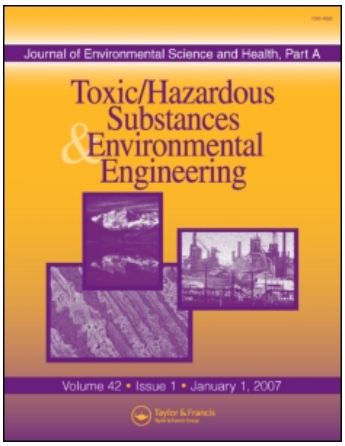

Journal of Environmental Science and Health, Part A
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597268

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Online Publication Date: 01 December 2008

To cite this Article Yu, Kewei, DeLaune, Ronald D., Devai, Istvan, Tao, Rui and Jugsujinda, Aroon(2008)'Total and methyl mercury in wetland soils and sediments of Louisiana's Pontchartrain Basin (USA)',Journal of Environmental Science and Health, Part A,43:14,1657 — 1662

To link to this Article: DOI: 10.1080/10934520802330008

URL: http://dx.doi.org/10.1080/10934520802330008

Journal of Environmental Science and Health Part A (2008) **43**, 1657–1662

ISSN: 1093-4529 (Print); 1532-4117 (Online)
DOI: 10.1080/10934520802330008

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Total and methyl mercury in wetland soils and sediments of Louisiana's Pontchartrain Basin (USA)

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Accumulation of methylmercury (MeHg) in aquatic biota is a primary toxicological concern associated with Hg contamination in the environment. This study reports total mercury (THg) and MeHg measurements in 11 swamp and 24 marsh soils/sediments in wetlands surrounding Lake Pontchartrain and Lake Maurepas located in Louisiana's Pontchartrain Basin. The salinity level ranged from fresh, brackish to salt water. Average THg content in the swamp soils/sediments (112.3 μg kg^{-1}, n = 10) was significantly higher ($P = 0.04$) than in the marsh soils/sediments (56.5 μg kg^{-1}, n = 24). The THg content in the marsh soils/sediments tended to decrease with salinity increase, probably due to geographical locations of the sampling sites with less Hg input in more saline regions. Average MeHg content in the soils/sediments was 1.3 μg kg^{-1} (n = 34), higher than reported values in the bottom sediments of Lake Maurepas (0.8 μg kg^{-1}, n = 27) and Lake Pontchartrain (0.6 μg kg^{-1}, n = 147). Average MeHg/THg ratio in the marsh soils/sediments (0.022) was considerably higher than in the swamp soils/sediments (0.012). Analysis of MeHg/THg ratio along the salinity gradient at the marsh soils/sediments show that the highest MeHg/THg ratio (up to 0.040, n = 5) was found at the fresh/brackish water sites, and the lowest (0.002, n = 1) at the salt water site. Results suggest that there was a greater potential for MeHg formation in wetland soils/sediments than in bottom sediments of adjacent lakes. Results suggest that wetland surrounding the lakes may be a potential source of MeHg to the aquatic food chain and significance is governed by area of the adjacent wetland.

Keywords: Total mercury, methyl mercury, wetlands, salinity, Pontchartrain.

Introduction

Mercury (Hg) is a widely distributed and persistent pollutant in the environment. Anthropogenic activities can cause Hg contamination by discharging into adjacent soils, sediments and water bodies and by atmospheric deposition.[1] Under oxidizing conditions Hg (II) is the dominant form. Under reducing conditions methylmercury (MeHg), a more toxic and mobile form than its precursor, can be formed.[2] It has been reported that sulfate-reducing bacteria are primarily responsible for the Hg methylation process in the environment.[3,4]

Accumulation of MeHg in aquatic biota is a primary toxicological concern related to Hg in the environment. Methylmercury can be biomagnified in food webs to levels that may potentially be hazardous to wildlife and humans through fish consumption.[2] Many water bodies in United States have been impacted by Hg contamination resulting elevated Hg concentrations in fish and wildlife, thus public health agencies have to issue advisories to the public on fish consumption.[5] Louisiana Department of Health and Department of Environmental Quality have issued advisories for a number of lakes statewide, which suggests the desirability of these lakes for fishing has decreased.[6]

