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Legacy and Fate of Mercury and Methylmercury in the Florida Everglades

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Mass inventories of total Hg (THg) and methylmercury (MeHg) and mass budgets of Hg newly deposited during the 2005 dry and wet seasons were constructed for the Everglades. As a sink for Hg, the Everglades has accumulated 914, 1138, 4931, and 7602 kg of legacy THg in its 4 management units, namely Water Conservation Area (WCA) 1, 2, 3, and the Everglades National Park (ENP), respectively, with most Hg being stored in soil. The current annual Hg inputs account only for 1−2% of the legacy Hg. Mercury transport across management units during a season amounts to 1% or less of Hg storage, except for WCA 2 where inflow inputs can contribute 4% of total MeHg storage. Mass budget suggests distinct spatiality for cycling of seasonally deposited Hg, with significantly lower THg fluxes entering water and floc in ENP than in the WCAs. Floc in WCAs can retain a considerable fraction (around 16%) of MeHg produced from the newly deposited Hg during the wet season. This work is important for evaluating the magnitude of legacy Hg contamination and for predicting the fate of new Hg in the Everglades, and provides a methodological example for large-scale studies on Hg cycling in wetlands.

Introduction

The Florida Everglades is a subtropical freshwater wetland ecosystem, spreading over 7000 km^2 and including four management units, namely the Arthur R. Marshall Loxahatchee National Wildlife Refuge (LNWR, also known as the Water Conservation Area 1, WCA 1), WCA 2, WCA 3, and the Everglades National Park (ENP) from north to south (Figure S1 of the Supporting Information, SI). The accumulation of elevated levels of mercury (Hg) in fishes and wildlife is a particular concern for this ecosystem (*1*). Efforts have been made to investigate source (*2*), transport (*3, 4*), transformation (reduction/oxidation and in particular methylation/demethylation) (*5*−*8*), and bioaccumulation of Hg in fish and wildlife (*9, 10*) in the Everglades. These efforts significantly advanced the understanding of individual processes of Hg transformation and transport in the Everglades.

An extensive system of canals, levees, and water control structures has interrupted slow overland sheetflow of water in the Everglades marsh and provided a conduit for pollutant transport across management units (*11*). In such a large ecosystem with subareas connected by water flows, investigations on the distribution and cycling of Hg at the ecosystem level are important. Examples of such investigations include constructing ecosystem-scale mass inventories for Hg present in the Everglades and calculating mass budgets for Hg newly input into the system. Mass inventories (including inputs, outputs, and storage) of total Hg (THg) and methylmercury (MeHg) would not only provide complete information on Hg mass distribution and transport, but also reveal the relative importance of each input and output pathway in Hg cycling. The benefit of establishment of mass budgets for newly deposited Hg would include identification of ecosystem components (e.g., surface water, soil, periphyton, and fish) where Hg will be retained. Such information obtained through constructing Hg mass inventories and mass budgets would be particularly useful for understanding the scope and magnitude of legacy Hg contamination and for predicting the fate of new Hg.

A few studies have surveyed the scope of Hg contamination in the Everglades (*11*−*14*), but more ecosystem-level information is needed to better understand Hg cycling in this complex ecosystem. Our previous study has constructed a mass budget for the seasonally deposited Hg at the scale of the entire Everglades, but management unit-level mass budgets remain lacking. The understanding of Hg cycling in each management unit will provide critical information for assessing the effects of Everglades restoration activity on mercury biogeochemistry in this ecosystem. In addition, a complete mass inventory of legacy Hg in the Everglades is currently not available.

On the basis of previous work, the objectives of this study were set to (1) evaluate the magnitude of legacy Hg contamination in the Everglades by constructing Hg mass inventories and (2) predict the fate of new Hg in each management unit by calculating a mass budget for newly deposited Hg during a season. By taking advantage of the probability-based sampling design and ecosystem-wide sampling of R-EMAP and combining R-EMAP with other data sets, we constructed mass inventories, including inputs, transport, outputs, and storage in ecosystem components, for THg and MeHg in each management unit of the Everglades. We further established management unit-level mass budgets for THg deposited during the 2005 dry and wet seasons and for MeHg produced from the seasonally deposited THg.

