Supporting Information

Legacy and Fate of Mercury and Methylmercury in the Florida Everglades

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There are total of 42 pages, 15 tables and 4 figures.

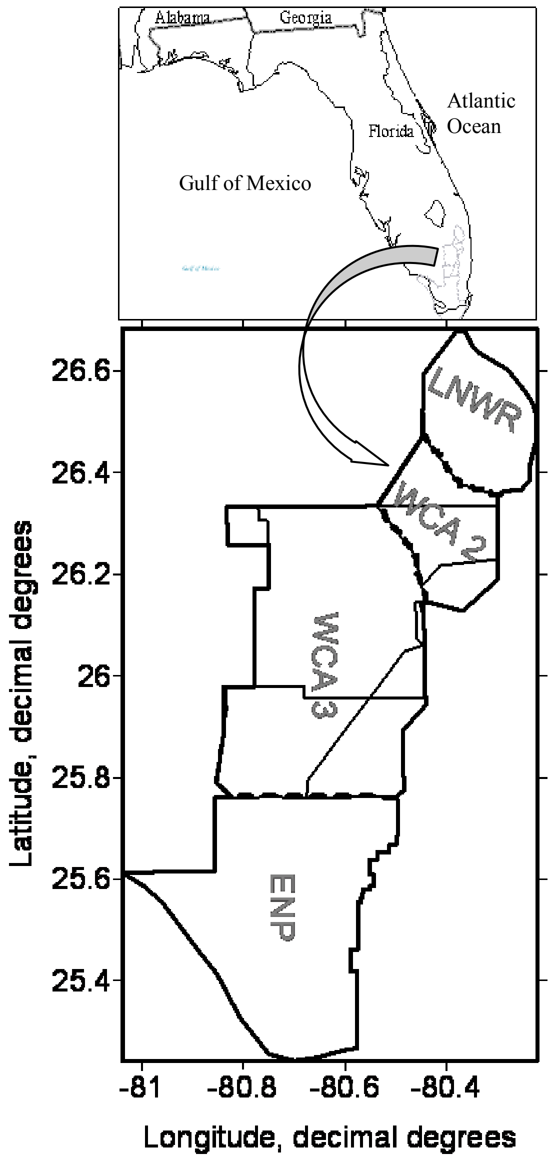

Fig. S1 A map showing four management units in the Florida Everglades. LNWR, Arthur R. Marshall Loxahatchee National Wildlife Refuge (also known as WCA 1); WCA 2 and WCA 3, Water Conservation Areas 2 and 3; ENP, Everglades National Park.

METHODS

Data Collection. Multiple datasets have been generated for Hg in the Everglades, originating from projects focusing on different aspects of Hg transport, transformation, and cycling. Examples of these datasets include the U.S. Environmental Protection Agency (EPA) Everglades Regional Environmental Monitoring and Assessment Program (R-EMAP), the USGS Aquatic Cycling of Mercury in the Everglades (ACME) project, the Mercury Deposition Network (MDN, http://nadp.sws.uiuc.edu/mdn/), and the DBHYDRO database maintained by South Florida Water Management District (SFWMD) (*1,2*).

Constructing mass inventories and mass budgets to characterize ecosystem-wide Hg cycling requires the combination and comprehensive analysis of these datasets. Data used in this study were collected from multiple datasets, including R-EMAP (*3*), ACME (*2*), MDN (*4*), and DBHYDRO (*5*). The R-EMAP datasets (in particular those generated in 2005) were used as the primary data source for constructing a mass inventory and a mass budget because R-EMAP adopted a probability-based, ecosystem-wide sampling design. The R-EMAP 2005 sampling occurred in May and November for the dry and wet seasons, respectively. Consistent with the R-EMAP schedule, we defined the 2005 dry season from mid November 2004 through mid May 2005 and the wet season from mid May 2005 through mid November 2005 during our calculations. Detailed information about sampling design, sampling protocols, analytical procedures, and original data for each dataset can be found elsewhere (*1-3,6*).

Mass Inventory of THg and MeHg. For each management unit, 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) were included in developing a mass inventory of THg and MeHg. The inputs and outputs were calculated for the whole 2005 wet season, while the storage, which was used to reflect a relative perspective of instantaneous mass among ecosystem components at the time of sampling, was calculated based on the R-EMAP 2005 November data.

Calculation of THg Inputs and Outputs. Mass of THg input through wet deposition was estimated based on the MDN data monitoring at site FL11 (Everglades National Park Research Center) while dry deposition was based on the estimation in the literature (*7*). The THg mass in water inflows and outflows were estimated by multiplying cumulative flows for the 2005 wet season by THg concentrations in the flows (obtained from the SFWMD DBHYDRO database and related reports). The mass of THg evasion was estimated by assuming an evasion rate of 2 $ng/m^2/h$ (same for all management units due to lacking spatial data) and 10 h per day during which evasion occurs (*8-13*). Tables S1-S4 list parameters used for calculating mass inventory of THg and MeHg in the four management units of the Everglades during the 2005 wet season. The values of parameters and the procedures used for the calculations of deposition, inputs via water inflows, outputs via water outflows, and evasion of THg can be found in the tables and table notes.

Calculation of Hg Mass Storage. Mass of THg stored in each ecosystem component in each management unit of the Everglades was estimated based on R-EMAP probability sampling design. In this sampling design, the probability of a sample being included in the sampling, expressed as inclusion probability density function, is known for every element in the population. Thus the amount of area that each sample point represents is predetermined (as the reciprocal of the inclusion probability density function). According to the Horvitz-Thompson Theorem (*1,14,15*), THg mass in each region was calculated following the equations below:

$$M_{SW}^{THg} = \frac{\sum_{i=1}^{n} \frac{C_{SW\ i}^{THg} \times d_{SW\,i}}{\pi_i}}{\sum_{i=1}^{n} (\frac{1}{\pi_i})} \times A \times 10^3 \quad (1)$$

$$M_{SD}^{THg} = \frac{\sum_{i=1}^{n} \frac{C_{SD\ i}^{THg} \times d_{SD\,i} \times BD_{SD\,i}}{\pi_i}}{\sum_{i=1}^{n} (\frac{1}{\pi_i})} \times A \times 10^6 \quad (2)$$

$$M_{FC}^{THg} = \frac{\sum_{i=1}^{n} \frac{C_{FC\ i}^{THg} \times d_{FC\,i} \times BD_{FC\,i}}{\pi_i}}{\sum_{i=1}^{n} (\frac{1}{\pi_i})} \times A \times 10^6 \quad (3)$$

$$M_{PE}^{THg} = \frac{\sum_{i=1}^{n} \frac{C_{PE\ i}^{THg} \times BM_{PE\,i}}{\pi_i}}{\sum_{i=1}^{n} (\frac{1}{\pi_i})} \times A \quad (4)$$

$$M_{FS}^{THg} = \frac{\sum_{i=1}^{n} \frac{C_{FS\ i}^{THg} \times W_{FS\,i} \times BM_{FS\,i}}{\pi_i}}{\sum_{i=1}^{n} (\frac{1}{\pi_i})} \times A \quad (5)$$

where M_{SW}^{THg}, M_{SD}^{THg}, M_{FC}^{THg}, M_{PE}^{THg}, M_{FS}^{THg} are THg mass (ng) stored in surface water, soil (top 10 cm layer only), flocculent detrital material (floc, which is a layer of suspended organic materials on top of the Everglades soil and contains mostly detritus from plants and algal inputs from periphyton), periphyton, and mosquitofish, respectively. $C_{SW\ i}^{THg}$, $C_{SD\ i}^{THg}$, $C_{FC\ i}^{THg}$, $C_{PE\ i}^{THg}$, $C_{FS\ i}^{THg}$ are THg concentration (ng/L for water and ng/g for other compartments) in surface water, soil, floc, periphyton, and mosquitofish at station *i*, respectively. $d_{SW\,i}$, $d_{SD\,i}$, $d_{FC\,i}$ are thickness (m) of surface water, soil (10 cm was taken for all stations), and floc at station *i*. BD_{SD_i} and $BD_{FC\,i}$ are bulk density (g/mL) of soil and floc at station *i*. $BM_{PE\,i}$ (g/m^2) and $BM_{FS\,i}$ (fish/m^2) are areal biomass of periphyton and mosquitofish at station *i*. $W_{FS\,i}$ (g) is average weight of mosquitofish analyzed for station *i*. π_i is inclusion probability density function of station *i* being included in the sampling design. A is (m^2) of a management unit. An average value was estimated for $BM_{PE\,i}$ or $BM_{FS\,i}$ for each management unit, since these parameters were not exclusively determined for all sampling stations during the 2005 R-EMAP sampling (Tables S1-S4).

Since the 2005 R-EMAP sampling did not include macrophyte data, THg mass in macrophyte was estimated based on the average THg concentration in macrophyte and the biomass of macrophyte reported in the literature (*1,16,17*). Due to the lack of macrophyte THg data for different management units, we used the same average THg concentration in macrophyte for all four management units. Tables S1-S4 list the average macrophyte THg for the Everglades and macrophyte biomass for each management unit and the literature or data sources used to estimate these parameters.

In the same manner, mass of MeHg input, output, and storage was also estimated. However, for MeHg cycling, we did not consider atmospheric deposition a direct source of MeHg input to the Everglades. This is because rainwater is typically low in MeHg, with inorganic Hg as the dominant form (*18*). Limited studies on MeHg in rainfall in South Florida also suggested that the MeHg concentrations in rainwater into the Everglades were extremely low (*18*). Instead, previous studies have shown that MeHg is primarily produced in situ in the Everglades, in particular in soil, floc, and periphyton (*19-21*). Although Hg methylation has been observed in some waters (*22,23*), MeHg production in the Everglades water column is negligible (*21*). Therefore, *in situ* production was considered the primary source of MeHg load in each management unit. Loss of MeHg through evasion was not considered in this study, since it is considered to be a minor pathway of MeHg input and output (*16*).

