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Spatial Variability in Mercury Cycling and Relevant Biogeochemical Controls in the Florida Everglades

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Spatial patterns in mercury cycling and bioaccumulation at the landscape level in the Everglades were investigated by collecting and analyzing multimedia samples for mercury species and biogeochemical characteristics from 228 randomly located stations. Higher total mercury (THg) in environmental compartments (surface water, soil, flocculent detrital material (floc), and periphyton) generally occurred in the northern and central Everglades, but higher THg in water and periphyton in the Everglades National Park was an exception. Multiple biogeochemical characteristics, such as surface water dissolved organic matter (DOC_{SW}), pH, chloride, and compositional properties of solid compartments (soil and floc), were identified to be important factors controlling THg distribution. Methylmercury (MeHg) was also higher in the northern Everglades for water, soil, and floc, but not for periphyton. Higher mosquitofish THg and bioaccumulation factor were observed in the central and southern Everglades, partially in accordance with periphyton MeHg distribution, but not in the "hot spot" areas of water, soil, or floc MeHg. The discrepancy in mercury bioaccumulation and mercury distribution in environmental compartments suggests that in addition to MeHg production, biogeochemical controls that make MeHg available to aquatic organisms, such as DOC_{SW} and compositional properties of soil and floc, are important in mercury bioaccumulation.

Introduction

Mercury (Hg) remains one of the major water quality concerns in the Florida Everglades. Efforts have been made to investigate the source (*1*−*3*), transport (*4*−*7*), transformation (especially methylation/demethylation) (*8*−*13*), and bioaccumulation (*14*−*16*) of Hg in the Everglades. However, biogeochemical controls on Hg cycling and bioaccumulation in the Everglades are not fully understood, in particular at the landscape level.

The Everglades is a subtropical freshwater wetland ecosystem, currently with four management units that span an area of over 5,500 km^2: Loxahatchee National Wildlife Refuge (LNWR), the Water Conservation Areas (WCA 2 and 3), and Everglades National Park (ENP) from north to south. In this large area, dikes, levees, roadways, urban development, and other landscape features alter water flow, habitat, and nutrient loading, resulting in spatial variations in corresponding ecological conditions (*17*). A variety of soil types are found in the Everglades, from organic peat with high organic matter in northern and central Everglades to calcitic mud (marl) with low organic matter in southern Everglades. The organic matter content in Everglades soil can range from <1 to 97%, depending on the geographic location (*17*). Nutrient loading from the northern Everglades Agricultural Area (EAA) and urban areas has affected the ecological conditions in the downstream WCAs and ENP in different ways. Nutrient (nitrogen and phosphorus), sulfate, and dissolved organic matter (DOC) concentrations generally bear a decreasing gradient from north to south (*17*). The progressive eutrophic impacts include altered periphyton communities, loss of water column dissolved oxygen, increased soil phosphorus content, and conversion of wet prairie and sawgrass plant communities to cattail. These collective changes impact the structure and function of the aquatic ecosystem.

Because of the spatial variability in ecological conditions, distinct spatial patterns can be expected for Hg distribution and cycling in the Everglades (*17*), which have been observed for localized areas (e.g., WCAs) (*7, 18, 19*) but remain unclear on a large scale (e.g., throughout the entire Everglades ecosystem) (*20*). Understanding large-scale and landscape patterns of Hg cycling is critical not only for revealing biogeochemical processes that are related to Hg cycling and bioaccumulation, but also for managing the Everglades ecosystem to achieve restoration goals of the Comprehensive Everglades Restoration Program (CERP). Our objective in this paper was to investigate biogeochemical factors that control Hg distribution and cycling by analyzing the spatial patterns of Hg distribution and related biogeochemical characteristics in the Everglades. The data were obtained during the U.S. Environmental Protection Agency (EPA) Everglades Regional Environmental Monitoring and Assessment Program (R-EMAP) phase III sampling events. The R-EMAP program sampled the entire Everglades freshwater marsh, excluding tree islands and shrubby sawgrass strands, by using a probability sampling design (*17*). This ecosystem-wide sampling design allowed us to explore the spatial patterns for Hg and biogeochemical parameters at the landscape level.