Methods

After collecting data from multiple data sets, including R-EMAP (*11*), ACME (*15*), MDN (*16*), and DBHYDRO (*17*) with R-EMAP being the primary data source, the following calculations were made: (1) Mass inventory of legacy THg

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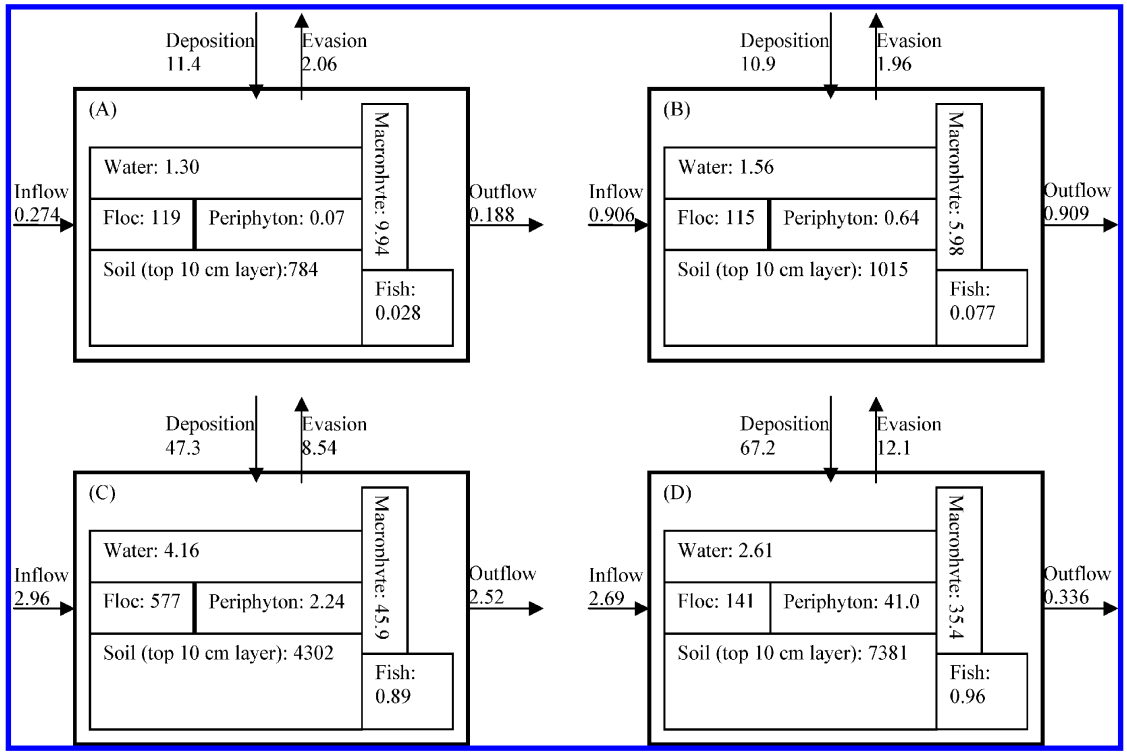

FIGURE 1. Mass inventory of THg in (A) WCA 1, (B) WCA 2, (C) WCA 3, and (D) ENP. Numbers (kg) accompanying arrows depict THg inputs and outputs during the 2005 wet season (May through November). Numbers (kg) inside each ecosystem component illustrate instantaneous THg mass stored in that component at the time of sampling (November 2005). Mosquitofish was abbreviated to fish in the figure.

and MeHg for each management unit, including inputs (atmospheric deposition, wet and dry, and water inflow), outputs (evasion into the atmosphere and water outflow), and storage in each ecosystem component (surface water, soil, flocculent detrital material (floc), periphyton, macrophyte, and mosquitofish); (2) Mass budget of newly deposited Hg during the 2005 dry and wet seasons for each management unit; (3) Uncertainty analysis associated with the calculations of mass inventory of legacy Hg and mass budget of new Hg; and (4) Fluxes of new Hg in each management unit (THg or MeHg mass divided by area of that management unit per season) for investigating the spatiality in Hg cycling. Detailed procedures for these calculations, including data source for each calculation (Tables S1−S8), are provided in SI and references therein (*12, 18−20*). Information about each data set, including sampling design, sampling protocols, analytical procedures, and original data, can be found in the literature related to that data set (*11, 12, 15, 21*).