Bioaccumulation and trophic transfer of MeHg in aquatic ecosystems can be influenced by level of inorganic Hg present and activity of Hg methylating organisms governed by a variety of environmental factors.[2,7] The reducing conditions required for Hg methylation process suggest lake sediments and wetlands can be sources of MeHg in aquatic environments. Although wetlands are well known as sinks of heavy metals including Hg, increasing evidences have suggested that wetlands may be a major source of MeHg to receiving waters.[8] Near-shore zones are of particular interest for enhanced MeHg biological uptake, because (1) MeHg is continuously supplied from wetlands, tributaries and in situ production in shallow sediments; (2) fish spend a large portion of their life time, spawning and feeding, in these regions.

This study reports total mercury (THg) and MeHg measurements in wetland soils/sediments of Louisiana's Pontchartrain Basin with salinity level ranging from fresh,

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Received August 21, 2007.

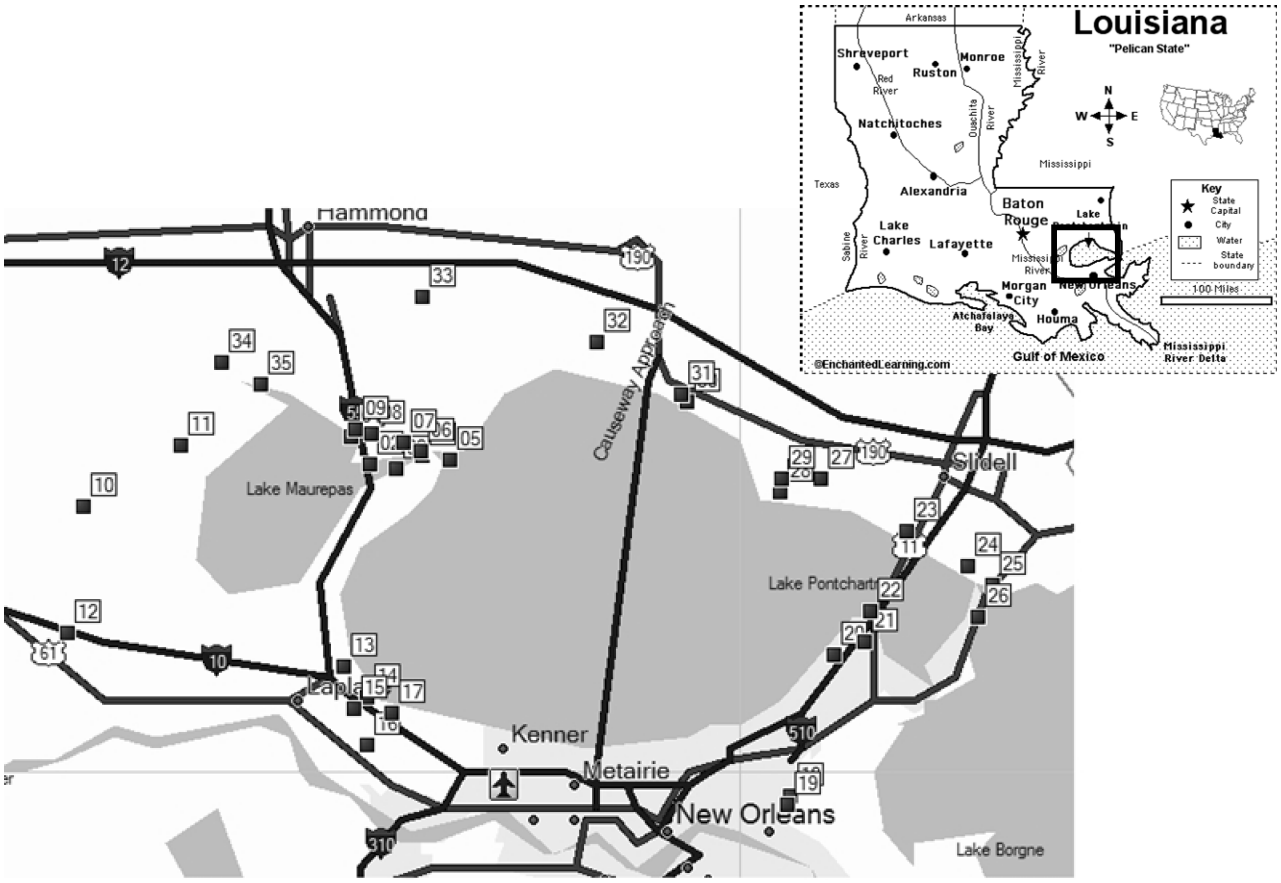

Fig. 1. Locations of wetland sampling sites surrounding Lake Pontchartrain and Lake Maurepas. The sampling locations were determined by GPS coordinates on site, and synthesized into map using MapSource (version 6.11.5, Garmin). Among the total 35 sampling sites, there are 11 swamp sites and 24 marsh sites.

brackish to salt water. The results were compared with the previously conducted THg and MeHg measurements in the lake bottom sediments of the Basin.

Material and methods

Sampling location

