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Wading bird guano contributes to Hg accumulation in tree island soils in the Florida Everglades



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ABSTRACT

Tree islands are habitat for wading birds and a characteristic landscape feature in the Everglades. A total of 93 surface soil and 3 soil core samples were collected from 7 degraded/ghost and 34 live tree islands. The mean Hg concentration in surface soils of ghost tree islands was low and similar to marsh soil. For live tree islands, Hg concentrations in the surface head region were considerably greater than those in mid and tail region, and marsh soils. Hg concentrations in bird guano ($286 \mu\text{g kg}^{-1}$) were significantly higher than those in mammal droppings ($105 \mu\text{g kg}^{-1}$) and plant leaves ($53 \mu\text{g kg}^{-1}$). In addition, Hg concentrations and $\delta^{15}\text{N}$ values displayed positive correlation in soils influenced by guano. During 1998–2010, estimated annual Hg deposition by guano was $148 \mu\text{g m}^{-2} \text{yr}^{-1}$ and ~ 8 times the atmospheric deposition.

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1. Introduction

The Florida Everglades is the largest freshwater wetland in the continental United States. High Hg concentrations in fish and wildlife have been of particular concern (Axelrad et al., 2006; Rumbold et al., 2007). The Hg concentrations in the majority of largemouth bass (*Micropterus salmoides*) from the Everglades protection area have exceeded the USEPA methylmercury (MeHg) criterion of 0.3 mg kg^{-1} in fish tissue since 1998 (Gabriel et al., 2010). The Hg in the Everglades originates from local, regional and global sources (Axelrad et al., 2006). Atmospheric deposition, primarily inorganic Hg, contributes over 95% of Hg input to the Everglades (Axelrad et al., 2006; Liu et al., 2008a). Deposition rates for atmospheric Hg vary between 15 and $25 \mu\text{g m}^{-2} \text{yr}^{-1}$ in southern Florida (NADP, 2007). Soil is the largest sink for Hg, with $\sim 80\%$ of seasonally deposited Hg being entrapped in soils (Liu et al., 2009, 2010).

Tree islands are a characteristic landscape feature in the Everglades. The total area of tree islands varies from 14% in the northern

Everglades to 3.8% in the central Everglades (Brandt et al., 2000; Wetzel, 2003; Wetzel et al., 2011). Tree islands in the central and southern Everglades typically consists of head, middle and tail regions (Mason and Valk, 2002). The head is the most elevated part of the island, containing the largest trees in terms of both height and trunk diameter. The middle and tail region of tree island usually are vegetated by low stature trees and a dense herbaceous understory including shrubs, forbs ferns, shrubs and sawgrass (Mason and Valk, 2002; Sah, 2004). In addition, most of wading bird nests are located in the head region of tree islands due to their favorable vegetation communities (Orem et al., 2003).

The Everglades includes four management units: Water Conservation Areas (WCA-1, WCA-2 and WCA-3) and the Everglades National Park (ENP). Tree islands in WCA-2A and 3A have suffered the most change and lost their elevation and woody vegetation due to persistent and deep flooding, and they are known as “ghost” islands (Hofmockel et al., 2008). Ghost tree islands have disappeared from the landscape and can be located only as scars in aerial photographs. The live tree islands are not degraded and have healthy vegetation community. Most of the live tree islands are located in WCA-1, 3A, 3B and ENP. Live tree islands have more woody vegetation and wading bird nests than ghost tree islands, especially in their head regions (Sklar et al., 2011).

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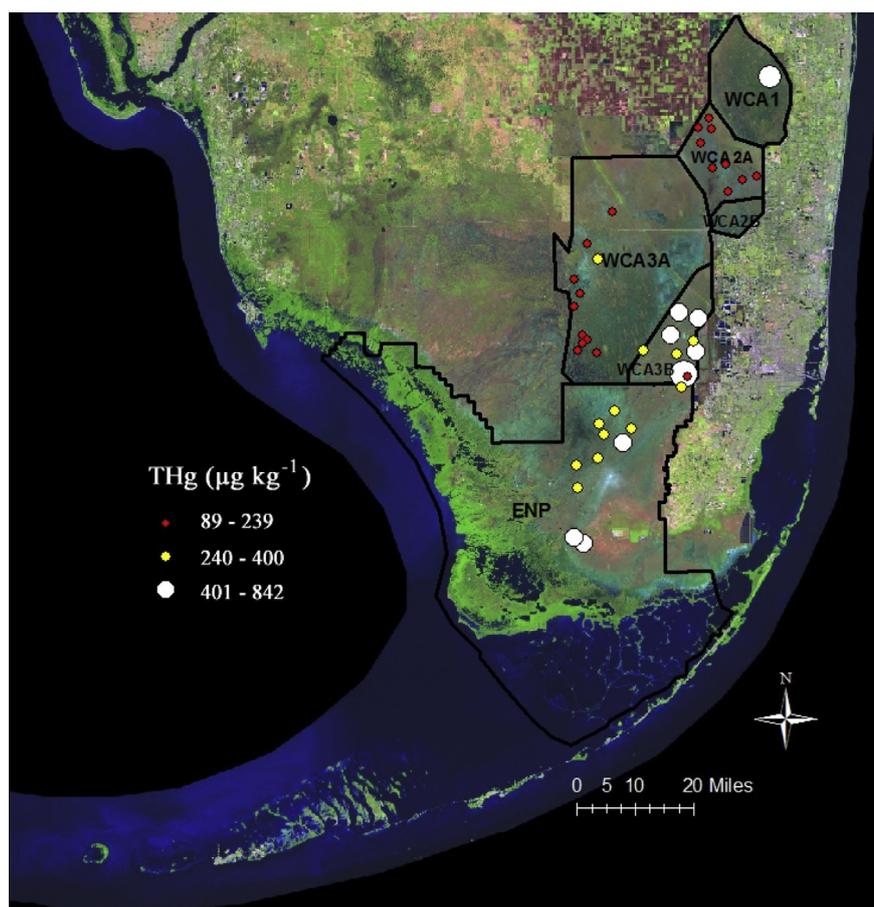


Fig. 1. Location of tree islands in the Everglades (white dots indicate tree islands with Hg hotspots).

