

Total Mercury and Methylmercury Response in Water, Sediment, and Biota to Destratification of the Great Salt Lake, Utah, United States

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S Supporting Information

ABSTRACT: Measurements of chemical and physical parameters made before and after sealing of culverts in the railroad causeway spanning the Great Salt Lake in late 2013 documented dramatic alterations in the system in response to the elimination of flow between the Great Salt Lake's north and south arms. The flow of denser, more-saline water through the culverts from the north arm (Gunnison Bay) to the south arm (Gilbert Bay) previously drove the perennial stratification of the south arm and the existence of oxic shallow brine and anoxic deep brine layers. Closure of the causeway culverts occurred concurrently with a multiyear drought that resulted in a decrease in the lake elevation and a concomitant increase in top-down erosion of the upper surface of the deep brine layer by wind-forced mixing. The combination of these events resulted in the replacement of the formerly stratified water column in the south arm with one that was vertically homogeneous and oxic. Total mercury concentrations in the deep waters of the south arm decreased by approximately 81% and methylmercury concentrations in deep waters decreased by roughly 86% due to destratification. Methylmercury concentrations decreased by 77% in underlying surficial sediment, whereas there was no change observed in total mercury. The dramatic mercury loss from deep waters and methylmercury loss from underlying sediment in response to causeway sealing provides new understanding of the potential role of the deep brine layer in the accumulation and persistence of methylmercury in the Great Salt Lake. Additional mercury measurements in biota appear to contradict the previously implied connection between elevated methylmercury concentrations in the deep brine layer and elevated mercury in avian species reported prior to causeway sealing.



INTRODUCTION

Mercury is a global pollutant that migrates through natural systems by complex transformation and transport processes that are, in large part, governed by redox conditions in the environment.¹ Mercury exists in nature as elemental mercury Hg(0), divalent mercury Hg(II), and organomercury compounds such as monomethylmercury (MeHg), the bioaccumulative form of mercury. Biomagnification of MeHg can result in potentially toxic levels of MeHg exposure to humans and wildlife through consumption of prey, causing detrimental neurological, behavioral, and reproductive effects.²

The methylation of inorganic mercury in the environment is predominantly biologically mediated, occurs in anoxic environments, and is carried out largely by sulfate-reducing bacteria.^{3–5} Methylation by iron-reducing bacteria and methanogens can also be important, and many other microbes have been shown to contain the *hgcAB* gene cluster responsible for mercury methylation.^{6–9} Abiotic parameters, such as dissolved organic matter (DOM), have also been shown to increase mercury

methylation under sulfidic conditions in lab experiments,^{5,10} and MeHg concentrations in aquatic sediments are often positively correlated with concentrations of dissolved organic carbon (DOC), although this is not always the case due to the complex effects of DOM on mercury's biogeochemical cycling via its role as a substrate for microbial respiration and its role in inorganic mercury complexation and chemical speciation.^{11–13}

The Great Salt Lake (GSL) is the largest terminal lake in the Western Hemisphere, and the Western Hemisphere Shorebird Reserve Network recognizes it as a habitat of hemispheric importance for millions of migratory birds (Figure 1). Over 1.4 million shorebirds use the GSL for breeding and staging areas, and over 7 million waterfowl utilize the GSL and its adjacent 1900 km² of freshwater and brackish wetlands during some

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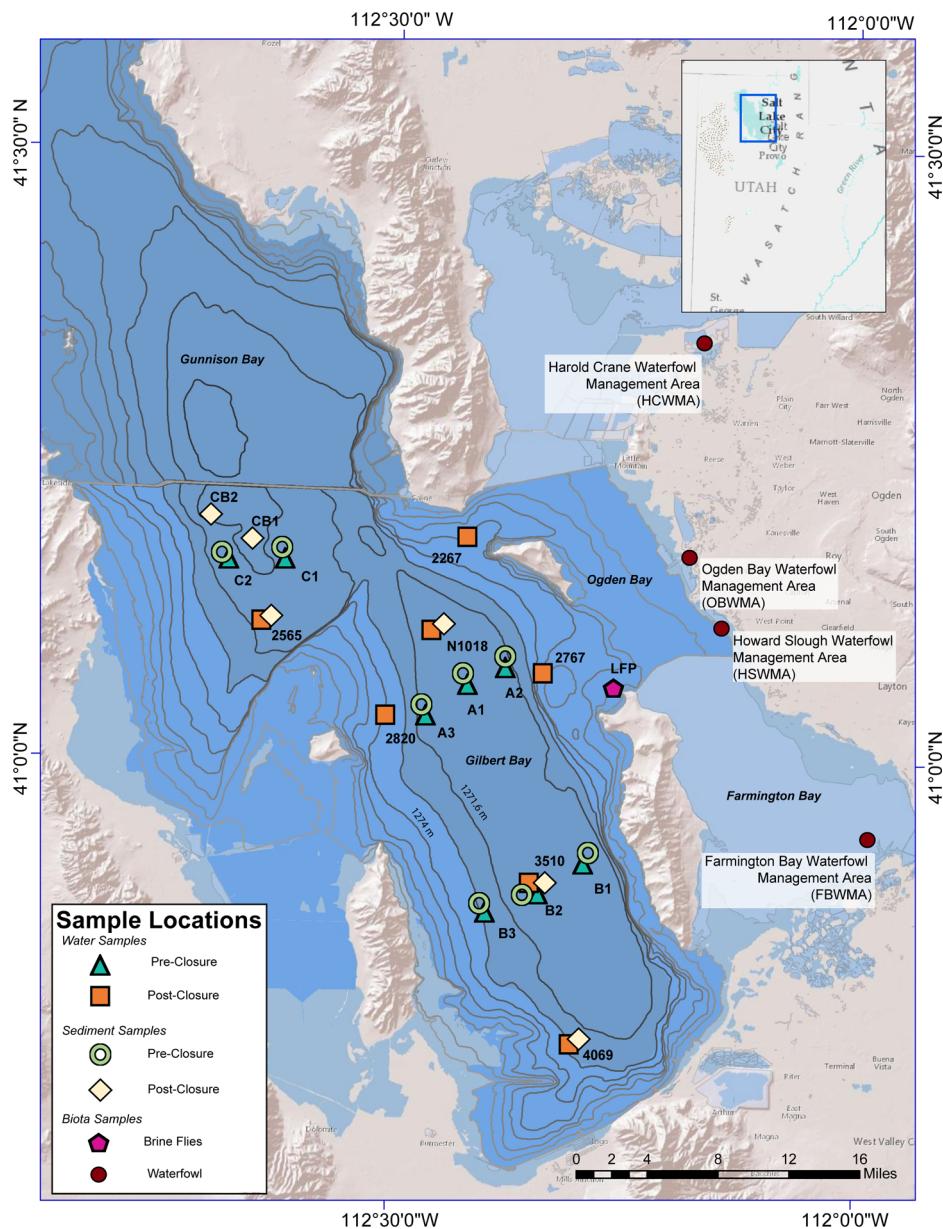


