, organic content, grain size, pH, redox, metal oxide and sulphide contents and bioturbation (Nedwell and Walker, 1995; Walker et al., 2014). In order to properly understand potential management options, such as dredging (e.g., Walker et al., 2013e) or ecological risks associated with contaminated sediments in Chaleur Bay, more information related sediment characteristics are required.

Repeat sediment sampling at stations along the same transect may be required following decommissioning of the chlor-alkali plant to evaluate whether the trend of decreasing THg sediment concentrations continues. Therefore, further monitoring of these shallow coastal sediments will be critical in order to determine potential ecological risk and bioavailability to marine biota.

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