



Isotopic evidence for distinct sources of mercury in lake waters and sediments



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ABSTRACT

Lakes are important ecosystems where mercury (Hg) may undergo extensive methylation and bioaccumulation in the aquatic food web. It is thus crucial to understand the sources of Hg in lake systems. Mercury isotope composition is a powerful tracer of its sources in the environment. However, previous studies on Hg isotope compositions in lake systems mainly focus on the aquatic organism and sediments, whereas lake water that provides the nutrients for the bottom of aquatic food chain has received much less attention. Here we report Hg isotope compositions in surface water and sediments of 8 freshwater lakes in Ontario, Canada. We determined significantly positive $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$ values in surface water of all lakes, which are similar to previously reported isotope signature of local atmospheric precipitation. In contrast, the isotope compositions of lake sediments show predominantly negative $\Delta^{199}\text{Hg}$ and negligible $\Delta^{200}\text{Hg}$ values that are similar to atmospheric elemental Hg(0) and terrestrial soils and plants. We conclude that the source of Hg in lake surface water is likely dominated by direct atmospheric precipitation, while the Hg in lake sediments is primarily derived from watershed inputs of terrestrial particles. The concurrent odd Hg isotope anomalies in surface waters suggest that methylmercury (MeHg) from the watershed wetland, together with Hg(II) derived from the catchment weathering is likely another Hg source to lake waters besides atmospheric deposition. We suggest that the different Hg sources and isotope compositions between lake water and sediments would result in different isotope signatures for MeHg that is produced in water column and in sediments. Therefore, our study not only elucidates the sources of Hg to lake systems, but also has implications in distinguishing between different MeHg sources and in tracing the bioaccumulation of MeHg in aquatic organism.

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1. Introduction

Mercury (Hg) is a globally-distributed metal pollutant. It is deposited into surface ecosystems after long-range atmospheric transport and may be converted to its most toxic form mono-methylmercury (MeHg) that is effectively bio-accumulated and magnified in aquatic food webs, posing significant threats to human health and aquatic biota. Being an important recipient of atmospheric deposition, aquatic systems play a critical role in the biogeochemical cycle of Hg (Mason et al., 1994; Morel et al., 1998). It is thus crucial to identify the linkage between the sources of Hg in aqueous environments (e.g. lakes) and Hg (mainly MeHg) bioaccumulated in aquatic biota. Although impressive progress has been made recently in understanding many aspects of Hg cycling, including the atmospheric deposition (Kirk et al., 2011;

Muir et al., 2009; Prestbo and Gay, 2009; Selvendiran et al., 2009), transport from land to water reservoirs (Buck et al., 2015; Cohen et al., 2004; Lepak et al., 2015; Meili, 1991), surface-atmosphere exchange (Feng et al., 2004; Lindberg et al., 2002; Poissant and Casimir, 1998; Poissant et al., 2000; Southworth et al., 2007; Zhu et al., 2011), and specie transformation (Amyot et al., 1997; Driscoll et al., 1994; Lindberg et al., 2002; O'Driscoll et al., 2006; Sellers et al., 2001; Ullrich et al., 2001), the sources and mechanisms involved in the aquatic Hg cycling still remain poorly understood (Bergquist and Blum, 2009; Krabbenhoft et al., 2002; Morel et al., 1998; Selin, 2009). For example, among many others, one important puzzling question is why Hg (MeHg) concentrations in most Chinese wild fish are generally lower (e.g. <500 ng/g of ww) than those in USA, Canada and northern European countries (>500 ng/g of ww) (Lin et al., 2012; Lindqvist et al., 1991; Pan et al., 2014; Zhang et al., 2014), given the fact that China has the highest Hg production, consumption and emission in the world (Selin, 2009).