Tree islands are an important habitat for wildlife, especially large populations of wading birds (Gawlik and Rocque, 1998). WCA-1, WCA-3 and ENP are the primary nesting regions for wading birds in the Everglades. In 2010, ENP supported the most nests (37%) followed by WCA-1 (34%) while WCA-3 supported the lowest number of nests (29%) (Cook and Kobza, 2010). Wading bird colonies located in central and south portion of the Everglades (WCA-3B and ENP) were at higher risk to Hg exposure than those colonies feeding elsewhere in the Everglades due to high Hg concentration in fish (Stober et al., 2001).

Recent research indicates that tree islands are biogeochemical hotspots that sequester nutrient within the Everglades (Wetzel et al., 2005, 2011). The concentration distribution of total Hg and MeHg across Everglades marsh soils has been documented (Arfstrom et al., 2000; Cohen et al., 2009; Liu et al., 2009), but the spatial distribution of Hg in tree islands soils is unknown. It is known that seabirds transport contaminants to their nesting sites via deposition of guano, carcasses, and shed feathers (Blais et al., 2005; Brimble et al., 2009; Evenset et al., 2007; Foster et al., 2011). A positive correlation between Hg concentrations and seabird populations was observed in Arctic ponds in Devon Island, with ponds most affected by seabird guano containing 25 times higher Hg concentrations than unaffected sediments (Blais et al., 2005). Deposition of wading bird guano has been hypothesized for nutrient accumulation in tree island soils in the Everglades, particularly phosphorus (Wetzel et al., 2011, 2005). Besides nutrients, wading birds in the Everglades may transport Hg to their nesting site via guano. This is because wading birds in the Everglades prey on invertebrates and fish from the marsh and

accumulate high Hg concentrations in their feathers and eggs (Herring et al., 2009). However, Hg deposition from wading bird guano in the Everglades tree island soils has not been reported.

Stable nitrogen isotope analysis is a powerful tool to document the trophic position of animals, as consumer $\delta^{15}\text{N}$ values display an average increase of 3.4‰ per trophic level along food chains (Post, 2002). Elevated concentrations of organic and metal pollutants associated with high $\delta^{15}\text{N}$ in soil and sediment affected by seabirds have been reported (Blais et al., 2005; Brimble et al., 2009; Foster et al., 2011). The correlation of $\delta^{15}\text{N}$ and Hg was used to identify trophic linkages and Hg transfer in organisms living near arctic seabird colonies (Choy et al., 2010). In addition, Hg concentrations and $\delta^{15}\text{N}$ values are significantly positively correlated in seabird-affected ponds (Blais et al., 2005; Evenset et al., 2007).

The objective of this study was to assess the spatial distribution of Hg in tree island soils and the influence of wading bird guano deposition on Hg accumulation in tree island soils. This was accomplished by 1) comparing Hg concentrations in tree island soils with and without wading bird guano, 2) investigating the relationship between Hg concentration and $\delta^{15}\text{N}$ values in tree island soils affected by wading bird guano, and 3) calculating the wading birds guano deposition rates to tree island soils during 1998–2010.

2. Materials and methods

2.1. Study site

Forty-one tree islands representing a broad range of sizes and hydrologic regimes in the Everglades were selected for this study. They were from 3 WCAs in the

Table 1

Hg concentrations and $\delta^{15}\text{N}$ values in soils, plant leaves, mammal droppings, bird bones and bird guano from the Everglades tree islands.

Statistic	Tree island soils		Others			
	All surface soils	Head soils	Plant leaves	Mammal droppings	Bird bones	Bird guano
Hg ($\mu\text{g kg}^{-1}$)						
<i>n</i>	93	34	11	9	5	5
Mean	209	280	53	105	89	286
std error	14	27	9	32	28	87
Min	79	89	20	22	41	109
Max	842	842	128	34	192	596
Median	155	258	49	64	55	267
$\delta^{15}\text{N}$ (‰)						
Mean	ND ^a	7.5	2.3	3.2		8.8
std error	ND	0.8	0.6	0.8		0.7
Min	ND	0.7	-3.7	0.9		7.8
Max	ND	13.9	9.4	9.0		10.6
Median	ND	9.0	2.1	2.3		8.4

^a ND: not determined, Hg ($\mu\text{g kg}^{-1}$) and $\delta^{15}\text{N}$ (‰) were expressed as dry weight.

northern and central Everglades and ENP in the south Everglades (Fig. 1). Seven ghost tree islands from WCA-2A and 34 live tree islands from WCA-1, WCA-3 and ENP were selected for this study (Fig. 1).

2.2. Sample collection

Soil samples were taken from 7 ghost and 34 live tree islands between 2009 and 2010. Transects were set up at the head, middle and tail in ghost and live tree islands from WCA-2, 3A and 3B. A total of 93 surface soil samples (top 10 cm) were collected. Not all tree islands had mid and tail sections, with several small tree islands having only a head section (WCA-1 and ENP). In addition to surface soils, three soil cores (50 cm long) were obtained from the head, mid and tail section of a live tree island in WCA-3B. The head region of the tree island had wading bird nests. Soil core samples were sectioned at 2 cm intervals for Hg analysis.