Table S1. Input parameters for calculating mass inventory of THg and MeHg in WCA 1 during the 2005 wet season

Parameter	Definition	Value	Reference
A	Area (m^2) of the region	5.72×10^8	(*24*)
$d_{SW\,i}$	Thickness (m) of surface water at station *i*	R-EMAP Phase III data	This study; (*3*)
$d_{SD\,i}$	Thickness (m) of soil at station *i*		
$d_{FC\,i}$	Thickness (m) of floc at station *i*		
$BD_{SD\,i}$	Bulk density (g/ml) of soil at station *i*		
$BD_{FC\,i}$	Bulk density (g/ml) of floc at station *i*		
$BM_{PE\,i}$	Areal biomass (g/m^2) of periphyton at station *i*	5.0	(*25,26*)
BM_{PK}	Areal biomass (g/m^2) of macrophyte	2380	(*16,17,27*)
$BM_{FS\,i}$	Areal biomass ($fish/m^2$) of mosquitofish at station *i*	4.80	(*28,29*)
$W_{FS\,i}$	Average weight (g) of mosquitofish analyzed for station *i*	R-EMAP Phase III data	This study; (*3*)
π_i	Inclusion probability of station *i*		
$C_{SW\ i}^{THg}$	THg concentration (ng/L) in water at station *i*		
$C_{SD\ i}^{THg}$	THg concentration (ng/g) in soil at station *i*		
$C_{FC\ i}^{THg}$	THg concentration (ng/g) in floc at station *i*		
$C_{PE\ i}^{THg}$	THg concentration (ng/g) in periphyton at station *i*		
$C_{FS\ i}^{THg}$	THg concentration (ng/g) in mosquitofish at station *i*		
$C_{SW\ i}^{MeHg}$	MeHg concentration (ng/L) in water at station *i*		
$C_{SD\ i}^{MeHg}$	MeHg concentration (ng/g) in soil at station *i*		
$C_{FC\ i}^{MeHg}$	MeHg concentration (ng/g) in floc at station *i*		
$C_{PE\ i}^{MeHg}$	MeHg concentration (ng/g) in periphyton at station *i*		
C_{PK}^{THg}	Average THg concentration (ng/g) in macrophyte	7.30 (ACME and REMAP data)	(*1,16*)
C_{PK}^{MeHg}	Average MeHg concentration (ng/g) in macrophyte	0.51 (ACME data)	(*16*)
M_{BD}^{THg}	THg deposition (ng) including wet and dry deposition[a]	1.14×10^{13}	MDN (*4*); (*7*)
M_{EV}^{THg}	THg evasion (ng)[b]	2.06×10^{12}	This study
V_{IF}	Volume (L) of water inflow[c]	2.28×10^{11}	DBHYDRO (*5*); (*30-32*)
V_{OF}	Volume (L) of water outflow[c]	1.96×10^{11}	
C_{IF}^{THg}	Average THg concentration (ng/L) in inflow water[d]	1.20	
C_{OF}^{THg}	Average THg concentration (ng/L) in outflow water[d]	0.96	
C_{IF}^{MeHg}	Average MeHg concentration (ng/L) in inflow water[d]	0.12	
C_{OF}^{MeHg}	Average MeHg concentration (ng/L) in outflow water[d]	0.28	

Table S2. Input parameters for calculating mass inventory of THg and MeHg in WCA 2 during the 2005 wet season

Parameter	Definition	Value	Reference
A	Area (m^2) of the region	5.44×10^8	(*24*)
$d_{SW\,i}$	Thickness (m) of surface water at station *i*	R-EMAP Phase III data	This study; (*3*)
$d_{SD\,i}$	Thickness (m) of soil at station *i*		
$d_{FC\,i}$	Thickness (m) of floc at station *i*		
$BD_{SD\,i}$	Bulk density (g/ml) of soil at station *i*		
$BD_{FC\,i}$	Bulk density (g/ml) of floc at station *i*		
$BM_{PE\,i}$	Areal biomass (g/m^2) of periphyton at station *i*	82	(*25,26*)
BM_{PK}	Areal biomass (g/m^2) of macrophyte	1507	(*16,17,27*)
$BM_{FS\,i}$	Areal biomass ($fish/m^2$) of mosquitofish at station *i*	17.4	(*28,29*)
$W_{FS\,i}$	Average weight (g) of mosquitofish analyzed for station *i*	R-EMAP Phase III data	This study; (*3*)
π_i	Inclusion probability of station *i*		
$C^{THg}_{SW\ i}$	THg concentration (ng/L) in water at station *i*		
$C^{THg}_{SD\ i}$	THg concentration (ng/g) in soil at station *i*		
$C^{THg}_{FC\ i}$	THg concentration (ng/g) in floc at station *i*		
$C^{THg}_{PE\ i}$	THg concentration (ng/g) in periphyton at station *i*		
$C^{THg}_{FS\ i}$	THg concentration (ng/g) in mosquitofish at station *i*		
$C^{MeHg}_{SW\ i}$	MeHg concentration (ng/L) in water at station *i*		
$C^{MeHg}_{SD\ i}$	MeHg concentration (ng/g) in soil at station *i*		
$C^{MeHg}_{FC\ i}$	MeHg concentration (ng/g) in floc at station *i*		
$C^{MeHg}_{PE\ i}$	MeHg concentration (ng/g) in periphyton at station *i*		
C^{THg}_{PK}	Average THg concentration (ng/g) in macrophyte	7.30 (ACME and REMAP data)	(*1,16*)
C^{MeHg}_{PK}	Average MeHg concentration (ng/g) in macrophyte	0.51 (ACME data)	(*16*)
M^{THg}_{BD}	THg deposition (ng) including wet and dry deposition[a]	1.09×10^{13}	MDN (*4*); (*7*)
M^{THg}_{EV}	THg evasion (ng)[b]	1.96×10^{12}	This study
V_{IF}	Volume (L) of water inflow[c]	9.44×10^{11}	DBHYDRO (*5*); (*30-32*)
V_{OF}	Volume (L) of water outflow[c]	9.09×10^{11}	
C^{THg}_{IF}	Average THg concentration (ng/L) in inflow water[d]	0.96	
C^{THg}_{OF}	Average THg concentration (ng/L) in outflow water[d]	1.00	
C^{MeHg}_{IF}	Average MeHg concentration (ng/L) in inflow water[d]	0.28	
C^{MeHg}_{OF}	Average MeHg concentration (ng/L) in outflow water[d]	0.10	

Table S3. Input parameters for calculating mass inventory of THg and MeHg in WCA 3 during the 2005 wet season

Parameter	Definition	Value	Reference
A	Area (m^2) of the region	2.44×10^9	(*24*)
$d_{SW\,i}$	Thickness (m) of surface water at station *i*	R-EMAP Phase III data	This study; (*3*)
$d_{SD\,i}$	Thickness (m) of soil at station *i*		
$d_{FC\,i}$	Thickness (m) of floc at station *i*		
$BD_{SD\,i}$	Bulk density (g/ml) of soil at station *i*		
$BD_{FC\,i}$	Bulk density (g/ml) of floc at station *i*		
$BM_{PE\,i}$	Areal biomass (g/m^2) of periphyton at station *i*	45	(*25,26*)
BM_{PK}	Areal biomass (g/m^2) of macrophyte	2656	(*16,17,27*)
$BM_{FS\,i}$	Areal biomass ($fish/m^2$) of mosquitofish at station *i*	26.6	(*28,29*)
$W_{FS\,i}$	Average weight (g) of mosquitofish analyzed for station *i*	R-EMAP Phase III data	This study; (*3*)
π_i	Inclusion probability of station *i*		
$C_{SW\ i}^{THg}$	THg concentration (ng/L) in water at station *i*		
$C_{SD\ i}^{THg}$	THg concentration (ng/g) in soil at station *i*		
$C_{FC\ i}^{THg}$	THg concentration (ng/g) in floc at station *i*		
$C_{PE\ i}^{THg}$	THg concentration (ng/g) in periphyton at station *i*		
$C_{FS\ i}^{THg}$	THg concentration (ng/g) in mosquitofish at station *i*		
$C_{SW\ i}^{MeHg}$	MeHg concentration (ng/L) in water at station *i*		
$C_{SD\ i}^{MeHg}$	MeHg concentration (ng/g) in soil at station *i*		
$C_{FC\ i}^{MeHg}$	MeHg concentration (ng/g) in floc at station *i*		
$C_{PE\ i}^{MeHg}$	MeHg concentration (ng/g) in periphyton at station *i*		
C_{PK}^{THg}	Average THg concentration (ng/g) in macrophyte	7.30 (ACME and REMAP data)	(*1,16*)
C_{PK}^{MeHg}	Average MeHg concentration (ng/g) in macrophyte	0.51 (ACME data)	(*16*)
M_{BD}^{THg}	THg deposition (ng) including wet and dry deposition[a]	4.73×10^{13}	MDN (*4*); (*7*)
M_{EV}^{THg}	THg evasion (ng)[b]	8.53×10^{12}	This study
V_{IF}	Volume (L) of water inflow[c]	1.65×10^{12}	DBHYDRO (*5*); (*30-32*)
V_{OF}	Volume (L) of water outflow[c]	1.58×10^{12}	
C_{IF}^{THg}	Average THg concentration (ng/L) in inflow water[d]	1.80	
C_{OF}^{THg}	Average THg concentration (ng/L) in outflow water[d]	1.60	
C_{IF}^{MeHg}	Average MeHg concentration (ng/L) in inflow water[d]	0.22	
C_{OF}^{MeHg}	Average MeHg concentration (ng/L) in outflow water[d]	0.12	