Mercury isotopes are an important tracer for the sources and pathways of Hg in the environment. Recent studies have discovered

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both mass-dependent fractionation (MDF, $\delta^{202}\text{Hg}$) and mass-independent fractionation (odd-MIF as $\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$, even-MIF as $\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$) for Hg isotopes in natural samples (Bergquist and Blum, 2007; Blum et al., 2013; Demers et al., 2013; Point et al., 2011; Sherman et al., 2010, 2015; Sonke, 2011; Sun et al., 2014). Most Hg transformation processes can induce MDF, and large variations of $\delta^{202}\text{Hg}$ (up to 20‰) have been determined in the nature (Chen et al., 2012; Demers et al., 2013; Feng et al., 2010; Ghosh et al., 2008; Rose et al., 2015; Sherman et al., 2015; Sonke, 2011; Zheng et al., 2015). Interestingly, MIF is only observed in a small number of processes, such as photoreduction, photodemethylation, abiotic dark reduction and evaporation (Bergquist and Blum, 2007; Estrade et al., 2009; Kritee et al., 2008; Zheng et al., 2007; Zheng and Hintelmann, 2010a). Except for photoreduction of thiol-bound Hg, photodegradation and photoreduction produce the most significant MIF that typically enriches odd Hg isotopes in the reactant, and are speculated to be responsible for the significant enrichment of ^{199}Hg and ^{201}Hg widely observed in aquatic biota (e.g., fish) (Bergquist and Blum, 2007; Blum et al., 2013, 2014; Point et al., 2011). However, this hypothesis has not been tested by direct determination of the isotope composition of Hg (and MeHg) in natural lake waters.

Atmospheric deposition is a primary pathway by which Hg enters the terrestrial ecosystems (Mason et al., 1994; Morel et al., 1998; Selin, 2009). Intriguingly, recent work has discovered MIF of even Hg isotopes (even-MIF) mainly in atmospheric samples, adding a new aspect to the Hg isotope systematics (Chen et al., 2012; Demers et al., 2013; Gratz et al., 2010; Huang et al., 2015; Rolison et al., 2013; Wang et al., 2015; Yuan et al., 2015). According to these studies, even-MIF of precipitation displayed an increase with latitude (Wang et al., 2015; Cai and Chen, 2016), with the highest $\Delta^{200}\text{Hg}$ values (up to 1.24‰) found in precipitation from southern Ontario, Canada (Chen et al., 2012). The mechanism of even-MIF is not yet understood (Eiler et al., 2014; Mead et al., 2013), but it has been speculated to result from the photo-related processes occurring in the tropopause (Chen et al., 2012). Though recent studies have observed significant $\Delta^{200}\text{Hg}$ in lake sediments, fish, soils, tree moss and seawater (Štrok et al., 2014; Kwon et al., 2015; Lepak et al., 2015; Cai and Chen, 2016; Zheng et al., in review-a), no data was reported for even-MIF in water samples from continental surface reservoirs.

In this study we measured Hg isotopic composition of surface waters and sediments collected from eight lakes in the central and southern Ontario, Canada (Tables 1 and 2, Fig. 1). Surface waters from three Chinese lakes were also measured for comparison (Table S1, Fig. S1). The objectives are 1) to characterize Hg isotope signatures in surface water and sediment of freshwater lakes; 2) to carry out a comparison between Hg isotope compositions in lake system with the published isotope data for atmospheric Hg deposition and terrestrial solid samples (e.g., soils and plants) and 3) to identify the sources of Hg in water and sediment of Canadian lakes using Hg isotope compositions.

2. Methods

2.1. Reagents and materials

All reagents (HCl, HNO_3 , BrCl, L-cysteine, $\text{NH}_2\text{OH}\cdot\text{HCl}$, SnCl_2) used in this study were analytical grade and prepared under Hg-free condition (Chen et al., 2010, 2012). All vessels were glass- or Teflon- made and cleaned with first BrCl, then HNO_3 and a last rinse of H_2O before use. NIST SRM3133 Hg and UM-Almadén Hg were used as reference standards and measured regularly for quality control (Chen et al., 2010).

