

# Source Identification of Florida Bay's Methylmercury Problem: Mainland Runoff Versus Atmospheric Deposition and *In situ* Production

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**Abstract** The first advisory to limit consumption of Florida Bay fish due to mercury was issued in 1995. Studies done by others in the late 1990s found elevated water column concentrations of both total Hg (THg) and methylmercury (MeHg) in creeks discharging from the Everglades, which had its own recognized mercury problem. To investigate the significance of allochthonous MeHg discharging from the upstream freshwater Everglades, we collected surface water and sediment along two transects

from 2000 to 2002. Concentrations of THg and MeHg, ranging from 0.36 ng THg/L to 5.98 ng THg/L and from <0.02 ng MeHg/L to 1.79 ng MeHg/L, were elevated in the mangrove transition zone when compared both to upstream canals and the open waters of Florida Bay. Sediment concentrations ranged from 5.8 ng THg/g to 145.6 ng THg/g and from 0.05 ng MeHg/g to 5.4 ng MeHg/g, with MeHg as a percentage of THg occasionally elevated in the open bay. Methylation assays indicated that sediments from Florida Bay have the potential to methylate Hg. Assessment of mass loading suggests that canals delivering stormwater from the northern Everglades are not as large a source as direct atmospheric deposition and *in situ* methylation, especially within the mangrove transition zone.

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## Introduction

The principal pathway for methylmercury (MeHg) exposure to humans is through the consumption of marine fish and fish products (Fitzgerald and Clarkson 1991; Sunderland 2007). This has added significance in Florida due to the combination of high mercury (Hg) levels in resident marine species and relatively high fish consumption rates (Moya et al. 2008). Florida's Department of Health (FDOH) has issued guidance advising the public to limit consumption of over 59 species of commercially and recreationally valued fish from coastal waters off Florida (FDOH 2009).

Fish consumption advisories are, of course, not unique to Florida. According to U.S. EPA (2007), 12 states have statewide advisories in their coastal waters due to mercury.

Yet, mercury levels appear to be particularly elevated in fish from Florida Bay. Surveys conducted in the late 1980s and early 1990s routinely found fish from northeastern Florida Bay to contain Hg concentrations exceeding 0.5 ppm and often 1.0 ppm (Adams and McMichael 2001; Strom and Graves 2001). More recent surveys continue to find mercury levels in fish from northeastern Florida Bay elevated relative to other parts of the bay (Evans and Crumley 2005), relative to other estuaries in south Florida (Adams et al. 2003) and relative to other estuaries along the Gulf of Mexico (Ache et al. 2000). Current fish consumption advisories are more restrictive for a number of species in the Florida Bay as compared to advisories for these same species inhabiting other coastal waters of Florida (FDOH 2009).

Early studies reported elevated water column concentrations of both total Hg (THg, all forms of Hg) and MeHg in canals and creeks that discharge from the Everglades into Florida Bay (Kannan et al. 1998; Lores et al. 1998). Additionally, Kang et al. (2000) documented a distinct spatial pattern in sediments with 2 to 6 times greater accumulation of THg in sediment cores downstream of one of the major flow paths into Florida Bay, Taylor Slough, relative to more open waters of the bay. They concluded that runoff was the dominant source of Hg to sediments near these sloughs. At this same time, methylation of mercury was thought to be inhibited in marine sediments that were highly sulfidic and overlain by highly saline waters (Barkay et al. 1997; Gilmour et al. 1998; Benoit et al. 1998; for review, see Langer et al. 2001), both of which were recognized problems in Florida Bay (Robblee et al. 1991; Carlson et al. 1994). It was therefore reasonable at that time to conclude that Florida Bay's mercury problem was simply an extension, through the discharge of allochthonous MeHg, of the well recognized mercury problem in the upstream freshwater Everglades.

This study was undertaken in 2000 to better understand the sources and distribution of MeHg in Florida Bay, especially in relation to discharges from the mainland watershed. This was and continues to be a fundamental concern because the South Florida Water Management District (SFWMD) and U.S. Army Corps of Engineers (USACE) are involved in massive collaborative restoration program in South Florida (Comprehensive Everglades Restoration Plan or CERP) that will alter the quantity, quality, timing, and distribution of water delivered to Florida Bay. Concerns have been raised that increased flows could exacerbate and further the expansion of the Hg problem from the freshwater Everglades (Kang et al. 2000; Strom and Graves 2001). Here, we present the results from the collection and analyses of surface water and sediments along two transects into northeast Florida Bay and at a reference site in the open bay. Flow data and estimates of atmospheric deposition to the watershed and directly to the

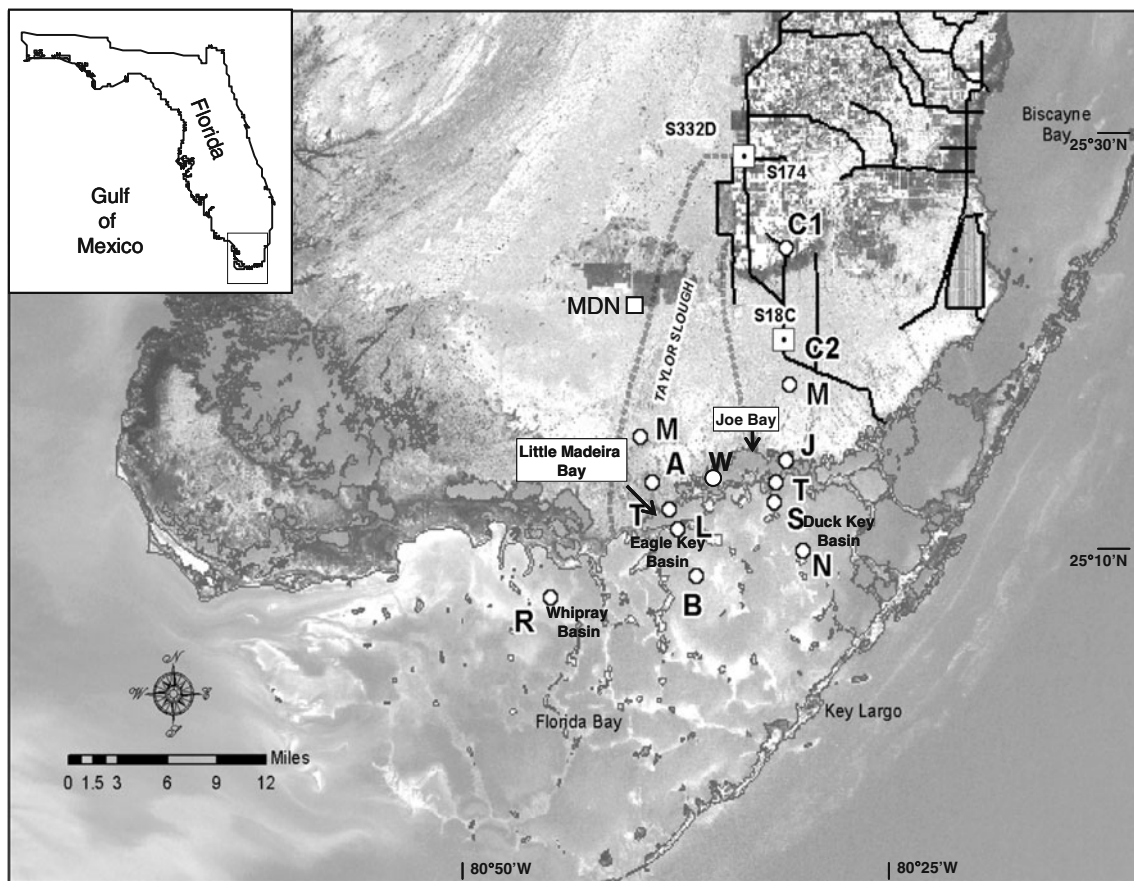
bay were used to quantify and prioritize the sources of MeHg and the processes that control its distribution in the bay. Because we found higher than expected levels of MeHg in sediments from the open bay, methylation assays were added to the study to assess *in situ* production.

## Study Area and Methods

Florida Bay is a large (2,256 km<sup>2</sup>) shallow (typical depths are 1 to 2 m) subtropical estuarine ecosystem, covered with fine-grained carbonate sediments, made up of basins separated by mud banks and mangrove islands (Fig. 1). It is bordered to the north by Everglades National Park (ENP) and to the south by the Florida Keys. Freshwater deliveries to the bay are regulated through water control structures with schedules developed by the SFWMD and USACE. Deliveries enter northeastern Florida Bay via Taylor Slough and the C-111 Canal. Taylor Slough has its source as discharge from the S-332D pump and S-174 gated spillway and direct rainfall (Fig. 1). Surface water in Taylor Slough and water that overtops the C-111 Canal downstream of the S-18C gated spillway flows south until the flow becomes channelized into 20 or more tidal creeks that flow into northeast Florida Bay. Wind and, to a lesser extent, tidal forcing of water through these creeks, combined with low ground elevations, allow for extensive inundation and mixing in the coastal wetlands and mangrove transition zone (Schaffranek et al. 2001). Freshwater from the Everglades drains also through the S-12 structures and flows into Shark River Slough and generally moves southwest towards Whitewater Bay and the Gulf of Mexico. After passing around Cape Sable, some of this water enters northwest Florida Bay and passes through the Florida Keys (Lee et al. 2002).

With its subtropical climate, extensive mangrove forests, occasional hypersalinity, shallow depths, which may allow for tighter pelagic–benthic coupling, carbonate sediments and its watershed devoid of industrial point sources, Florida Bay differs from other estuaries where extensive mercury studies have previously been carried out (e.g., Long Island Sound, Chesapeake Bay, San Francisco Bay).