Louisiana's Pontchartrain drainage basin or watershed covers 12,173 km^2. The Basin encompasses land in 16 Louisiana parishes and 4 Mississippi counties. This vast ecological system includes lakes, rivers, bayous, forest, swamps and marshes. Lake Pontchartrain and Lake Maurepas are two of the major water bodies in the Basin. It is a habitat for numerous species of fish, birds, mammals, reptiles and plants. It is also the most densely populated portion of Louisiana with almost 1.5 million people residing immediately around Lake Pontchartrain.

Measurements of THg and MeHg from bottom sediment in Lake Pontchartrain and Lake Maurepas have previously been conducted and reported.[9] In this study, sediment samples (top 15 cm) were collected from 35 wetland sites surrounding Lake Pontchartrain and Lake Maurepas (Fig. 1). The 11 swamp sites are located to west side of Lake Maurepas and southwest bank of Lake Pontchartrain. The 24 marsh sites are spread around Lake Pontchartrain. Vegetation and hydrological conditions at each sampling site were recorded during the sampling. Water salinity level was estimated according to established vegetation, which reflects long-term salinity level at each sampling site (Table 1). The sediment samples were stored in glass containers at −20°C in laboratory before analysis.

Sample analysis

Total Hg (both organic and inorganic) was measured by cold vapor technique according to EPA method # 245.1, 245.5 and 7471A using a LabAnalyzer 254 (Hg Instruments GmbH Analytical Technologies, Karlsfeld, Germany). Mercury in the sample was first reduced to its elementary state by a reductant ($SnCl_2$). A stream of air produced by a built-in membrane pump striped the Hg from the sample into the optical cell. The Hg concentration in the cell was determined by measuring light absorption at a wavelength of 253.7 nm. The UV-light source was controlled by a reference beam and the UV-detector was

Table 1. Description of the sampling sites and THg and MeHg measurement results in the soils/sediments.