Results and Discussion

Mass Inventory of THg. Our mass inventory (Figure 1) suggests that atmospheric deposition is the primary source of THg inputs to the Everglades, while THg inputs through water inflows account for about 2−8% of total THg inputs, which agrees with previous studies (*12, 13, 22, 23*). This is true for the entire Everglades ecosystem as well as for localized areas on the scale of the management units. Even for the WCAs, which are adjacent to the Everglades Agricultural Area (EAA) and receive runoff from EAA, THg inputs through water inflows are relatively small (e.g., 7.6% for WCA 2 and 5.9% for WCA 3, respectively), in comparison to atmospheric inputs. The mercury amount in the water outflow of WCA1 and WCA2 did not match the mercury amount in the water inflow to WCA2 and WCA3, respectively. This is because the water inputs to WCA 2 and WCA 3 are not coming solely from WCA 1 and WCA 2, respectively. It seems that THg transport between different management units indeed occurs, albeit in a minor amount. For instance, about 3 kg of THg entered into WCA 3 and ENP through water inflows during the 2005 wet season.

The THg mass inventory suggested that THg loss from the Everglades ecosystem is less than THg inputs. Mercury removal via water outflows accounts for a small proportion of THg inputs (1.6, 7.7, 5.0, and 0.5% for WCA 1, WCA 2, WCA 3, and ENP, respectively). Evasion releases a considerable amount of THg out of the ecosystem, but still a small fraction of THg inputs (around 17.5%). These results indicate that the Everglades ecosystem is a sink for THg, where most of the THg inputs (either from atmospheric deposition or from water inflows) will be accumulated. Our calculation of THg mass storage in ecosystem components indicates that, as a result of accumulation of THg inputs, significant amount of THg has been stored in the system, in particular in soil and floc. The Everglades soil (top 10 cm layer) stored from 784 kg of THg for WCA 1 to 7381 kg of THg for ENP, while the THg masses in floc can be up to 10−15% of soil THg amounts. The mass storage of THg in periphyton, macrophyte, and mosquitofish was small (1% or less). THg storage in soil greatly exceeded THg mass in any other component as well as any THg input or output. The mass storage of THg in ecosystem components we estimated agrees well with previous studies (*12, 14*), except for mosquitofish for which greater THg mass was obtained in this study, probably due to higher fish biomass used here.

Table S9 of the SI summarizes the total input, output, and legacy of THg and MeHg in the four management units. Compared to THg masses stored in the ecosystem (914 kg for WCA 1, 1138 kg for WCA 2, 4931 kg for WCA 3, and 7602 kg for ENP), the amount of annual THg inputs (including deposition and water inflow) to the Everglades is relatively small, amounting to 1−2% of the legacy THg. This disparity underscores the importance of understanding the cycling of legacy Hg when investigating Hg contamination in the Everglades.