From February 2000 through September 2002, surface water and sediment (and biota, results will be reported elsewhere) were collected along two transects into northeastern Florida Bay (Fig. 1). The first begins with two sampling locations in the C-111 Canal (sites C1 and C2 near the S-18C water control structure) and extends south through the freshwater marsh (site M), includes a site at the mouth of a creek flowing into northeastern Joe Bay (site J), at the mouth of another creek flowing into western Joe Bay (site W), out through Trout Creek (site T), Stump Pass (site S) and then the open bay near the Nest Keys (site N). The

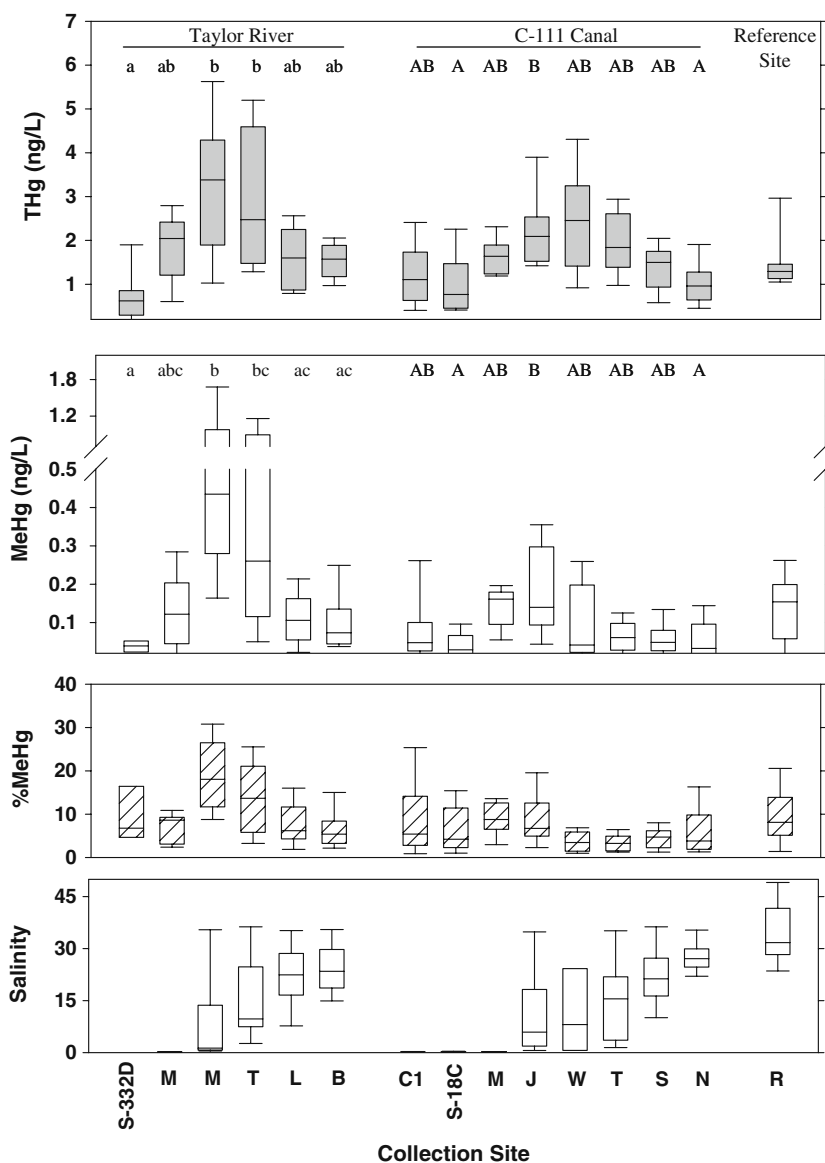


**Fig. 1** Map of Florida Bay and its watershed showing sampling stations along two transects and at a single reference site (see text for description of sampling sites). The Mercury Deposition Network (MDN) site is also shown

second transect follows the flow path of Taylor Slough, beginning in the freshwater marsh (site M), then at Argyle Henry Pond (site A) on Taylor River, the mouth of Taylor River (site T) as it discharges in Little Madeira Bay, the opening of the embayment (site L) and finally out in the open bay (B) of Eagle Key Basin (Fig. 1). An additional site in the open bay of Whipray Basin, located away from these discharges (Fig. 1), was sampled as a reference site (site R). This site occasional becomes hypersaline (Fig. 2); however, at times, this basin can receive discharge plumes from the Terrapin Bay area. Surface water was collected three times in 2000 and on a quarterly basis in 2001 and 2002, through to September (total  $n=10$ ). Because wind forcing is more dominant than the tidal signature in northeastern Florida Bay, samples were collected from mid-depth independent of tidal condition (i.e., ebb or flood). Water depths at sampling sites averaged  $222\pm 72$  cm (mean $\pm$ 1SD) in canals,  $18\pm 6$  cm in freshwater marshes and  $137\pm 46$  cm in sub-tidal areas. At the time of sample collection, temperature, conductivity/salinity, turbidity, and dissolved oxygen were measured at the bottom, mid-depth, and near surface of each location using a YSI 6920 multi-parameter water quality sonde (YSI Inc., Yellow Springs, OH).

Collection of water followed strict ultra-trace “clean hands–dirty hands” techniques (EPA Method 1669; EPA 821/R-95-027). At each location, duplicate water samples were collected for both filtered and unfiltered constituents. Unfiltered total mercury (THg) and methylmercury (MeHg) were collected separately in 250- or 500-mL bottles (re-usable Teflon bottles or single-use off-the-shelf glass bottles) using a peristaltic pump and Teflon sampling trains. Separate filtered samples (THgF and MeHgF) were filtered through new 0.45- $\mu$ m capsule filters (Meissner Filtration Products, Inc.) without filter pre-washing. These filters are used by SFWMD in its permit-mandated Hg-monitoring program after they had been found not to be a source of Hg contamination during a side-by-side comparison with another brand of filter (unpublished SFWMD report, May 1998). All re-usable equipment and bottles coming into contact with sample water were pre-cleaned (i.e., acid-washed, in a mercury-clean lab and transported to and from the sampling site in double resealable freezer bags). Sampling trains, bottles, and filters were triple rinsed with ambient water prior to each sample collection. Water samples were transported from the field on ice and immediately shipped overnight to the analytical laboratory.

**Fig. 2** Box plots showing the median (*line within box*), the 25th/75th percentile (outer boundaries of box) and the 10th/90th percentile (*whiskers*) concentrations of unfiltered total mercury (*THg*, *top panel*), methylmercury (*MeHg*), percent of THg as MeHg (*%MeHg*) and salinity (*lower panel*) of surface water samples collected quarterly from February 2000 through September 2002 (i.e., 10 sampling events unless site was inaccessible; S-332D data from SFWMD monitoring program, *n*=9). Sites with similar letter designations within a transect did not differ significantly



Sediment samples were collected from marshes, canals (often only thin sediment over rock), and sub-tidal areas on five occasions (roughly semi-annually in 2000 and 2001, and once in 2002). When dry-down prevented airboat access to marsh sites, efforts were made to collect samples at a later date when access was again possible. At each location, cores (top 4 cm) were collected in triplicate by driving a clean butyrate core tube into the sediment using a gloved hand. Tubes were then capped at the top to create a vacuum and withdrawn. Water above the sediment layer was carefully decanted and excess sediment, representing material deeper than 4 cm discarded. The three cores were then extruded into a labeled resealable polyethylene freezer bag and kept on ice for transport to the processing lab. Before and after each use, coring tube, caps, and all sampling utensils were rinsed a minimum of three times

with ambient water. Sediment samples were processed on the day of collection to minimize MeHg loss or formation by first removing large rocks and plant debris (e.g., roots, sticks, etc.) both living and dead using gloved hands. Composited sediments were then homogenized using a stainless steel blender, forming a single sediment sample for each location, and immediately frozen in a labeled amber glass jar. Samples were shipped frozen to the analytical lab for determination of THg, MeHg, and percent solids and concentrations reported on a dry weight basis).

From February 2000 through August 2000, analyses of THg and MeHg in surface water and MeHg in sediments were carried out by Frontier GeoSciences (FGS, Seattle, WA). THg determination in sediments collected prior to August 2000 was done by the Florida Department of Environmental Protection Chemistry Laboratory (Tallahas-



see, Florida). After August 2000, all analyses were carried out by CEBAM Analytical, Inc. (Seattle, WA). Water analyses followed EPA method 1631 (Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry; EPA-821-R-99-005) for THg determination and draft EPA method 1630 (Methylmercury in Water and Tissues by Distillation, Extraction, Aqueous Phase Ethylation, Purge and Trap, Isothermal GC Separation, Cold Vapor Atomic Fluorescence Spectrometry) for MeHg determination. THg in sediment was determined by subjecting samples to HNO<sub>3</sub> digestion at 95 °C for 4 h. The divalent inorganic mercury, Hg(II), in the digestate was then reduced using SnCl<sub>2</sub> and Hg<sup>0</sup> purged and trapped and quantified by Cold Vapor Atomic Fluorescence Spectrometry. MeHg in sediment was first leached using a 4 N HNO<sub>3</sub>–1 M CuSO<sub>4</sub> solution and then extracted with CH<sub>2</sub>Cl<sub>2</sub>. The CH<sub>2</sub>Cl<sub>2</sub> was then evaporated to back extract MeHg into distilled–deionized water (DDW). The back extract was then analyzed for MeHg using methods described above for surface water. The different contract laboratories achieved similar method detection limits (MDLs). MDLs achieved by CEBAM were 0.2 ng THg/L and 0.02 ng MeHg/L for water and 0.5 ng THg/g and 0.05 ng MeHg/g wet sediments.

#### Quality Assurance

An inter-laboratory comparative study using split samples was not done when switching contract laboratories. The contract laboratories did perform well; however, in intercomparison exercises analyzing total mercury and methylmercury in water (Niu and Tittle 2003). A comparison of concentrations reported by the three laboratories for different matrices collected over different times suggested no inter-laboratory differences for sediment but possible differences for water (Table 1). The observed differences between labs may, however, simply represent real environmental trends over time. Results of laboratory and field quality control (QC) check samples reported by the primary contract laboratory

(i.e., CEBAM) are presented in Table 2. Maintaining DDW systems without measureable inorganic Hg was a challenge for all three laboratories. This was an issue because DDW served as blank-water for all field QC check samples and there were several instances where laboratory contamination falsely indicated QC failures for THg (Table 2). However, QC check samples rarely contained measureable amounts of MeHg (Table 2), which is not a common laboratory contaminant. Given this and that all equipment were triple rinsed with ambient water prior to each sample collection, quality control was deemed satisfactory.

#### Methylation Rates in Intact Cores

In June 2001 and September 2002, duplicate cores were collected from each location using 30-cm, pre-cleaned butyrate, or polycarbonate tubes and incubated for methylation rate determination. Core tubes were driven into the sediment to the desired depth using a gloved hand. Upon reaching the depth of approximately 15 cm, the tube containing the sediment and overlying water was capped at the top with a rubber stopper, withdrawn from the sediments, and the bottom capped. Cores were transported to the field lab, while maintaining upright orientation, and injected with isotopes within 6 h of collection. Cores were injected with Hg(II) spiking solution (100 ng <sup>202</sup>Hg(NO<sub>3</sub>)<sub>2</sub>/mL) through injection ports (predrilled holes covered with silicone) spaced at 1-cm intervals using a hypodermic needle. It is important to note that the isotope solution was not pre-equilibrated with water from the sampling sites. Each port received 1 ml of spiking solution (to match ambient concentrations of bulk sediments). Following injections, ports were covered with electrical tape. Core tubes were then incubated for 24 h in the dark in a closed cooler placed in the shade outside of the field lab. Temperature in the coolers ranged from 29 °C to 36 °C, which reflected the ambient conditions over the course of the two incubation periods. Following incubation, overlying water was decanted from each tube and cores were extruded and cut to desired

**Table 1** Comparison of median (and maximum) concentrations reported by the different contract laboratories for different analyte–matrix combinations

Analyte–matrix combination	FGS <sup>a</sup>	FDEP <sup>b</sup>	CEBAM <sup>c</sup>
THg in filtered water (ng/L)	0.78 (2.65) <i>n</i> =48		1.50 (2.75) <i>n</i> =44
MeHg in filtered water (ng/L)	0.078 (0.86) <i>n</i> =48		0.050 (0.25) <i>n</i> =44
THg in sediment (ng/g dry)		18 (63) <i>n</i> =9	17 (145) <i>n</i> =41
MeHg in sediment (ng/g dry)	0.245 (2.33) <i>n</i> =9		0.225 (2.65) <i>n</i> =41