Table S4. Input parameters for calculating mass inventory of THg and MeHg in ENP during the 2005 wet season

Parameter	Definition	Value	Reference
A	Area (m^2) of the region	3.37×10^9	(*24*)
$d_{SW\,i}$	Thickness (m) of surface water at station *i*	R-EMAP Phase III data	This study; (*3*)
$d_{SD\,i}$	Thickness (m) of soil at station *i*		
$d_{FC\,i}$	Thickness (m) of floc at station *i*		
$BD_{SD\,i}$	Bulk density (g/ml) of soil at station *i*		
$BD_{FC\,i}$	Bulk density (g/ml) of floc at station *i*		
$BM_{PE\,i}$	Areal biomass (g/m^2) of periphyton at station *i*	739	(*25,26*)
BM_{PK}	Areal biomass (g/m^2) of macrophyte	1438	(*16,17,27*)
$BM_{FS\,i}$	Areal biomass ($fish/m^2$) of mosquitofish at station *i*	14.5	(*28,29*)
$W_{FS\,i}$	Average weight (g) of mosquitofish analyzed for station *i*	R-EMAP Phase III data	This study; (*3*)
π_i	Inclusion probability of station *i*		
$C_{SW\,i}^{THg}$	THg concentration (ng/L) in water at station *i*		
$C_{SD\,i}^{THg}$	THg concentration (ng/g) in soil at station *i*		
$C_{FC\,i}^{THg}$	THg concentration (ng/g) in floc at station *i*		
$C_{PE\,i}^{THg}$	THg concentration (ng/g) in periphyton at station *i*		
$C_{FS\,i}^{THg}$	THg concentration (ng/g) in mosquitofish at station *i*		
$C_{SW\,i}^{MeHg}$	MeHg concentration (ng/L) in water at station *i*		
$C_{SD\,i}^{MeHg}$	MeHg concentration (ng/g) in soil at station *i*		
$C_{FC\,i}^{MeHg}$	MeHg concentration (ng/g) in floc at station *i*		
$C_{PE\,i}^{MeHg}$	MeHg concentration (ng/g) in periphyton at station *i*		
C_{PK}^{THg}	Average THg concentration (ng/g) in macrophyte	7.30 (ACME and REMAP data)	(*1,16*)
C_{PK}^{MeHg}	Average MeHg concentration (ng/g) in macrophyte	0.51 (ACME data)	(*16*)
M_{BD}^{THg}	THg deposition (ng) including wet and dry deposition[a]	6.72×10^{13}	MDN (*4*); (*7*)
M_{EV}^{THg}	THg evasion (ng)[b]	1.21×10^{13}	This study
V_{IF}	Volume (L) of water inflow[c]	1.68×10^{12}	DBHYDRO (*5*); (*30-32*)
V_{OF}	Volume (L) of water outflow[c]	2.40×10^{11}	
C_{IF}^{THg}	Average THg concentration (ng/L) in inflow water[d]	1.60	
C_{OF}^{THg}	Average THg concentration (ng/L) in outflow water[d]	1.40	
C_{IF}^{MeHg}	Average MeHg concentration (ng/L) in inflow water[d]	0.12	
C_{OF}^{MeHg}	Average MeHg concentration (ng/L) in outflow water[d]	0.10	

Notes for Tables S1-S4:

[a] Dry and wet deposition were obtained from reference (*7*) and monitoring data from Mercury Deposition Network (MDN, http://nadp.sws.uiuc.edu/mdn/) site FL11 (Everglades National Park Research Center) (*4*), respectively. There are other MDN stations, e.g., FL34, FL04 and FL11, in a north-south transect through the 4 management units in the Everglades and Hg wet deposition varies among these stations due to concentration and rainfall differences. There would be slight differences in THg input when changing MDN stations. However, changing location for Hg wet deposition has no effect on the calculations of Hg masses stored in the ecosystem components, since Hg mass storage and Hg input were calculated separately in the mass inventory model. Therefore, we used only FL11 for Hg wet deposition input, since this site has the longest period of record monitoring mercury in wet deposition. It should be noted that the landfall of several hurricanes (e.g., Katrina in August and Rita in September) in 2005 significantly affected the functioning of Hg monitoring at station FL11. It was observed that, during these severe storm events, the collectors recorded significant precipitation with little THg or were non-functioning (*33*). Therefore, in consideration of the impacts of hurricanes on Hg monitoring devices, the monitoring results of Hg wet deposition in 2005 downloaded from the MDN network should be viewed with caution.

[b] Mass of THg evasion was estimated by assuming an evasion rate of 2 ng/m^2/h, 10 h per day during which evasion occurs, and 180 days for the 2005 wet season (*8-13*). After calculation, the Hg evasion rate for the 2005 wet season was 3.6 µg/m^2/season. If the same evasion rate was used to calculate Hg evasion for the whole year of 2005 (previous studies show similar evasion rates for both the dry and wet seasons, Table S15), the annual Hg evasion rate would be 7.2 µg/m^2/yr. The processes of THg evasion, including production of dissolved gaseous Hg, Hg^0 oxidation, and

Hg^0 emission, are affected by a variety of factors such as temperature, solar radiation, and water depth (*8,11,12,34*). The assumed evasion rate of 2 ng/m^2/h was based upon the previous studies. A series of studies have directly measured gaseous Hg fluxes over open water surface and estimated the evasion rates of Hg from water in the Everglades (*11,12,35,36*). The results of these studies (Table S15) suggest that the Hg evasion rates are around 1-3 ng/m^2/h (daytime), with Hg evasion at night being negligible (compared to daytime). For instance, Lindberg et al. (*11*) measured a direct evasion flux of Hg from Everglades water of averaging 2.7 ng/m^2/h. in addition, based on the measured dissolved gaseous mercury concentrations, Krabbenhoft et al. (*8*) calculated an annual evasion rate of Hg from the Everglades of 2.2 μg/m^2/year. It should be noted that Hg transpiration through macrophyte was not included in our models. Different from Hg evasion from open water surface, Hg transpiration transfers Hg in sediment to the atmosphere through a series of processes, including reduction of Hg^{2+} to Hg^0 in the rhizosphere, uptake of Hg^0 by macrophyte roots, and transport of Hg^0 from macrophyte to the atmosphere (*9,10,12,13,35*). In our models, only the top 10 cm layer of sediment was considered inside the system, whereas the sediment below 10 cm depth was considered outside the system. Since Hg transpiration transfers Hg in sediment to the atmosphere, macrophyte roots may absorb Hg from the sediment deeper than 10 cm. In this case, it is not appropriate to include Hg transpiration in our models, because this process does not transport Hg out of the system. This is the primary reason why Hg transpiration was not included in our models. Additional consideration of not including Hg transpiration in our model is because it is difficult to estimate the Hg transport through transpiration. In order to estimate Hg transpiration, the types and coverage of macrophytes in each management unit and the capability of each type of macrophyte transferring Hg need to be known, yet they remain lacking for most areas of the Everglades.

[c] The water inflows and outflows were calculated based on the monthly inflows and outflows of major structures entering and leaving each management unit (Table S13), which were recorded in the DBHYDRO database and reported in the South Florida Environmental Report (2006 and 2007) (*31,32*). The inflows and outflows occurred during the period from May 2005 to November 2005 were summed up to calculate the flows for the 2005 wet season. It should be noted that the outflow for the ENP was estimated based on the results of water flows to Florida Bay and the Keys from the Everglades. Flows from Taylor River Slough, C111 structure, Shark River Slough, and three major creeks that flow into the bay, Trout Creek and Taylor Creek (both flowing into the eastern bay) and McCormick Creek (flowing into the central bay), were used for calculation (*30*).

[d] The average concentrations of THg or MeHg in water inflows and outflows were calculated by averaging THg or MeHg concentrations monitored for each structures during the period of the 2005 wet season defined in this paper, which were recorded in the DBHYDRO database. It should be noted that not all structures were monitored for THg or MeHg.

Mass Budget of Seasonally Deposited THg and MeHg Produced from Seasonally Deposited THg. A model, based on Hg compartmentalization between water and the other ecosystem component, was adapted to predict the fate of seasonally deposited Hg for each management unit. This model was previously used to estimate the fate of seasonally deposited Hg on the Everglades-wide scale (*37*). For each management unit, THg fractions entering each ecosystem component post deposition were calculated following Equation 6. Of all Hg deposited during the dry (or wet) season, a small portion was methylated into MeHg, which was estimated according to MeHg to THg ratio (*f*) in soil, floc, and periphyton. MeHg fractions retained by each ecosystem component after being produced were calculated following Equation 7.

$$M_{BD}^{THg} = \Delta C_{SW}^{THg} \times (V_{SW} + R_{SD}^{THg} M_{SD} + R_{FC}^{THg} M_{FC} + R_{PE}^{THg} M_{PE} + R_{PK}^{THg} M_{PK} + BAF_{FS}^{THg} M_{FS} + V_{OF}) + M_{EV}^{THg} + M_{SL}^{THg} \quad (6)$$

$$M_{PD}^{MeHg} = \Delta C_{SW}^{MeHg} \times (V_{SW} + R_{SD}^{MeHg} M_{SD} + R_{FC}^{MeHg} M_{FC} + R_{PE}^{MeHg} M_{PE} + R_{PK}^{MeHg} M_{PK} + BAF_{FS}^{MeHg} M_{FS} + V_{OF}) + M_{SL}^{MeHg} \quad (7)$$

where *M*, *C*, and *V* refer to mass, concentration, and volume, respectively. *ΔC* represents the increase in THg or MeHg concentration due to the input of seasonally deposited Hg. *R* is the distribution ratio of Hg between water and other ecosystem components. For each parameter, the subscript denotes the ecosystem component (SW: surface water; SD: soil; FC: floc; PE: periphyton; PK: macrophyte; FS: mosquitofish) or transport pathway (OF: outflow; EV: evasion; SL: soil loss), while the superscript denotes the Hg species. Tables S5-S8 list the values of these parameters and the procedures and data sources used to estimate these parameters. The left-hand sides of the equations are bulk deposition (BD) of THg or total MeHg produced (PD), while the right-hand sides represent THg or MeHg amounts redistributed into each ecosystem component plus transport. After *M*, *V,* and *R* were calculated, ΔC_{SW}^{THg} and ΔC_{SW}^{MeHg} were obtained from Equations 6 and 7 and *ΔC* for the other ecosystem components were calculated based on the definition of R. After all ΔC are obtained, the Hg mass and fraction entering each ecosystem component were then computed. Detailed information on model development, definition of each parameter, and calculations can be found elsewhere (*37*).