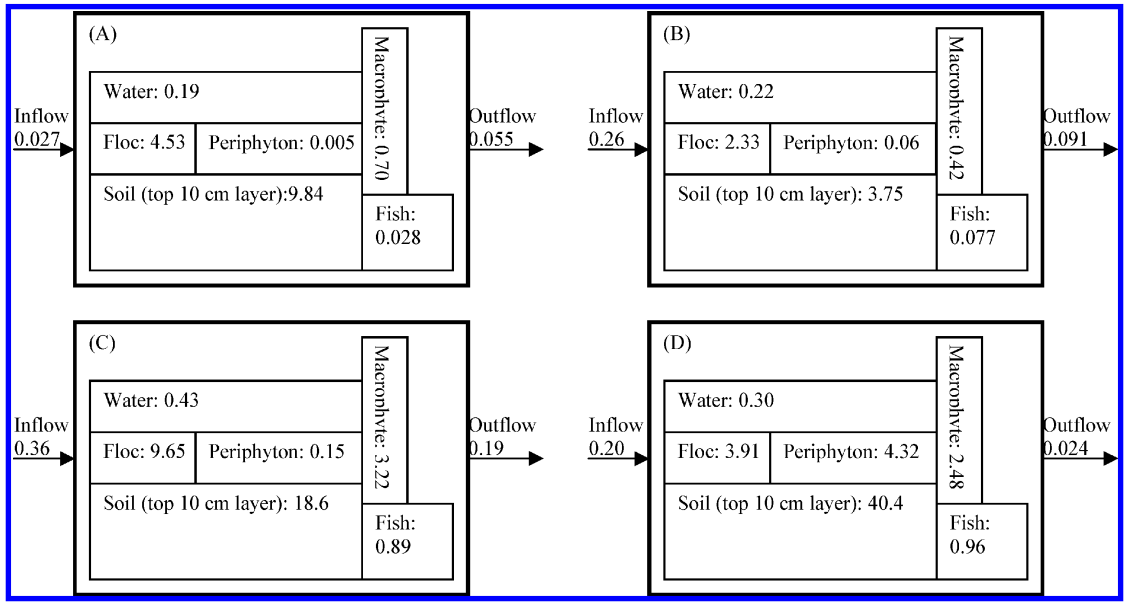

FIGURE 2. Mass inventory of MeHg in (A) WCA 1, (B) WCA 2, (C) WCA 3, and (D) ENP. Numbers (kg) accompanying arrows depict MeHg inputs and outputs during the 2005 wet season (May through November). Numbers (kg) inside each ecosystem component illustrate instantaneous MeHg mass stored in that component at the time of sampling (November 2005). Mosquitofish was abbreviated to fish in the figure.

Mass Inventory of MeHg. The calculation of MeHg masses showed that, similar to THg, soil and floc are major places where MeHg is stored (Figure 2). Floc plays an important role in MeHg cycling. Except for ENP, a significant amount of MeHg is stored in floc, with 4.53, 2.33, and 9.65 kg calculated for WCAs 1, 2, and 3, respectively. These values are about 50% of the MeHg mass in soil for each corresponding management unit, indicating the importance of floc in MeHg cycling. For ENP, 3.91 kg of MeHg is entrapped in the floc layer, which was less than 10% of MeHg mass in soil. Although the absolute amount of MeHg in floc in ENP is of a comparable order of magnitude in comparison to the WCAs, the fraction of floc MeHg against total MeHg storage is much lower for ENP. One possible reason for this distinction could be the difference in floc thickness. It was observed that floc in ENP (with a median of 0.83 cm) is significantly thinner than in the WCAs (medians ranging from 2.83 to 4.17 cm). Periphyton generally stored a minor amount of MeHg (0.005 to 0.15 kg or less than 2% of soil MeHg for WCAs), except for ENP where 4.32 kg (or more than 10% of soil MeHg) was stored. Macrophyte stored about 5% of total MeHg in each management unit. The mass storage of MeHg estimated here was comparable with a previous study (*12*), although floc MeHg mass appeared higher in this work. It should be noted that there is a great deal of variability in MeHg mass estimation for different years (with varying water and floc depth and periphyton biomass), which makes comparing different studies difficult in some cases.

Both inputs through water inflow and outputs through water outflow of MeHg are rather small (1% or less), compared to MeHg masses that are being stored in the ecosystem (Table S1 of the SI). One exception to this is MeHg inputs through inflows to WCA 2, which is about 4% of total MeHg storage in WCA 2. This increased MeHg input through water inflow could be related to the fact that WCA 2 receives runoff from EAA. Nonetheless, in situ production is generally the predominant source of MeHg that is cycled in the Everglades. After being produced in soil, floc, and periphyton, MeHg can be transported into the water column and/or transferred into the food web. The cycling of MeHg (from production to bioaccumulation) inside the ecosystem determines the magnitude of MeHg bioaccumulation in fish.

Fate of Seasonally Deposited THg. The results of mass budget estimation for newly deposited THg for the wet season are illustrated in Figure 3, while the mass budget of MeHg produced from the new Hg in Figure S4 of the SI. The results for the dry season are illustrated in Figures S2 (THg) and S3 (MeHg) of the SI. For all four management units, most seasonally deposited THg (around 80%) is retained in soil (Figures 3 and S2 of the SI), agreeing well with the mass inventory results, which showed that soil is the largest sink for THg storage, and with previous studies (*12, 24*). After deposition, THg redistribution into other ecosystem components was relatively small, except for floc in the wet season, which retained considerable fractions of newly deposited Hg, particularly in WCAs (ranging from 3.0 to 6.5%). This could be due to greater floc thickness and thus mass in the wet season, with wet season floc mass being an order of magnitude higher than dry season (Tables S5−S8 of the SI). Evasion and soil loss are two important pathways of Hg removal from the system, with 10 and 8% of seasonally deposited THg being removed through these two pathways, respectively. Compared to evasion and soil loss, water outflow is a minor pathway of THg removal for all four management units, accounting for only a very small proportion of THg deposited (0.004−0.24%).