<sup>a</sup> Water collected during the 2nd and 3rd quarters of 2000; first set of sediments (bay only)

<sup>b</sup> First set of sediments (bay only)

<sup>c</sup> Water collected during the 2nd and 3rd quarters in 2001; sediments collected throughout the remaining study

**Table 2** Results of laboratory and field quality control check samples reported by the primary contract laboratory

QC	Matrix	Check sample	THg	MeHg	
Laboratory	Water	Mean recovery in matrix spikes	96% ( $n=82$ ) >75 and <113%	96% ( $n=43$ ) >75 and <113%	
		Mean relative percent difference (RPD) between matrix spikes	2.5% ( $n=29$ ) <11%		
		Mean RPD between laboratory duplicates	6.4% ( $n=12$ ) <20%	8.7% ( $n=25$ ) <25%	
		Mean recovery in certified reference material (CRM) and performance evaluation (PE) samples analyzed	96% ( $n=9$ ) >91% and <101%	98% ( $n=13$ ) >83 and <107%	
		Sediment	Mean recovery in matrix spike	99% ( $n=10$ ) >88% and <124%	98% ( $n=8$ ) >80% and 107%
			Mean RPD between matrix spike duplicates	0.8% ( $n=2$ ) <1%	1% ( $n=1$ pair)
			Mean RPD between laboratory duplicates	7.6% ( $n=7$ ) <14%	14.8% ( $n=7$ ) <42%
		Mean recovery in CRM analyzed along with sediments <sup>a</sup>	102% ( $n=4$ ) >99% and <106%	100% ( $n=6$ ) >93% and <107%	
Field	Water <sup>b</sup>	Proportion of Field QC check samples that exceeded the laboratory's practical quantitation limit <sup>c</sup>			
		Trip blanks ( $n=13$ )	33%	0%	
		Pre-cleaned equipment blanks ( $n=13$ )	69%	8%	
		Field-cleaned equipment blanks ( $n=12$ )	58%	8%	
		Field blanks ( $n=14$ )	50%	7%	
	Sediment	Mean RPD between field splits of homogenized sediments	43% ( $n=3$ pairs)	18% ( $n=1$ )	
		Mean RPD between field duplicates	68% ( $n=4$ )	32% ( $n=4$ )	

<sup>a</sup> CRMs included NIST2709, BCR580, IAEA142, and NRCC DORM-2; PE water samples: WS-68 and WS-98

<sup>b</sup> Because water samples (of filtered and unfiltered) were collected in duplicate (i.e., serial duplicates), field QC duplicates were unnecessary

<sup>c</sup> From February 2000 through August 2000, during which time FGS was the contract laboratory, THg exceeded the MDL in 56% of field QC samples (i.e., 9 of 16; 3 TB, 3 EB1, 2 EB2 and 1 FB); however, none of these exceeded the laboratory's PQL. During that time, MeHg never exceeded the Lab's MDL in any field QC check samples ( $n=16$ )

horizons. In June 2001, cores were cut into two sections representing the upper 4 cm horizon and 4–10 cm horizon. In September 2002, cores were cut into three sections representing the upper 1 cm, 1–4 cm and 4–10 cm horizons. Sections were placed in labeled, double resealable freezer bags, and frozen pending shipment to the U.S. Geological Survey (USGS) Laboratory in Middleton, Wisconsin. The net amount Me<sup>202</sup>Hg formed from the <sup>202</sup>Hg(II) addition was determined using methods described elsewhere (Hintelmann and Evans 1997). In brief, following extractions from the solid phase similar to that described above, and in the case of Me<sup>202</sup>Hg GC separation, isotopes were measured by continuous-flow cold vapor generation inductively coupled plasma mass spectrometry (ICP-MS). Bulk organic matter was also determined gravimetrically at the USGS lab as weight loss upon ignition (LOI).

USGS reported recoveries from CRM IAEA-405 averaged 90% for THg (range, 77% to 106%;  $n=8$ ) and 74% for MeHg (range, 57% to 86%;  $n=8$ ). The average coefficient of variation (CV) among eight sets of laboratory triplicate analyses was 17% for excess Me<sup>202</sup>Hg (range of 5% to 35%), 18% for excess T<sup>202</sup>Hg (range from 5% to 27%), and 3% for LOI (range from 1% to 6%). In 2001, subsamples of

five of the cores (all from the non-homogenized 4 cm to 10 cm horizon) were submitted blind to USGS for duplicate analyses as QC check samples. The relative percent differences (RPDs) between duplicate sediments averaged 54% for Me<sup>202</sup>Hg (4% to 138%), 55% for T<sup>202</sup>Hg (6% to 152%), and 9% for %LOI (4% to 18%), indicating incomplete homogenization.

#### Sources of Data for Estimating Mass Loads

Fluxes were calculated as the product of concentrations of Hg species measured in the discrete grab samples multiplied by the instantaneous flow measured using hydro-acoustic current meters or, if at a water control structure, algorithms based on hydraulic characteristics of the structure (i.e., SFWMD's FLOW program calculates water discharge based on upstream and downstream stage elevations, geometry of the spillway or pump revolutions per minute and size of pump and gear ratio between the engine and impeller). Daily mean discharges were obtained from databases maintained by SFWMD and USGS (Hittle et al. 2001; Levesque 2004) for locations and structures identified in Table 3.

**Table 3** Source of hydrometeorological data used to estimate mass loads

Parameter	Location or station	Source
Daily mean discharges at SFWMD Structures	S-332D	SFWMD
	S-175	SFWMD
	Flow out of C-111 Canal = estimated as S-18C minus S-197	SFWMD
Coastal discharges	Taylor River just downstream of Argyle Henry	USGS
	Mouth of Taylor River	USGS
	Joe Bay 2E (i.e., Northeastern Creek)	USGS
	Joe Bay 8 W (i.e., Western Creek)	USGS
	Trout Creek	USGS
Rainfall	Taylor Slough Basin = average rainfall at P-37, Craighead Pond and Taylor River	ENP
	C-111 Basin = average rainfall at S-18C and ENP Joe Bay (ENPJB)	SFWMD and ENP
	Little Madeira Bay = ENP Little Madeira Bay Station (ENPLM)	ENP
	Joe Bay = average of ENP Trout Creek Station (ENPTC) and ENP Joe Bay (ENPJB)	ENP
	Duck Key/Nest Key Basin = ENP Duck Key Station (ENPDC)	ENP
	Eagle Key Basin = average of ENP Butternut Key (ENPBK), and ENP Little Madeira Bay Station (ENPLM).	ENP
	Whipray Basin = ENP Whipray Basin Station (ENPWB)	ENP

It should be noted that vertical profiles in conductivity/salinity and temperature, taken at the time of the grab samples, always indicated a well-mixed water column. Gradients in concentration profiles across the transverse plane of the river or creek cannot be discounted; however, any error in load estimate would likely to be insignificant in the very narrow rivers and creeks. Concentrations of THg and MeHg in surface water in the canal at S-332D were collected (unfiltered only) as part of SFWMD's quarterly monitoring program under Project Code HGOS (which began in December 2000). Both THg and MeHg concentrations at S-18C were also supplemented with data from SFWMD's quarterly monitoring program. Daily concentrations of Hg species were estimated using linear interpolation between the quarterly sampling events.

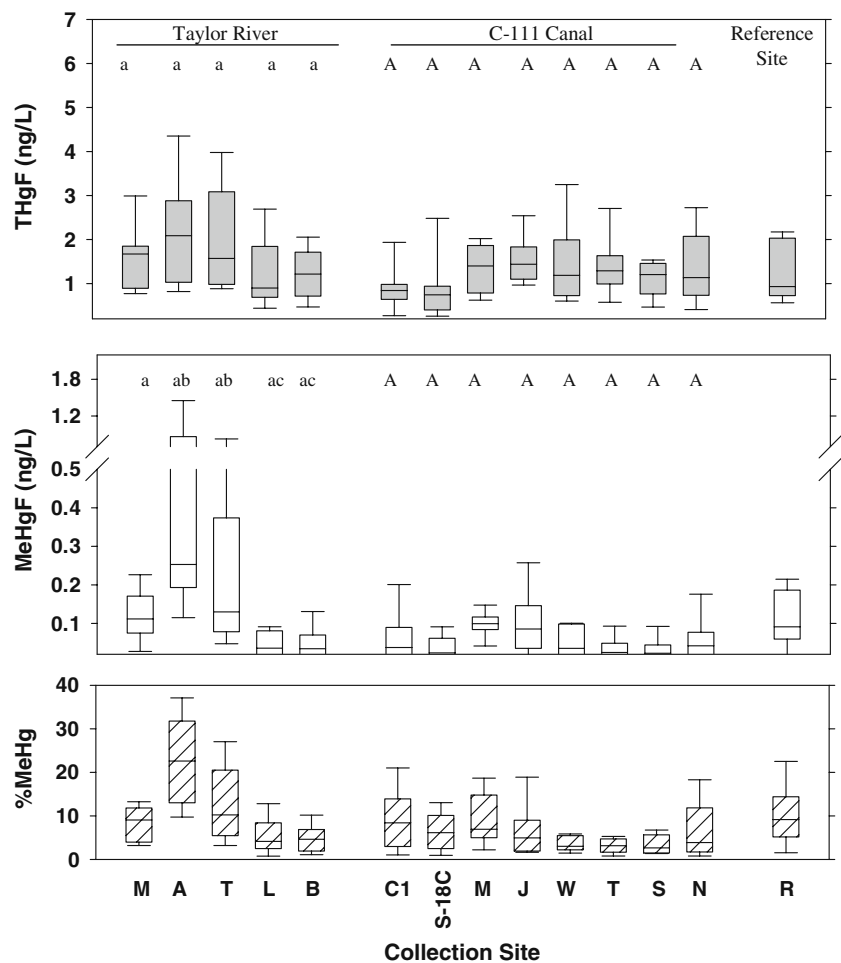
Atmospheric deposition of THg was based on bulk rainfall samples collected weekly under the protocols of the National Atmospheric Deposition Program's Mercury Deposition Network (MDN) at the Beard Research Center in ENP. Beginning in July 2001, samples collected at this station were also analyzed for MeHg (i.e., a monthly composite sample representing 10% of each of the weekly samples). Several of the values of THg concentrations and two of the MeHg concentrations in the bulk rainfall samples were considered suspect and replaced with interpolated values. Rainfall accumulations were estimated from daily rainfall accumulations collected at 10 gauges maintained by ENP (Table 3). Basin areas were derived from Nuttle (2002) and Cosby et al. (2003).