2.2. Sampling processes

A total of eleven lakes were chosen for Hg isotope study, with eight headwater lakes in the Dorset and Kawartha forest regions of southern-central Ontario, Canada, on the southern margin of Canadian

Table 1
Lake characteristics, mercury concentration and isotopic compositions in lake and stream waters (ON, Canada).

	Date	T°C	pH	DOC (mg/L)	Hg (ng/L)	Recovery (%)	$\delta^{202}\text{Hg}$ (‰)	$\Delta^{199}\text{Hg}$ (‰)	$\Delta^{200}\text{Hg}$ (‰)	$\Delta^{201}\text{Hg}$ (‰)	$\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$	A_c (km ²)	A_0 (km ²)	Latitude (N)	Longitude (W)	Max. depth (m)	Average depth (m)
Lakes																	
Harp (H1)	10/07/10	25	7.4	4.2	0.98	104	-1.49 ± 0.10	1.00 ± 0.04	0.28 ± 0.03	0.63 ± 0.06	1.73	5.09	0.71	45.23	79.07	37.5	13.3
Harp (H2)	22/09/10	17	7.3	4.3	0.61	96	-1.59 ± 0.10	1.02 ± 0.04	0.18 ± 0.03	0.70 ± 0.06	1.46	5.09	0.71	45.23	79.07	37.5	13.3
Plastic	10/07/10	26	6.6	2.2	0.55	93	-1.96 ± 0.10	1.16 ± 0.04	0.08 ± 0.03	0.65 ± 0.06	1.78	0.89	0.32	45.10	78.48	16.3	8
Fairy	16/07/10	26	7.7	4.4	1.19	96	-1.75 ± 0.10	0.97 ± 0.04	0.18 ± 0.03	0.59 ± 0.06	1.64	46.6	7.12	45.19	79.11	74	22
Dikie	23/09/10	16	6.5	6.1	1.15	93	-1.83 ± 0.10	0.73 ± 0.04	0.13 ± 0.03	0.59 ± 0.06	1.24	4.49	0.94	45.09	79.05	12	5
Stoney	12/07/10	27	8.5	4.6	0.51	95	-1.31 ± 0.10	1.91 ± 0.04	0.26 ± 0.03	0.84 ± 0.06	2.27	167	28.2	44.35	78.30	32	6
Coon	25/09/10	17	7.6	6.0	0.51	97	-1.27 ± 0.04	0.78 ± 0.02	0.21 ± 0.04	0.52 ± 0.11	1.50	2.92	0.53	44.36	78.12	18	2.9
Julian	25/09/10	17	8.2	6.5	0.25	98	-1.73 ± 0.10	0.58 ± 0.04	0.22 ± 0.03	0.47 ± 0.06	1.51	3.24	0.78	44.36	78.09	12	4.8
Moir	27/09/10	18	8.4	6.2	0.28	95	-1.82 ± 0.10	0.95 ± 0.04	0.18 ± 0.03	0.63 ± 0.06	1.91	579	8.7	44.30	77.27	8	3
Stream																	
Stream	23/09/10	13	6.0	3.5	1.04	98	-2.22 ± 0.15	0.21 ± 0.07	0.15 ± 0.05	0.11 ± 0.09							

DOC, dissolved organic carbon; A_c , lake catchment area; A_0 , lake surface area; Max. depth, maximum depth of lake water; Avg. depth, average depth of lake water; error bars are 2 σ external standard deviations for samples with multiple measurements, or $\pm 0.10\%$, $\pm 0.04\%$, $\pm 0.03\%$, and $\pm 0.06\%$ for $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$, $\Delta^{200}\text{Hg}$ and $\Delta^{201}\text{Hg}$ of samples measured only one time. Recovery, the percentage of Hg recovered from the preconcentration (measured by MC-ICP-MS) relative to that of the initial solution (measured on Tekran 2600) for the same sample.

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