Fate of MeHg Produced from Seasonally Deposited Hg. In contrast to THg, which exhibits a relatively uniform pattern among management units in terms of distribution within ecosystem components, the distribution of MeHg is rather complicated. Soil is still the largest sink, with about 80−90% of MeHg produced from newly deposited Hg being retained in soil in the dry season (Figure S3 of the SI). However, in the wet season, the fractions of MeHg retained by soil decreased significantly, in particular in WCAs, where the fractions ranged from 48% for WCA 2 to 68% for WCA 1 (Figure S4 of the SI). Corresponding with these decreases in soil MeHg fractions, the fractions of MeHg retained by floc increased to 16% in the wet season from about 0.1−4% in the dry season. Macrophyte represented another important sink for MeHg, in particular during the wet season (with about 10% of MeHg produced from seasonally deposited Hg being taken up by macrophyte). The greater proportion of MeHg found in floc and macrophytes during the wet season

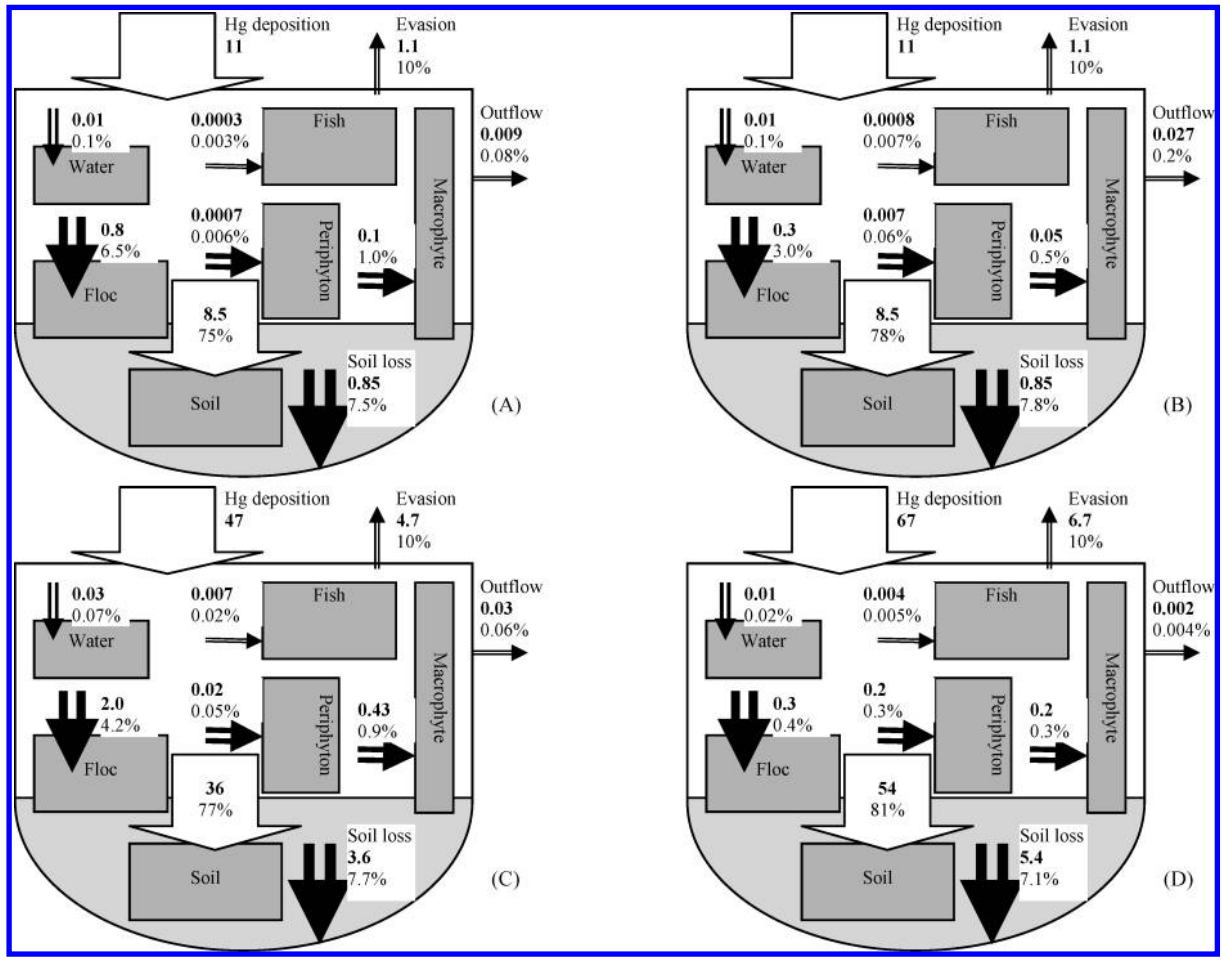

FIGURE 3. Mass (kg) and fraction (%) of THg entering each ecosystem component or leaving the system through output pathways after being deposited into the Everglades in the wet season in 2005. Arrows do not project the actual transport pathways. Mosquitofish was abbreviated to fish in the figure. (A) WCA 1; (B) WCA 2; (C) WCA 3; and (D) ENP.

may be the result of higher biological activity, productivity, and methylation during these warmer months (*7, 24, 25*). Soil loss was the major pathway of MeHg loss from the system, with 5−9% of MeHg, depending on management unit, being removed through this pathway. Water outflow was generally a minor pathway of MeHg loss, except for WCA 2, where 3.6 and 10% of MeHg produced from seasonally deposited Hg was lost through this pathway during the dry and wet season, respectively.

Uncertainty of Calculated Results. The results of uncertainty analysis are illustrated in Tables S10−S12 of the SI. The uncertainties associated with the calculated mass storage of THg and MeHg in the ecosystem components are usually less than 30%, except for floc for which uncertainties ranged about from 30 to 57%. The uncertainties associated with the predicated THg mass entering each ecosystem component after being deposited are about 20−30%, but can be higher (50−60%) for WCA 2. For the MeHg mass predicated to be redistributed into each ecosystem component after being produced from the seasonally deposited Hg, higher uncertainties are present, ranging from 30 to 100%.