## Results and Discussion

This study was undertaken to better understand the sources and distribution of MeHg in Florida Bay. More specifically, the objective was to determine if Florida Bay's mercury problem was simply an extension, through the discharge of allochthonous MeHg, of the well recognized mercury problem in the upstream freshwater Everglades. Surface water sampling along the two transects did not, however, show unidirectional gradients of either THg or MeHg concentrations decreasing away from Everglades water control structures into Florida Bay (Figs. 2 and 3), as might have been expected. Instead, sampling revealed a pattern of elevated THg and MeHg concentrations within the mangrove transition zone of both Taylor Slough and C-111 Basins (Figs. 2 and 3). As the discussion below will explain, this pattern in water column concentrations together with concentrations observed in sediments and results from methylation assays indicate that direct atmospheric deposition of THg to the basins and *in situ* methylation might be important.

Concentrations of THg in unfiltered surface water collected along the Taylor Slough transect differed among sites (Kruskal–Wallis one-way ANOVA on ranks,  $H=25.1$ ,  $df=5$ ,  $p<0.001$ ), with the sites within the mangrove transition zone (Argyle Henry and Taylor River) having higher concentrations than other sites (Dunn's pairwise comparisons  $p<0.05$ , Fig. 2). More importantly, concentrations of MeHg differed also along this transect ( $H=28.8$ ,  $df=5$ ,  $p<0.001$ ) with Argyle Henry again having higher concentrations than

**Fig. 3** Box plots of filtered total mercury (THgF), filtered methylmercury (MeHgF) and % MeHg of surface water samples collected contemporaneously with unfiltered samples. Sites with similar letter designations within a transect did not differ significantly. Note, filtered samples were not collected at S-332D as part of the SFWMD monitoring program



all other sites except Taylor River and the freshwater marsh ( $p > 0.05$ , Fig. 2). Argyle Henry typically also had the highest percent of THg as MeHg (%MeHg) in both unfiltered and filtered samples, ranging between 9% and 31% (Figs. 2 and 3). Concentrations of MeHg at Taylor River were higher than at S-332D but similar to the other sites (Fig. 2). This pattern was replicated along the second transect. Concentrations of THg and MeHg differed along the C-111 Canal transect ( $H = 24.6$ ,  $df = 7$ ,  $p < 0.001$ , and  $H = 20.9$ ,  $df = 7$ ,  $p = 0.004$ , respectively), with concentrations of both significantly higher in the mangrove transition zone than concentrations either at the upstream control structure, S-18C, or the downstream open bay site (Nest Keys, Site N, Fig. 2). Pairwise comparisons between the reference site located in the open bay of Whipray Basin and other sites (Dunn's Method) revealed no significant difference in either average THg or MeHg concentrations ( $p > 0.05$ ). Moreover, the reference site had the third highest % MeHg after Argyle Henry and Taylor River (Fig. 2).

Dissolved species (i.e., operationally defined as passing through a 0.45- $\mu\text{m}$  filter) dominated over particle-bound species at most sites and exhibited much less variability (Figs. 2 and 3). On average, dissolved forms accounted for

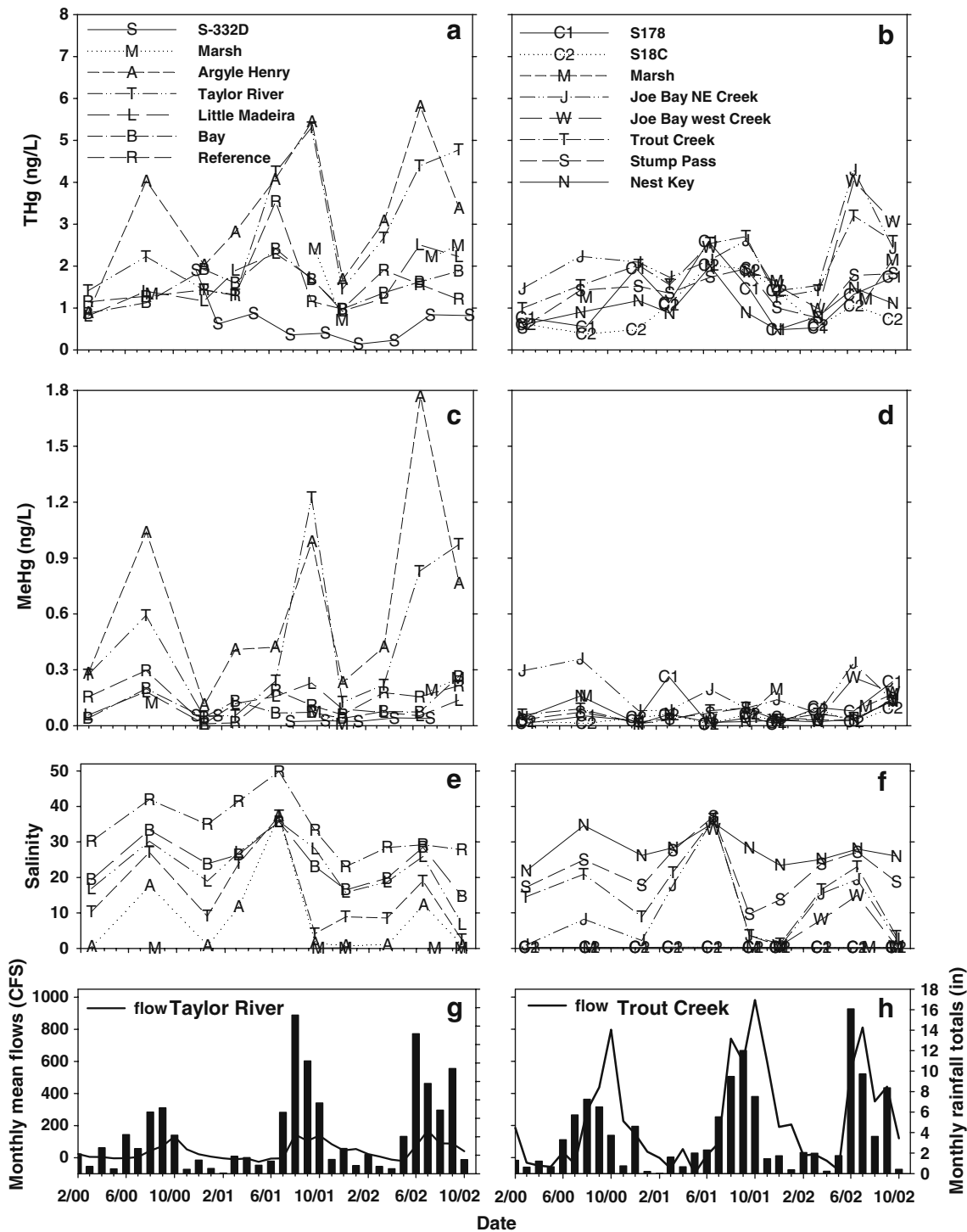
80% and 73% of the total THg and MeHg, respectively. Concentrations of dissolved THg did not differ along either the Taylor Slough transect (ANOVA,  $F = 1.97$ ,  $df = 4$ ,  $p = 0.12$ ) or the C-111 Canal transect ( $H = 13.45$ ,  $df = 7$ ,  $p = 0.06$ ). Although dissolved MeHg concentrations differed along the C-111 Canal transect ( $H = 14.76$ ,  $df = 7$ ,  $p = 0.04$ ), pairwise comparisons revealed no significant differences between individual sites ( $p > 0.05$ ). Dissolved MeHg concentrations also differed along the Taylor Slough transect ( $H = 26.2$ ,  $df = 4$ ,  $p \leq 0.001$ ). Pairwise comparisons along this transect revealed statistically significant between-site differences with Argyle Henry and Taylor River differing from the open bay sites but not the marsh site (Fig. 3; note, filtered samples were not collected at S-332 D).

While dissolved species generally dominated, much of the variation observed in the unfiltered concentrations resulted from the variability in the particulate or filterable fractions of THg and MeHg (i.e., calculated as the difference between average concentration of duplicate unfiltered and filtered samples). This was often observed in the mangrove transition zone where the Everglades runoff mixes with saline bay waters (Figs. 4 and 5) and may



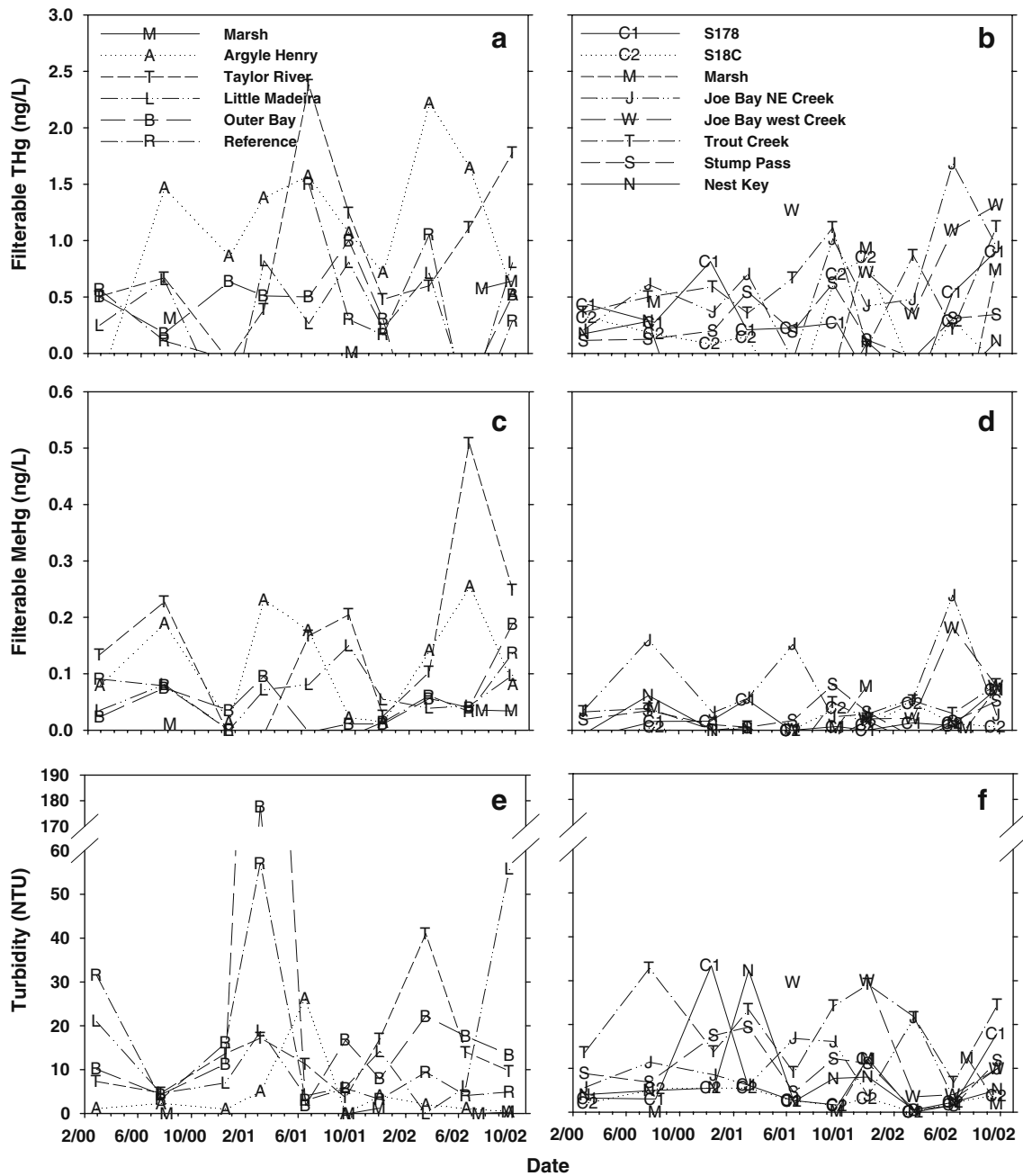
be attributable to a decrease in solubility, flocculation of negatively charged particles, or re-suspension. Surprisingly, filterable THg concentrations did not covary with water column turbidity (Pearson correlation coefficient,  $r=0.12$ ,