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Materials And Methods

Sampling and Sample Analysis. The R-EMAP phase III sampling was conducted in 2005 at 228 (109 in May and 119 in November) randomly selected stations distributed throughout the entire Everglades freshwater marsh (Figure S1 in Supporting Information (SI)). The media sampled at each site included surface water (SW), pore water (PW), soil (SD), flocculent detrital material (floc, FC), periphyton (PE), and Eastern mosquitofish (*Gambusia holbrooki*) (FS). These samples were measured for Hg (THg and MeHg) as well as a long list of biogeochemical parameters (see SI). The concentration of a parameter in a specific compartment is expressed as an abbreviation of that parameter labeled with the compartment name throughout the text below, e.g., DOC_{SW} for DOC in surface water (see Table S1 for the list of acronyms used). The detailed sampling and analytical procedures for each matrix can be found in the SI and related references (*17, 21−23*).

Data Processing. We first examined the spatial patterns of Hg and ancillary parameters in each environmental compartment using contour maps produced by Surfer (Version 8, Golden Software, Golden, CO). These contour maps were generated by a kriging method in which a value for each node of the grid was estimated using the linear variogram model (no nugget effect).

We then defined two parameters to interpret the observed Hg spatial patterns. The distribution ratio (R) of Hg between soil, floc, or periphyton and surface water was calculated as R (L/kg) = Hg in soil, floc, or periphyton (dry weight based)/Hg in surface water (filtered through a 105-μm screen). R was used to describe the compartmentalization process (distribution and transport between water and other environmental compartments) of Hg. The MeHg fraction (f = MeHg/THg) was used to represent the capacity for MeHg production based on previous studies showing that f is a valid indicator of MeHg production, at least in the WCA areas (*10, 24*). Detailed information on defining R and f can be found in the SI. Multiple regressions between MeHg concentration and parameters f and R were carried out with SPSS (Version 12 for Windows, SPSS Inc., Chicago, IL) for each environmental compartment to explore the explanatory effect of MeHg production and compartmentalization on MeHg distribution.

We further identified biogeochemical parameters affecting the compartmentalization of Hg in the Everglades by conducting multiple regressions between R and selected parameters, for both THg and MeHg. Screening biogeochemical parameters included in the regression model was done by comparing the differences in spatial patterns of R and biogeochemical parameters using contour maps. These comparisons were done by overlaying two contour maps and examining the differences in distribution of two variables on the contour map. All correlation and regression analyses were conducted on log-transformed data.

Results And Discussion

THg Spatial Patterns. The contour maps of THg and MeHg distribution in each environmental compartment (surface water, soil, floc, and periphyton) and mosquitofish are illustrated in Figures S2−S4. As can be seen from Figure S2, the spatial patterns of THg distribution in the Everglades are complicated, varying among environmental compartments and seasons. There is no uniform "hot spot" area (area of high concentration) where THg concentrations in all environmental compartments are high. The general pattern is that the "hot spot" areas with high THg occur in the northern Everglades (LNWR and WCAs), but surface water and periphyton in ENP are exceptions. High water THg occurred in WCA 2 and LNWR during both seasons and in the central part of the ENP during the dry season. Soil and floc have similar spatial patterns of THg distribution, particularly during the wet season when three "hot spot" areas occurred in LNWR, WCA 2, and southwestern WCA 3 at approximately the same locations for both compartments. Contrary to THg in soil and floc, high periphyton THg was observed in both northern and southern Everglades.