Uncertainty Analysis. Uncertainty analysis was conducted for the models used for calculating mass inventory of legacy Hg and mass budget of new Hg, for the purpose of evaluating the precision of calculated results. The principles described in the U.S. Guide to the Expression of Uncertainty in Measurement (GUM) were applied to estimate the uncertainty for calculated THg and MeHg mass storage and mass budget (*38*). Here we used the relative standard error (*RSE*) to estimate the relative uncertainty (U_r). For calculations of mass storage, the uncertainty was estimated based on the GUM and following the Horvitz-Thompson Theorem (*1,14,15*), which provides general estimating formulae for the accompanying variance expressions, in addition to the means.

$$U_r = 100 \times \frac{\sqrt{n} \times \sqrt{\dfrac{\sum_{i=1}^{n} (\dfrac{Z_i - \widehat{Z}}{\pi_i})^2}{n-1}}}{\sum_{i=1}^{n} \dfrac{Z_i}{\pi_i}} \qquad (8)$$

where Z_i, expressed as a function of measured parameters that are used to calculate Hg storage in the ecosystem components,

$= C_{SW\,i}^{Hg} \times d_{SW\,i}$ for water

$= C_{SD\,i}^{Hg} \times d_{SD\,i} \times BD_{SD\,i}$ for soil

$= C_{FC\,i}^{Hg} \times d_{FC\,i} \times BD_{FC\,i}$ for floc

$= C_{PE\,i}^{Hg} \times BM_{PE\,i}$ for periphyton

$= C_{FS\,i}^{Hg} \times W_{FS\,i} \times BM_{FS\,i}$ for fish

$\widehat{Z}$ is the mean of Z_i and calculated as follows

$$\widehat{Z} = \frac{\sum_{i=1}^{n} \frac{Z_i}{\pi_i}}{\sum_{i=1}^{n} (\frac{1}{\pi_i})} \quad (9)$$

For mass budget of newly deposited Hg, the uncertainty largely comes from the variations in *R* values (for both THg and MeHg) as well as in *f* values (for MeHg only) which were used during the calculation. Mean values of *R* and *f* were used to calculate Hg mass compartmentalized into each ecosystem component for each management unit, which would inevitably lead to the uncertainty in model output because the Everglades is highly spatial in ecological conditions, even in the same management unit. The relative uncertainty of individual *R* and *f* was calculated as follows

$$U_{ri} = RSE_i \quad (i = 1, 2, \cdots m) \quad (10)$$

The total relative uncertainty (U_{rT}) for Hg mass into each ecosystem component was estimated for the combined uncertainty as the square root of the sum of the squares of individual relative uncertainty for each *R* and *f*

$$U_{rT} = \sqrt{\sum_{i=1}^{m} (U_{ri})^2} = \sqrt{\sum_{i=1}^{m} (RSE_i)^2} \quad (11)$$

It should be noted that, during both mass inventory and mass budget calculations, the inputs (e.g., atmospheric deposition and water inflows) and outputs (e.g., outflows, evasion, and soil loss) of THg and MeHg were estimated from the values reported in the literature. Also, concentrations of THg and MeHg in macrophyte and areal biomass of macrophyte were taken from the USGS monitoring data and other reports. For these data, uncertainty analysis was not performed here due to insufficient data. An arbitrary estimate is that the uncertainties associated with these data should be of comparable magnitude to the uncertainties estimated here for other parameters.

Comparison of Cycling of Newly Deposited Hg in Different Management Units of the Everglades. After Hg masses were calculated, Hg fluxes were derived by dividing THg or MeHg mass per season by area of a management unit for all management units and then compared to investigate the spatiality of Hg cycling. The comparison of Hg fluxes was carried out by comparing their confidence intervals (CIs), which were constructed following uncertainty analysis of Hg mass into the ecosystem components.

Same as Hg mass compartmentalized into each ecosystem component, the uncertainty for calculated Hg fluxes comes from the variations in R values (for both THg and MeHg) as well as in f values (for MeHg only) for each sampling station. The U_{rT} for Hg flux into each ecosystem component equals to U_{rT} for Hg mass. Hence the confidence intervals of calculated Hg fluxes were constructed based on the equation below.

$$CI = F_c \pm t \times \frac{s}{\sqrt{n}} = F_c \pm t \times F_c \times U_{rT} = F_c \times (1 \pm t \times U_{rT}) \qquad (12)$$

where F_c is the calculated flux of Hg into one ecosystem component, t is t-distribution critical value (=1.37 for 83% confidence limit), and U_{rT} is the total relative uncertainty for each calculated Hg flux. Here we used 83% confidence intervals to compare two Hg fluxes, which approximates a test at the significance level of P = 0.05 (*39*). The criterion to assess whether or not two Hg fluxes are significantly different is whether or not two confidence intervals overlap. If two 83% confidence intervals do not overlap, then the two Hg fluxes are significantly different from one another at the P=0.05 level.

Table S5. Input parameters for estimating mass budget of Hg newly deposited to WCA 1 of the Everglades during the 2005 dry and wet seasons

Parameter	Definition	Value		Reference
		Dry season	Wet season	
M_{BD}^{THg} (ng)	THg deposition, = wet + dry THg deposition[a]	2.840×10^{12}	1.14×10^{13}	MDN (*4*); (*7*)
V_{SW} (L)	Surface water volume, = water depth[b]× WCA 1 area[c]	1.07×10^{11}	3.14×10^{11}	This study; R-EMAP; (*24*)
M_{SD} (g)	Soil dry mass, = soil thickness[d] × WCA 1 area × bulk density[b]	3.88×10^{12}	3.64×10^{12}	
M_{FC} (g)	Floc dry mass, = floc thickness[b] × WCA 1 area × bulk density[b]	1.66×10^{10}	3.83×10^{11}	
M_{PE} (g)	Periphyton dry mass, = areal biomass[e] × area	2.86×10^{9}	2.86×10^{9}	(*1,25,26,40*); This study
M_{PK} (g)	Macrophyte dry mass, = areal biomass[f] × area	9.08×10^{11}	1.36×10^{12}	(*16,17,27*); This study
M_{FS} (g)	Mosquitofish mass in wet weight, = areal biomass[g]× weight[h] × area	3.38×10^{8}	3.89×10^{8}	(*28,29*); This study
M_{EV}^{THg}	THg evasion[i]	10% of THg deposited		This study
V_{OF} (L)	Water outflow[j]	6.73×10^{10}	1.96×10^{11}	DBHYDRO (*5*); (*30-32*)
R_{SD}^{THg} (L/g)	Distribution ratio of THg between water and soil[b]	40.3	51.9	This study; R-EMAP
R_{FC}^{THg} (L/g)	Distribution ratio of THg between water and floc[b]	32.2	43.1	
R_{PE}^{THg} (L/g)	Distribution ratio of THg between water and periphyton[b]	12.3	5.08	
R_{PK}^{THg} (L/g)	Distribution ratio of THg between water and macrophyte[k]	2.0	3.3	This study; ACME; R-EMAP
BAF_{FS}^{THg} (L/g)	Bioaccumulation factor of THg in mosquitofish[b]	7.19	18.8	This study; R-EMAP
R_{SD}^{MeHg} (L/g)	Distribution ratio of MeHg between water and soil[b]	10.1	5.03	
R_{FC}^{MeHg} (L/g)	Distribution ratio of MeHg between water and floc[b]	7.84	11.5	
R_{PE}^{MeHg} (L/g)	Distribution ratio of MeHg between water and periphyton[b]	1.89	4.78	
R_{PK}^{MeHg} (L/g)	Distribution ratio of MeHg between water and macrophyte[k]	1.10	2.4	This study; ACME
BAF_{FS}^{MeHg} (L/g)	Bioaccumulation factor of MeHg in mosquitofish[b]	113.9	178.8	This study; R-EMAP
M_{PD}^{MeHg} (ng)	MeHg produced in soil, floc, and periphyton[l]	6.81×10^{10}	1.73×10^{11}	
M_{SL}^{THg}	THg loss due to the loss of top 10 cm soil[m]	10% of THg in top 10 cm		This study
M_{SL}^{MeHg}	MeHg loss due to the loss of top 10 cm soil[m]	10% of MeHg in top 10 cm		This study

Table S6. Input parameters for estimating mass budget of Hg newly deposited to WCA 2 of the Everglades during the 2005 dry and wet seasons