Figure 1. Locations sampled in the Great Salt Lake for water, sediment, and biota. Squares represent sample locations from 2007 to 2012, corresponding to conditions when the DBL was present. Triangles represent sample locations in 2015–2016 following causeway closure and disappearance of the DBL. Circles show locations of waterfowl harvest in 2014 and 2015. The brine fly sampling location in 2012–2015 at Lady Finger Point is indicated by a star. Bathymetric contours are from Baskin.⁴⁹ Shading represents typical salinities (percent total dissolved solids) observed in deep waters during 2007–2012, with gray corresponding to approximately 28%, darker blue corresponding to approximately 15%, and the light blue representing salinities less than 5%. Thus, the darker shading in the south arm indicates the approximate extent of the deep brine layer at the beginning of this study prior to its disappearance.

portion of their biannual migration. The GSL avian community feeds on aquatic organisms adapted to highly saline conditions, which includes brine shrimp (*Artemia franciscana*), brine fly (*Ephydria* spp.), water boatman (*Corixidae*) and numerous algal species.¹⁴ In 2007, human consumption advisories were placed on three duck species (Cinnamon Teal, Northern Shoveler, and Common Goldeneye) at the GSL based on elevated mercury levels in breast muscle tissue exceeding the EPA screening value (0.3 µg/kg, wet weight).^{15–17}

The construction of a railroad causeway in 1959 restricted water flow between the north arm (Gunnison Bay) and the south arm (Gilbert Bay).¹⁵ Because the south arm receives nearly all surface water inflows of freshwater to the GSL, the

north arm is evaporatively concentrated to a greater extent than the south arm, yielding brine in the north arm (250–280 g/L) that is 1.4 to 1.6 times more saline than the south arm (110–180 g/L), the latter being 3–5 times more saline than seawater (33–36 g/L).^{18,19} Culverts in the causeway allowed bidirectional flow, with shallow water flowing from the south arm to the north arm and deeper, denser water flowing from the north to the south arm. This flow of dense brine from the north arm, along with limited wind-driven mixing, resulted in saline-based density-driven stratification and the formation of a monomictic limnion, herein referred to as the deep brine layer (DBL), in the south arm that persisted 6–7 m below the surface of the lake, and that was not subject to annual turnover.^{15,18–20} The

DBL was anoxic with elevated DOC (59–88 mg/L) and sulfide (7–29 mg/L),^{18,22,23} high activities of sulfate-reducing bacteria,²⁴ and elevated MeHg (20 to 32 ng/L) and total mercury (HgT) (38–80 ng/L), with a high percentage of HgT existing as MeHg.²² Several studies^{20,25–27} have implicated the elevated MeHg in the DBL as a potential source of elevated mercury in biota, possibly via mixing into the oxic upper brine layer, where uptake into the food chain could subsequently occur.²¹ However, a previous laboratory study²³ reported no difference in the HgT concentration of brine shrimp grown for 14 days in a water column where a DBL was absent compared to a stratified water column where an anoxic DBL was present. They reported that brine shrimp grown in water created by mixing different ratios of the upper brine layer and DBL accumulated Hg in proportion to the Hg/particulate organic carbon (POC) ratio, such that at least the short-term effect of mixing in water from the DBL was a decrease in the HgT concentration in the brine shrimp.

Mixing between the DBL and upper brine layer in the south arm of the GSL is believed to be relatively limited, with previous research²⁸ indicating that the pools of dissolved inorganic nitrogen (DIN) and total dissolved phosphorus (TDP) in the shallow brine layer are only weakly coupled to those in the deep brine layer. However, despite persistent stratification in the GSL, Beisner et al.²⁹ reported evidence that limited mixing between the DBL and shallow brine layer occurred during conveyance of the DBL from the northern to southern areas of the south arm, and Jones and Wurtsbaugh²³ estimated that 40% of the DBL was entrained into the shallow brine layer annually. Such mixing would allow for the transfer of MeHg from the DBL to the overlying shallow brine layer, and by extension, into the base of the food chain and eventually to brine fly larvae and brine shrimp that serve as the diet for many avian species.^{23,27,28,30,31} Such an indirect connection was inferred by Johnson et al.²² based on elevated Hg concentrations previously reported in birds (eared grebes)³² and brine flies²⁶ on the southwestern side of Gilbert Bay (where the DBL existed at depth in the most proximal water body), as well as due to the temporal correspondence of reduced Hg(II)-methylation rates in the DBL and reduced Hg burdens among aquatic invertebrates (brine shrimp^{30,33} and brine flies).²⁶

In November 2012 and December 2013, two railway causeway culverts that allowed limited water flow between the north and south arms of the GSL were closed due to deteriorating structural integrity. These closures provided the opportunity to determine the geochemical response of the system to elimination of flow between the north and south arms and destratification and disappearance of the DBL in the south arm (described below). This event also allowed us to quantify how HgT and MeHg concentrations in the water column, surficial sediment, and biota responded to the disappearance of the DBL, thus providing insight into the role of the DBL in the biogeochemical cycling of mercury in the GSL.