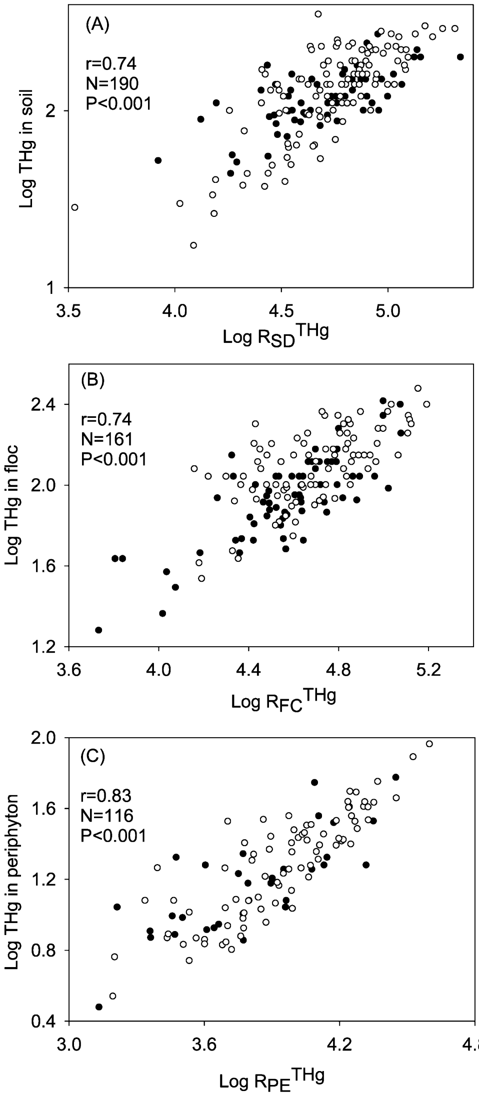

FIGURE 1. Correlations between distribution ratios (*R*) and THg concentrations in (A) soil, (B) floc, and (C) periphyton in the Everglades. Filled and open circles are data obtained during the 2005 dry and wet season, respectively.

The spatial variations of THg must be related to spatial differences in Hg compartmentalization processes which transport Hg from one environmental compartment to another and thus redistribute Hg among different compartments. Strong positive correlations were observed between R and the concentration of THg for soil, floc, and periphyton (Figure 1), illustrating the effectiveness of R in describing Hg retention by a compartment. Any biogeochemical parameter that affects R would also influence THg concentrations in a compartment.

The results of multiple regression analysis between R of THg and biogeochemical characteristics are listed in Table 1. The high r^2 (0.63−0.82) and low P (<0.001) values for these regressions suggest that the compartmentalization of THg in the Everglades was influenced by multiple factors, including surface water characteristics such as DOC_{SW}, Cl_{SW}, pH_{SW}, $TURB_{SW}$, and $COND_{SW}$, and compositional characteristics of solid compartments such as $AFDW_{SD}$, MC_{SD}, $AFDW_{FC}$, and

TABLE 1. Multiple Regression Results for the THg Distribution Ratio (*R*) with Selected Biogeochemical Parameters in the Everglades[a]

dependent	independent	standardized coefficient (β)	r^2	significance
R_{SD}^{THg}	DOC_{SW}	−0.63	0.63	$P < 0.001$
	$AFDW_{SD}$	0.74		
	MC_{SD}	0.47		
	Cl_{SW}	−0.95		
	pH_{SW}	−0.66		
	$COND_{SW}$	1.3		
R_{FC}^{THg}	DOC_{SW}	−0.83	0.82	$P < 0.001$
	$AFDW_{FC}$	0.62		
	Cl_{SW}	−0.14		
	$TURB_{SW}$	0.32		
	$COND_{SW}$	0.38		
	$CHLA_{FC}$	−0.42		
R_{PE}^{THg}	DOC_{SW}	−0.95	0.63	$P < 0.001$
	Cl_{SW}	1.6		
	$COND_{SW}$	−1.2		

[a] The magnitudes of the standardized coefficient (β) can be used to approximately evaluate the relative importance of the biogeochemical parameters. A minus sign before β indicates a negative correlation between the dependent and the independent variables. See SI for definition of acronyms.

$CHLA_{FC}$. Different parameters played different roles, depending on the compartment. For soil, in addition to $AFDW_{SD}$ and MC_{SD}, DOC_{SW}, Cl_{SW}, pH_{SW}, and $COND_{SW}$ were important parameters affecting the compartmentalization of THg between water and soil. These six parameters could explain 63% of the variance of R_{SD}^{THg}. For floc, DOC_{SW}, Cl_{SW}, $TURB_{SW}$, and $COND_{SW}$, together with $AFDW_{FC}$ and $CHLA_{FC}$ explained 82% of the variance of R_{FC}^{THg}. Since compositional characteristics were not determined for periphyton, only surface water parameters were included in the regression model of R_{PE}^{THg} versus biogeochemical parameters. Three surface water parameters, DOC_{SW}, Cl_{SW}, and $COND_{SW}$, had a significant effect on R_{PE}^{THg} variation, accounting for 63% of the variance. The standardized coefficients (β) in Table 1 represent the independent contributions of each biogeochemical parameter to the prediction of *R*. For each equation, they are directly comparable to one another and can be used to approximately evaluate the relative importance of the biogeochemical parameters included in that equation. A larger β associated with a parameter (regardless of sign) suggests that parameter contributes more to the variation of the corresponding *R*. A negative β means that *R* decreases with increase in that parameter. DOC_{SW}, Cl_{SW}, and $COND_{SW}$ appeared in all three regression models, suggesting that they are critical surface water parameters correlated with the compartmentalization processes of THg in the Everglades. Under certain circumstances, other surface water parameters may also play a role on THg compartmentalization, as revealed by the effect of pH_{SW} on THg compartmentalization between water and soil and $TURB_{SW}$ on floc.

