



Mercury empirical relationships in sediments from three Ontario lakes

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ARTICLE INFO

Article history:

Received 30 June 2009

Received in revised form 17 December 2009

Accepted 17 December 2009

Available online 6 February 2010

Keywords:

Sediment
Lake
Mercury
Redox potential
Organic carbon
Sediment bulk density

ABSTRACT

Total mercury (THg), methyl mercury (MeHg), total organic carbon (TOC), sediment bulk density (SBD), redox potential (Eh) and percent fines measurements were made on sediment cores collected along transects from littoral to profundal depths in Harp, Dickie, and Blue Chalk lake located on the Canadian Shield near Dorset, Ontario, Canada to determine whether empirical relationships exist among these sediment properties.

MeHg was positively correlated with THg in all sediments with a MeHg:THg ratio (0.004 ± 0.004) comparable to other uncontaminated profundal lakes. MeHg, MeHg:THg and TOC decreased with sediment depth within the core for all lakes, whereas THg only showed a decrease in Harp Lake. MeHg:THg ratio in surficial sediments was positively correlated with Eh and negatively correlated with TOC [$\text{MeHg:THg} = -0.009 * \text{TOC} (\%) + 0.001 * \text{Eh} (\text{mV}) - 1.902$, $p = 0.026$]; whereas THg was positively correlated with TOC [$\log \text{THg} (\text{ppb}) = 0.026 * \text{TOC} (\%) + 1.400$, $p < 0.0001$].

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1. Introduction

All lakes, whether deep or shallow, typically tend to show declining redox potentials in the hypolimnion as summer progresses. Low redox potentials (e.g., -118 mV for sulfate-reducers) in the hypolimnion and surficial sediment tend to promote mercury methylation by anaerobic bacteria (Regnell and Ewald, 1997). Anaerobic bacteria, including the sulfate and iron-reducers, are thought to be responsible for methylating mercury (Hg) using bioavailable Hg^{2+} (King et al., 2002; Kerin et al., 2006). The methyl mercury (MeHg) formed biomagnifies through foodwebs and toxic effects are observed in biota at higher trophic levels, even in locations removed from industrial sources (O'Driscoll et al., 2005). Some studies have found a correlation between MeHg and total mercury (THg) in sediment (Regnell and Ewald, 1997), while others have not (Lambertsson and Nilsson, 2006).

Concentrations of MeHg and THg in sediments are known to show high spatial and temporal (MeHg) variability, even at remote locations (Lindeberg et al., 2006) and within a lake (Campbell et al., 2003). For example, Mills et al. (2009) found THg in the surficial sediments of 172 Ontario lakes to range from 19.7 to 501 ppb dw (mean: 224 ppb dw). Sediment depths within the core >25 cm in these lakes had THg values which ranged from 11.6 to 271 ppb dw (mean: 107 ppb dw). Boszke et al. (2003) found the MeHg:THg ratio

to vary from 0.0065 ± 0.0034 near the surface to 0.0001–0.0005 at greater core depths.

Croston et al. (1996) found that temporal and spatial abundance of sediment MeHg reflected variations in redox potential (Eh), Hg availability for methylation, and the nature of bacterial populations. An increase in Eh (-200 mV to $+50$ mV) has been found to result in a decrease in MeHg concentration at the sediment–water interface (DeLaune et al., 2004). Several other environmental variables have also been reported to show a correlation with THg and/or MeHg in sediments, such as bacterial abundance, acid volatile sulfides, organic matter, Fe, Mn and Al hydroxides, chlorides, and temperature (e.g., Boszke et al., 2003; Drott et al., 2007; Kainz et al., 2003; Roulet et al., 2001). THg and MeHg have also been found to be positively correlated with organic matter in some sediments, but not in others (e.g., significantly (Regnell and Ewald, 1997; Lambertsson and Nilsson, 2006; Regnell et al., 2001 (upper lake); He et al., 2007 (surficial sediments)), weakly (Kamman et al., 2005; Regnell et al., 2001 (lower lake)), and non-significantly (Kannan et al., 1998; He et al., 2007 (sediment cores))).