Spatiality in THg and MeHg Cycling. For fluxes of seasonally deposited THg, spatial differences among management units mainly lie in between ENP and WCAs (Table 1). Although there are no significant differences in THg fluxes of seasonal deposition into soil among the 4 management units, levels of THg entering the other components (e.g., water, floc, periphyton, and mosquitofish) are different between ENP and WCAs. A distinct spatial characteristic of the fate of seasonally deposited THg is that the THg entered water and floc at a significantly lower rate in ENP than in the WCAs. For example, the water THg flux was 4.0 ng/m^2/season in the 2005 wet season for ENP, but it was 14−25 ng/m^2/season for the WCAs. For floc, the THg flux into ENP in the 2005 wet season was 87 ng/m^2/season, while it was 598−1302 ng/m^2/season for the WCAs. Possible reasons for these spatial variations include (1) lower water depth and lower floc thickness in ENP than in the WCAs, which would result in decreased capacity of THg storage in water and floc per unit area in ENP, compared to WCAs; (2) differences in the effects of floc on redox conditions, benthic-pelagic coupling, and dissolved gaseous Hg production and evasion rate between ENP and WCAs; and (3) differences in DOC quantity and quality and inorganic nutrients between ENP and WCAs, which would affect Hg compartmentalization between ecosystem components. These results suggested the importance of water levels in determining Hg cycling and fate in the Everglades. In addition to water and floc, the seasonal THg fluxes into periphyton and mosquitofish were also different between ENP and WCAs, but with complicated patterns. For instance, for the wet season, periphyton THg flux was 52 ng/m^2/season for ENP, which was greater than that for WCAs (1.1−12 ng/m^2/season). For the dry season, the spatial patterns in periphyton THg fluxes changed from the wet season, with WCA 2 being significantly higher in periphyton THg than the other management units.