Parameter	Definition	Value		Reference
		Dry season	Wet season	
M_{BD}^{THg} (ng)	THg deposition, = wet + dry THg deposition[a]	2.70×10^{12}	1.09×10^{13}	MDN (*4*); (*7*)
V_{SW} (L)	Surface water volume, = water depth[b]× WCA 2 area[c]	1.69×10^{11}	4.02×10^{11}	This study; R-EMAP; (*24*)
M_{SD} (g)	Soil dry mass, = soil thickness[d] × WCA 2 area × bulk density[b]	5.68×10^{12}	5.86×10^{12}	
M_{FC} (g)	Floc dry mass, = floc thickness[b] × WCA 2 area × bulk density[b]	4.75×10^{10}	4.02×10^{11}	
M_{PE} (g)	Periphyton dry mass, = areal biomass[e] × area	1.47×10^{11}	4.46×10^{10}	(*1,25,26,40*); This study
M_{PK} (g)	Macrophyte dry mass, = areal biomass[f] × area	5.46×10^{11}	8.20×10^{11}	(*16,17,27*); This study
M_{FS} (g)	Mosquitofish mass in wet weight, = areal biomass[g]× weight[h] × area	1.16×10^{9}	1.34×10^{9}	(*28,29*); This study
M_{EV}^{THg}	THg evasion[i]	10% of THg deposited		This study
V_{OF} (L)	Water outflow[j]	3.90×10^{11}	9.09×10^{11}	DBHYDRO (*5*); (*30-32*)
R_{SD}^{THg} (L/g)	Distribution ratio of THg between water and soil[b]	46.7	49.5	This study; R-EMAP
R_{FC}^{THg} (L/g)	Distribution ratio of THg between water and floc[b]	54.9	27.6	
R_{PE}^{THg} (L/g)	Distribution ratio of THg between water and periphyton[b]	12.3	5.08	
R_{PK}^{THg} (L/g)	Distribution ratio of THg between water and macrophyte[k]	2.0	3.3	This study; ACME; R-EMAP
BAF_{FS}^{THg} (L/g)	Bioaccumulation factor of THg in mosquitofish[b]	14.2	20.2	This study; R-EMAP
R_{SD}^{MeHg} (L/g)	Distribution ratio of MeHg between water and soil[b]	1.45	0.74	
R_{FC}^{MeHg} (L/g)	Distribution ratio of MeHg between water and floc[b]	9.08	3.77	
R_{PE}^{MeHg} (L/g)	Distribution ratio of MeHg between water and periphyton[b]	1.89	4.78	
R_{PK}^{MeHg} (L/g)	Distribution ratio of MeHg between water and macrophyte[k]	1.10	2.4	This study; ACME
BAF_{FS}^{MeHg} (L/g)	Bioaccumulation factor of MeHg in mosquitofish[b]	77.0	115.5	This study; R-EMAP
M_{PD}^{MeHg} (ng)	MeHg produced in soil, floc, and periphyton[l]	4.85×10^{10}	4.25×10^{10}	
M_{SL}^{THg}	THg loss due to the loss of top 10 cm soil[m]	10% of THg in top 10 cm		This study
M_{SL}^{MeHg}	MeHg loss due to the loss of top 10 cm soil[m]	10% of MeHg in top 10 cm		This study

Table S7. Input parameters for estimating mass budget of Hg newly deposited to WCA 3 of the Everglades during the 2005 dry and wet seasons

Parameter	Definition	Value		Reference
		Dry season	Wet season	
M_{BD}^{THg} (ng)	THg deposition, = wet + dry THg deposition[a]	1.17×10^{13}	4.73×10^{13}	MDN (*4*); (*7*)
V_{SW} (L)	Surface water volume, = water depth[b]× WCA 3 area[c]	7.15×10^{11}	1.78×10^{12}	This study; R-EMAP; (*24*)
M_{SD} (g)	Soil dry mass, = soil thickness[d] × WCA 3 area × bulk density[b]	3.07×10^{13}	2.60×10^{13}	
M_{FC} (g)	Floc dry mass, = floc thickness[b] × WCA 3 area × bulk density[b]	1.70×10^{11}	1.71×10^{12}	
M_{PE} (g)	Periphyton dry mass, = areal biomass[e] × area	2.37×10^{11}	1.07×10^{11}	(*1,25,26,40*); This study
M_{PK} (g)	Macrophyte dry mass, = areal biomass[f] × area	4.20×10^{12}	6.29×10^{12}	(*16,17,27*); This study
M_{FS} (g)	Mosquitofish mass in wet weight, = areal biomass[g]× weight[h] × area	7.75×10^{9}	8.92×10^{9}	(*28,29*); This study
M_{EV}^{THg}	THg evasion[i]	10% of THg deposited		This study
V_{OF} (L)	Water outflow[j]	5.79×10^{11}	1.58×10^{12}	DBHYDRO (*5*); (*30-32*)
R_{SD}^{THg} (L/g)	Distribution ratio of THg between water and soil[b]	62.2	73.2	This study; R-EMAP
R_{FC}^{THg} (L/g)	Distribution ratio of THg between water and floc[b]	42.3	60.0	
R_{PE}^{THg} (L/g)	Distribution ratio of THg between water and periphyton[b]	7.06	11.8	
R_{PK}^{THg} (L/g)	Distribution ratio of THg between water and macrophyte[k]	2.0	3.3	This study; ACME; R-EMAP
BAF_{FS}^{THg} (L/g)	Bioaccumulation factor of THg in mosquitofish[b]	24.5	41.6	This study; R-EMAP
R_{SD}^{MeHg} (L/g)	Distribution ratio of MeHg between water and soil[b]	6.02	2.57	
R_{FC}^{MeHg} (L/g)	Distribution ratio of MeHg between water and floc[b]	15.3	11.6	
R_{PE}^{MeHg} (L/g)	Distribution ratio of MeHg between water and periphyton[b]	4.36	8.63	
R_{PK}^{MeHg} (L/g)	Distribution ratio of MeHg between water and macrophyte[k]	1.10	2.4	This study; ACME
BAF_{FS}^{MeHg} (L/g)	Bioaccumulation factor of MeHg in mosquitofish[b]	241.2	489.2	This study; R-EMAP
M_{PD}^{MeHg} (ng)	MeHg produced in soil, floc, and periphyton[l]	1.79×10^{11}	2.19×10^{11}	
M_{SL}^{THg}	THg loss due to the loss of top 10 cm soil[m]	10% of THg in top 10 cm		This study
M_{SL}^{MeHg}	MeHg loss due to the loss of top 10 cm soil[m]	10% of MeHg in top 10 cm		This study

Table S8. Input parameters for estimating mass budget of Hg newly deposited to ENP of the Everglades during the 2005 dry and wet seasons

Parameter	Definition	Value		Reference
		Dry season	Wet season	
M_{BD}^{THg} (ng)	THg deposition, = wet + dry THg deposition[a]	1.67×10^{13}	6.72×10^{13}	MDN (*4*); (*7*)
V_{SW} (L)	Surface water volume, = water depth[b]× ENP area[c]	2.4×10^{11}	1.32×10^{12}	This study; R-EMAP; (*24*)
M_{SD} (g)	Soil dry mass, = soil thickness[d] × ENP area × bulk density[b]	9.72×10^{13}	1.11×10^{14}	
M_{FC} (g)	Floc dry mass, = floc thickness[b] × ENP area × bulk density[b]	6.07×10^{10}	5.47×10^{11}	
M_{PE} (g)	Periphyton dry mass, = areal biomass[e] × area	2.22×10^{11}	2.49×10^{12}	(*1,25,26,40*); This study
M_{PK} (g)	Macrophyte dry mass, = areal biomass[f] × area	3.23×10^{12}	4.84×10^{12}	(*16,17,27*); This study
M_{FS} (g)	Mosquitofish mass in wet weight, = areal biomass[g]× weight[h] × area	6.00×10^{9}	6.91×10^{9}	(*28,29*); This study
M_{EV}^{THg}	THg evasion[i]	10% of THg deposited		This study
V_{OF} (L)	Water outflow[j]	1.50×10^{11}	2.4×10^{11}	DBHYDRO (*5*); (*30-32*)
R_{SD}^{THg} (L/g)	Distribution ratio of THg between water and soil[b]	32.2	47.6	This study; R-EMAP
R_{FC}^{THg} (L/g)	Distribution ratio of THg between water and floc[b]	22.5	52.2	
R_{PE}^{THg} (L/g)	Distribution ratio of THg between water and periphyton[b]	5.7	6.9	
R_{PK}^{THg} (L/g)	Distribution ratio of THg between water and macrophyte[k]	2.0	3.3	This study; ACME; R-EMAP
BAF_{FS}^{THg} (L/g)	Bioaccumulation factor of THg in mosquitofish[b]	25.9	50.1	This study; R-EMAP
R_{SD}^{MeHg} (L/g)	Distribution ratio of MeHg between water and soil[b]	2.0	2.5	
R_{FC}^{MeHg} (L/g)	Distribution ratio of MeHg between water and floc[b]	4.4	11.9	
R_{PE}^{MeHg} (L/g)	Distribution ratio of MeHg between water and periphyton[b]	3.1	6.7	
R_{PK}^{MeHg} (L/g)	Distribution ratio of MeHg between water and macrophyte[k]	1.10	2.4	This study; ACME
BAF_{FS}^{MeHg} (L/g)	Bioaccumulation factor of MeHg in mosquitofish[b]	225.6	742.3	This study; R-EMAP
M_{PD}^{MeHg} (ng)	MeHg produced in soil, floc, and periphyton[l]	2.41×10^{11}	3.49×10^{11}	
M_{SL}^{THg}	THg loss due to the loss of top 10 cm soil[m]	10% of THg in top 10 cm		This study
M_{SL}^{MeHg}	MeHg loss due to the loss of top 10 cm soil[m]	10% of MeHg in top 10 cm		This study

Notes for Tables S5-S8:

[a] Dry and wet deposition were obtained from reference (*7*) and monitoring data from Mercury Deposition Network (MDN, http://nadp.sws.uiuc.edu/mdn/) site FL11 (Everglades National Park Research Center), respectively. See the notes for Tables S1-S4 for detailed information.