Published studies that examine within-lake variability in MeHg and THg concentrations in sediments have largely focused on historic sedimentary profiles (e.g., Hines et al., 2004). Others have attempted to analyze the horizontal (i.e., transect from littoral to profundal region of lake) and vertical (i.e., sediment cores) profiles of sediment in remote lakes simultaneously (e.g., Campbell et al., 2003; He et al., 2007). Studies that report a correlation coefficient (r) or significance value ($p < 0.05$) for relationships rarely include information regarding the corresponding equation (or at least the slope of the relationship), information that could be useful for other applications (i.e., models).

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Most of the lakes studied to date have generally been near industrial sources of Hg (e.g., Marvin et al., 2004; Kerfoot et al., 2004), although some were from more distant locations (e.g., Wiener et al., 2006; Allan et al., 2001). Here we provide data for three relatively pristine shield lakes receiving inputs from atmospheric deposition, direct runoff and streams within the drainage basin. The main objective of this study was to determine whether observations made in these three lakes could produce empirical relationships applicable to comparable lakes in other regions (e.g., shield lakes with low conductance). Horizontal (surficial sediment) and vertical (sediment cores) relationships (and corresponding equations) among sediment MeHg, THg, organic content, bulk density, percent fines and Eh were examined and compared with data from other locations.

2. Methods

Harp (45°23'N; 79°08'W), Dickie (45°09'N; 79°05'W) and Blue Chalk (45°12'N; 78°56'W) are well-studied lakes near Dorset, Ontario, Canada (Table 1) that are situated on the Canadian Shield (Fig. 1). As a result, all of these lakes are low in buffering capacity and calcium content. Blue Chalk (surface area = 52.4 ha; max depth = 23 m) and Dickie (93.6 ha; 12 m) are seasonally stratified with anoxic conditions developing later in the summer (Fig. 2). Harp Lake (71.4 ha; 37.5 m) does exhibit thermal stratification (~4 m, Fig. 2A), but anaerobic conditions in the water column do not occur in this relatively deep lake (Fig. 2D). Studies have shown that summer hypolimnetic oxygen (from 1975 to 1995) seldom rises above 1 mg L⁻¹ in Dickie Lake, but is consistently over 4 mg L⁻¹ in Harp Lake (Quinlan et al., 2005; Eimers et al., 2006).

2.1. Sample collection

Temperature (Fig. 2A–C) and dissolved oxygen (Fig. 2D–F) depth profiles in the water column were measured on-site routinely (bi-weekly: Harp and Dickie Lake; monthly: Blue Chalk Lake) throughout the summer using standard Ontario Ministry of the Environment (MOE) field protocols with a YSI model 58 oxygen/temperature meter (Dorset Environmental Science Centre, Dorset, Ontario). Sediment cores (4 or 5) were collected in June and August 2005 with an open-barreled Kajak–Brinkhurst-type gravity corer modified to have an auto suction-seal and a percussion knocker to extend penetration from Harp, Dickie, and Blue Chalk Lake (Table 2). The corer was rinsed with lake water to prevent cross-contamination as cores were sampled along transects from littoral to profundal region of each lake along the main axis. Lower oxygen levels were anticipated in the profundal sites in late August. At each site, water column depth was determined using marked line on rope attached to sediment corer and the location was recorded using a Garmin portable 12-channel Global Positioning System (GPS). Variation in lake basin slope (expressed as %) between

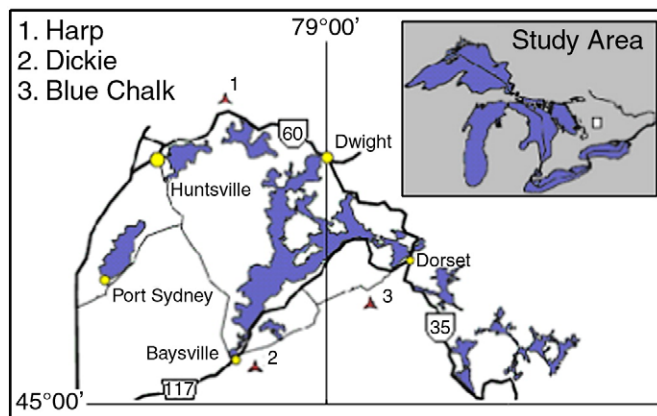


Fig. 1. Location of study area and lakes.

sediment core sampling locations along each transect was calculated as the difference between site depths divided by the distance between samples.