Bird guano samples were from mixed species of wading birds from several tree islands in WCA-3A and 3B, the species included great egrets (*Ardea alba*), white ibis (*Eudocimus albus*), snowy egrets (*Egretta thula*), little blue herons (*Egretta caerulea*) and tri-colored herons (*Egretta tricolor*). The bone samples of wading birds consisted of great egrets (*Ardea alba*) and white ibis (*Eudocimus albus*). Plant leaves and excrement from other animals were also collected to compare their Hg concentrations with wading bird guano. The plant leaves collected from tree islands including wax myrtle (*Morella cerifera*), coco plum (*Chrysobalanus icaco*), willow (*Salix caroliniana*), button bush (*Cephalanthus occidentalis*) and hard fen (*Blechnum* sp.). Mammal droppings samples were from white-tail deer (*Odocoileus virginianus*), eastern cottontail rabbit (*Sylvilagus floridanus*), feral pig (*Sus scrofa*), black bear (*Ursus americanus*) and raccoons (*Procyon lotor*). Mammal identification was based on dropping appearance and previous reports on wildlife surveys for tree islands in the Everglades.

2.3. Hg deposition via wading bird guano

The wading bird colony (Hidden) located in WCA-3 of the Everglades was selected. Five dominant species of wading birds, including great egrets (*A. alba*), white ibis (*E. albus*), snowy egrets (*E. thula*), tricolored herons (*E. tricolor*), and little blue herons (*E. caerulea*) were selected to calculate Hg deposition via guano. The numbers of wading bird nesting in the colony between 1998 and 2012 were based on South Florida Wading Bird Reports (1998–2010). The area (11,000 m²) of the colony was estimated with aerial imagery. Dry weight (kg) guano per nestling specie was estimated from a previous study (Frederick and Powell, 1994). Average Hg concentration in mixed dry guano samples was used to calculate guano Hg deposition (Table 1).

2.4. Hg analysis and quality assurance

Total Hg concentrations in soils, plant leaves, mammal dropping, wading bird guano and bones were digested with 7:3 (v/v) concentrated HNO₃/H₂SO₄ acids at 110 °C overnight according to EPA Method 1631 and analyzed using hydride generation atomic fluorescence spectrometry (HGAFS). Soil, plant leaves, mammal droppings, wading bird guano and bones were oven-dried (60 °C) and ground to fine powder (<2 mm) before digestion. Hg concentrations in all samples were calculated based on dry weight. Strict quality control procedures were followed during sample analysis. Quality control samples, including two method blanks, matrix spikes, lab duplicates, and certified reference material SRM 2709, were analyzed every 30 samples. In all method blanks, total Hg concentrations were below detection limits

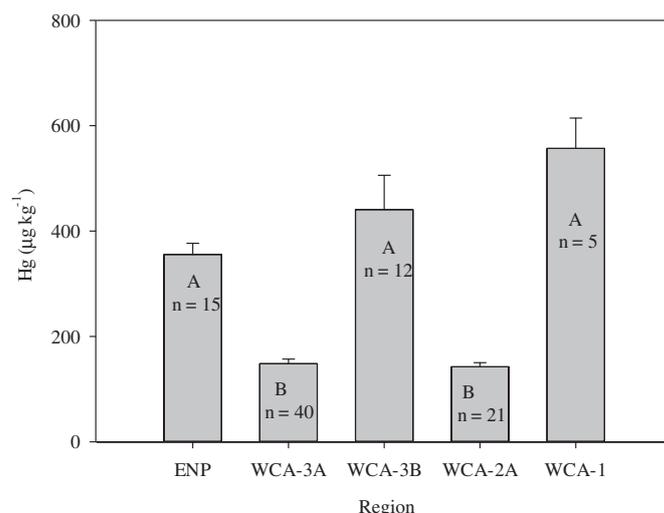


Fig. 2. Mean soil Hg concentrations in surface soils of tree islands from the three Water Conservation Areas (WCAs) and Everglades National Park (ENP) (Error bars represent standard deviation of the mean. Different letters indicate significant differences in Hg concentrations at $\alpha = 0.05$. *n* = number of soil samples).

of HGAFS (0.01 $\mu\text{g L}^{-1}$ for water and 2 $\mu\text{g kg}^{-1}$ for soil). Mean coefficients of variation for duplicate samples were low (6.70–13.2%) and recoveries for matrix spikes or certified reference material were within the acceptable ranges (80–120%). Instrument performance was checked by running an intermediate calibration standard for every 15 samples. All calibration standard checks were within the acceptable range (90–110%).

2.5. Ancillary parameter analysis

For soil $\delta^{15}\text{N}$ (the ratio of ¹⁵N to ¹⁴N, defined as $\delta^{15}\text{N}$ with a unit ‰) analysis, ~1 mg of the fine powder was wrapped into tin capsules and analyzed using a Carlo Erba elemental analyzer interfaced with a Finnigan MAT Delta Plus XP stable isotope ratio mass spectrometer at the Department of Geological Science, Florida State University, USA. The $\delta^{15}\text{N}$ values were reported in standard delta notation relative to atmospheric N. Analytical precision (based on replicate analyses of laboratory standards processed with each batch of samples and on sample replicates) was $\pm 0.1\%$. Soil organic matter (SOM) was determined by loss on ignition at 550 °C overnight and total nitrogen (TN) was determined by combustion using a CNS analyzer (Elementar Analysensysteme GmbH, Hanau, Germany). Total phosphate was determined by combusting 200 mg of oven-dried soil at 550 °C for 4 h and dissolving the ash in 6 M HCl.