$p=0.19$ ,  $n=123$ ); neither did filterable MeHg ( $r=0.069$ ,  $p=0.44$ ,  $n=123$ ). The open bay site on the C-111 transect, Nest Key (Site N), had the lowest filterable fraction of both THg and MeHg (Fig. 5). Occasionally, average concentrations in



**Fig. 4** Time series of water column concentrations of THg, MeHg (values represent the mean of sequentially collected duplicate, unfiltered samples) and salinity (mid-depth). SFWMD monitoring at S-332D consisted of a single sample for THg and MeHg and no

salinity. Bottom panel shows flows and rainfall (shown as bars) measured at Taylor River (Taylor Slough Basin transect) and Trout Creek (C-111 Basin transect)



**Fig. 5** Time series of filterable THg and MeHg (i.e., average concentration of duplicate unfiltered minus average concentration of duplicate filtered samples)

paired filtered samples exceeded average concentrations in paired unfiltered samples at this site (these bottles were collected sequentially, not as split samples). As previously observed in unfiltered concentrations, filtered concentrations of THg and MeHg at the open bay reference site in Whipray Basin did not significantly differ from concentrations at other sites (Dunn's Method,  $p > 0.05$ ).

The concentrations reported here are within ranges reported by two previous short-term studies in Florida Bay by Kannan et al. (1998) and Lores et al. (1998). In

general, water column concentrations of THg were similar to results from other coastal areas that did not have obvious point sources, whereas MeHg concentrations tended to be higher (Benoit et al. 1998; Mason et al. 1999; Lacerda et al. 2001; Guentzel and Tsukamoto 2001). The patterns observed in water column concentrations of both dissolved and filterable forms across the transects were consistent also with spatial patterns observed in other estuaries (Benoit et al. 1998; Guentzel and Tsukamoto 2001), including a study done in another mangrove-dominated estuary where

THg and reactive Hg were enriched in the mangrove zone relative to open bay waters (Lacerda et al. 2001).

### Mass Load Estimates

Seasonal mass loads of THg and MeHg were approximated as the sum of instantaneous fluxes (in grams per wet or dry season) and are summarized in Tables 4 and 5. The results must be interpreted cautiously, however, due to the uncertainties introduced by: 1) estimating daily concentrations using linear interpolation between quarterly sampling events, 2) the reliance on measured deposition at a single MDN station and, 3) estimating MeHg deposition based on measurements of monthly composites from weekly rainfall samples. Despite these uncertainties, several points are evident from examining Tables 4 and 5.

### Total Mercury

Direct wet atmospheric deposition of THg to the Taylor Slough Basin was two orders of magnitude greater than inputs from the canal via the S-332D pump (Tables 4 and 5). Similarly, direct wet atmospheric loading of THg to the C-111 Basin was 66% higher than loads delivered by the C-111 Canal. This is in agreement with previous reports (Stober et al. 1996, 1998; Fink et al. 1999) that atmospheric deposition dominates over surface water inflows as the source of

inorganic Hg to the Everglades. It should be noted that, while the focus here is on wet-only deposition, dry deposition likely adds significantly (30–60% of wet deposited) to the overall atmospheric load (FDEP 2003). When inputs from S-332D pump were combined with direct atmospheric deposition, THg loads to Taylor Slough were greater than the loads to the C-111 Basin (i.e., when inputs from the canal were combined with direct atmospheric deposition, Tables 4 and 5). Comparison of the two basins suggests land-based runoff loads of THg at the mouth of Taylor River were similar to or slightly greater than loads in the two creeks discharging into Joe Bay (i.e., Northeast Creek and Western Creek combined, Table 4). Both Little Madeira and Joe Bay have inflows in addition to those monitored, and Western Creek likely includes flows originating in Taylor Slough. Direct atmospheric deposition of THg to these embayments was similar (Table 5) and was larger than loads in monitored rivers and creeks (Tables 4 and 5). Because flow was not measured at the boundary of Little Madeira Bay with outer bay, loads could not be estimated in surface flows to the open bay in Eagle Key Basin. Flows were measured, however, at Trout Creek where much of Joe Bay discharges to Trout Cove and the open bay. The large THg loads that passed through Trout Creek (Table 4) reflected high water flows and modest mercury concentrations. Nevertheless, the dominant THg loads to the open bay were from direct atmospheric deposition (Tables 4 and 5).

**Table 4** Surface water discharges and seasonal mass loads of total mercury (THg) and methylmercury (MeHg) at various locations by water-year (wet season is May–October, dry season is from November

through April of the following year). Sampling began in February 2000 (late into the 2000 dry season)

Season	Location	Surface Flows (M <sup>3</sup> xMillions)			THg (g)			MeHg (g)		
		2000	2001	2002	2000	2001	2002	2000	2001	2002
Wet	Canal at S-332D	112	109	62	N/A <sup>a</sup>	42	51	N/A <sup>a</sup>	2	2
	Argyle Henry Pond (Site A)	2	3	3	8	13	15	2	2	4
	Taylor River Mouth (Site T)	18	26	27	34	124	125	6	26	25
	Canal at S18C (Site C2)	142	127	114	112	145	93	2	5	8
	Joe Bay NE Creek (Site J)	N/A <sup>b</sup>	11	11	N/A <sup>b</sup>	26	39	N/A <sup>b</sup>	1	3
	Joe Bay Western Creek (Site W)	N/A <sup>b</sup>	23	21	N/A <sup>b</sup>	45	76	N/A <sup>b</sup>	1	5
Dry	Trout Creek (Site T)	104	175	145	182	427	433	6	14	11
	Canal at S-332D		10	69		N/A <sup>c</sup>	18		N/A <sup>c</sup>	2
	Argyle Henry Pond (Site A)		3	6		8	13		1	2
	Taylor River Mouth (Site T)		5	15		7	30		1	4
	Canal at S18C (Site C2)		14	60		15	52		0	2
	Joe Bay NE Creek (Site J)		N/A <sup>b</sup>	12		N/A <sup>b</sup>	18		N/A <sup>b</sup>	1
	Joe Bay West Creek (Site W)		N/A <sup>b</sup>	4		N/A <sup>b</sup>	6		N/A <sup>b</sup>	0
Trout Creek (Site T)		29	73		58	108		1	3	

<sup>a</sup> Not available because WQ sampling began at S-332D on 11/12/2000

<sup>b</sup> Not available because USGS began monitoring flows at Joe Bay Northeastern and Western creeks on 7/19/2001

<sup>c</sup> Not available because grabs were not collected during periods of no flow

**Table 5** Seasonal rainfall totals and loads from wet atmospheric deposition to select basins by water-year

Season	Basin (area in km <sup>2</sup> )	Rainfall (M <sup>3</sup> xMillions)			THg (g)			MeHg (g)		
		2000	2001	2002	2000	2001	2002	2000	2001	2002
Wet	Taylor Slough (214)	156	261	237	2,273	4,865	3,376	N/A	3	17
	Little Madeira (10)	6	11	8	73	214	105	N/A	<1	<1
	Open Bay—Eagle Key Basin (97)	66	99	74	937	1,734	985	N/A	1	4
	C-111 Marsh (130)	90	122	112	1,353	2,008	1,546	N/A	1	6
	Joe Bay (15)	8	12	12	113	211	164	N/A	<1	1
	Open Bay (181)	129	164	130	2,118	3,153	1,792	N/A	2	6
	Open bay—Whipray Basin (39)	26	42	28	377	699	359		<1	1
Dry	Taylor Slough (214)		36	50		259	472		N/A	4
	Little Madeira (10)		1	2		10	18		N/A	0
	Open Bay—Eagle Key Basin (97)		18	22		132	183		N/A	3
	C-111 Marsh (130)		17	21		94	164		N/A	3
	Joe Bay (15)		2	2		11	14		N/A	<1
	Open Bay (181)		34	37		200	294		N/A	5
	Open bay—Whipray Basin (39)		6	8		43	79		N/A	1

N/A not available, MeHg determination of MDN samples began on 7/10/2001

These findings are consistent with the conclusions of Kang et al. (2000) that runoff contributes as much as 80% of the total flux of THg to the sediments near the river sloughs but less than 20% to sediments in remote areas of Florida Bay (i.e., where the majority must be from atmospheric deposition). The present loading assessment suggests, however, that the source of the THg near river sloughs was direct atmospheric deposition to the basins immediately upstream of the embayments and not from canals routing runoff from further north. Recognition of boundary conditions of the watershed and the ratio of its land surface area to estuary water surface area must be considered when comparing loading assessments. The latter should provide an indication of the importance of indirect versus direct atmospheric deposition.

#### Methylmercury

As in other deposition assessments (for review, see Hall et al. 2005), only a small percentage of the atmospheric deposition of THg was in the methyl form (less than 1% in wet season and less than 2.6% in dry season, Table 5). MeHg loads in surface water runoff were also relatively small compared to THg loads in runoff (Tables 4 and 5). Unlike THg, MeHg loads in rivers and creeks to the two embayments were larger than atmospheric loads (Tables 4 and 5) and differed greatly between the two basins with larger loads coming down Taylor River (Table 4). Furthermore, unlike THg, MeHg loads through Trout Creek dominated loads from direct atmospheric deposition to northeastern Florida Bay (Tables 4 and 5). This is likely to

be the case also for Eagle Key Basin downstream of Little Madeira Bay.

#### Seasonality

Not surprisingly, inputs from both direct atmospheric deposition and from runoff were much reduced during the dry season (Tables 4 and 5). Estimates of atmospheric deposition may be underestimated, however, because they do not include dry deposition, which may be less seasonal. Concentrations of both THg and MeHg were highest during the summer wet season with heavy rainfall and peak discharges (Fig. 4). Quarterly sampling, however, prevented identification of any time lag from the onset of rains (i.e., direct deposition of “new” inorganic Hg) or peak discharges (i.e., indirect deposition and runoff) and concentration pulses in THg or MeHg. Routine mercury monitoring for more than 8 years at water control structures in the freshwater Everglades shows flows and, more importantly, concentrations of THg and MeHg peak during the late summer months of July–September, corresponding to the rainy season (Rumbold et al. 2007a).