[b] Median value determined in the R-EMAP Phase III study was used for calculation, except for floc thickness in the dry season where the mean was used because the median was zero.

[c] The areas of WCA 1, 2, 3, and ENP were set to 572, 544, 2370, and 3368 km^2, respectively (*24*).

[d] Soil thickness was set to 10 cm taking into account that only surface soil is likely involved in Hg distribution during a period of a season (about 6 months).

[e] Periphyton areal biomass was set to 5.0, 270, 100, and 66 g/m^2 for WCA 1, 2, 3, and ENP respectively in the dry season while 5.0, 82, 45, and 739 g/m^2 in the wet season (*1,25,26,40*).

[f] Macrophyte areal biomass was set to 1587, 1005, 1771, and 959 g/m^2 for WCA 1, 2, 3, and ENP respectively in the dry season while 2380, 1507, 2656, and 1438 g/m^2 in the wet season (*16,17,27*).

[g] The areal biomass (density) of mosquitofish was assumed, based on the ALFISH model outputs, to be 4.80, 17.4, 26.6, and 14.5 $fish/m^2$ for WCA 1, 2, 3, and ENP, respectively (*1*), which was the same for the dry and wet seasons (*28,29*). The fish biomass estimates here are somewhat different than in our previous studies. We previously estimated a mosquitofish biomass of 3.5 and 14.5 $fish/m^2$ during the dry and wet season, respectively, for the entire Everglades. In the current studies, we need to estimate fish biomass for each of the 4 management units of the Everglades (WCAs 1, 2, 3, and ENP), instead of the whole Everglades. In this case, we think the

outputs of the ALFISH model, which is a model developed to assess the spatial pattern of fish densities through the greater Everglades freshwater marshes, would be more accurate. Since it is difficult to accurately estimate fish biomass, these estimates might underestimate mosquitofish biomass, which would result in underestimate of Hg storage in mosquitofish. In addition, it should be noted that this study estimated Hg storage in mosquitofish only, among all fish and wildlife that could accumulate Hg. The Hg storage in mosquitofish estimated here should only be viewed as Hg stored in mosquitofish, rather than Hg storage in fish in general. Our intention was to select mosquitofish as an example to demonstrate how to calculate Hg storage in higher-trophic level biological components (e.g., fish and wildlife). Mosquitofish was selected because we had ecosystem-wide mosquitofish weights and Hg concentrations in mosquitofish, which made it possible to estimate mosquitofish Hg storage. It is difficult to obtain sufficient data of ecosystem-wide Hg concentrations and biomasses for larger predatory fish and wildlife and thus to estimate the overall Hg storage in fish and wildlife. However, our estimates suggest that Hg storage in mosquitofish is lower than in soil and floc by several orders of magnitude. Therefore, compare to Hg storage in other ecosystem components (e.g., soil and floc), Hg storage in higher-trophic level biological components could be significantly less. The overall Hg storage in fish and wildlife, which would be (possibly several-fold) higher than our mosquitofish Hg storage, should have minimal effect on the relative distribution patterns of Hg mass storage among the ecosystem components.

[h] The median mosquitofish weights determined in the R-EMAP Phase III were used for calculation.

[i] Currently information is not available regarding the fraction of newly deposited (new) versus legacy (old) Hg among all Hg evaded out of this ecosystem. Our calculations in mass inventory

model of legacy Hg (assuming 2 $ng/m^2/h$ of evasion rate) suggest the annual mass flux of THg evolved from surface water is about 35% of the annual flux from atmospheric deposition. But the mass flux of THg evolved includes both the new and old Hg, and the new Hg input amounts only 1-2% of the old Hg already present in the Everglades. Considering the new Hg might be more reactive in evasion, we assumed that 10% of THg newly deposited to Everglades during a season (being same for the dry and wet season) would be evaded out of the system during the same time period. We further tested the sensitivity of mass budget model on evasion rate by changing the evasion rate of new Hg. If the evasion rate of new Hg is changed from 10% to 5%, the Hg mass entering each ecosystem component due to seasonally deposited Hg would increase by 5.5%. If the evasion rate of new Hg is changed from 10% to 20%, the Hg mass entering each ecosystem component due to seasonally deposited Hg would decrease by 11%.

[j] The water outflows were calculated based on the monthly outflows of major structures leaving each management unit, which were recorded in the DBHYDRO database and reported in the South Florida Environmental Report (2006 and 2007) (*31,32*) (see Table S1-S4).

[k] To calculate R_{PK}^{THg} and R_{PK}^{MeHg}, macrophyte THg concentrations were assumed to be 4.5 and 7.3 ng/g for the dry and wet season, respectively, based on previous R-EMAP and USGS ACME results (*1,16*). Macrophyte MeHg was assumed to account for 7% of THg based on the results of USGS analyses for cattail and sawgrass samples from the WCAs (*16*).

[l] In the Everglades, MeHg is produced primarily in soil, floc, and periphyton (*19-21*). For each component, the MeHg produced from seasonally deposited Hg was estimated as the product of the THg distributed into that component during that season and the median MeHg to THg ratio in that component. We did not take into account MeHg production in the water column, although Hg methylation, including photoproduction, has been observed in some other waters (*22,23,41*).

We also did not take into account MeHg input from atmospheric deposition. Measurements of MeHg in precipitation are sparse and the very limited previous studies have shown that the concentration of MeHg in rain is small (*42,43*). Previous studies have shown that MeHg concentrations in South Florida rain are generally considered environmentally insignificant (*44,45*).

[m] This portion of Hg loss was considered to be caused by the loss of top 10 cm layer soil due to a variety of processes, e.g., erosion, subsidence, and downward movement. Since limited information is available regarding these processes, we assumed that 10% of Hg in the upper 10 cm layer will be lost accompanying soil loss during the time period of one season. The sensitivity of mass budget model on this parameter was tested by changing the Hg loss rate. If the soil loss of new Hg is changed from 10% to 5%, the Hg mass entering each ecosystem component due to seasonally deposited Hg would increase by 4.7%. If the soil loss of Hg is changed from 10% to 20%, the Hg mass entering each ecosystem component due to seasonally deposited Hg would decrease by 8.2%.

Table S9 THg and MeHg inventory in WCA 1, WCA 2, WCA 3, and ENP. The inputs (kg) and outputs (Kg) are for the 2005 wet season (May to November), while the legacy illustrates instantaneous mass (kg) stored in all ecosystem components (including surface water, soil, floc, periphyton, macrophyte, and mosquitofish) at the time of sampling (November 2005) as well as Hg mass per unit area (kg/km^2).

Compartment	In	Out	Total accumulated	Legacy Hg	Legacy Hg / area
			THg		
WCA 1	11.7	2.2	9.4	914	1.6
WCA 2	11.8	2.9	8.9	1138	2.1
WCA 3	50.3	11.1	39.2	4931	2.1
ENP	69.9	12.4	57.5	7602	2.3
			MeHg		
WCA 1	0.027	0.055	0.028	15	0.027
WCA2	0.26	0.091	0.17	6.8	0.012
WCA 3	0.36	0.19	0.17	32	0.014
ENP	0.20	0.024	0.18	51	0.015

