Science of the Total Environment 511 (2015) 489–500

Contents lists available at ScienceDirect

Science of the Total Environment

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

Total- and methyl-mercury concentrations and methylation rates across the freshwater to hypersaline continuum of the Great Salt Lake, Utah, USA

William P. Johnson [a,*], Neil Swanson [a], Brooks Black [a], Abigail Rudd [b], Greg Carling [c], Diego P. Fernandez [a], John Luft [d], Jim Van Leeuwen [d], Mark Marvin-DiPasquale [e]

[a] Department of Geology and Geophysics, University of Utah, Salt Lake City, UT 84112, United States
[b] Brooks-Rand LLC, 4415 6th Ave NW, Seattle, WA 98107, United States
[c] Department of Geological Sciences, Brigham Young University, Provo, UT 84602, United States
[d] State of Utah Division of Wildlife Resources, 1594 W North Temple, Suite 2110, Box 146301 Salt Lake City, UT 84114, United States
[e] United States Geological Survey, Menlo Park, CA 94025, United States

HIGHLIGHTS

- Highest Great Salt Lake methyl mercury concentrations occur in deep brine layer.
- Deep brine layer is proximal to highest reported mercury burdens reported in birds.
- Methylation rates in the deep brine layer are lowest during mid-summer.
- Mid-summer is when mercury burdens are reported lowest in aquatic invertebrates.

GRAPHICAL ABSTRACT

ARTICLE INFO

Article history:
Received 25 August 2014
Received in revised form 27 December 2014
Accepted 27 December 2014
Available online 7 January 2015

Editor: Mae Mae Sexauer Gustin

Keywords:
Aqueous geochemistry
Limnology
Toxic elements
Trace elements

ABSTRACT

We examined mercury (Hg) speciation in water and sediment of the Great Salt Lake and surrounding wetlands, a locale spanning fresh to hypersaline and oxic to anoxic conditions, in order to test the hypothesis that spatial and temporal variations in Hg concentration and methylation rates correspond to observed spatial and temporal trends in Hg burdens previously reported in biota. Water column, sediment, and pore water concentrations of methylmercury (MeHg) and total mercury (THg), as well as related aquatic chemical parameters were examined. Inorganic Hg(II)-methylation rates were determined in selected water column and sediment subsamples spiked with inorganic divalent mercury (^{204}Hg(II)). Net production of Me^{204}Hg was expressed as apparent first-order rate constants for methylation (k_{meth}), which were also expanded to MeHg production potential (MPP) rates via combination with tin reducible 'reactive' Hg(II) (Hg(II)$_R$) as a proxy for bioavailable Hg(II). Notable findings include: 1) elevated Hg concentrations previously reported in birds and brine flies were spatially proximal to the measured highest MeHg concentrations, the latter occurring in the anoxic deep brine layer (DBL) of the Great Salt Lake; 2) timing of reduced Hg(II)-methylation rates in the DBL (according to both k_{meth} and MPP) coincides with

* Corresponding author.
E-mail address: william.johnson@utah.edu (W.P. Johnson).

http://dx.doi.org/10.1016/j.scitotenv.2014.12.092

reduced Hg burdens among aquatic invertebrates (brine shrimp and brine flies) that act as potential vectors of Hg propagation to the terrestrial ecosystem; 3) values of k_{meth} were found to fall within the range reported by other studies; and 4) MPP rates were on the lower end of the range reported in methodologically comparable studies, suggesting the possibility that elevated MeHg in the anoxic deep brine layer results from its accumulation and persistence in this quasi-isolated environment, due to the absence of light (restricting abiotic photo demethylation) and/or minimal microbiological demethylation.

1. Introduction

1.1. Reported mercury burdens in higher biota

The Great Salt Lake (GSL), located in northwestern Utah, USA (Fig. 1), is the largest terminal lake in the Western Hemisphere, and an important ecosystem for millions of migratory birds. It is recognized as a site of hemispheric importance by the Western Hemisphere Shorebird Reserve Network, with over 1.4 million shorebirds using the GSL and surrounding wetlands for breeding and staging areas (Aldrich and Paul, 2002), and over seven million waterbirds utilizing the GSL and its associated wetlands during some portion of their biannual migration (Cline et al., 2011). Human consumption advisories are in place for three GSL duck species among seven examined (Scholl and Ball, 2005, 2006), based on breast muscle tissue mercury (Hg) concentrations exceeding the EPA screening value of 0.3 mg-kg^{-1} ww (USEPA, 2000). Several important spatial and temporal trends are suggested by existing studies of elevated mercury concentrations among avian species at GSL:

1) Hg concentrations in multiple migratory species increase during the fall season; e.g., eared grebes, which consume primarily brine shrimp from GSL during the fall molting period showed factor of three increased median liver Hg concentrations during the 3–5 month fall molting period (Naftz et al., 2008a), and eared grebe blood Hg concentrations that spent the fall of 2006 on GSL were shown to be higher in November than September, and were greater for adults relative to juveniles (Vest et al., 2008; Conover and Vest, 2009a). A trend towards elevated levels of liver Hg concentrations in autumn-collected waterfowl at Ogden Bay wetlands was observed in 2008 (Cline et al., 2011); however, it was not determined whether these birds arrived on the lake with that exposure or if they were exposed to Hg via the GSL open-water food chain, or in the wetlands.
2) Blood Hg concentrations in eared grebes that spent the fall of 2006 on GSL were higher at Stansbury Island on the west side of Gilbert Bay (Fig. 1) (10.1 ± 2.6, $n = 30$) relative to Antelope Island on the east side of Gilbert Bay (4.3 ± 0.5 mg kg^{-1}, $n = 30$) (Conover and Vest, 2009);
3) Elevated Hg concentrations vary inter-annually among avian species, as indicated by results collected in 2008 (Cline et al., 2011), where the mean Hg concentrations in adult breast muscle did not exceed the EPA screening level of 0.3 mg kg^{-1} ww, except for adults of one species only during the spring season only. This contrasts with the preceding studies showing highly elevated Hg burdens, and suggests inter-annual variation over several year periods (2005–2008).

1.2. Reported Hg concentrations in the aquatic system

At approximately the same time that high Hg concentrations were recognized in some waterfowl on the GSL in 2007, exceptionally high MeHg concentrations were found in the anoxic deep brine layer (DBL) of the GSL (Naftz et al., 2008), ranging beyond 30 ng-L^{-1}. The DBL occupies the deepest portions of the GSL at depths (during the study) from approximately 6.5 to 9 m below the surface (Baskin, 2005; Diaz et al., 2009). The DBL arises from a strong salinity contrast between the north and south arms of the GSL, which are separated by a railroad causeway. Higher salinity water flows from the north to the south arm through breaches in the causeway (and the permeable fill material), and pools in the south arm of the lake. This high salinity bottom water is not subject to annual turnover because of the strong and persistent density differences between the upper and lower water bodies (Naftz et al., 2008; Gwynn, 2002; Loving et al., 2002). Since MeHg is the bioaccumulative form of Hg (Baeyens et al., 2003; Mason et al., 2006), these high MeHg concentrations in the DBL indicate a possible connection to elevated Hg in waterfowl.

Reactive gaseous mercury (RGM) is produced within the GSL basin on the basis that a regular diel pattern in RGM concentrations was observed regardless of season and boundary layer height (Peterson and Gustin, 2008). Measured cumulative annual riverine Hg load to the GSL (~6 kg) (Naftz et al., 2009) was far less than measured cumulative annual atmospheric deposition load (~36 kg) (Peterson and Gustin, 2008). With respect to aquatic geochemical processes, the DBL has unique characteristics relative to typical surface water bodies, including: a) anoxia (Gwynn, 2002; Diaz et al., 2009b) high activity levels of sulfate reducing bacteria (Naftz et al., 2009; Ingvorsen and Brandt, 2002);and c) high organic carbon content ca. 60–90 mg-L^{-1} (Diaz et al., 2009, Supporting information); all of which have been associated with MeHg production (King et al., 2000; Sunderland et al., 2006; Graham et al., 2012). The above characteristics suggest that the DBL may have uniquely high Hg(II)-methylation rates relative to other water bodies.

The eventual propagation of Hg from the DBL to shallow portions of GSL is suggested by direct measurement of at least limited mixing that occurs during wind events (Beisner et al., 2009), and by high north-to-south velocities measured in the DBL (Beisner et al., 2009) that suggest relatively rapid convection and imply eventual re-entrainment of DBL into the shallow brine layer at the south end of the GSL. Water column depths in the overall GSL system range from ~9 m in the main body of the GSL to approximately 1–1.5 m in the impounded wetlands and shallow portions of the freshwater-influenced bays to (Fig. 1). The impounded wetlands experience diel cycles in water column pH, dissolved oxygen (DO), and trace elements (e.g., selenium, antimony, manganese), with the greatest diel swings occurring during summer (Dicataldo et al., 2010; Carling et al., 2011). Despite diel and seasonal variations, wetland surface water, pore water, and sediment show major and trace element chemistries that occupy unique locations in non-metric multidimensional scaling (NMS) space. These locations in NMS space correspond to proximity of the wetland to, for example, metropolitan effluent and hypersaline GSL water (Carling et al., 2013). The Impounded wetlands contain freshwater, whereas pore water in the western-most Sheetflow wetlands have elevated salinity due to periodic encroachment by saline water from Farmington Bay, a freshwater-influenced bay of GSL. Pintail, an impounded pond at the north end of the system that is groundwater fed, shows elevated salinity relative to other freshwater wetlands in the system (Carling et al., 2013).

1.3. Reported Hg burdens in lower biota

The above-described Hg burdens in multiple avian species and exceptional MeHg concentrations in at least one aquatic settling highlight the need to understand potential pathways of Hg bioaccumulation in this hydrologic system that also has a constrained food web. Specifically, salinities >120 g L^{-1} in most parts of the GSL exclude predacious fish, so