Site	*Wetland*	*Vegetation*	*Hydrology*	*Salinity*	*GPS coordinates*	*THg* $\mu g\ kg^{-1}$	*MeHg* $\mu g\ kg^{-1}$
1	Marsh	*Sagittaria lancifolia*	3 to 5	F	N30.3147, W90.4169	138.6	9.4
2	Marsh	*Sagittaria lancifolia*	Saturated	F/B	N30.2883, W90.3957	64.0	3.6
3	Marsh	*Sagittaria lancifolia*, *Panicum hemitomon*	Saturated	F/B	N30.2844, W90.3678	53.6	2.9
4	Marsh	*Sagittaria lancifolia*	3 to 5	F/B	N30.2967, W90.3397	59.1	4.3
5	Marsh	*Spartina patens*	3 to 5	B	N30.2921, W90.3101	47.6	0.3
6	Marsh	*Sagittaria lancifolia*, *Typha angustifolia*	5 to 8	F	N30.3002, W90.3413	57.6	0.8
7	Marsh	*Sagittaria lancifolia*	Saturated	F	N30.3087, W90.3600	49.3	0.2
8	Marsh	*Sagittaria lancifolia, Polygonum spp.*	3 to 5	F	N30.3171, W90.3940	41.1	0.2
9	Marsh	*Sagittaria lancifolia*, *Panicum hemitomon*	5 to 8	F	N30.3203, W90.4112	87.0	1.7
10	Swamp	Tupelo Gum (*Nyssa aquatica*)	10 to 13	F	N30.2495, W90.7029	74.8	1.1
11	Swamp	Bald Cypress (*Taxodium distichum*), Tupelo Gum (*Nyssa aquatica*)	Flooded	F	N30.3067, W90.5981	50.9	0.4
12	Swamp	Tupelo Gum (*Nyssa aquatica*) (very anaerobic sediment)	Flooded	F	N30.1323, W90.7197	68.5	11.4
13	Swamp	Palmetto (*Sabal minor*), Willow (*Salix nigra*), Tupelo Gum (*Nyssa aquatica*)	Flooded	F	N30.1016, W90.4235	110.7	1.3
14	Marsh	*Phragmites communis*	Saturated	F	N30.0726, W90.3989	71.6	0.1
15	Swamp	Willow (*Salix nigra*)	Flooded	F	N30.0618, W90.4129	99.8	1.1
16	Swamp	Mixed vegetations	7 to 10	F	N30.0286, W90.3987	49.2	1.4
17	Swamp	Bald Cypress (*Taxodium distichum*) (dead trees)	Saturated	B	N30.0576, W90.3720	119.7	2.0
18	Marsh	*Spartina patens, Spartina alterniflora*	Flooded	B/S	N29.9811, W89.9459	82.7	0.1
19	Marsh	*Spartina alterniflora*	Flooded	S	N29.9737, W89.9488	58.8	0.1
20	Marsh	*Spartina patens, Typha angustifolia*	5 to 8	F/B	N30.1115, W89.8983	71.4	0.7
21	Marsh	*Spartina alterniflora,* Three-cornered grass (*Scirpus olneyi*), Bulrush (*Scirpus maritimus*)	5 to 8	B/S	N30.1240, W89.8654	35.5	0.9
22	Marsh	*Spartina patens, Spartina alterniflora*	5 to 8	B/S	N30.1531, W89.8590	54.9	0.5
23	Marsh	*Spartina patens*	5 to 8	B	N30.2264, W89.8214	55.2	1.1
24	Marsh	*Spartina patens*	15	B	N30.1945, W89.7550	54.4	0.1
25	Marsh	*Spartina patens, Scirpus olneyi*	15	B	N30.1761, W89.7291	19.9	0.4
26	Marsh Marsh	Bulrush (*Scirpus maritimus*), *Spartina patens*	10 to 13	B	N30.1464, W89.7444	41.59	1.15
27	Marsh	Mixed vegetation	5 to 8	F	N30.2757, W89.9118	54.0	1.2
28	Marsh	*Spartina patens*, Bulrush (*Scirpus maritimus*)	5	B	N30.2625, W89.9563	57.8	1.4
29	Marsh	*Typha angustifolia* and others	5 to 8	F/B	N30.2746, W89.9550	40.8	0.3
30	Marsh	*Spartina patens, Spartina alterniflora,* Three-cornered grass (*Scirpus olneyi*)	5 to 10	B/S	N30.3464, W90.0563	8.7	0.5
31	Swamp	Bald Cyprus (*Taxodium distichum*), Tupelo Gum (*Nyssa aquatica*), Elephantsear (*Colocasia antiquorum)*	Saturated	F	N30.3532, W90.0622	90.9	1.2
32	Marsh	*Sagittaria lancifolia*, *Typha angustifolia*	10	F	N30.4015, W90.1519	51.4	0.2
33	Swamp	Bald Cypress (*Taxodium distichum*), Tupelo Gum (*Nyssa aquatica*), *Panicum hemitomon*	Flooded	F	N30.4434, W90.3390	64.1	0.7
34	Swamp	Bald Cypress (*Taxodium distichum*), Tupelo Gum (*Nyssa aquatica*)	30 to 60	F	N30.3829, W90.5547	174.4	1.1
35	Swamp	Tupelo Gum (*Nyssa aquatica*)	8 to 10	F	N30.3632, W90.5124	288.9	1.3