It is not surprising that DOC_{SW} played an important role in the compartmentalization of THg between water and other compartments in the Everglades. DOC has a strong binding affinity for Hg and Hg is known to be present mainly as complexes with DOC in water (*4, 25*−*31*). We have observed significant correlations ($P < 0.001$) between surface water THg and DOC in the Everglades (*22*), in accordance with many other studies where THg concentrations are positively correlated with DOC in surface water of lakes (*32*) and rivers (*33*). Everglades water is typically rich in DOC, ranging from 5 to 50 mg/L (with an average of about 20 mg/L), according to our monitoring results (*21*). The DOC in Everglades water would form complexes with Hg and thus retain Hg in the water column, hindering Hg from being redistributed into other compartments (soil, floc, or periphyton). This could explain our observed negative correlations between DOC_{SW} and *R*.

The influence of Cl_{SW} on THg compartmentalization could be ascribed to the competition of Cl^- for binding sites of solid particles and the formation of mercury chloride ($HgCl_2$), which both reduce Hg adsorption onto particles and soil and retains Hg in the water column. Previous studies have shown that increasing Cl^- from 10^{-5} to 10^{-4} M sharply reduced Hg adsorption on clay particles (*34*). The concentrations of Cl^- in the Everglades (ranging from 20 to 1000 mg/L, approximately in order of 10^{-4} to 10^{-2} M) are thus expected to result in less Hg being redistributed into soil and floc, as revealed by the negative correlations between Cl_{SW} and *R*. The generally high Cl^- concentrations in the Everglades favor the formation of uncharged $HgCl_2$ (*24*). For periphyton, the formation of neutral $HgCl_2$ species would increase the bioavailability of Hg for algal uptake since this uptake is likely a passive diffusion process (*24*). Therefore a positive correlation between Cl_{SW} and *R* was observed for periphyton, since periphyton is rich in living algae that could take up Hg from the water column. In agreement with our results, the facilitation of algae uptake of Hg by chloride has been previously reported for marine and freshwater algal populations (*35, 36*).

In addition to surface water parameters, effects of the compositional characteristics of solid compartments (soil and floc) on THg compartmentalization are also expected. For example, $AFDW_{SD}$ and MC_{SD} were positively correlated with *R* for Everglades soil. $AFDW_{SD}$ and MC_{SD} measured two important components of the soil compartment. $AFDW_{SD}$ indicated the content of soil organic matter while MC_{SD} (expressed as % ash) measured the content of soil mineral substances including Fe and Mn oxides. Both components are known to have a strong affinity to bind Hg and thus result in the redistribution of Hg from the water column to solid compartments. Therefore, $AFDW_{SD}$ and MC_{SD} were positively correlated with *R* for Everglades soil, indicating more Hg would be entrapped in the soil compartment with increasing $AFDW_{SD}$ and MC_{SD}.

MeHg Spatial Patterns. The contour maps of MeHg in each compartment (water, soil, floc, and periphyton) are illustrated in Figure S3. For water, soil, and floc, the MeHg "hot spot" areas usually occurred in the northern Everglades, including LNWR, WCA 2, and the north part of WCA 3. Similar to THg, the locations of the "hot spot" areas for one compartment were not duplicated in other compartments. The spatial patterns for periphyton were significantly different from the other three compartments. The northern Everglades was observed to have moderately high periphyton MeHg concentrations, while a "hot spot" area with high MeHg occurred in ENP during both seasons and in the northern WCA 3 during the wet season.