For fluxes of MeHg produced from seasonally deposited THg (Table 2), there were basically no spatial differences among different management units in the dry season. Two exceptions were that MeHg flux to floc was significantly lower in ENP than in WCA 3 and that periphyton MeHg flux for WCA 3 was higher than for WCA 1. During the wet season,

TABLE 1. THg Fluxes to Each Ecosystem Component (μg/m^2/season for Soil and ng/m^2/season for the Other Components) Generated from Hg Seasonally Deposited into the Different Management Units of the Everglades in 2005[a]

component	ENP	WCA 3	WCA 2	WCA 1
		dry season		
water	0.31 (0.11−0.51)[a]	1.5 (1.0−2.0)[b]	2.5 (1.1−4.0)[b]	2.7 (1.4−4.1)[b]
soil	4.0 (1.2−6.8)[a]	4.0 (2.7−5.4)[a]	4.0 (1.6−6.3)[a]	4.0 (1.6−6.4)[a]
floc	1.8 (0.49−3.0)[a]	15 (10−20)[b]	39 (15−63)[b]	14 (6.4−21)[b]
periphyton	1.6 (0.44−2.8)[ab]	3.5 (2.1−4.9)[a]	27 (9.0−45)[c]	0.9 (0.4−1.0)[b]
macrophyte	8.2 (3.0−13)[a]	17 (12−23)[a]	16 (7.1−25)[ab]	45 (24−67)[b]
Mosquitofish	0.20 (0.058−0.34)[ab]	0.40 (0.26−0.54)[b]	0.25 (0.088−0.41)[ab]	0.062 (0.029−0.095)[a]
		wet season		
water	4.0 (2.9−5.1)[a]	14 (11−18)[b]	22 (7.1−36)[bc]	25 (19−31)[c]
soil	16 (11−21)[a]	15 (11−19)[a]	16 (5.0−27)[a]	15 (11−19)[a]
floc	87 (60−114)[a]	829 (616−1042)[b]	598 (151−1045)[b]	1302 (939−1665)[b]
periphyton	52 (35−69)[a]	10 (7.5−13)[b]	12 (2.2−22)[b]	1.1 (0.8−1.5)[c]
macrophyte	60 (43−77)[a]	181 (139−223)[b]	98 (32−163)[ab]	198 (150−247)[b]
Mosquitofish	1.1 (0.72−1.4)[a]	3.0 (2.2−3.8)[b]	1.5 (0.41−2.5)[abc]	0.58 (0.41−0.74)[ac]

[a] Numbers in parentheses are the 83% confidence intervals of the calculated values. For each ecosystem component, differences among units (at $p = 0.05$ significance level) are denoted by the letters accompanying the numbers. If two fluxes are labeled by different letters, then they are significantly different.

TABLE 2. MeHg Fluxes (ng/m^2/season) to Each Ecosystem Component Generated from MeHg Produced from Hg Newly Deposited into the Different Management Units of the Everglades during the 2005 Dry or Wet Season[a]

component	ENP	WCA 3	WCA 2	WCA 1
		dry season		
water	0.078 (−0.013−0.17)[a]	0.25 (0.14−0.36)[a]	1.4 (0.045−2.8)[a]	0.29 (0.046−0.52)[a]
soil	64 (−23−150)[a]	65 (34−95)[a]	69 (−7.3−145)[a]	105 (−2.3-211)[a]
floc	0.088 (−0.029−0.20)[a]	0.91 (0.44−1.4)[b]	3.6 (−0.21−7.4)[ab]	0.35 (0.021−0.68)[ab]
periphyton	0.22 (−0.070−0.51)[ab]	0.36 (0.19−0.54)[a]	2.3 (−0.35−5.0)[ab]	0.01 (0.0−0.03)[b]
macrophyte	0.72 (−0.12−1.6)[a]	1.7 (0.96−2.5)[a]	2.3 (0.074−4.5)[a]	3.0 (0.49−5.6)[a]
Mosquitofish	0.44 (−0.086−0.97)[a]	0.66 (0.34−0.98)[a]	0.75 (−0.01−1.5)[a]	0.10 (0.01−0.20)[a]
		wet season		
water	0.38 (0.24−0.52)[a]	1.4 (0.91−1.9)[b]	3.5 (−0.28−7.2)[ab]	3.5 (0.84−6.2)[b]
soil	81 (47−116)[a]	52 (32−72)[a]	37 (−12−86)[a]	204 (7.3−402)[a]
floc	1.9 (1.1−2.7)[a]	16 (9.2−22)[b]	13 (−2.7−29)[ab]	49 (6.3−92)[b]
periphyton	4.8 (2.9−6.8)[a]	0.72 (0.45−1.0)[b]	1.8 (−0.37−4.1)[abc]	0.2 (0−0.3)[c]
macrophyte	5.8 (3.7−7.9)[a]	13 (8.2−17)[b]	9.4 (−0.77−20)[ab]	21 (5.0−37)[ab]
Mosquitofish	1.5 (0.92−2.1)[a]	3.4 (2.1−4.7)[b]	1.3 (−0.15−2.8)[ab]	0.77 (0.16−1.4)[a]