Hydrology: saturated–no standing water; flooded–standing water less than 3 cm; others–actual depth of standing water. Water depth is in cm. Salinity level: fresh (F) <1‰; fresh/brackish (F/B); brackish (B) 4–8‰; brackish/salt (B/S); salt (S) 8–12‰.

thermostatically stabilized in order to maintain an extremely stable baseline. Heating of the optical cell prevented sensitivity decrease associated with water vapor. Thus, a stable and accurate calibration was achieved using this method.

Sample preparation for MeHg analysis was performed based on the method of Alli et al.[10] and Cai et al.[11] Methylmercury analysis was performed using a gas chromatography-atomic fluorescence spectrometry (GC-AFS) system. The system consists of an integrated gas

Table 2. Statistics of THg and MeHg measurements in the wetland soils/sediments.

Wetland	*N*	*Analysis*	*Mean*	*Range*	*SD*	*Median*
Swamp	10	THg (μg kg^{-1})	112.3	49.2 to 288.9	72.4	95.3
		MeHg (μg kg^{-1})	1.1	0.4 to 2.0	0.4	1.2
		MeHg/THg	0.012	0.004 to 0.027	0.007	0.011
Marsh	24	THg (μg kg^{-1})	56.5	8.7 to 138.6	24.6	54.7
		MeHg (μg kg^{-1})	1.3	0.1 to 9.4	2.1	0.6
		MeHg/THg	0.022	0.001 to 0.072	0.022	0.016
Total	34	THg (μg kg^{-1})	72.9	8.7 to 288.9	50.2	57.7
		MeHg (μg kg^{-1})	1.3	0.1 to 9.4	1.7	1.0
		MeHg/THg	0.019	0.001 to 0.072	0.020	0.012

Note: One site (# 12) was treated as an outlier with 68.5 μg THg kg^{-1}, 11.4 μg MeHg kg^{-1}, and MeHg/THg ratio 0.166, and not included in the above statistics. This fresh water swamp forest sampling site showed much more reducing condition than the other sites (Table 1).

chromatography (HP 6890, Agilent Inc., USA) coupled to a Merlin Hg fluorescence detector system (Model 10.023, PS Analytical Ltd., UK) via a pyrolysis oven maintained at 810°C. A Megabore fused silica analytical column (15 m long, 0.53 mm i.d. J&W Scientific Inc., USA) coated with a 1.5 μm film thickness of DB-1 (J&W Scientific Inc., USA) was used in the analysis. The GC oven temperature was maintained at 50°C for 1.0 min, and programmed at 30°C/min to 140°C holding for 3.0 min, then finally programmed at 30°C/min to 250°C holding for 3.0 min. A split/splitless injector was used in the splitless mode and maintained at 200°C. The carrier gas flow was 4.0 mL/min of high-purity argon, and make-up gas flow was 120 mL/min of high purity argon. The column eluate was passed through a pyrolyzer (Thermolyne 21100 Tube Furnace) via a deactivated fused silica tubing into the Merlin Hg fluorescence detector system with sheath gas flow 200 mL/min of argon for Hg detection. A real time chromatographic control and data acquisition system (Hewlett-Packard ChemStation, Agilent Inc., USA) was interfaced with the GC and AFS system. Quantitative MeHg analysis in the samples was determined using a 5-point (between 0.2 and 10.0 ppb) calibration curve with linear regression forced to zero.[12]

A sub-sample of the fresh soil/sediment was dried at 105°C to a constant weight for determining moisture content. All data are presented based on dry weight.

Calculation and statistical analysis

Statistical analysis was conducted using SAS (V8 for Windows, SAS Institute Inc. Cary, NC, USA). The level of significance was chosen at $\alpha = 0.05$.