The spatial patterns of MeHg are related to two processes: MeHg production and the compartmentalization of MeHg after production. Comparing Figure S2 with Figure S3, we found that the MeHg "hot spot" areas did not always match THg "hot spots", indicating spatial variability in MeHg production and/or compartmentalization in the Everglades. Since *f* and *R* characterize MeHg production and compartmentalization processes, respectively, the spatial variations in MeHg concentrations should be accounted for by *f* and *R*. The results of multiple regressions between MeHg concentration and *f* and *R* confirmed the effect of MeHg production and compartmentalization on MeHg distribution, as evidenced by the strong correlation between MeHg and *f* and *R* (Table 2). Together, *f* and *R* explained 83, 82, and 65%

TABLE 2. Multiple Regression Results for MeHg Concentration versus MeHg Fraction (*f*) and Distribution Ratio (*R*) for Everglades Surface Water, Soil, Floc, and Periphyton[a]

dependent	independent	standardized coefficient (β)	r^2	significance
$MeHg_{SW}$	f_{SD}^{MeHg}	0.41	0.78	$P < 0.001$
	R_{SD}^{MeHg}	−0.57		
	f_{FC}^{MeHg}	0.33		
	R_{FC}^{MeHg}	−0.36		
	f_{PE}^{MeHg}	0.17		
	R_{PE}^{MeHg}	−0.083		
$MeHg_{SD}$	f_{SD}^{MeHg}	0.72	0.83	$P < 0.001$
	R_{SD}^{MeHg}	0.23		
$MeHg_{FC}$	f_{FC}^{MeHg}	0.66	0.82	$P < 0.001$
	R_{FC}^{MeHg}	0.36		
$MeHg_{PE}$	f_{PE}^{MeHg}	0.61	0.65	$P < 0.001$
	R_{PE}^{MeHg}	0.33		

[a] As MeHg in the surface water was assumed to be transported from soil, floc, and periphyton, $MeHg_{SW}$ was regressed against *f* and *R* of these three compartments. See SI for definition of acronyms.

TABLE 3. Multiple Regression Results for the MeHg Distribution Ratio (*R*) with Selected Biogeochemical Parameters in the Everglades[a]

dependent	independent	standardized coefficient (β)	r^2	significance
R_{SD}^{MeHg}	DOC_{SW}	−0.11	0.46	$P < 0.001$
	$AFDW_{SD}$	0.092		
	MC_{SD}	0.12		
	Eh_{PW}	0.34		
	pH_{SW}	−0.46		
R_{FC}^{MeHg}	DOC_{SW}	−0.42	0.52	$P < 0.005$
	$AFDW_{FC}$	0.22		
	pH_{SW}	−0.39		
	DO_{SW}	0.51		
	$CHLA_{FC}$	−0.16		
R_{PE}^{MeHg}	DOC_{SW}	−0.32	0.66	$P < 0.005$
	Cl_{SW}	2.2		
	$COND_{SW}$	−2.5		

[a] See SI for definition of acronyms.

of the variance in MeHg concentration for soil, floc, and periphyton, respectively. The standardized coefficients were higher for *f* than for *R*, suggesting that MeHg production, in comparison to MeHg compartmentalization, is more important in determining the spatial patterns of MeHg concentrations. For surface water, $MeHg_{SW}$ strongly correlated with *f* and *R* of MeHg in soil, floc, and periphyton, with 78% of variance explained. This strong correlation indicates that MeHg production in these compartments and subsequent compartmentalization processes are factors controlling the spatial variations of water MeHg. This result agrees with previous studies that suggested that MeHg in surface water is more likely to be transported from soil, floc, and periphyton than to be produced in situ (*22, 37*). Soil and floc played more important roles in determining spatial patterns of water MeHg, in comparison to periphyton, as indicated by higher β for the first two compartments. The similar values of β for *f* and *R* (regardless of sign) suggest that production in solid compartments and compartmentalization are equally important in determining spatial variation of water MeHg, which differed from the cases for solid MeHg.