METHODS

Water and Sediment Sampling and Analysis. Periodically from October 2015 to December 2016, water column chemical and physical conditions were characterized at seven locations across the south arm of GSL (Figure 1). Water samples were collected at five stations (2267, 2565, 2767, N1018, 2820, 3510, and 4069) at 0.2 m below lake surface and

0.5 m above lake bottom, referred to as shallow and deep samples, respectively. The data from these samples collected after the disappearance of the DBL were compared to the previous data collected from the shallow and deep brine layers from May 2007 to December 2012, prior to causeway closure when the DBL was present^{22,34} (Figure 1). Water-column temperature, specific conductance, pH, and dissolved oxygen (DO) were measured in the field using a YSI Professional Series Quatro probe that was calibrated within 12 h prior to sampling.²² The YSI probe corrects for temperature but not salinity. Thus, the DO values presented have a positive bias and are qualitative but are still useful for accurately denoting oxic versus anoxic conditions. Sulfide was measured in filtered water samples in the field immediately after collection using a photometric method (V-2000 Multianalyte LED Photometer and Vacu-vials, CHEMetrics).²²

Clean hands—dirty hands protocol was followed during sample collection and analysis.³⁵ Unfiltered Hg water samples were collected by peristaltic pump using acid-washed PTFE tubing into precleaned FLPE bottles. Bottles used to collect anoxic water samples were filled to overflowing to minimize headspace. Filtered water samples were passed through a 0.45 μm pore size, preacid-rinsed capsule filters in the field (Geotech Environmental). After collection, water samples for HgT and MeHg analyses (unfiltered) were stored on ice in the field and acidified to 0.5% using trace-metal-grade sulfuric acid for preservation the same day as sampling. All samples were then refrigerated and analyzed within 2 weeks of collection.

HgT samples were oxidized by amendment to 5% BrCl at least 24 h prior to analysis. This higher BrCl concentration relative to established protocols that call for 1% BrCl was necessary³⁶ to fully oxidize the high levels of dissolved organic matter (75.4 ± 11.1 mg/L) in these water samples (as described below). HgT concentrations in water were determined via reduction with SnCl₂, purge and trap onto gold traps, and thermal desorption, with quantification by cold vapor atomic fluorescence spectroscopy (CVAFS) using a MERX-T automated system (Brooks Rand) using established techniques.³⁶ Mean laboratory blank concentrations were 0.07 ± 0.008 ng/L HgT. HgT matrix spike recovery averaged $98.3 \pm 3.2\%$ ($n = 3$). MeHg concentration in water was measured after distillation with ammonium pyrrolidine dithiocarbamate (APDC), followed by aqueous-phase ethylation, purge and trap onto tenax traps, thermal desorption, pyrolytic decomposition, and CVAFS detection using a MERX-M automated system (Brooks Rand) using established techniques.³⁷ MeHg blanks averaged 0.15 ± 0.10 ng/L. Because no certified reference material exists for MeHg in water, MeHg matrix spikes were included in each distillation; MeHg spike recoveries ($n = 2$) averaged $93 \pm 29\%$. Water samples for dissolved organic carbon (DOC) analysis were placed on ice in the field then placed in a refrigerator upon return to the laboratory. DOC was measured in water samples (TOC-5000a, Shimadzu) within 1 week of sample collection using EPA method 1684.³⁸

Surficial sediment was sampled at 11 sites (A1, A2, B1, B2, C1, C2, CB, 2565, 3510, 4069, and N1018) in the GSL (Figure 1) and were collected by peristaltic pump using acid-washed PTFE tubing into precleaned 500 mL FLPE bottles filled to overflowing to minimize headspace and stored on ice in the field. The preferred method for examining vertical variations involves collecting a sediment core that is subsequently sectioned. However, because our focus was geographic variations in surficial sediment, we employed a faster method

to collect sediment. The method composites surficial sediment across depths of a few centimeters. Naftz et al.³⁹ reported 40% variation of HgT concentration within the top 2 cm of GSL sediment. Previous studies²² using this method showed relatively low variability in HgT and MeHg in surficial sediment, indicating that collected sediment was representative.

Subsamples were oven-dried at 105 °C for 12 h and reweighed to determine water content and allow conversion between wet weight (ww) and dry weight (dw) concentrations (without salt correction). HgT was extracted from sediment by digestion in a 7:3 mixture of HNO₃/H₂SO₄ (trace metal grade) at 80 °C for 6 h, followed by amendment to 5% BrCl.³⁶ MeHg was leached from sediment with a mixture of potassium bromide, sulfuric acid, and copper sulfate, extracted into methylene chloride, and back-extracted into water, followed by aqueous-phase ethylation, purge-and-trap, thermal desorption, pyrolytic decomposition, and CVAFS detection according to established techniques.^{36,37,41} Certified reference materials (CRMs) for HgT (MESS-3) and MeHg (CC-580) in sediment were also analyzed, with recoveries averaging 99% and 119%, respectively.