#### Differences Between Terrestrial Watersheds

Taylor Slough Basin had smaller inputs from water management canals but larger inputs from atmospheric deposition, owing to larger surface area, than the C-111 Basin (Tables 4 and 5). More importantly, median and seasonal maximal concentrations of THg and MeHg were much greater in surface water within the mangrove

transitional zone of Taylor Slough than C-111 Basin (Figs. 2, 3, and 4). These greater concentrations when combined with slightly larger flows yielded larger loads of both THg and MeHg delivered to Little Madeira Bay by Taylor River than delivered to Joe Bay by the Northeastern and Western Creeks (Table 4). The differences in loads of allochthonous MeHg (produced in the upstream marshes and the mangrove transition zones) to the two embayments appear to be significant. This may explain the greater MeHg biomagnification observed in the biota of Little Madeira Bay compared to Joe Bay (Cantillo et al. 1997; Goodman et al. 1999; D. Evans, NOAA).

As Hg species enter the estuaries and travel through the numerous gradients that occur there (e.g., gradients in salinity, pH, nutrients, organic matter, etc.), they are subject to numerous competitive physico-chemical processes such as ion–ligand exchange, adsorption–desorption, colloid aggregation, and degradation of particulate organic matter (for reviews, see Horvat et al. 1999; Conaway et al. 2003 and references therein). Clearly, differences in quality of stormwater runoff, especially as it relates to carrier phases, such as dissolved organic matter (DOM), will cause Hg species to behave differently among estuaries and within the same estuary. Although it was beyond the scope of the present study, a more thorough investigation of these processes may explain the differences between these two watersheds and improve our understanding of Hg distribution in Florida Bay.

### Sediment

Concentrations in sediments collected from the bay and upstream marshes and canals ranged from 5.8 to 145.6 ng THg/g dry weight and from 0.05 to 5.4 ng MeHg/g dry weight (Figs. 6 and 7). The percent of THg that occurred as MeHg in sediments ranged from 0.1% to 88.9% (Figs. 6 and 7). Although the latter seems implausibly high, there was no evidence of analytical error. When data were pooled over time, both sediment-THg and sediment-MeHg differed significantly among sites on the Taylor Slough transect ( $H=19.8$ ,  $df=4$ ,  $p<0.001$  and  $H=16.8$ ,  $df=4$ ,  $p=0.002$ , respectively). Pairwise comparisons along this transect revealed that sediments from the open bay site (Site B in Eagle Key Basin) differed significantly from both Argyle Henry and Taylor River in THg and from the freshwater marsh and Taylor River sediments in MeHg; no other pairwise comparisons were significant. Both sediment-THg and sediment-MeHg differed also along the C-111 Canal transect ( $H=24.3$ ,  $df=6$ ,  $p<0.001$  and  $H=13.7$ ,  $df=6$ ,  $p=0.03$ , respectively). Here, sediments from Stump Pass differed in THg only from sediments from S-178 and Joe Bay creek ( $p<0.05$ ); no other pairwise comparisons in either THg or MeHg (suspected spurious MeHg value from

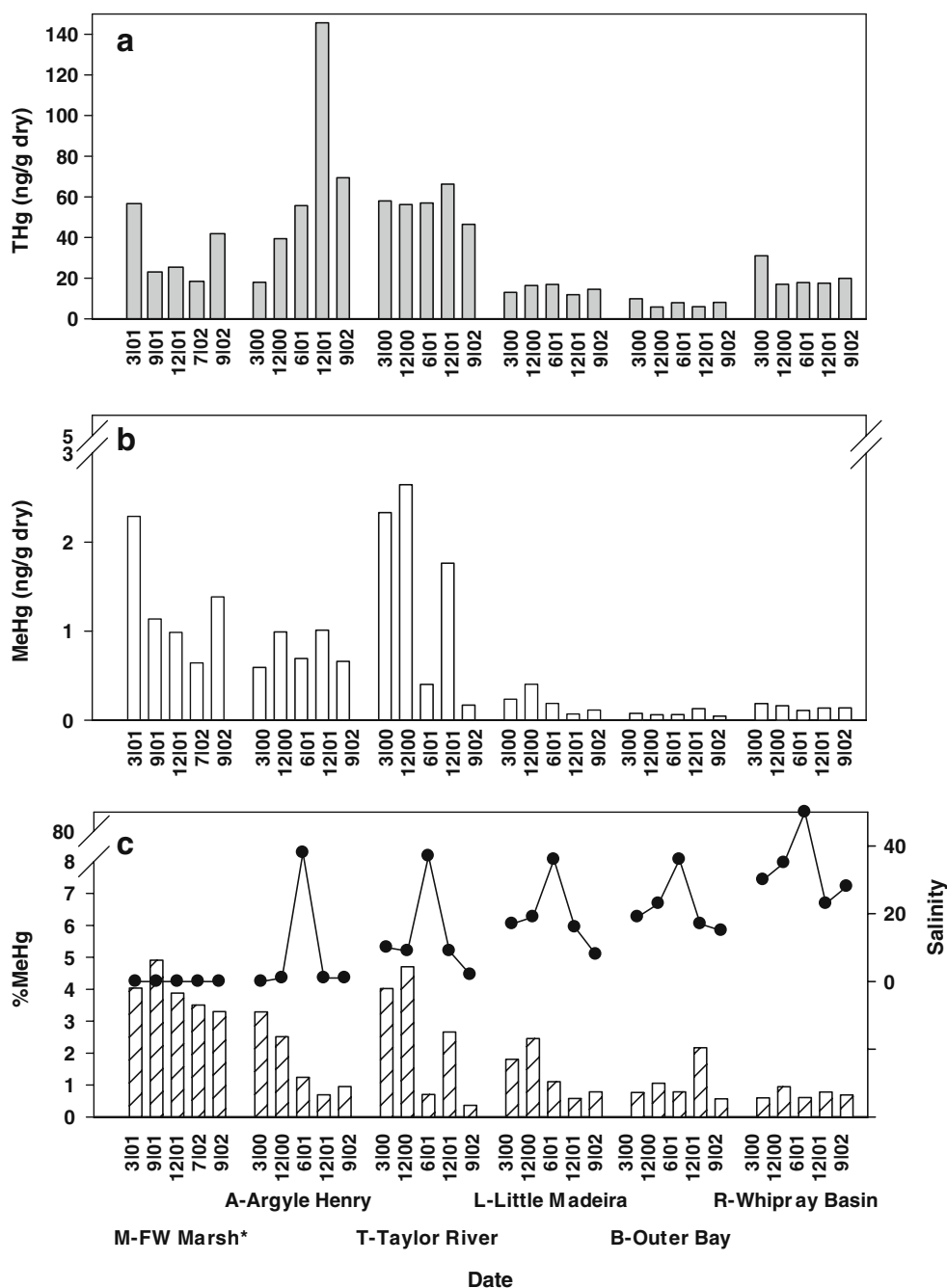
the marsh site was excluded) were statistically significant ( $p>0.05$ ). When compared to all other sites, sediments from the reference site differed only from the Taylor Slough marsh site in MeHg content (Dunn's post-hoc,  $p<0.05$ ); no other pairwise comparisons in MeHg or THg were significant. Notice also that sediments collected from the open bay near the Nest Keys (i.e., saline end-member of the C-111 transect), contained up to 1.8 ng MeHg/g, which constituted almost 8% of the THg present. Median %MeHg in sediments at this open bay site was 3.9%, which was similar to the percentages seen in sediments from the freshwater marsh sites. When pooled across sites, %MeHg in sediments was not correlated with the salinity of bottom water (Pearson Coefficient= $-0.16$ ,  $df=60$ ,  $p=0.214$ ; excluded the likely spurious %MeHg).

Comparisons of Hg levels in these sediments with levels in freshwater marl sediments from the lower Everglades, muck sediments from the northern Everglades, or sediments from other estuaries must be done cautiously because concentrations should first be normalized to total organic carbon or bulk density (note, bulk density was not measured in the present study, and %LOI was measured only in later cores). With this caveat in mind, sediment-THg concentrations appeared similar to values for other sediments collected from Florida Bay (Kannan et al. 1998; Kang et al. 2000) but lower than concentrations reported for sediments collected from the northern Everglades (Scheidt and Kalla 2007; Cohen et al. 2009) and from other estuaries, especially estuaries known to be contaminated by localized industrial activity (Bloom et al. 1999; Mason et al. 1999; Mason and Lawrence 1999; Hammerschmidt et al. 2004). Alternatively, sediment-MeHg concentrations were occasionally elevated compared to values previously reported for Florida Bay (Kannan et al. 1998) and other coastal marine sediments (Benoit et al. 1998; Bloom et al. 1999; Mason and Lawrence 1999; Hammerschmidt et al. 2004). The %MeHg in the bay sediments, especially from the Nest Keys, was high as compared to sediments from the freshwater Everglades collected as part of the present study and other studies (Gilmour et al. 1998; Rumbold et al. 2007b). The unusually high %MeHg was due, in part, to the low THg concentrations in these sediments compared to other areas, especially compared to contaminated sediment from highly urbanized estuaries or those near heavy industry (Bloom et al. 1999; Mason et al. 1999; Mason and Lawrence 1999; Hammerschmidt et al. 2004).

At the time the present study began, a limited number of studies, often involving estuaries or Fjords contaminated from urbanization or industrial activities (Gagnon et al. 1996; Bloom et al. 1999; Mason and Lawrence 1999) had documented MeHg in pore water and solid-phase sediment. Yet, as stated earlier, mercury methylation had been thought to be inhibited in marine sediments that were highly sulfidic