2.6. Statistical analysis

The Hg distribution patterns in surface soils of tree islands were examined by a probabilistic model, which assumed the log transformed data are all uncorrelated. Comparison of Hg concentrations in different regions of ghost and live tree islands soils was performed using the student's *t*-test, and comparison of Hg concentrations and $\delta^{15}\text{N}$ values in head, mid and tail section in soil core samples was performed by the paired *t*-test. Regression analyses between Hg concentrations and soil property (TP, TN, SOM, $\delta^{15}\text{N}$ and bulk density) were also conducted. All correlation analyses were conducted on log-transformed data. The normality test (Shapiro–Wilk statistic) and homogeneity variance test (Levene's test) indicated all variables met the assumption of normality and homogeneity after log transformation. Comparisons of Hg concentrations in plants, mammal dropping, wading bird guano and bones in tree islands were performed by the student's *t*-test. A *p* value less than 0.05 was considered significant. All statistical analyses were carried out with SAS 9.3 or SigmaPlot 11.0.

3. Results and discussion

3.1. Spatial Hg distributions in tree islands soils were different from marsh soils

Surface soil samples (*n* = 93) were collected from 41 tree islands across the Everglades. The soil Hg concentrations ranged from 79 to 842 $\mu\text{g kg}^{-1}$, averaging 209 $\mu\text{g kg}^{-1}$ (Table 1). The mean soil Hg

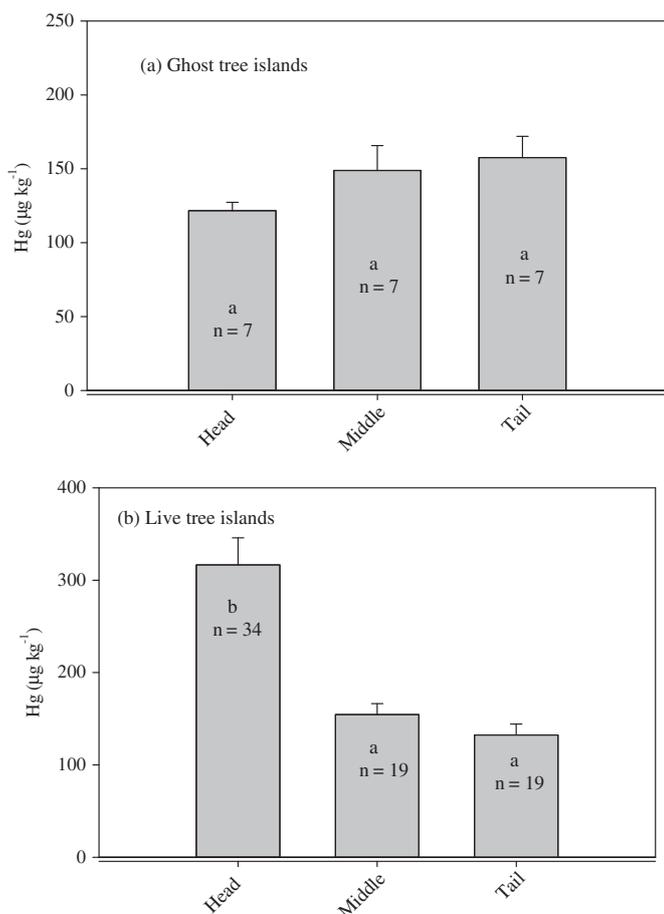


Fig. 3. Mean soil Hg concentrations in the head, middle and tail of ghost and live tree islands (Surface soil samples were collected from 7 ghost tree island in WCA-2A and 34 live tree islands in WCA 1, 3A, 3B and ENP. Error bars represent standard deviation of mean. Different letters indicate significant differences in Hg concentrations at $\alpha = 0.05$. $n =$ number of soil samples).

concentrations from tree islands in WCA-1 ($557 \pm 58 \mu\text{g kg}^{-1}$), WCA-3B ($441 \pm 65 \mu\text{g kg}^{-1}$) and ENP ($356 \pm 21 \mu\text{g kg}^{-1}$) were significantly higher than those in WCA-2A ($142 \pm 12 \mu\text{g kg}^{-1}$) and WCA-3A ($165 \pm 24 \mu\text{g kg}^{-1}$) (Fig. 2). A total of 9 Hg hotspots (defined as $\text{Hg} \geq 400 \mu\text{g kg}^{-1}$) were identified in the head regions, of which 5 were in WCA-3B, 3 were in ENP, and 1 in WCA-1. The highest soil Hg concentration ($840 \mu\text{g kg}^{-1}$) was observed in WCA-3B whereas Hg concentrations in 7 ghost tree islands soils from WCA-2A were all $< 400 \mu\text{g kg}^{-1}$ (Fig. 1).