Biota Collection and Analyses. Adult brine flies (*Ephydria* spp.) were collected from Lady Finger Point on Antelope Island of the GSL (Figure 1) from spring to fall of 2012 to 2015. Flies were collected with nets, transferred into polypropylene tubes, placed on ice in the field, and frozen back in the lab the same day. A total of 76 waterfowl were harvested in mid-September of 2014, and 99 were harvested in late August and early September of 2015 from the GSL and surrounding wetlands by the Utah Division of Wildlife Resources. The waterfowl were harvested from the Farmington Bay, Ogden Bay, Howard Slough, and Harold Crane Waterfowl Management Areas (Figure 1). Ducks were harvested with shotguns using nonlead shot then frozen. The age of each bird was determined by physical characteristics: examining rectrices, wing and body plumage, and cloacal characters. The species sampled were Northern Shovelers (*Anas clypeata*) (DBL present: $n = 16$; DBL absent: $n = 16$), Mallard (*Anas platyrhynchos*) (DBL present: $n = 16$; DBL absent: $n = 21$), Gadwall (*Anas strepera*) (DBL present: $n = 15$; DBL absent: $n = 22$), and Cinnamon Teal (*Anas cyanoptera*) (DBL present: $n = 23$; DBL absent: $n = 31$). Tissue samples were thawed, a scalpel was used to remove the skin, and breast muscle tissue was harvested and stored in a Whirl-Pak (NASCO) polyethylene bag and frozen. Breast muscle was analyzed because it is the most likely tissue to be consumed by duck hunters.

Both brine fly and waterfowl samples were freeze-dried and homogenized prior to analysis, and thus, all HgT concentrations in biota are reported on a dry-weight basis. Brine flies were digested in Teflon vials with 10 mL of a 2:1 mixture of trace-metal-grade nitric and sulfuric acids. Samples were allowed to predigest at room temperature for 1 h and were then heated to 100 °C for 4 h. Following the digestion, samples were amended to 1% BrCl. All digestions included at least two digestion blanks and two certified reference materials (TORT-2 and DORM-3, National Research Council Canada), each digested in duplicate. Aliquots of the digested samples were measured by oxidation with BrCl, reduction with SnCl₂, purge-and-trapping using dual-stage gold trap amalgamation, and quantification by CVAFS.³⁶ The average daily HgT detection limit, based on 3 times the standard deviation of digestion blanks, was 2.3 ng g⁻¹, assuming a 100 mg sample. Recoveries for HgT in the biota certified reference materials averaged

101.4% ± 7.8% ($n = 88$). The duck muscle tissue was analyzed using thermal decomposition, amalgamation, and atomic absorption spectrophotometry using a DMA-80 (Milestone) following established protocols.⁴² Muscle tissue samples were only analyzed for HgT because previous studies have shown that >95% of Hg in most bird muscle is MeHg.⁴³ Each analysis run included each of the two certified reference materials (TORT-2 and DORM-3) analyzed in duplicate, with recoveries of HgT ranging from 88% to 123%. Analytical precision of the HgT measurements was assessed by analyzing eight samples in triplicate, and the average percent relative standard deviation was 5.2%.

The HgT concentrations in the waterfowl were log-transformed prior to statistical analysis to meet the assumptions of parametric statistics. The data were analyzed using multifactor ANOVA using a crossed design. While we did not anticipate a priori that the sex of the waterfowl would have any effect on their HgT concentrations, we initially included sex in the statistical model, which included the following five variables as fixed factors: duck species, age, year, site, and sex. The fully crossed model could not be run due to missing cells because not all sites were sampled both years. We therefore used a reduced model made by removing the five-way and four-way interactions.⁴⁴ In this model none of the interactions involving sex, nor sex as a main factor, were significant ($p > 0.49$ in all cases). Given this result along with our a priori assumption regarding the importance of sex, this factor was removed from the model. The multifactor ANOVA using a crossed design with the fixed factors duck species, age, year, and site was then reduced by removing the four-way interaction and the three-way interaction age × site × year due to the missing cells, as described above.

RESULTS

Water Quality Parameters. Before 2013, prior to closure of the causeway culverts, shallow waters in the south arm were oxic (DO = 9.48 ± 0.70 mg/L), whereas deep waters in areas where the DBL exists were anoxic (DO = 0.05 ± 0.02 mg/L) (Figure 2). After causeway closure, mean DO was 9.1 ± 3.4 mg/L in shallow waters and 7.2 ± 5.2 mg/L in all deep waters. Prior to culvert closure, DO concentrations were significantly higher in surface waters than in the DBL (t test, $p \ll 0.001$), whereas there was no significant difference in DO between shallow and deep waters ($p = 0.52$) two years after the culverts were closed. There was no change in DO in shallow waters following culvert closure ($p = 0.81$).

Under pre-closure conditions (before 2013), mean pH in shallow (8.35 ± 0.71) and deep waters (7.62 ± 0.12) differed significantly (t test, $p = 0.01$). However, 2 years after the culvert closures (late 2015–2016), mean pH was 8.27 ± 0.40 and 8.09 ± 0.20 for shallow and deep waters, respectively, with no significant difference ($p = 0.39$) (Figure 2). Under pre-closure conditions, mean sulfide concentrations in filtered shallow waters (1.3 ± 1.0 mg/L) were significantly lower ($p = 0.001$) than sulfide concentrations in filtered deep waters (19.2 ± 7.6 mg/L). After culvert closure, there was no longer a difference ($p = 0.51$) between mean sulfide concentrations in filtered shallow waters (0.04 ± 0.02 mg/L) and deep waters (0.05 ± 0.01 mg/L) (Figure 2).