Results and discussion

Previously conducted measurements in bottom sediment of open water bodies or lakes in the Pontchartrain Basin showed that both THg and MeHg decreased with salinity increase, in the order of Lake Maurepas > Lake Pontchartrain > Lake Borgne/Chandeluer Sound. Average THg and MeHg contents in bottom sediments were 98.0 and 0.8 μg kg^{-1} in Lake Maurepas (n = 27), and 67.0 and 0.6 μg kg^{-1} in Lake Pontchartrain (n = 147), respectively.[9] Results of THg and MeHg measurements in soils/sediments of swamps and marshes surrounding Lake Maurepas and Lake Pontchartrain are summarized in Table 2. The swamp soils/sediments showed significantly higher THg content than the marsh soils/sediments ($P = 0.04$), and than the lake bottom sediments within the Basin. This could be due to the swamp sampling sites are mostly located in the western part of the Basin, which would be more likely affected by inland anthropogenic activities or due to higher clay content of swamp sediment, which retains Hg. Average THg content in the 34 wetland soils/sediments was 72.9 μg kg^{-1}, within the range of THg content in the bottom sediments of Lake Maurepas (98.0 μg kg^{-1}) and Lake Pontchartrain (67.0 μg kg^{-1}).

Results of the MeHg measurements in the wetland soils/sediments showed larger variation than the THg measurements, indicating complex nature of environmental factors controlling Hg methylation process (Table 2). Average MeHg content in the surrounding wetland soils/sediments (1.3 μg kg^{-1}) was higher than in the bottom sediments of the lakes (0.8 and 0.6 μg kg^{-1} for Lake Maurepas and Lake Pontchartrain, respectively). The average MeHg/THg ratio in the 34 wetland soils/sediments was 0.019, substantially higher than in the lake bottom sediments (<0.010). The results suggest that there was a greater potential for MeHg formation in wetland soils/sediments than lake bottom sediments. Reducing conditions with redox potential less than −200 mV[7,13] is required for the activity of sulfate-reducing bacteria that are responsible for Hg methylation.[4,14] Wetland soils/sediments is well known for its high organic matter (OM) content, especially elevated dissolved organic matter (DOC) concentration,[15] which can support developing anaerobic conditions required for Hg methylation process. The fresh water swamp site (#12) showed much more reducing conditions than the other sites, which was probably the major cause for the measured extraordinary high Hg methylation activity (11.4 μg MeHg kg^{-1} and MeHg/THg ratio 0.166). In addition, wetland vegetations

can prevent solar radiation to the wetland soils/sediments and DOC in water can also attenuate light, which would retard photo-destruction of MeHg.[16] Although wetlands are generally known to be sinks of heavy metals including Hg, they tend to be net source of MeHg, and wetland runoff may have an immediate impact on the MeHg level in adjacent water bodies, especially the near-shore area.[8]

In this study average MeHg contents in the marsh and swamp soils/sediment were not significantly different ($P = 0.66$). However, the ratio of MeHg/THg was considerably higher in the marsh soils/sediments (0.022) than in the swamp soils/sediments (0.012) because of the lower THg content in the marsh soils/sediments (Table 2). The current study did not provide information on sediment characteristics controlling Hg methylation activity, such as redox potential, OM and DOC in the soils/sediments and standing water, which deserves future investigation. However, salinity gradient among the sampling sites could partially explain the difference in Hg methylation activity between the swamp and marsh sites. Results of THg and MeHg are summarized according to salinity gradient, and are presented in Figure 2 and 3.

Only one swamp sampling site showed brackish water vegetations, and all the other swamp sites were classified as fresh water system (Fig. 2). The ratio of MeHg/THg in the swamp soils/sediments was generally 0.010 to 0.020. The marsh sampling sites covered a wide range of salinity condition, from fresh water to salt water (Fig. 3). The THg content in the wetland soils/sediments showed significant decrease with salinity increase ($P = 0.05$, n = 34), similar to reported values for lake bottom sediment results. Geographical location of the sampling site is probably the major reason for the THg distribution with less Hg input in more saline regions. The linear correlation between THg and MeHg was not statistically significant ($P = 0.12$, n = 34), indicating variations of Hg methylation activity in different environment. Similar to the swamp soils/sediments, the ratio of MeHg/THg in the marsh soils/sediments was also in a range of 0.010 to 0.020 at the fresh water and brackish water sites. The ratio of MeHg/THg at the fresh water/brackish water sites was up to 0.040, the highest of all salinity levels. Meanwhile the single salt water marsh sediment sampled for this study showed the lowest MeHg/THg ratio (0.002). This result agrees well with a reported finding that in anaerobic conditions low salinity (4‰) favored Hg methylation, but high salinity (25‰) inhibited it.[7] It is worthwhile to mention that salinity is only one of the numerous factors governing MeHg formation, and its impact on Hg cycle should be integrated with other key characteristics of the soils/sediments and waters.