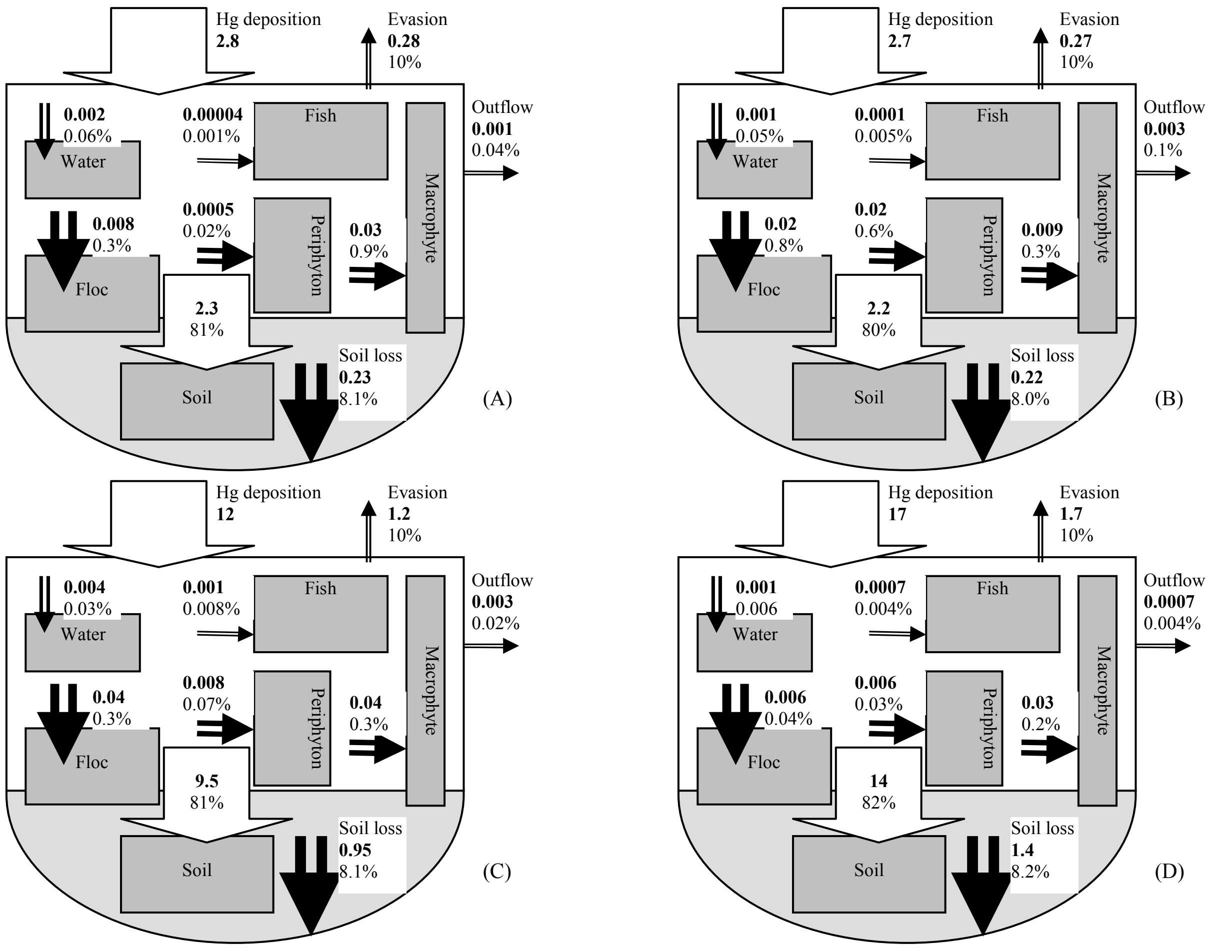

Fig. S2 Mass (kg) and fraction (%) of THg entering each ecosystem component or leaving the system through output pathways after being deposited into the Everglades in the dry season in 2005. (A) WCA 1; (B) WCA 2; (C) WCA 3; and (D) ENP.

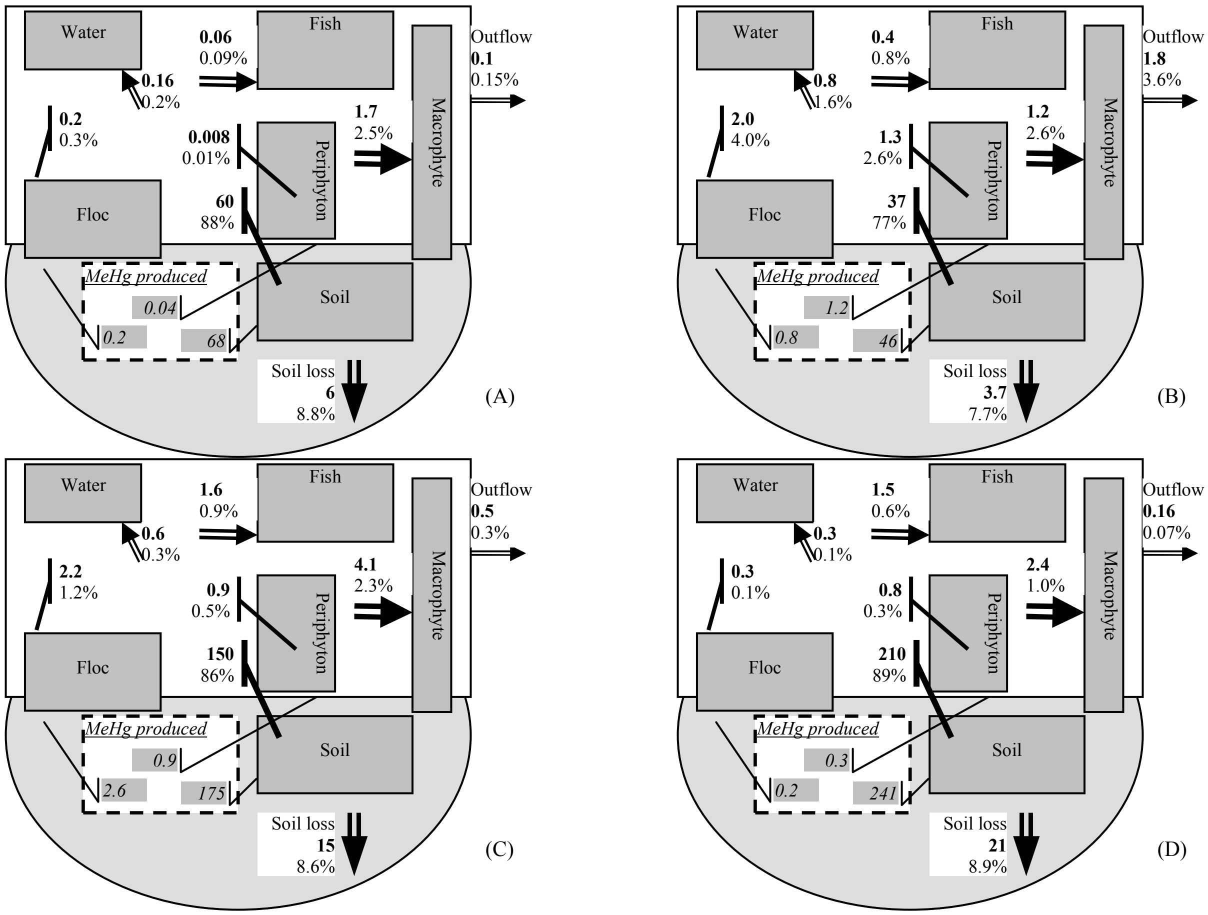

Fig. S3 Mass (g) and fraction (%) of MeHg entering each ecosystem component or leaving the system through output pathways after being produced (from seasonally deposited Hg) in the Everglades during the 2005 dry season. MeHg produced is shown in a rectangle with dashed line, with filled callouts linked to respective compartments. MeHg retained in soil, floc, or periphyton after redistribution is shown by line callouts. Arrows do not project the actual transport pathways. (A) WCA 1; (B) WCA 2; (C) WCA 3; and (D) ENP.

Table S10 Mass storage of THg and MeHg and associated uncertainty (relative standard error, %) in WCA 1, WCA 2, WCA 3, and ENP in the Everglades. THg and MeHg masses stored in each ecosystem component and corresponding uncertainties are estimated for the 2005 wet season sampling (November 2005).

		THg				MeHg			
		WCA 1	WCA 2	WCA 3	ENP	WCA 1	WCA 2	WCA 3	ENP
Water	Mass storage (kg)	1.3	1.6	4.2	2.6	0.19	0.22	0.43	0.30
	Uncertainty (%)	17	17	7.3	11	33	19	12	14
Soil	Mass storage (kg)	784	1015	4302	7381	9.8	3.8	18	40
	Uncertainty (%)	10	15	4.1	6.7	37	29	12	18
Floc	Mass storage (kg)	119	115	577	141	4.5	2.3	9.6	3.9
	Uncertainty (%)	42	37	30	31	57	57	30	30
Periphyton	Mass storage (kg)	0.067	0.64	2.2	41	0.0050	0.057	0.15	4.3
	Uncertainty (%)	25	25	10	14	28	28	11	19
Mosquitofish	Mass storage (kg)	0.028	0.077	0.89	0.96				
	Uncertainty (%)	17	25	11	11				

Table S11 Mass budget and corresponding uncertainty analysis of newly deposited Hg during the 2005 dry or wet season. Illustrated in the table are THg masses (kg) entering each ecosystem component after being deposited into the Everglades and associated uncertainties (U_{rT}, expresses as relative standard error, %) for these calculated THg masses.

	WCA 1		WCA 2		WCA 3		ENP	
	THg mass	U_{rT}	THg mass	U_{rT}	THg mass	U_{rT}	THg mass	U_{rT}
Dry season								
Water	0.002	35	0.001	41	0.004	23	0.001	47
Soil	2.3	44	2.2	43	9.5	25	14	51
Floc	0.008	39	0.021	45	0.036	25	0.006	53
Periphyton	0.001	39	0.015	49	0.008	29	0.005	53
Mosquitofish	0.00004	39	0.0001	47	0.001	26	0.001	52
Wet season								
Water	0.014	18	0.012	49	0.034	17	0.014	21
Soil	8.5	20	8.5	52	36	18	54	22
Floc	0.74	20	0.32	54	2.0	19	0.29	23
Periphyton	0.001	21	0.007	60	0.024	19	0.18	24
Mosquitofish	0.0003	21	0.001	53	0.007	20	0.004	23

Table S12 Mass budget and corresponding uncertainty analysis of MeHg produced from newly deposited Hg during the 2005 dry or wet season in the Everglades. Illustrated in the table are MeHg masses (g) entering each ecosystem component after being produced and associated uncertainties (U_{rT}, expresses as relative standard error, %) for these calculated MeHg masses.

	WCA1		WCA2		WCA3		ENP	
	MeHg mass	U_{rT}	MeHg mass	U_{rT}	MeHg mass	U_{rT}	MeHg mass	U_{rT}
Dry season								
Water	0.16	61	0.76	71	0.59	32	0.26	85
Soil	60	74	37	81	154	35	214	99
Floc	0.20	68	2.0	77	2.2	38	0.30	97
Periphyton	0.01	69	1.3	84	0.86	35	0.75	96
Mosquitofish	0.06	69	0.41	73	1.6	35	1.5	87
Wet season								
Water	2.0	55	1.9	79	3.3	25	1.3	26
Soil	117	70	20	95	124	28	273	31
Floc	28	63	7.1	88	37	30	6.4	30
Periphyton	0.09	58	1.0	88	1.7	28	16	29
Mosquitofish	0.44	58	0.73	81	8.1	27	5.0	28

Table S13 Structures used for calculating THg and MeHg input and output through water inflows and outflows for each management unit of the Everglades.