Before causeway closure, mean concentration of DOC in the shallow waters was 49 ± 12 mg/L compared to 75 ± 11 mg/L in the pre-closure DBL, with these being significantly different ($p = 0.005$). After causeway closure, mean DOC concentrations

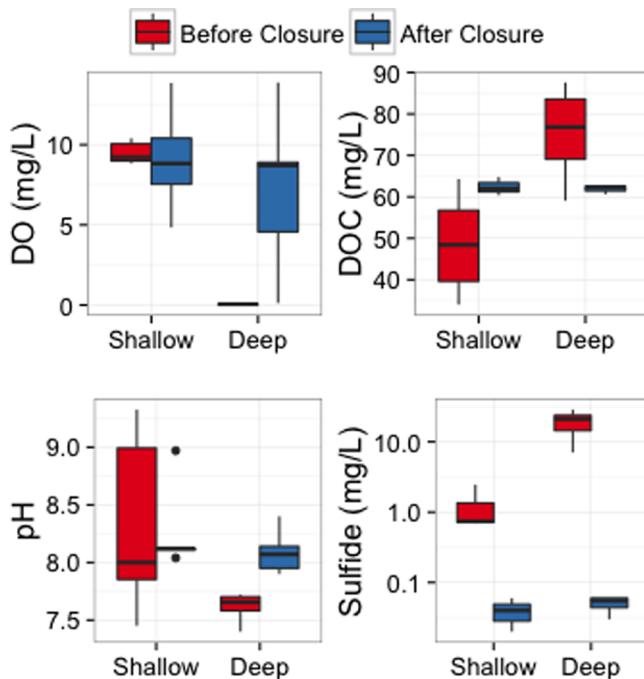


Figure 2. Boxplots of dissolved oxygen (DO), pH, sulfide, and DOC concentrations in the shallow and deep waters of the GSL with DBL present (2007–2012) and after DBL disappearance (2015–2016). DBL-present: shallow ($n = 9$) and deep ($n = 6$); DBL-absent: shallow ($n = 5$) and deep ($n = 5$). The middle line represents the median, the outer edges of filled boxes represent first and third quartiles, and the stems represent data within 1.5 of the interquartile range as an estimate of the 95% confidence interval.

in shallow waters (61.9 ± 1.2 mg/L) and deep waters (62.4 ± 2.2 mg/L) were no longer significantly different ($p = 0.75$) (Figure 2). Depth profiles developed using data from the USGS and the State of Utah for DO, temperature, and specific conductance (SC) demonstrate the disappearance of the deep brine layer after causeway closure, as evidenced by the loss of the sharp interface in these parameters between shallow and deep waters (Figures S1–S5).

Mercury Concentrations in Water and Surficial Sediment. Prior to causeway closure (before 2013), mean HgT concentrations in unfiltered waters were 44.8 ± 15.5 and 6.15 ± 5.2 ng/L in the deep and shallow brine layers, respectively (Figure 3), which were significantly different ($p \ll 0.001$). Mean unfiltered MeHg concentrations in water were 21.4 ± 9.0 and 1.2 ± 1.6 ng/L in the deep and shallow brine layers, respectively, yielding a significant difference ($p \ll 0.001$) between shallow and deep waters for MeHg prior to culvert closure (Figure 3). Following culvert closure (late 2015–2016), mean unfiltered HgT concentrations were 8.4 ± 6.1 and 4.6 ± 2.1 ng/L in deep and shallow samples, respectively, and mean unfiltered MeHg concentrations were 3.0 ± 4.5 and 0.8 ± 0.65 ng/L in deep and shallow samples, respectively. These post-closure results showed no significant difference between deep and shallow samples for unfiltered HgT ($p = 0.12$) or MeHg ($p = 0.24$). Thus, following culvert closure and disappearance of the DBL, concentrations of unfiltered HgT and MeHg decreased in deep waters of the south arm by 81% ($p \ll 0.001$) and 86% ($p \ll 0.001$), respectively. The concentration of unfiltered HgT and MeHg in surface water decreased slightly by 25% and 34%, respectively. However, neither of these measured decreases were statistically significant ($p = 0.13$ and p

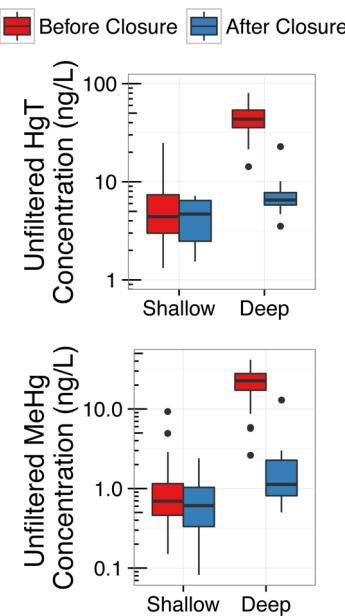


Figure 3. Unfiltered HgT and unfiltered MeHg concentrations from shallow and deep water samples from before (2007–2012) and after (2015–2016) the culvert closures in the GSL causeway and disappearance of the DBL. Unfiltered HgT concentrations before closure ranged from 1.3 to 24.8 ng/L in shallow waters ($n = 36$) and from 14.2 to 80.2 ng/L in deep waters ($n = 28$). Unfiltered MeHg concentrations before closure ranged from 0.15 to 9.3 ng/L in shallow waters ($n = 36$) and 2.63 to 41.6 ng/L in deep waters ($n = 27$). Unfiltered HgT concentrations after closure ranged from 1.54 to 7.2 ng/L in shallow waters ($n = 36$) and 3.5 to 22.8 ng/L in deep waters ($n = 8$). Unfiltered MeHg concentrations after closure ranged from 0.08 to 2.4 ng/L in shallow waters ($n = 16$) and 0.50 to 13.0 ng/L in deep waters ($n = 7$). The middle line in each box represents the median, outer edges of filled boxes represent the first and third quartiles, and stems represent data within 1.5 of the interquartile range as an estimate of the 95% confidence interval.

= 0.19, respectively). Prior to culvert closure, the fraction of HgT composed of MeHg in the deep brine layer was 47%,²² whereas following causeway closure this fraction decreased to 35% in deep waters.

Average HgT and MeHg concentrations in surface sediment underlying the DBL in the south arm before culvert closure were 103 ± 80 ng/g (dw) and 0.87 ± 0.54 ng/g (dw), respectively (Figure 4). Average HgT and MeHg concentrations in surface sediment underlying the DBL following causeway sealing were 117 ± 105 ng/g (dw) and 0.20 ± 0.39 ng/g (dw), respectively. While HgT concentrations in surface sediment did not change ($p = 0.75$) in response to causeway closure, MeHg concentrations in surface sediments decreased by roughly 77% ($p < 0.001$) during this same time period.