Hg concentrations in the marsh soils were generally higher in the north and central Everglades (WCA-1 and 2, 3) and lower in the southern regions (ENP) (Cohen et al., 2009; Liu et al., 2009; Stober et al., 2001). Higher atmospheric deposition of Hg in the northern Everglades leads to higher Hg levels in the north Everglades marsh soil (Arfstrom et al., 2000; Cohen et al., 2009). If atmospheric deposition were the only source of Hg to the Everglades, then soils in tree islands and marshes should have received similar amounts of Hg, and Hg spatial distribution in different hydrological regions (WCAs and ENP) of the tree island and marsh soil should be similar. Compared to marsh soils, the Hg concentrations in tree island soils followed a different pattern with high values in the northern and southern Everglades (WCA-1, WCA-3B and ENP). These observations indicated that in addition to atmospheric deposition, the high Hg concentrations in tree island soils may have derived from other sources. In addition, normal probability plots for surface soil Hg did

not fit a straight line (data not shown), suggesting more than one source of Hg in these samples. Deposition of wading bird guano and other animals' droppings enriched with Hg is a possible source of Hg accumulation in tree island soils.

3.2. Hg concentrations in surface soils from ghost tree islands were low

The Hg concentrations from the 7 ghost tree island soils ($n = 21$) from WCA-2A ranged from 89 to $265 \mu\text{g kg}^{-1}$, averaging $142 \mu\text{g kg}^{-1}$, which was similar to marsh soils ($146 \mu\text{g kg}^{-1}$) (Cohen et al., 2009; Liu et al., 2008b). Cohen et al. observed Hg values of 2–917 $\mu\text{g kg}^{-1}$, averaging $162 \mu\text{g kg}^{-1}$ (Cohen et al., 2009). Liu et al. (2008b) reported Hg concentrations in the Everglades marsh soils range from 9.3 to $350 \mu\text{g kg}^{-1}$, averaging $130 \mu\text{g kg}^{-1}$. In addition, the average bulk density of the ghost tree island soils (0.10 g cm^{-3}) was similar to the surrounding slough (0.08 g cm^{-3}), and was significantly lower than those in live tree islands in WCA-3A ($0.3\text{--}0.6 \text{ g cm}^{-3}$) (Sklar et al., 2011). The mean soil Hg concentrations in the head, mid and tail regions were 122, 149 and $157 \mu\text{g kg}^{-1}$. There was no significant difference in soil Hg concentrations among head, mid and tail regions ($p < 0.05$) (Fig. 3a). The head regions of ghost tree islands in WCA-2A mostly had no woody vegetation, with the most common species being sawgrass and herbaceous species (Sklar et al., 2011). The absence of woody vegetation and dominance of sawgrass in the head region of ghost tree islands led to the absence of wading birds colonies and guano deposition. These observations indicated that the absence of wading bird guano could contribute to the low Hg concentrations in surface soils of ghost tree islands. Other process such as flooding and fire could also lead to low Hg concentration in the degraded tree islands, which need further study.

3.3. Soils from head regions of live tree islands were enriched with both Hg and $\delta^{15}\text{N}$

The Hg concentrations in 34 live tree island soils ($n = 72$) from WCA-1, 3A, 3B and ENP ranged from 79 to $842 \mu\text{g kg}^{-1}$, averaging $234 \mu\text{g kg}^{-1}$. The mean Hg concentrations in the head region ($317 \mu\text{g kg}^{-1}$) from live tree islands were significantly higher than those in mid ($155 \mu\text{g kg}^{-1}$, $p < 0.01$) and tail ($132 \mu\text{g kg}^{-1}$, $p < 0.01$) regions (Fig. 3b). In addition, the Hg concentration ($317 \mu\text{g kg}^{-1}$) in the head region of live tree islands was significantly higher than that of ghost tree islands ($122 \mu\text{g kg}^{-1}$). Besides surface soils from 34 live tree islands, we also compared Hg concentrations in three soil core samples (50 cm) from the head, mid and tail regions of a live tree island in WCA-3B. The head region of this live tree island is particularly conducive for wading bird reproduction and large numbers of nests are concentrated in this region. The mean Hg concentrations in the head region in the surface 10 cm were $589 \mu\text{g kg}^{-1}$. In addition, the mean Hg concentrations in the head region soil core ($389 \mu\text{g kg}^{-1}$) were significantly higher than that in mid ($124 \mu\text{g kg}^{-1}$, $p < 0.01$) and tail region ($106 \mu\text{g kg}^{-1}$, $p < 0.01$) (Fig. 4a).

In addition to high Hg, the $\delta^{15}\text{N}$ values in soils from the head regions of live tree islands were also high. Marsh soil and plants are depleted in $\delta^{15}\text{N}$, with a range from -1 to 3‰ (Sklar et al., 2011). The mean $\delta^{15}\text{N}$ value of head surface soils from live tree islands was 7.5‰ (Table 1). The $\delta^{15}\text{N}$ values in the head region (9.7‰) in the core were significantly higher than that in mid (5.5‰ , $p < 0.01$) and tail region (4.9‰ , $p < 0.01$) (Fig. 4b). The $\delta^{15}\text{N}$ of guano samples collected close to wading bird colonies in the Everglades ranged from 7.8 to 10.6‰ , averaging 8.8‰ (Table 1). The high $\delta^{15}\text{N}$ values of live tree island soils may result from guano deposition from wading birds. Other sources cannot lead to the observed $\delta^{15}\text{N}$ enrichment

