Marine Pollution Bulletin xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

Baseline

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

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ARTICLE INFO

Article history:
Received 20 December 2015
Received in revised form 7 March 2016
Accepted 16 March 2016
Available online xxxx

Keywords:
Chlor-alkali plant
Total mercury (THg)
Sediment
Ecological risk

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 bioavilability of Hg that could pose ecological risks to marine biota.

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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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http://dx.doi.org/10.1016/j.marpolbul.2016.03.033
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Please cite this article as: Walker, T.R., Mercury concentrations in marine sediments near a former mercury cell chlor-alkali plant in eastern Canada, Marine Pollution Bulletin (2016), http://dx.doi.org/10.1016/j.marpolbul.2016.03.033

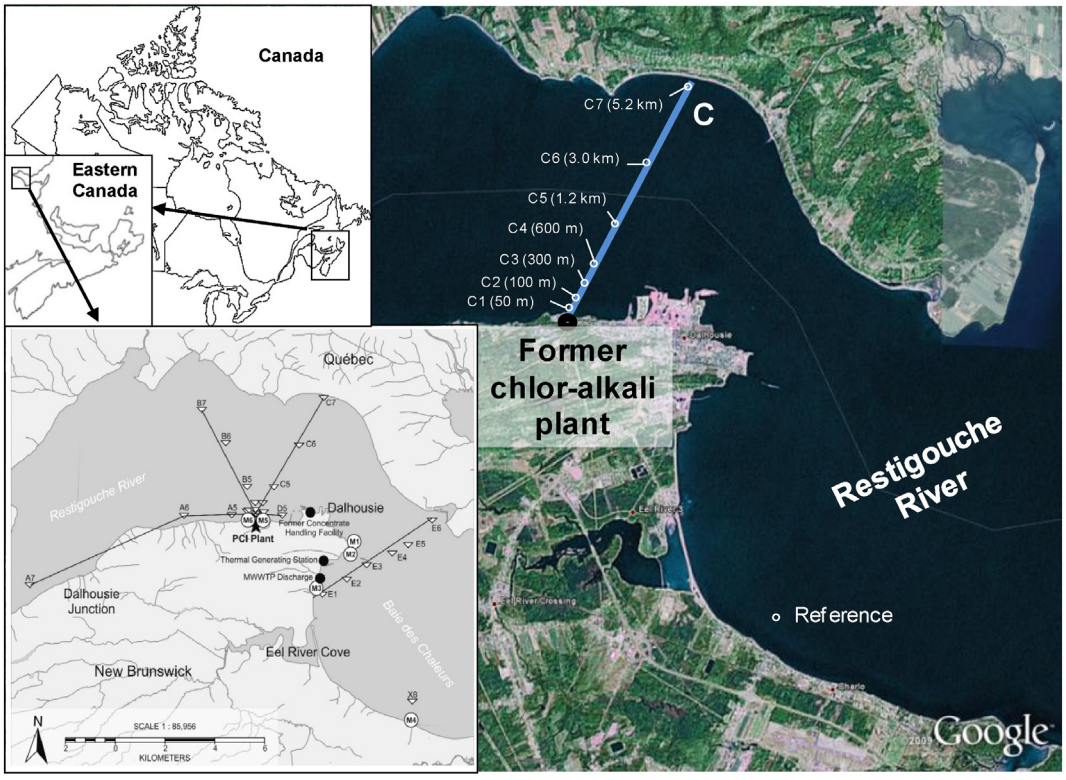

Fig. 1. Sediment sampling stations in Chaleur Bay, New Brunswick (2011). Top inset indicates where this study was conducted within Canada. Bottom inset indicates sediment sampling stations used in 2001 (adapted from Garron et al., 2005) (Google Maps 2015).

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 ($p < 0.05$) 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

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