Mercury Concentrations in Biota. Mean HgT concentrations in adult brine flies were 440 ± 110 ng/g (dw) and 530 ± 200 ng/g (dw) for DBL-present versus DBL-absent conditions, respectively, with this increase being statistically significant ($p \ll 0.001$). Brine fly Hg burdens separated by month (Figure 5) show higher HgT concentrations under DBL-absent conditions during May to August.

Mean HgT concentrations in all waterfowl were 440 ± 480 ng/g (dw) for pre-closure versus 410 ± 380 ng/g (dw) for post-closure conditions, respectively. Waterfowl Hg concentrations by species (Figure 6) show mean HgT concentrations in pre-closure versus post-closure samples were 480 ± 230 ng/g

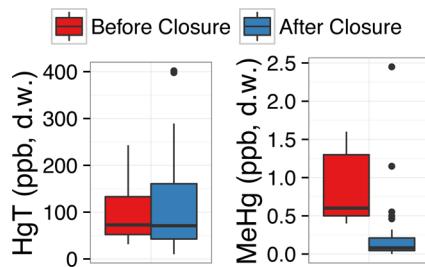


Figure 4. Box plots of HgT and MeHg concentrations in surficial sediment collected from sites underlying the DBL when the DBL was present in 2012 ($n = 6$ and $n = 6$, respectively) and after its disappearance in 2015–2016 ($n = 36$ and $n = 47$, respectively). The middle line in each box represents the median, outer edges of filled boxes represent the first and third quartiles, and stems represent data within 1.5 of the interquartile range as an estimate of the 95% confidence interval.

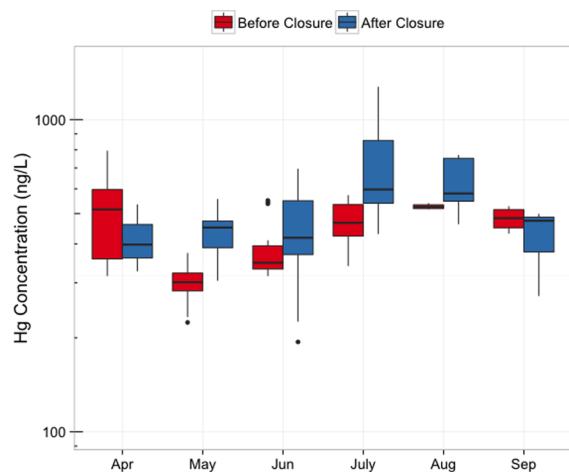


Figure 5. Concentrations of HgT in adult brine flies by month when the DBL was present (2012–2013, $n = 102$) and after disappearance of the DBL (2014–2015, $n = 112$). The middle line in each box represents the median, outer edges of filled boxes represent the first and third quartiles, and stems represent data within 1.5 of the interquartile range as an estimate of the 95% confidence interval.

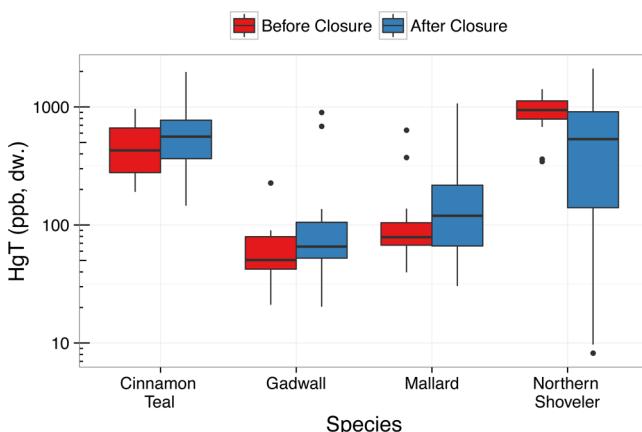


Figure 6. Total mercury concentrations in muscle tissue of waterfowl when the DBL was present (September, 2014) vs when absent (August–September, 2015). The middle line in each box represents the median, outer edges of filled boxes represent the first and third quartiles, and stems represent data within 1.5 of the interquartile range as an estimate of the 95% confidence interval.

Table 1. Multifactor ANOVA Results for Log-Transformed HgT in Waterfowl Muscle Tissue Using a Crossed Experimental Design That Included the Four Fixed Factors of Duck Species, Age, Site, and Year^a

source	type III SS	degrees of freedom (df)	mean square	F ratio	p value
duck species	8.749	3	2.920	25.16	<0.001
age	0.798	1	0.798	6.89	0.010
site	0.841	3	0.280	2.42	0.070
year	0.092	1	0.920	0.79	0.375
age × site	0.351	3	0.117	1.01	0.392
duck species × age	1.378	3	0.459	3.96	0.010
age × year	0.567	1	0.567	4.89	0.029
duck species × site	3.257	8	0.407	3.51	0.001
site × year	0.055	2	0.280	0.24	0.788
duck species × year	0.878	3	0.293	2.53	0.061
duck species × age × site	0.298	5	0.060	0.52	0.764
duck species × age × year	0.775	3	0.258	2.23	0.088
species × site × year	0.579	2	0.289	2.50	0.086
intercept	206.4	1	206.4	1780	<0.001
error	14.0	121	0.116		

^aThe model was run after the removal of the four-way interaction and one three-way interaction due to missing cells.

(dw) versus 690 ± 480 ng/g (dw) (Cinnamon Teal), 67 ± 49 ng/g (dw) versus 140 ± 220 ng/g (dw) (Gadwall), 130 ± 160 ng/g (dw) versus 230 ± 270 ng/g (dw) (Mallard), and 920 ± 300 ng/g (dw) versus 640 ± 590 ng/g (dw) (Northern Shovelers).