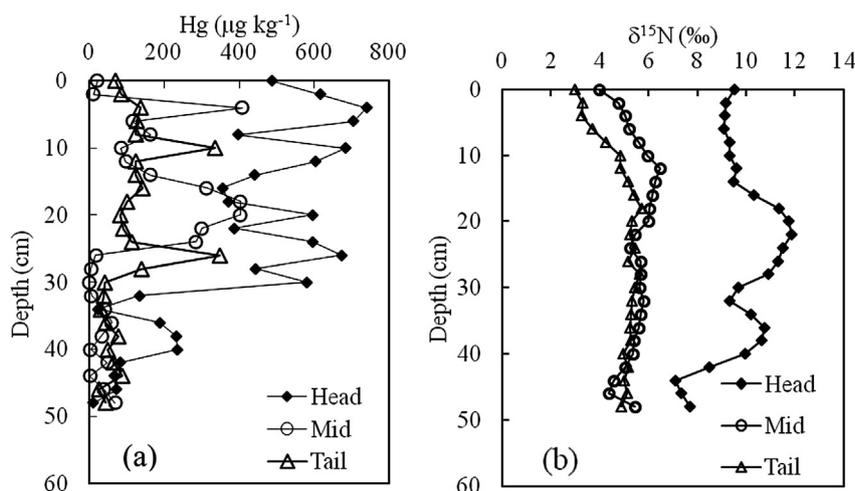


Fig. 4. Hg concentrations (a) and $\delta^{15}\text{N}$ values (b) in soil cores from a live tree island in WCA-3B.

in the soils due to their low trophic positions (e.g., tree leaves and mammal droppings) or low population size of higher trophic level predators (e.g., bears) (Axelrad et al., 2013). Hence, $\delta^{15}\text{N}$ was used as a tracer for wading bird guano in this study. The high $\delta^{15}\text{N}$ values in the head regions of live tree islands indicated the presence of wading bird guano in those soils.

In our study, $\delta^{15}\text{N}$ was positively correlated with soil Hg concentrations in surface samples of live tree island ($R^2 = 0.52$, $p < 0.01$) and the core soil samples ($R^2 = 0.27$, $p < 0.01$) from a live tree island with wading birds colony (Table 2; Fig. 5 a, b). Soil Hg concentrations and $\delta^{15}\text{N}$ values were not significantly correlated in the mid and tail regions ($R^2 = 0.01\text{--}0.18$, $p > 0.01$), which were not influenced by wading bird guano (Fig. 5c, d). These results indicated that high Hg concentrations in the soil may have been influenced by bird guano. To support this hypothesis, we analyzed the Hg concentrations in surface soils ($n = 5$) from a live tree island in WCA-1, which is known to be nested by great egrets. The mean Hg concentrations in those soils were high, averaging $454 \mu\text{g kg}^{-1}$, which supported our hypothesis.

3.4. Hg deposition via wading bird guano

Wading birds can accumulate Hg in their feathers, bones and guano. The guano and bone samples in this study represented mixed wading bird species, including great egrets, snowy egrets, white ibis, little blue herons and tri-colored herons. These species feed on invertebrates and fish in the Everglades and are positioned near the top of the aquatic food chain. Hg biomagnifies as it progressively moves up the food chains of the Everglades. The Hg concentration in wading bird guano, bones and mammal dropping were 286, 89 and $105 \mu\text{g kg}^{-1}$, respectively (Table 1). The Hg

concentrations in wading bird guano were significantly higher than those in wading bird bones and mammal droppings (Fig. 6, $p < 0.05$).

The Hg concentrations in tree island plants (wax myrtle, coco plum, willow, button bush and hard fen) were low, ranging from 20 to $128 \mu\text{g kg}^{-1}$, averaging $53 \mu\text{g kg}^{-1}$. The Hg concentrations in plant leaves were higher than those in cattail and sawgrass leaves ($1.0\text{--}13 \mu\text{g kg}^{-1}$) (Stober et al., 2001). Assuming Hg concentration of $53 \mu\text{g kg}^{-1}$ in the leaves, and dry mass in aboveground litterfall of $500 \text{g m}^{-2} \text{yr}^{-1}$ (Guentzel et al., 1998), the calculated annual Hg deposition was $27 \mu\text{g m}^{-2} \text{yr}^{-1}$. This is comparable to the wet Hg deposition in southern Florida ($19 \mu\text{g m}^{-2} \text{yr}^{-1}$ in 2006) (NADP, 2007). Hence, Hg deposition through plant leaves in tree islands in the Everglades may not be a significant source.

Considering the high Hg concentrations in wading bird guano and the large population of wading birds in tree islands, we calculated the guano Hg deposition in a tree island with a wading bird colony. Based on measured average Hg concentration in guano, guano excretion rates and wading bird nest numbers of the Hidden colony (data not shown), wading bird guano Hg deposition rate was as high as $790 \mu\text{g m}^{-2} \text{yr}^{-1}$ in 2009 (more than 42 times the average atmospheric deposition rate). The average guano Hg deposition for 1998–2010 was $148 \mu\text{g m}^{-2} \text{yr}^{-1}$ (~ 8 times that average atmospheric deposition rate). The estimated wading guano Hg deposition in tree islands suggests that wading birds play an important role in Hg redistribution within the Everglades ecosystem, and Hg deposition from wading bird guano at densely populated colonies can be high.

Our calculation provided a conservative estimate about guano Hg deposition to the tree island soils in the Everglades. The historical Hg deposition by guano could be much higher than $148 \mu\text{g m}^{-2} \text{yr}^{-1}$. This is because the Hg concentrations in the Everglades fish and wading birds have undergone a significant decline since 1990s. Hg concentrations in fish from WCAs have declined by $\sim 40\text{--}80\%$ over the past decade (Axelrad et al., 2008). Thus Hg concentrations in wading bird guano before 2010 could be much higher than $286 \mu\text{g kg}^{-1}$. In addition, the Hg deposition rate by wading bird guano at tree islands could change among different years and locations due to the variability of guano Hg concentrations and nest numbers. Unlike inorganic Hg in atmosphere deposition, MeHg concentration in wading bird guano could be high. This was because MeHg is the major form of Hg in fish, and bird may accumulate MeHg through food chain. For example, the seabird guano from Xisha Island, China contains 45% MeHg, which

Table 2
Correlation analyses (R^2 , probability of significance) between Hg and selected soil property in live tree island surface and core samples.