As with THg, multiple biogeochemical factors were identified to have significant effects on MeHg compartmentalization and played varying roles depending on compartment (Table 3). It should be noted that the biogeochemical parameters influencing MeHg compartmentalization mentioned below also controlled MeHg production, as identified by multiple regression. These results were not shown as we focused the current study on Hg compartmentalization. Besides surface water parameters (DOC_{SW} and pH_{SW}) and soil compositional characteristics ($AFDW_{SD}$ and MC_{SD}), Eh_{PW} was also partially correlated with the spatial variation in MeHg retention, with 46% of the variance of R_{SD}^{MeHg} explained by these five parameters. For floc, 52% of the variance of R_{FC}^{MeHg} was explained by DOC_{SW}, pH_{SW}, DO_{SW}, $AFDW_{FC}$, and $CHLA_{FC}$. Three surface water parameters, DOC_{SW}, Cl_{SW}, and $COND_{SW}$, explained 66% of the variance of R_{PE}^{MeHg}. Surface water DOC was again a critical parameter in determining the spatial patterns of MeHg, as evidenced by its appearance in all three regression models.

The effect of DOC_{SW} on MeHg compartmentalization could result from strong complexation of DOC with MeHg, as is the case of THg. DOC strongly binds both inorganic Hg and MeHg. Our previous studies have shown that DOC could have stronger binding affinity with MeHg than with THg (*22, 38*). The complexation of DOC with MeHg would retain the latter in the water column, resulting in the negative correlation between DOC_{SW} and *R* for all three compartments.

It is expected that other surface water chemistry parameters, e.g., pH_{SW}, played a role in determining the spatial patterns of MeHg in the Everglades. pH can affect both MeHg production and compartmentalization of MeHg after production. It is generally accepted that MeHg production increases with decreasing pH (*30*). Meanwhile, decreasing pH also could result in decreasing association of MeHg with DOM and inorganic colloids present in surface water (*24, 30*). Since most MeHg present in Everglades water is bound by DOM and/or inorganic colloids, this decreasing association would thus lead to an increasing *R* for MeHg, as evidenced by the inverse relationship between pH and R_{SD}^{MeHg} or R_{FC}^{MeHg} (Table 3).

Hg Bioaccumulation. The "hot spot" areas for mosquitofish THg occurred in ENP during the dry season and in central WCA 3 and ENP during the wet season (Figure S4). The "hot spot" areas for bioaccumulation factor (BAF) also occurred in central WCA 3 and ENP during the wet season, but did not completely match the "hot spot" areas for mosquitofish THg. In the dry season, BAF was high in central WCA 3, again inconsistent with mosquitofish THg.

Comparison of Figures S3 and S4 shows that during the dry season extremely high mosquitofish THg occurred in a low-MeHg area in ENP where MeHg in all environmental compartments (surface water, soil, floc, and periphyton) were low, rather than in any of the "hot spot" areas of these compartments, e.g., WCA 2 for SW or northern WCA 3 for floc. During the wet season, the "hot spot" for mosquitofish THg in ENP coincides with that for periphyton MeHg, while the one in WCA 3 occurred in a moderate MeHg area. The occurrence of high mosquitofish THg and BAF in the central and southern Everglades, rather than in the northern Everglades where MeHg "hot spots" occurred for environmental compartments, suggests that strong Hg bioaccumulation requires not only high MeHg concentration, but also an appropriate combination of biogeochemical controls that make MeHg available to aquatic organisms.

Correlation analysis revealed that mosquitofish THg was significantly correlated with periphyton MeHg ($r = 0.465$, $P < 0.001$) (Figure 2), but not with water, soil or floc MeHg (data not shown). This positive correlation is expected given the fact that periphyton can be consumed by mosquitofish as a direct food source or, if not consumed directly, is integral

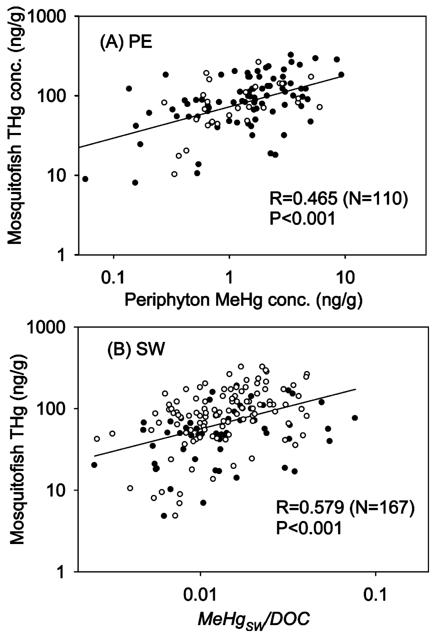

FIGURE 2. Correlation between mosquitofish THg and (A) periphyton MeHg, and (B) DOC-normalized surface water MeHg. Filled and open circles are data obtained in the dry and wet season, respectively.