There was a significant effect ($p \ll 0.001$) of species on duck HgT concentration (Figure 6) according to a multifactor ANOVA analysis using a crossed experimental design including the fixed factors: duck species, age, site, and year (Table 1). Northern shovelers and cinnamon teal had the highest HgT concentrations, followed by mallards and gadwalls with the lowest HgT concentrations. Tukey pair-wise post hoc comparisons demonstrated that HgT concentrations in northern shovelers and cinnamon teal were not significantly different ($p = 0.99$), while HgT in both mallards and gadwalls were significantly different from all other duck species ($p < 0.013$ in all cases). There was no significant difference ($p = 0.38$) in HgT in ducks for pre-versus post-closure (Figure 6 and the Supporting Information), nor was there any significant difference in duck HgT concentrations by site ($p = 0.07$) (Figure S8).

The significant interaction ($p = 0.01$) between duck species and age (Figure S6) was due to the effect of age on HgT concentration not being the same in all duck species. Using planned contrasts, we found that there was a significant decrease in HgT between juveniles (hatching year) versus adults (after hatching year) for northern shovelers ($p = 0.011$) and mallards ($p = 0.016$), but there was no effect of age on HgT in cinnamon teal ($p = 0.48$) or gadwalls ($p = 0.59$) (Figure S6). This significant interaction ($p = 0.029$) between duck age and year (Figure S7) was due to a minor difference in HgT concentrations between 2014 and 2015 that varied slightly between the two age classes. However, this interaction did not affect our ability to interpret the main effect described above.

■ DISCUSSION

The disappearance of the major contrast in DO, pH, and sulfide concentrations between shallow and deep waters of the south arm that existed prior to culvert closure (Figure 2), along with the observed disappearance of measurable sulfide and the increase in DO to saturated levels across the water column of the GSL, demonstrate disappearance of the DBL in 2014, such that a single oxic brine occupied the water column from surface to bottom after closure. The significant variance in DO in the shallow waters reflects daily and seasonal changes in photosynthesis.⁴⁵ In contrast, variance in DO in deep waters after causeway closure reflects differences in water column depth (persistence of DBL in deeper sites), and thus differences in the timing of the disappearance of the DBL at each site (Figures S1–S4). Although the timing of the changes in water chemistry and disappearance of the DBL suggests a response to culvert closure and loss of bidirectional flow, drought conditions and lower lake levels allowed wind-driven mixing at depth to erode the upper surface of the DBL and thus also contributed to the shift from a stratified to nonstratified system.

The observed change in geochemical regime and destratification was accompanied by decreases in the concentrations of unfiltered HgT and MeHg in deep waters of the GSL of 81% and 86%, respectively (Figures 2 and 3). Elevated Hg methylation rates and MeHg concentrations have previously been reported in the anoxic hypolimnions of freshwater lakes during seasonal stratification^{46,47} as well as below the interface of the suboxic hypolimnion of the Black Sea.⁴⁸ The dramatic decrease in MeHg in deep waters of the GSL following transient destratification are consistent with changes documented for seasonally anoxic hypolimnions of freshwater lakes.⁴⁷ Our result provide additional insight into the potential response of freshwater lacustrine and marine systems following destratification and reoxygenation of anoxic bottom waters.

The volume of the DBL prior to causeway sealing was estimated by integration between the bathymetric surface⁴⁹ and the maximum elevation of the DBL surface (1272.6 m, which is about 2.5 m above the deepest point of the GSL) using 3D analyst tools in ArcGIS.⁴³ Multiplying the resulting DBL volume (1.00 km^3) by the average pre-closure concentrations of HgT and MeHg in the DBL yielded approximately $45 \pm 16 \text{ kg HgT}$ and $21 \pm 9 \text{ kg MeHg}$ in the DBL prior to its disappearance. Based on time series depth profile data of DO, specific conductance, and temperature collected by the USGS (Figures S2 and S5), we estimate that the elevation of the DBL decreased from approximately 1272.5 to 1271.4 m between December 2013 and June 2014 following the culvert closures, yielding a decrease in the volume of the DBL of approximately 0.69 km^3 . The corresponding water and mercury fluxes for DBL mixing into the upper brine layer averaged across these 6 months are approximately $3.8 \times 10^6 \text{ m}^3 \text{ day}^{-1}$, $0.17 \text{ kg HgT day}^{-1}$, and $0.082 \text{ kg MeHg day}^{-1}$. This simple calculation results in an annual flux from the DBL (when present and being eroded due to decreasing lake level) to the upper brine layer of $63 \pm 22 \text{ kg HgT}$ and $30 \pm 13 \text{ kg MeHg}$, which is roughly twice what was estimated for a steady-state flux by Jones and Wurtsbaugh.²³

Mixing of HgT and MeHg from the DBL into the upper brine was clearly not conservative (Figure 3) because HgT and MeHg concentrations in surface water also decreased, although these changes were not statistically significant ($p = 0.14$ and 0.19 , respectively). This is in contrast to the 3.2 ng/L ($\sim 75\%$)

increase in HgT in surface waters that would have occurred had the HgT lost from the DBL been redistributed across the entire water column (see the Supporting Information). The MeHg lost from deep waters of the GSL did not occur through sedimentation, as demonstrated by the concurrent 77% decrease in the concentration of MeHg in sediment previously underlying the DBL (Figure 4). If all of the HgT lost from deep waters of the GSL had partitioned onto the top 1 cm of underlying sediment, this would have resulted in an increase in their HgT concentration of less than 5 ng/g ($\sim 5\%$). We would not have had the statistical power to quantify a change this small given the spatial variability that existed for HgT in sediment both before and after disappearance of the DBL, and instead we measured no change ($p = 0.75$) in the concentration of HgT in sediment (Figure 4). If the Hg lost from deep waters was not removed by particle settling or partitioning into underlying sediments, the Hg loss may have occurred via volatilization of Hg(0) to the atmosphere or redistribution onto sediments distal to where the DBL had been present, although neither of these were measured.