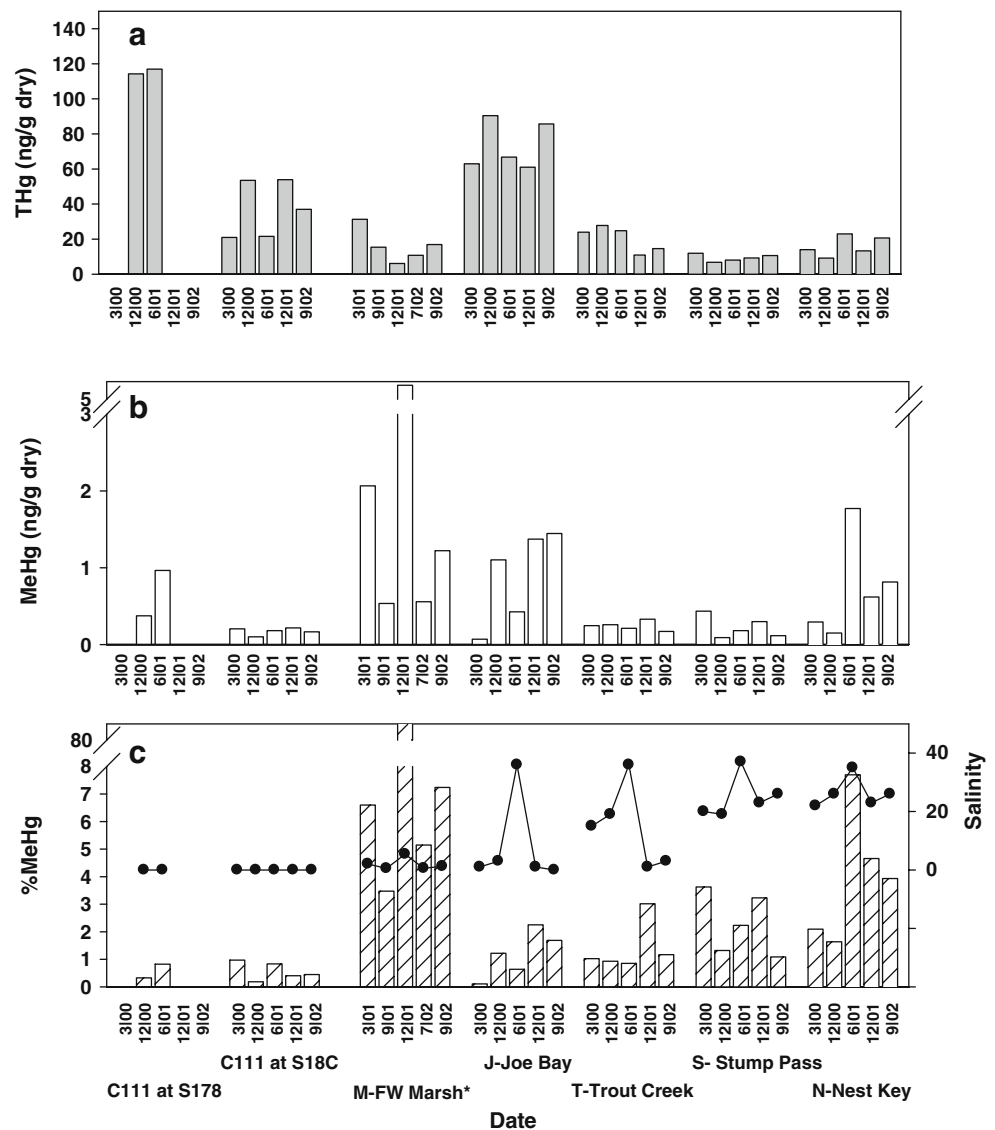
**Fig. 6** Bar graph showing concentrations of THg (*top panel*), MeHg (*middle panel*) and percent of THg as MeHg (*bottom panel*) in composite samples of sediment ( $n=3$ ) collected along Taylor Slough transect and at the reference site on a roughly semi-annual basis from late February 2000 through September 2002; note, freshwater marsh sites were sampled on an irregularly basis depending on airboat access. Salinity measured just above bottom is also shown (*bottom panel*)



and overlain by highly saline waters (Gilmour and Henry 1991; Barkay et al. 1997; Benoit et al. 1998; for review, see Langer et al. 2001). Benoit et al. (1999) provide evidence that charged mercury-sulfide complexes can dominate under highly sulfidic conditions, thereby reducing the bioavailability of Hg(II) and inhibiting methylation. Factors controlling Hg methylation have been separated into two general groups according to whether they affect the bioavailability of Hg(II) or affect the activity of the methylating bacteria (for review, see Heyes et al. 2006). However, some factors, such as sulfur, can control methylation through both processes.

Although sulfide was not determined in the sediments from the present study, sediments of much of Florida Bay are known to be highly sulfidic. Carlson et al. (1994) reported sulfide often >1 mM but as high as 13 mM in sediment pore water of Florida Bay. They concluded that these values resulted from a combination of rapid microbial sulfate reduction and the low Fe content and limited capacity of carbonate sediments to precipitate sulfide. Florida Bay is also known to frequently become hypersaline. Both hypersalinity and high sulfides have been implicated in seagrass die-offs over large areas of Florida Bay (Robblee et al. 1991; Carlson et al. 1994). Yet, as

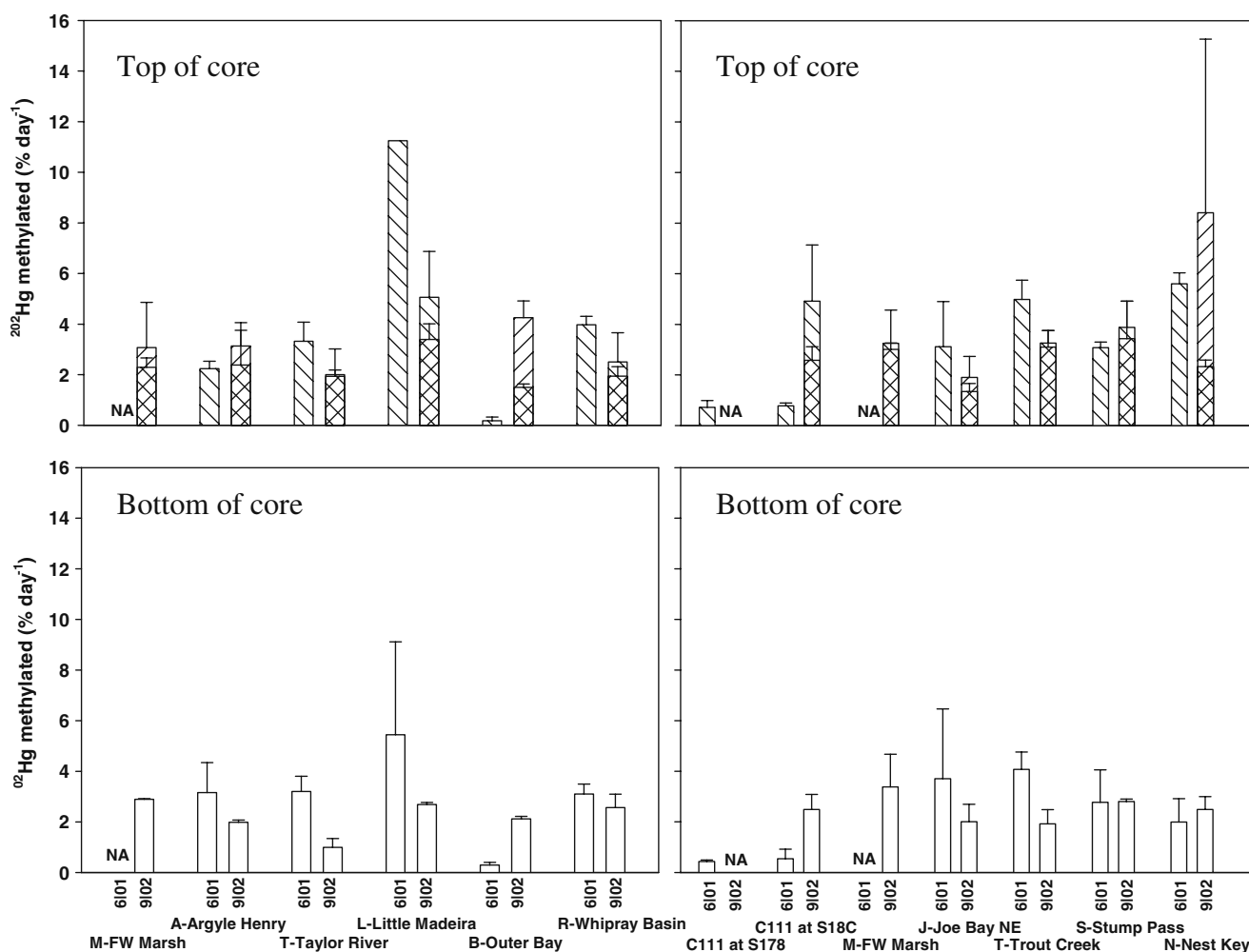
**Fig. 7** Bar graph showing concentrations of THg (*top panel*), MeHg (*middle panel*) and percent of THg as MeHg (*bottom panel*) in sediment collected along C-111 Canal transect; freshwater marsh sites were sampled on an irregularly basis depending on airboat access. Salinity measured just above bottom is also shown (*bottom panel*)



shown here, the percentage of the THg as MeHg in bay sediments was similar to the percentages seen in sediments from the freshwater marsh sites. This was unexpected based on information available at that time.

Because MeHg concentrations and %MeHg were unexpectedly high in bay sediments, methylation assays were added to the study. The median rate of  $^{202}\text{Hg}$  methylation was  $2.7\%$  day $^{-1}$  in the 0- to 4-cm horizon of cores taken in June 2001, with rates ranging as high as  $11.2\%$  day $^{-1}$ . The maximum  $^{202}\text{Hg}$  methylation rate occurred in a core taken at the outlet of Little Madeira Bay, Site L. Regrettably, there was a problem during the analysis of the top horizon from the duplicate core, and there was no confirmation of this unusually high conversion rate. The lower horizon (4 to 10 cm) from both cores exhibited variable but still relatively high rates of conversion ( $1.8\%$  day $^{-1}$  to  $9.1\%$  day $^{-1}$ ). More importantly,  $^{202}\text{Hg}$  methylation rates in cores from the open bay both from the Nest Keys and Whipray Basin were as high or higher

(with good agreement between duplicate cores) than rates in cores from freshwater canal sites (note, marshes were inaccessible in June 2001, Fig. 8). Median rates of  $^{202}\text{Hg}$  methylation was only slightly lower (at  $2.3\%$  day $^{-1}$ ) in lower horizons (4 to 10 cm) of cores taken in June 2001. In September 2002, incubated cores were cut into three sections with a median rate of  $^{202}\text{Hg}$  methylation of  $2.8\%$  day $^{-1}$  in the top 1-cm horizon,  $2.7\%$  day $^{-1}$  in 1- to 4-cm horizon and  $2.3\%$  day $^{-1}$  in the lower 4- to 10-cm horizon. The highest conversion during the September 2002 sampling event occurred in sediments from the Nest Keys ( $15\%$  day $^{-1}$ ); however, this time, the duplicate core exhibited a much lower methylation rate ( $2\%$  day $^{-1}$ ). Nonetheless, cores from several other sites, including Little Madeira Bay and Whipray Basin, again exhibited conversion rates as high or higher than cores from the freshwater marshes, with good agreement between duplicates (Fig. 8).  $^{202}\text{Hg}$  methylation rates in the present study were similar to or higher than values reported in



**Fig. 8** Percent  $^{202}\text{Hg}$  methylation during a 24-h incubation of cores collected along transects (bar and whisker represent average and upper range of duplicate cores). Cores were cut into two sections in 2001: 0

to 4 cm (\\ \\ top panel) and >4 to 10 cm (bottom panel) and three sections in 2002: 0 to 1 cm (/ / fill pattern), >1 to 4 cm (\\ \\ fill pattern) and >4 to 10 cm (bottom panel)

incubation studies of other estuarine sediments (Hammerschmidt and Fitzgerald 2004). Although the methylation assays provide evidence that sediments from Florida Bay have the potential to methylate Hg, caution must be exercised when interpreting these results. First, there are a number of factors, such as incubation duration, pre-equilibration, and spike concentration that can influence measurement of true methylation potential (for review, see Heyes et al. 2006). Furthermore, because demethylation was not measured in the present study, we cannot gauge the importance of net *in situ* methylation as a source of MeHg to biota.