[a] Numbers in parentheses are the 83% confidence intervals of the calculated values. For each ecosystem component, differences among units (at $p = 0.05$ significance level) are denoted by the letters accompanying the numbers. If two fluxes are labeled by different letters, tnen they are significantly different.

MeHg fluxes in ENP were significantly different than in WCA 3 for all ecosystem components. MeHg fluxes to soil, water, floc, macrophyte, and mosquitofish were 52, 1.4, 16, 13, and 3.4 ng/m^2/season in WCA 3 while they were 81, 0.38, 1.9, 5.8, and 1.5 ng/m^2/season in ENP during the wet season. These data suggest that less MeHg was retained in soil while more MeHg was transported into water, floc, macrophyte, and mosquitofish in WCA 3, in comparison to ENP. Again, the differences in water depth and thus in floc thickness between ENP and WCA 3 played an important role in MeHg cycling. The greater water depth and floc thickness in WCA 3 would allow more retention of MeHg in water and floc while less in soil per unit area in WCA 3, compared to ENP. However, for periphyton, MeHg flux in ENP (4.8 ng/m^2/season) was significantly higher than in WCA 3 (0.72 ng/m^2/season), which has important implications on MeHg bioaccumulation because periphyton can serve as the base of food web in the Everglades (*10, 26*).

Environmental Implications. This work detailed a methodological example of conducting large-scale investigations on biogeochemical cycling of Hg in wetland ecosystems. The data processing techniques employed here for constructing mass inventory and mass budget of THg and MeHg could be applied to other systems. The results revealed some relative distribution patterns of legacy Hg mass storage among the ecosystem components in the Everglades, which would provide implications for understanding Hg cycling in other wetlands.

In addition, this work has important implications for adaptive management of Hg contamination in the Everglades, and on the ongoing restoration of this ecosystem. Our results suggest that in general small amounts of Hg are transported in water from one management unit to another in the Everglades. The greatest transport occurs in WCA 2 where water inflow (mainly from EAA and WCA 1) could contribute 4% of total MeHg storage in the system. Despite appearing to play a minor role in Hg cycling under current conditions, the across-unit transport of Hg should be taken into account in stormwater management and in Everglades restoration. One component of the Comprehensive Everglades Restoration Plan (CERP) is to increase water deliveries to the Everglades (*27*). Such activities could alter the water flows and thus Hg transport across management units. For instance, for WCA 2, MeHg inflow would be close to 10% of total MeHg storage, if water inflow increases by 2-fold, with other conditions being unchanged.

Mass storage of Hg in the Everglades greatly exceeds annual inputs to the system, with the latter accounting for only 1−2% of the legacy Hg. Consequently, the Hg already entrapped in the system and its cycling among the ecosystem

components play an important role in fish Hg contamination. Since Hg cycling is closely related to specific ecological conditions (e.g., water depth, floc depth, and abundance of periphyton) in a management unit, alterations in these ecological conditions would influence Hg transport and transformation. The restoration activities, e.g., increasing water flows, could alter the ecological conditions and thus change the compartmentalization and bioaccumulation of Hg in this ecosystem.

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Supporting Information Available

Figures showing a map of the Everglades and mass budget results for the dry season, detailed calculation procedures, and tables listing legacy Hg inventory, uncertainty analysis results, and geochemical characteristics of the Everglades. This material is available free of charge via the Internet at http://pubs.acs.org.

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