Soils	TP	SOM	BD	TN	$\delta^{15}\text{N}$ (‰)
Surface soil ^b	0.43, <0.01 ^a	no, no	0.11, 0.30	0.10, 0.08	0.52, <0.01 ^a
Head ^c	0.13, 0.08	0.15, 0.06	0.05, 0.30	0.10, 0.13	0.27, <0.01 ^a
Mid ^c	0.21, 0.02	0.04, 0.37	<0.01, 0.98	0.19, 0.03	0.08, 0.18
Tail ^c	<0.01, 0.88	<0.01, 0.74	0.06, 0.25	<0.01, 0.58	0.01, 0.57

^a Significant at $p < 0.01$.

^b Indicated surface soils from 34 live tree island soils in WCA-1, 3A, 3B and ENP.

^c Indicated core soils from a live tree island in WCA-3B; TP = total phosphate, SOM = soil organic matter, BD = bulk density, TN = total nitrogen.

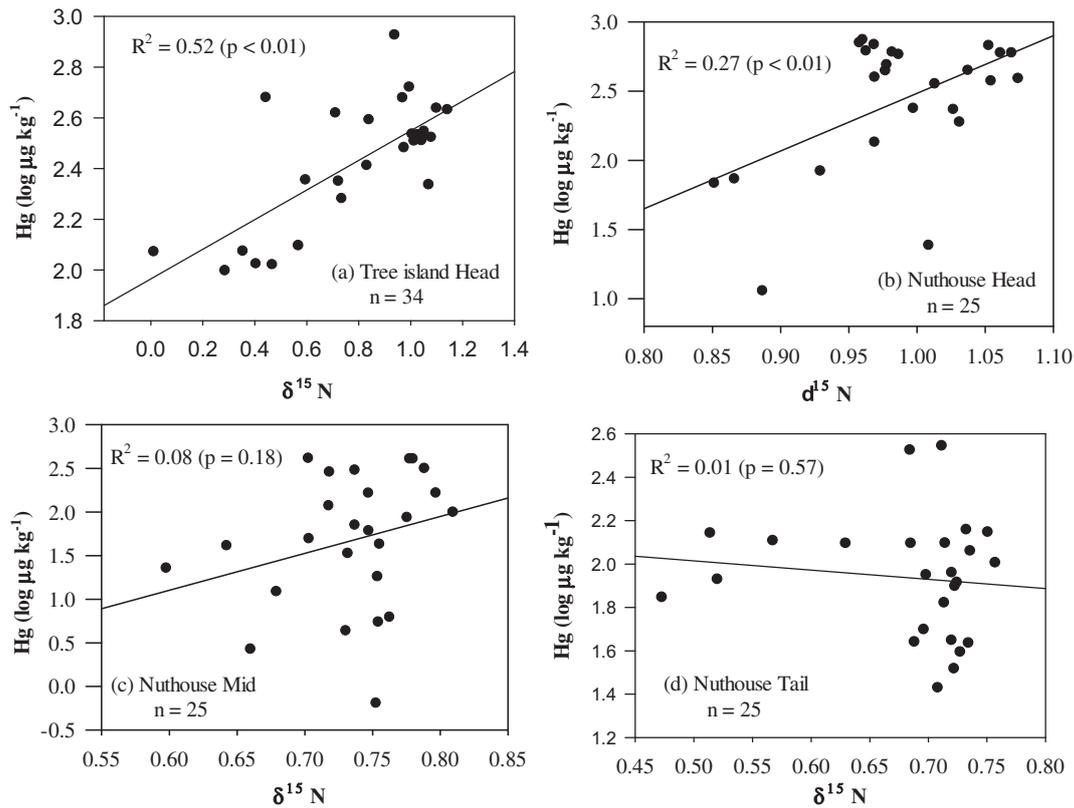


Fig. 5. Correlations of Hg concentrations with $\delta^{15}\text{N}$ values in tree island soils: (a) surface soils from head regions of 34 live tree islands, and core samples from (b) the head, (c) middle, and (d) tail in a live tree island with a bird colony (n = number of soil samples).

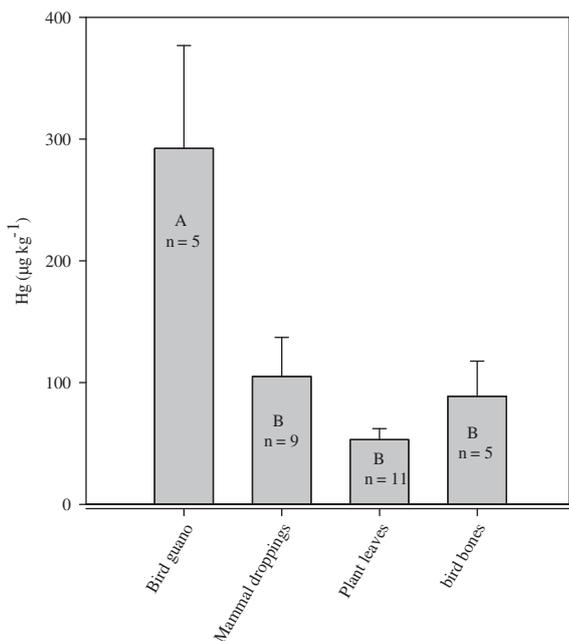


Fig. 6. Mean Hg concentrations in the mammal dropping, plant leaves, bird guano and bones from tree islands (Error bars represent standard deviation of mean. Different letters indicate significant differences in Hg concentrations at $\alpha = 0.05$. n = number of soil samples).

is identified as the major source of MeHg in ornithogenic coral sediments (Chen et al., 2012). The dominant form of Hg in the Everglades marsh soil is inorganic Hg (~99%), mean MeHg concentration was very low ($1.4 \mu\text{g kg}^{-1}$) (Liu et al., 2008b). Hence wading bird guano could elevate MeHg concentration in tree island soils.