Since initiation of the present study, a growing number of studies have reported mercury methylation in marine sediments, even where sulfides are in the millimolar range (King et al. 2000; Langer et al. 2001; for reviews, see Sunderland et al. 2006). Several non-exclusive explanations have been offered for *in situ* methylation in high sulfide environments including: 1) vertical migration of the oxycline and redox transition zone

induced by tidal pumping or by daytime peaks in photosynthesis (Langer et al. 2001), 2) wind induced or current driven mixing of the sediments (Sunderland et al. 2004) or via bioturbation (Hammerschmidt et al. 2004), 3) dissolved Fe scavenging of sulfides (Hammerschmidt and Fitzgerald 2004), and 4) creation of oxidized microenvironments by roots (Marvin-DiPasquale et al. 2003).

Many of these same processes, individually or in combination, could explain the methylation rates observed in the present study. For instance, Langer et al. (2001) observed methylation in sulfidic sediments (>28 mM) and posited that it occurred in the upper zone of the oxycline in surface sediments, particularly after inundation of the salt marsh with oxygenated water during flood stage. Net MeHg production would be enhanced under these conditions. Later, water with high MeHg concentrations would drain from the salt marsh as the tide ebbed. An analogous scenario was offered as an explanation for unprecedented MeHg concentrations drain-

ing from a (sulfate contaminated) freshwater stormwater treatment area where frequent use of large pumps inadvertently mimicked tidal pumping and caused water levels to fluctuate greatly, which in turn likely resulted in vertical migration of the redox transition zone (Rumbold and Fink 2006). Further evidence that tidal pumping can be significant in estuarine systems is provided by the data of Lacerda et al. (2001), who showed surface and pore water rich in THg and reactive Hg draining from mangrove transition zone of Sepetiba Bay, Brazil during the ebb tide. Such a scenario would easily explain the high MeHg and high THg in the mangrove transition zone of Florida Bay, which owing to its gentle slope, has extensive areas of coastal wetlands that can be inundated during wind-driven surges, seasonal sea-level fluctuations and the relatively minor tidal variations that occur in northeastern Florida Bay.

Langer et al. (2001) raise the possibility that the oxycline might also be affected during daytime peaks in photosynthesis by benthic algae producing near super-saturated oxygen concentrations at the sediment surface. This could be important also in Florida Bay because benthic microalgae are a major component of autotrophic biomass and have been shown to have high photosynthetic rates (Cornwell et al. 2003; Armitage et al. 2006); sea grasses and other submerged aquatic vegetation are also likely to play a significant role in the oxycline and biogeochemistry.

Others (Gagnon et al. 1996; Hammerschmidt and Fitzgerald 2004) have noted the importance of Fe in scavenging sulfides allowing Hg methylation by reduction of sulfide inhibition. Florida Bay sediments are generally low in Fe, and this process may not be as important here. Chambers et al. (2001) reports relatively high levels of reduced sulfur compounds but generally low levels of Fe in bulk sediments. Chambers et al. (2001) did conclude, however, that Fe availability, for sulfide sequestration, was higher in northeastern Florida Bay.

Sunderland et al. (2004) suggests that physical mixing of the active sediment layer may introduce more bioavailable inorganic mercury to deeper sediments and enhance also the transfer of sulfate and carbon potentially stimulating the methylating activity of sulfate-reducing bacteria (SRB). This highlights the second group of factors controlling methylation—those affecting the activity of the methylating bacteria (Heyes et al. 2006), including sulfate and metabolizable organic matter (Cossa et al. 1988; Guentzel and Tsukamoto 2001; Hammerschmidt and Fitzgerald 2004; Lambertsson and Nilsson 2006). The fine carbonate mud of Florida Bay can be readily resuspended by wind mixing in the shallow open waters. This might also explain why there was little difference in  $^{202}\text{Hg}$  methylation between upper and lower sediment horizons.

In the present study, organic matter content, estimated by loss on ignition (LOI), ranged from 4% to 84% (median

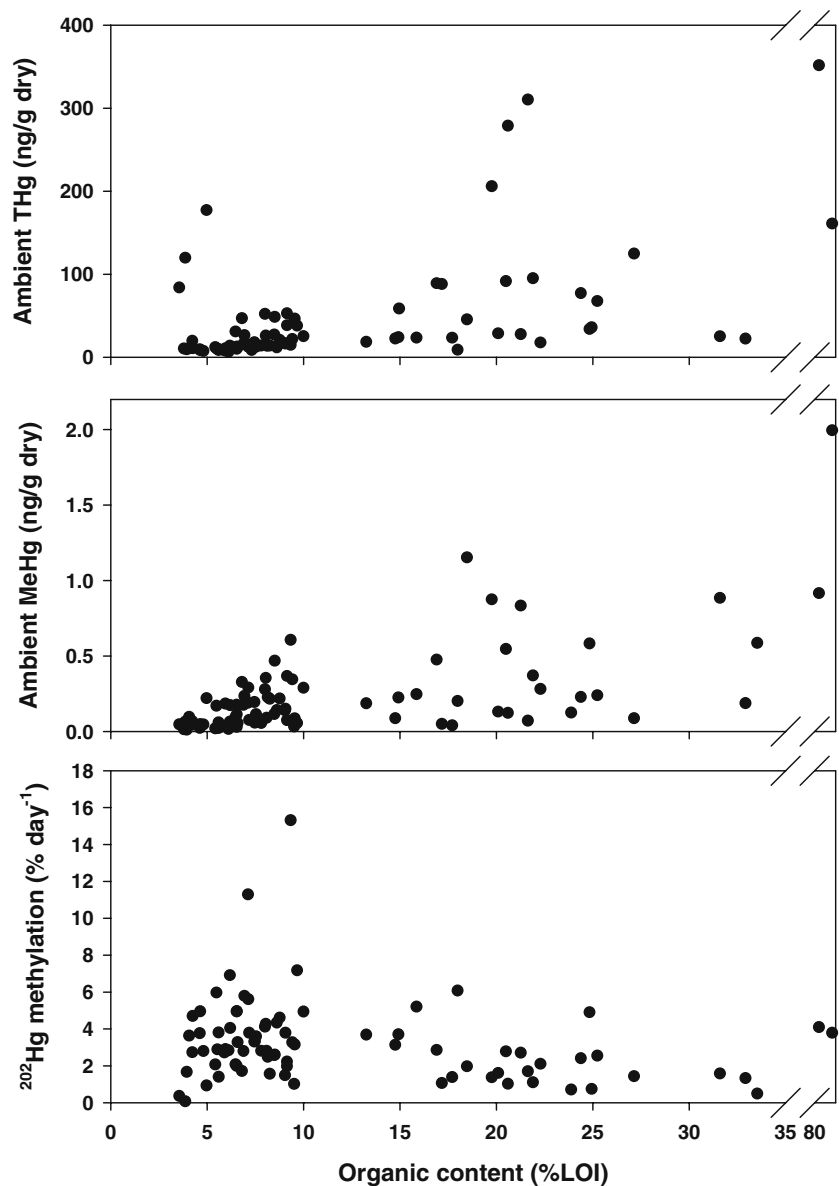
was 9%). The LOI was generally highest at Argyle Henry pond, Site A, and lowest in the open bay of Eagle Key Basin (i.e., saline end-member of Taylor Slough Transect, Site B). We found a positive correlation between organic matter and ambient THg concentration ( $r=0.57$ ,  $p<0.001$ ,  $n=74$ ) and ambient MeHg concentration ( $r=0.69$ ,  $p<0.001$ ,  $n=74$ ). This is in concurrence with the results of Mason and Lawrence (1999) who reported that THg and MeHg covaried with sediment organic matter. Alternatively, we found  $^{202}\text{Hg}$  methylation only correlated poorly with organic matter ( $r=-0.12$ ,  $p=0.28$ ; top horizons only, Fig. 9). Visual inspection of the data in Fig. 9 hints at a possible non-linear relationship between  $^{202}\text{Hg}$  methylation and organic matter, with an inhibitory effect in sediments with both low and high organic content (i.e., %LOI). Similar to the results reported here, Hammerschmidt and Fitzgerald (2004) also observed a reduction of methylation in sediments with relatively higher organic matter. Interestingly, in addition to stimulating the bacteria, several investigators (Le Roux et al. 2001; Hammerschmidt et al. 2004; Sunderland et al. 2006) suggest that the quantity and quality of DOM in pore waters may also influence the bioavailability of Hg(II) for methylation.

In addition to the remaining uncertainties regarding methylation potential, we lack also quantitative information on the amount of autochthonous MeHg entering the food web, which is the defining question. MeHg in sediments may enter the food web through efflux from sediment pore waters (both diffusive and advective), by movement of benthic organisms into the water column, or by direct grazing on surface sediments and benthic organisms. Rates of these processes are affected by many factors (Mason et al. 2006). Consequently, the importance of *in situ* MeHg production in Florida Bay will remain unclear until benthic flux measurements and benthic-pelagic trophic transfer studies are undertaken.

#### Management Relevant Conclusions

This study provides evidence that mercury methylation can occur in sediments of Florida Bay. Therefore, we need a more complete understanding of the fluxes and bioavailability of inorganic mercury as a factor, among many, that control *in situ* methylation rates. Concerns have been raised regarding restoration efforts inadvertently worsening existing mercury problems in other estuaries (Mason and Lawrence 1999; Hammerschmidt and Fitzgerald 2004). If, as it appears, THg and MeHg loads in canals delivering water from the Everglades and northern urban areas are not as significant as atmospheric loading and *in situ* methylation, then increasing freshwater flows in canals to Florida Bay in an effort to hydrologically restore the Everglades and Florida Bay ecosystems is unlikely to have a significant

**Fig. 9** Relationship between organic content (%LOI) of sediments from top of cores (i.e., 0- to 4-cm collected in 2001 and 0- to 1-cm and 1- to 4-cm horizons collected in 2002) and ambient THg (*top panel*), ambient MeHg (*middle panel*) and percent  $^{202}\text{Hg}$  methylation in incubated cores (*bottom panel*)



impact on inputs. Based on current data, we cannot meaningfully predict, however, whether restoration-induced changes (including increased deliveries of allochthonous DOM) will enhance or diminish methylation rates or subsequent biomagnification. At a minimum, continued monitoring of mercury levels in bay fish seems prudent. Because Hg levels are already high, small increases could result in even more restrictive fish consumption advisories (i.e., upgrading “limited consumption” to “no consumption”). Thus, monitoring Hg biomagnification in gamefish may not alert us to changes in time to take corrective action. Moreover, simply monitoring Hg levels in fish would not provide the information to discriminate which influential factor was altered and led to increased methylation or biomagnification. Information needed to make sound management actions

would require detailed process level monitoring. One area that is clearly deserving of more investigation is the underlying differences in sediment or water quality that results in lower exports from the C-111 Basin.

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