Our findings suggest that the DBL, when present, acts as a sort of cap that promotes the accumulation of MeHg in both surficial sediment and the DBL. We speculate that removal of the DBL allowed for the more-facile export of MeHg from the sediment into surface waters, which in turn facilitated the loss of MeHg from the system, such as via photodemethylation.⁵⁰ It is unclear if the disappearance of the DBL also resulted in a decrease in net Hg methylation in surficial sediment due to the change in redox conditions and other parameters or if the decrease in MeHg observed in the lake sediment was purely due to increased transport of MeHg out of the surficial sediment. Sedimentation rates during the last 50 years in the GSL reported in previous studies are on the order of $0.027 \text{ g cm}^{-2} \text{ year}^{-1}$ or roughly 0.1 cm year^{-1} for linear sedimentation rate.^{39,40} This would result in approximately 0.3 cm of material being added to the surficial sediment during the roughly 3 year period of our study following the disappearance of the DBL, or only a 10% increase in the top 3 cm of sediment that is composited and collected by our sampling method. Hence, increased sedimentation of material low in MeHg is an unlikely mechanism to explain the 77% decrease in the concentration of MeHg in surficial sediment we documented based on mass balance considerations, especially given that the concentration of HgT in these sediments did not change over the same time period.

Concentrations of HgT in adult brine flies (*Ephedra* spp.) collected along the south arm of the GSL were higher for post-versus pre-closure conditions during four of the six warmest months (Figure 5). Brine fly development progresses through larval and pupal stages in the lake before the adult brine flies emerge. Brine flies in all three stages of their life cycle are present at the GSL throughout the year, with larval densities lowest and pupal densities highest in June when adult emergence approaches its annual maximum.⁵¹ There are 1 to 2 generations of brine flies per year in the GSL, indicating that brine flies sampled in one year were not part of the same cohort as those sampled in another year. On average, adult *Ephydria* survive for less than a week, and unlike other species in the *Ephydria* genus, feeding by adult brine flies of the *Ephydria* species at the GSL is thought to be limited due to the inadequate availability of suitable food.^{51–53} If *Ephydria* at the GSL do not feed extensively as adults, then most Hg bioaccumulation from their diet occurs primarily during the

larval stage. The loss of sulfide from the water column may have caused a shift from mercury-sulfide complexation to mercury-chloride complexation, thus affecting bio-uptake by brine fly larvae and their periphyton prey. However, the extent to which the changes in HgT concentrations in the brine flies reflect changes in chemical complexation or other changes due to the disappearance of the DBL versus other seasonal or interannual influences is unclear.

None of the four avian species sampled exhibited a statistically significant change in HgT when samples collected were compared before and after the disappearance of the DBL. Northern shoveler (*A. clypeata*), which had the highest HgT concentrations in this study, feed higher on the food web and move further west in the fall and winter than the other species in this study, feeding primarily on aquatic invertebrates from open water regions of the GSL, principally brine shrimp (*Artemia*), brine shrimp cysts, brine fly larvae, and water boatman (*Corixidae*) similar to the diet of northern shoveler reported for other areas of the United States.^{54,55} The other duck species sampled in our study have more generalized feeding habits and lower trophic status. Cinnamon teal (*A. cyanoptera*) mainly feed on submerged aquatic vegetation, emergent and aquatic invertebrate larvae and pupae, and seeds in wetlands and shallower water along the lake^{54,56,57} but switch to feeding on the open water of the GSL in autumn.⁵⁴ Gadwall (*A. strepera*) and mallard (*A. platyrhynchos*) had the lowest HgT concentrations and both primarily feed on freshwater plants and seeds in wetlands rather than the open GSL, with minimal consumption of invertebrates.^{58–60}

Given the diet of the cinnamon teal, gadwall, and mallard and the fact that most of their foraging does not occur in the open waters of the GSL, a change in the concentration of HgT or MeHg in the open waters of the south arm would not be expected to necessarily be reflected in the Hg concentration in these ducks. However, the diet and greater utilization of open water of the GSL for foraging by northern shoveler might be expected to make it more likely that any changes in HgT or MeHg concentrations or cycling in the south arm would be reflected in the HgT levels in this avian species. While we measured dramatic decreases in the concentrations of HgT and MeHg in deep waters of the south arm due to the disappearance of the DBL, there was no statistically significant change in the concentration of HgT or MeHg in unfiltered surface waters (Figure 3), and similarly, there was no measurable change in the concentration of HgT in northern shoveler (Figure 6). The dramatic decrease in HgT and MeHg in deep waters of the GSL did not result in a decrease in HgT in northern shoveler, which may suggest a lack of the previously implied indirect connection between elevated HgT and MeHg in the DBL and elevated mercury in avian and other species at the GSL. Alternatively, this lack of observed HgT decrease in biota concomitant with HgT and MeHg decrease in the deep waters of the GSL following disappearance of the DBL may reflect the possibility that the inferred indirect link (which must propagate through the upper brine layer) may be temporally slow and may yet still manifest itself. Even prior to causeway sealing, significant interannual variability in avian HgT concentrations were observed.^{16,17,61} Despite the baseline variability, the consistency of pre- versus post-closure avian HgT concentrations suggest an absence of direct connection between elevated avian HgT and elevated MeHg/HgT in the DBL. It is unclear whether other avian species that feed

primarily in the open waters of the south arm, such as eared grebes or phalaropes, would have indicated a connection.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b05790.

Tables showing summaries of waterfowl mercury concentration and the mass balance results. Figures showing water column DO profiles, profiles of specific conductance, water column temperature profiles, water column conductance, interaction plots, and mercury concentrations in waterfowl. (PDF)

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