to their food web (*15, 39*). This correlation could explain why the "hot spot" areas of mosquitofish THg and periphyton MeHg in ENP occurred at the same location during the wet season. However, despite the lack of direct correlations with mosquitofish THg, water MeHg might still contribute to Hg bioaccumulation in mosquitofish; in addition to a food web source, mosquitofish could accumulate Hg via direct uptake from the water. We normalized water MeHg to DOC concentration to provide a measure of bioavailable dissolved MeHg in Everglades surface water. We observed a strong positive correlation between mosquitofish THg and DOC-normalized water MeHg (Figure 2), indicating that water MeHg contributes to Hg in mosquitofish, but is regulated by DOC.

The results of multiple regression of BAF versus biogeochemical characteristics revealed that DOC_{SW}, pH_{SD}, $H2S_{PW}$, $AFDW_{FC}$, and MC_{FC} play significant roles in Hg bioaccumulation, with 57% of variance in BAF explained ($P < 0.001$). The standardized coefficients were greatest for $AFDW_{FC}$ and MC_{FC} (0.99 and 1.5, respectively) and lowest for $H2S_{PW}$ (−0.30), with intermediate negative values for DOC_{SW} and pH_{SD} (−0.56 and −0.62, respectively).

Floc appeared to play an important role in Hg bioaccumulation, despite a lack of direct correlation between floc MeHg and mosquitofish THg. In fact, floc, as nonconsolidated biogenic detrital matter, is an important food web component for Everglades invertebrates and fish (*40*). The lack of correlation between floc MeHg and mosquitofish THg suggests that floc is not a direct diet for mosquitofish, at least in some areas of the Everglades. However, this cannot completely prevent floc MeHg from being transferred into mosquitofish. The Everglades food web is complicated and varies with spatial location and season. Mosquitofish have opportunistic food habits and will feed on other food sources, such as invertebrates, in addition to periphyton (*15*). These invertebrates could feed on floc and subsequently transfer floc MeHg into the mosquitofish food web. The positive correlation between BAF and $AFDW_{FC}$ or MC_{FC} could be the result of this indirect transfer of floc MeHg to mosquitofish, since higher organic matter or mineral substance content would retain more MeHg in floc.

The inclusion of DOC_{SW} in the multiple regression model indicates again that DOC is important in regulating Hg cycling and bioaccumulation. The complexation of MeHg by DOC would likely limit the bioavailability of MeHg for bioaccumulation, as evidenced by a number of studies (*28, 31, 32, 41−44*). Additionally, DOC complexation could lower MeHg partitioned onto the solid material (e.g., floc and periphyton) that mosquitofish eats and retain MeHg in water. Since BAF has water MeHg in the denominator, DOC could lower BAF by increasing water MeHg. We also observed previously a significant negative correlation between BAF and DOC_{SW} in the Everglades ($r = -0.639$, $p < 0.001$) (*22*). Thus, the minus sign before DOC_{SW} in the multiple regression model agrees with previous studies.

Discrepancies were clearly observed for spatial distributions between THg and MeHg as well as between environmental Hg and mosquitofish Hg in the Everglades, indicating that biogeochemical factors and processes are important in controlling Hg compartmentalization and bioaccumulation. Multiple regression analysis revealed that multiple biogeochemical characteristics, including both surface water chemistry and solid (e.g., soil and floc) composition, simultaneously played a role in determining spatial variability in Hg distribution, compartmentalization, and bioaccumulation. These results suggest that for the Everglades, which is a spatially heterogeneous ecosystem with dramatically varying ecological conditions, a uniform single-variable model cannot capture the complexity of the influences of biogeochemical characteristics on the compartmentalization of Hg in the Everglades. Therefore, for such complex ecosystems as the Everglades, direct correlation of Hg with a single biogeochemical parameter should be conducted with caution, since the effects of other factors need to be considered simultaneously.

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Supporting Information Available

A table listing definitions of acronyms used for presenting Hg and biogeochemical parameters, a sampling map, contour maps of THg, MeHg, and BAF spatial distribution, and sampling and analysis procedures. This material is available free of charge via the Internet at http://pubs.acs.org.

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