Cores were capped and transported to a lab at the Dorset Environmental Science Centre where duplicate samples from 5 cm slices were taken to a sediment depth of 25 cm. Cores were sectioned under N₂ and placed (with no headspace) in air-tight plastic 50 mL centrifuge tubes. The Eh (mV) was measured on each core with an Orion model 520 meter as it was being extracted by inserting a silver/silver chloride Eh probe into the sediment. Samples were then transported in a cooler to the National Wildlife Research Centre (Environment Canada, Ottawa, Ontario) and freeze-dried.

2.2. Laboratory analyses

Sediment bulk density (SBD, g cm⁻³) was determined by weighing sediment before and after freeze-drying with a water content (WC) conversion described by Jepsen et al. (2001) (i.e., $SBD = (SD * WD) / (WD + (SD - WD) * WC)$, which assumes sediment (SD, 2.6 g cm⁻³) and water (WD, 1 g cm⁻³) densities. Percent total organic carbon (TOC, %) was determined using weight loss on ignition method (USGA, 1993) using a muffle furnace set at 440 °C for 12 h.

For sediment MeHg analyses, a sodium thiosulfate clean-up was used on extracts of duplicate sediment samples (to quench any residual chemical reactions). MeHg was then isolated as bromide derivatives by acidic KBr and CuSO₄ with subsequent extraction into a small volume (100 μL) of dichloromethane (Cai et al., 1997). Separation method used avoids artifact MeHg formation, as described in Bloom et al. (1997). Determination of MeHg was done at the University of Ottawa by capillary gas chromatography coupled with atomic fluorescence spectrometry (GC-AFS) as described by Cai et al. (1997). Analysis of a MeHg estuarine sediment standard reference material (ERM-CC580, IRMM) was within the reported range (75 ± 4 ppb).

Sediment THg was determined at the National Wildlife Research Centre using a high temperature combustion Hg analyzer (AMA-254, Altec, Czech Republic) equipped with an ASS-254 autosampler (Weech et al., 2004). Quality assurance included the use of standard reference materials (LKSD-1, NBS-1646b, LKSD-3) and sample replicates (n = 39). The standard reference materials used were estuarine sediment NBS-1646b [65.2 ± 11.7 ppb (n = 3)], lake sediment LKSD-1 [104.9 ± 8.2 ppb (n = 46)], and lake sediment LKSD-3 [284.3 ± 18.1 ppb (n = 7)]. LKSD-3 was analyzed at the start of each run, and LKSD-1 was analyzed after every 5th sample. Measurements of THg in all of the SRMs were within 10% of certified values [NBS-1646b = 63 ± 12 ppb (National Bureau of Standards, 1982); LKSD-1 = 110 ppb (Geological Survey of Canada, 1990); LKSD-3 = 290 ppb

Table 1
Lake property data (Dorset Research Centre, Dorset, ON).

	Blue Chalk	Dickie	Harp
<i>Physical properties</i>			
Surface area (ha)	52.4	98.6	71.4
Mean water column depth (m)	8.5	5	13.3
Maximum water column depth (m)	23	12	37
Lake water volume (× 10 ⁵ m ³)	44.7	46.7	95.1
Watershed area (km ²)	1.9	5	5.4
<i>Chemical properties</i>			
Dissolved organic carbon (mg L ⁻¹)	1.8	5.2	3.7
Total phosphorus (μg L ⁻¹)	5	7.5	6.3
Conductivity (μS cm ⁻¹)	28.4	33.2	37.4
pH	6.7	5.9	6.4

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