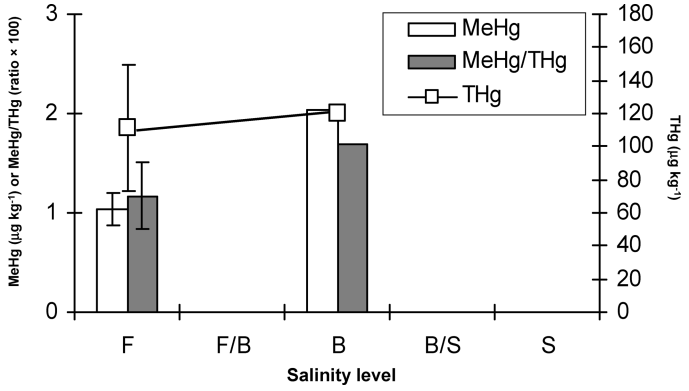

Fig. 2. Measurement of THg and MeHg in the swamp soils/sediments. Among the 10 sites included, 9 are in fresh water and 1 in brackish water. The fresh water swamp site (#12) not included in this figure was treated as an outlier with 68.5 μg THg kg^{-1}, 11.4 μg MeHg kg^{-1}, and MeHg/THg ratio 0.166, respectively. Data represent means of the measurement with standard deviation as error bars.

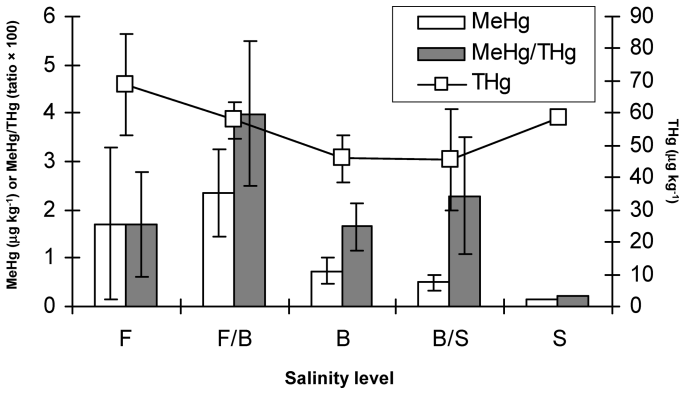

Fig. 3. Measurement of THg and MeHg in the marsh soils/sediments. Among the 24 sites, 8 are in fresh (F) water, 5 in fresh/brackish (F/B) water, 6 in brackish (B) water, 4 in brackish/salt (B/S) water, and 1 in salt (S) water. Data represent means of the measurements with standard deviation as error bars.

Methylmercury contamination has a global concern in which form Hg accumulates in fresh water fish, and consequently poses the greatest thread to wildlife and humans through food chain. Wetlands have been known as general sinks for most contaminants including Hg, but wetlands are often a source of MeHg due to its intrinsic reducing environment, as demonstrated in this study. There are evidences showing that water discharged from wetlands is generally enriched in MeHg, comparing to natural precipitation or runoff water from uplands.[17] Thus, the concentration of MeHg in water bodies will be significantly affected by the area of adjacent wetlands in the watershed or catchments.[18,19] Results of this study also show that the wetland sites surrounding the lakes may be a potential MeHg source to the aquatic organisms and contribute to the bioaccumulation of MeHg in the Basin wide food chain.

Acknowledgment

The research was supported by the Louisiana Water Resource Research Institute (Project Number: 2005LA-2B).

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