Management unit	Inflow Structures	Outflow Structures
WCA 1	G310, G301, G300, G251, S362, ACME1, ACME2	S10, G94, S39, G301
WCA 2	S10, S7, G335	S34, S38, S11
WCA 3	S11, S8, S9, S140, S150, S190	S12, S31, S142, S333, S343, S344, G69
ENP	S12, S18, S174, S332, S333-S334	C111, Taylor River Slough, Shark River Slough, Trout Creek, Taylor Creek, McCormick Creek

Table S14 Selected geochemical characteristics in the Everglades. DO: dissolved oxygen (mg/L); COND: conductivity (μmhos/cm); TURB: turbidity (NTU); TP: total phosphorus (μg/L for water and μg/g for soil and floc); TN: total nitrogen (mg/L for water and % for soil and floc); DOC: dissolved organic carbon (mg/L); AFDW: ash free dry weight (g/g).

		WCA 1		WCA 2		WCA 3		ENP	
		Median	Range	Median	Range	Median	Range	Median	Range
Dry season									
Surface Water	DO	3.3	0.85-4.9	5.6	2.5-9.1	3.9	0.98-10	4.8	1.9-8.2
	COND	196	125-374	722	526-1243	494	148-807	584	443-3696
	pH	6.1	5.3-7.0	7.5	7.1-8.1	7.3	6.5-8.1	7.3	7.0-7.5
	TURB	6.3	0.4-110	0.7	0.4-7.5	1.6	0.1-22	9.5	1.6-70
	TP	11	7.7-29	9.8	7.3-13.0	9.5	4.2-68	12	8.1-51
	TN	0.3	0.1-0.6	0.4	0.1-0.7	0.39	0.23-1.2	0.8	0.4-1.7
	DOC	25	20-33	31	21-50	20	11-29	22	18-49
Soil	pH	6.0	5.5-7.0	7.2	7.0-7.5	7.0	6.4-7.6	7.9	6.6-8.4
	TP	540	290-1100	390	240-1200	440	160-1400	280	57-690
	TN	3.2	2.6-3.9	2.8	1.4-3.3	3.1	1.2-4.7	1.1	0.21-3.5
	AFDW	0.07	0.04-0.12	0.13	0.11-0.68	0.16	0.08-0.65	0.77	0.13-0.95
Floc	TP	860	400-2000	620	310-860	560	140-1200	440	240-580
	TN	3.8	3.0-4.7	3.0	2.4-3.1	3.2	1.1-4.5	2.8	1.8-3.6
	AFDW	0.08	0.05-0.11	0.16	0.11-0.44	0.12	0.07-0.68	0.25	0.16-0.59
Wet season									
Surface Water	DO	3.1	1.1-7.4	4.2	1.7-8.0	4.3	1.2-410	6.7	1.5-85
	COND	152	107-945	1041	444-1423	445	321-760	410	72-642
	pH	6.5	5.7-7.1	7.4	7.1-7.9	7.2	6.7-7.7	7.5	6.9-8.0
	TURB	3.1	0.6-12	0.8	0.5-3.3	0.7	0-2.3	1	0-14
	TP	9.3	7.3-95	9.2	6.4-42	7.2	3.7-21	6.1	4.0-20
	TN	0.75	0.35-1.3	1.1	0.81-1.6	0.58	0.19-1.0	0.36	0.15-1.0
	DOC	19	15-33	31	18-45	16	11-25	12	4.6-20
Soil	pH	5.9	5.3-7.0	7.1	6.6-7.3	6.7	6.1-7.3	7.1	6.4-7.5
	TP	395	260-1400	515	110-960	435	200-840	305	120-730
	TN	3.1	2.4-4.4	3.3	0.58-3.7	3.3	0.94-4.4	1.3	0.46-3.8
	AFDW	0.94	0.56-0.96	0.85	0.21-0.90	0.83	0.18-0.95	0.25	0.09-0.91
Floc	TP	605	390-1300	655	400-1800	610	160-1300	370	190-1600
	TN	4.3	3.4-4.5	3.1	2.5-4.3	3.4	1.3-4.4	2.9	1.2-3.7
	AFDW	0.90	0.82-0.95	0.78	0.66-0.87	0.87	0.30-0.91	0.68	0.30-0.90

Table S15 Mercury evasion rates in the Florida Everglades reported in the literature. ENR: Everglades nutrients removal area; WCA: Water conservation area.

Site	Period	Time	Hg evasion	Number of Measurements	Reference
Direct measurement of daily mercury fluxes over water surfaces (ng/m^2/h)					
ENR overall	Year	Day+Night	1.2	459	(*11,13*)
ENR overall	Year	Day	2.7	212	(*11,12*)
ENR overall	Year	Night	0.2	247	(*11*)
WCA-2	Summer	Day	1.8	72	(*11*)
WCA-3	Summer	Day	2.7	5	(*11*)
ENR	Spring	Day	2.1	~50	(*11,35*)
ENR	Summer	Day	3.4	31	(*11*)
ENR	Fall	Day	1.3	59	(*11*)
ENR	Winter	Day	2.8	105	(*11*)
Estimated annual mercury evasion rate over water surfaces (µg/m^2/yr)					
Estimation from measurement of daily mercury fluxes					
ENR	Year	Day+Night	3.0	-	(*36*)
ENR	Year	Day+Night	2.0-6.0	-	(*35*)
Estimation from measurement of dissolved gaseous mercury					
WCA-2	Year	Day+Night	2.2	~60	(*8*)
Modeling results from Everglades Mercury Cycling Model (E-MCM)					
ENR	Year	Day+Night	2.0	-	(*46*)

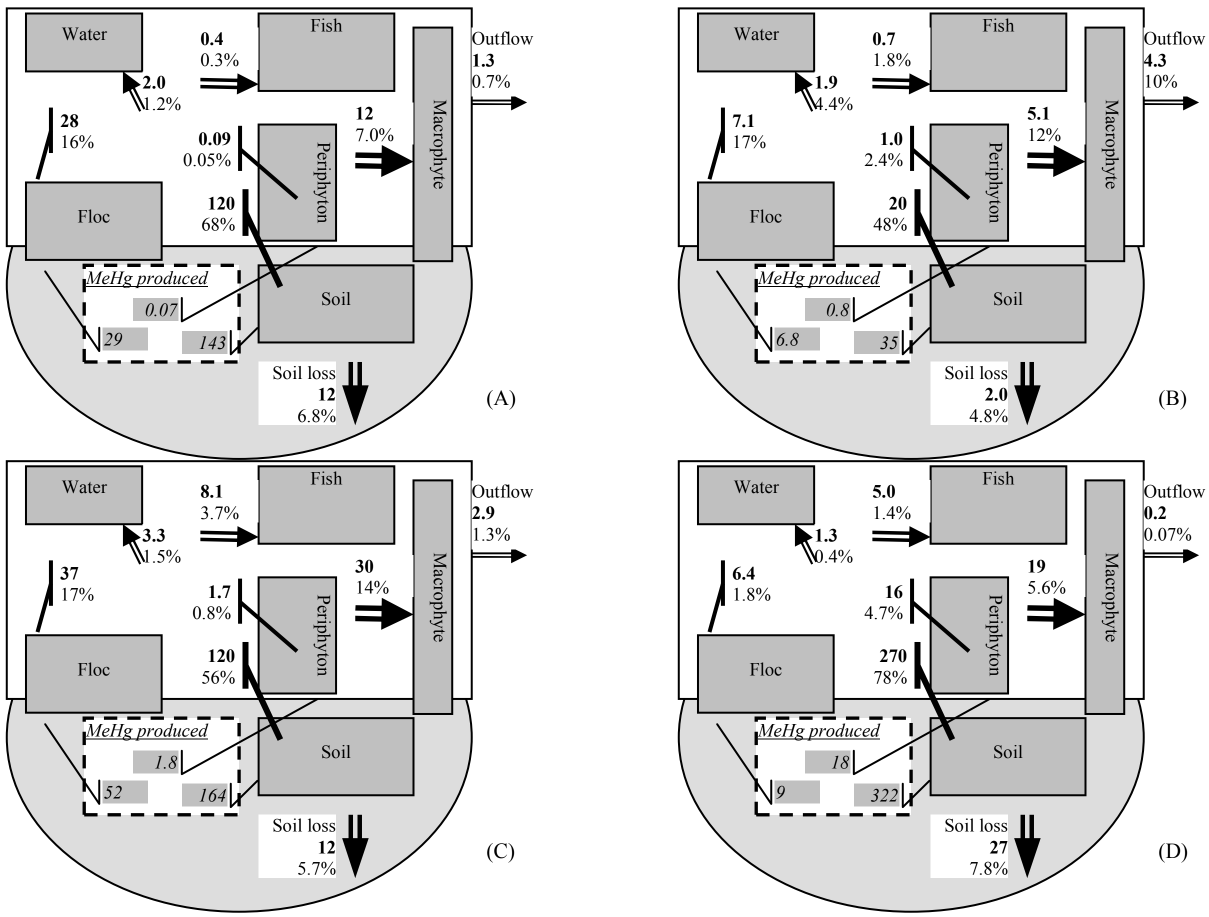

Fig. 4 Mass (g) and fraction (%) of MeHg entering each ecosystem component or leaving the system through output pathways after being produced (from seasonally deposited Hg) in the Everglades during the 2005 wet season. MeHg produced is shown in a rectangle with dashed line, with filled callouts linked to respective compartments. MeHg retained in soil, floc, or periphyton after redistribution is shown by line callouts. 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.

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