4. Environmental implications

Our results provide novel information on Hg concentrations in soils, plants, mammal droppings and wading bird guano in tree islands in the Everglades. Guano deposition by wading birds played an important role in transporting Hg to tree island soils. These findings have important implications for Hg management in the Everglades. The head regions of live tree islands are important habitats for animals (alligators, deer, bear, and panther), especially during the wet season. Although wading bird guano deposition redistributed only a small fraction of the Hg in the Everglades, Hg deposition at sites with large numbers of nests could be high. Hg concentrations and deposition rates of guano may differ between regions, years and species. The contribution of wading bird guano to the relative Hg budget in the Everglades could be applied to other wetland ecosystems. Future research should evaluate the level of highly toxic MeHg in tree island soils, and Hg bioavailability in wading bird guano and tree island soils, and assess the potential ecological health risks from Hg contamination in tree island soils.

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References

- Arfstrom, C., Macfarlane, A.W., Jones, R.D., 2000. Distributions of mercury and phosphorous in Everglades soils from water conservation area 3A, Florida, U.S.A. *Water Air Soil Pollut.* 121, 133–159.
- Axelrad, D.M., Atkeson, T.D., Pollman, C.D., Lange, T., 2006. Chapter 2B: Mercury Monitoring, Research and Environmental Assessment in South Florida. South Florida Environmental Report. South Florida Water Management District, West Palm Beach, FL.
- Axelrad, D.M., Lange, T., Gabriel, M., Atkeson, T.D., Pollman, C.D., Orem, W.H., Scheidt, D.J., Kalla, P.L., Frederick, P.C., Gilmour, C.C., 2008. Chapter 3B: Mercury and Sulfur Monitoring, Research and Environmental Assessment in South Florida. South Florida Environmental Report. South Florida Water Management District, West Palm Beach, FL.
- Axelrad, D.M., Pollman, C.D., Gu, B., Lange, T., 2013. Chapter 3B: Mercury and Sulfur Environmental Assessment for the Everglades. South Florida Environmental Report. South Florida Water Management District, West Palm Beach, FL.
- Blais, J.M., Kimpe, L.E., McMahon, D., Keatley, B.E., Mallory, M.L., Douglas, M.S.V., Smol, J.P., 2005. Arctic seabirds transport marine-derived contaminants. *Science* 309, 445.
- Brandt, L.A., Portier, K.M., Kitchens, W.M., 2000. Patterns of change in tree islands in Arthur R. Marshall Loxahatchee national wildlife refuge from 1950 to 1991. *Wetlands* 20, 1–14.
- Brimble, S.K., Foster, K.L., Mallory, M.L., Macdonald, R.W., Smol, J.P., Blais, J.M., 2009. High arctic ponds receiving biotransported nutrients from a nearby seabird colony are also subject to potentially toxic loadings of arsenic, cadmium, and zinc. *Environ. Toxicol. Chem.* 28, 2426–2433.
- Chen, Q., Liu, X., Xu, L., Sun, L., Yan, H., Liu, Y., Luo, Y., Huang, J., 2012. High levels of methylmercury in guano and ornithogenic coral sand sediments on Xisha Islands, South China Sea. *Arch. Environ. Contam. Toxicol.* 63, 177–188.
- Choy, E.S., Gauthier, M., Mallory, M.L., Smol, J.P., Douglas, M.S.V., Lean, D., Blais, J.M., 2010. An isotopic investigation of mercury accumulation in terrestrial food webs adjacent to an Arctic seabird colony. *Sci. Total Environ.* 408, 1858–1867.
- Cohen, M.J., Lamsal, S., Osborne, T.Z., Bonzongo, J.C.J., Newman, S., Reddy, K.R., 2009. Soil total mercury concentrations across the Greater Everglades. *Soil Sci. Soc. Am. J.* 73, 675–685.
- Cook, M.I., Kobza, M., 2010. South Florida Wading Bird Report. South Florida Water Management District, West Palm Beach, FL.
- Evenset, A., Carroll, J., Christensen, G.N., Kallenborn, R., Gregor, D., Gabrielsen, G.W., 2007. Seabird guano is an efficient conveyor of persistent organic pollutants (POPs) to Arctic lake ecosystems. *Environ. Sci. Technol.* 41, 1173–1179.
- Foster, K.L., Kimpe, L.E., Brimble, S.K., Liu, H., Mallory, M.L., Smol, J.P., Macdonald, R.W., Blais, J.M., 2011. Effects of seabird vectors on the fate, partitioning, and signatures of contaminants in a high arctic ecosystem. *Environ. Sci. Technol.* 45, 10053–10060.
- Frederick, P.C., Powell, G.V.N., 1994. Nutrient Transport by Wading Birds in the Everglades, Everglades: the Ecosystem and Its Restoration. St. Lucie Press, Delray Beach FL, pp. 571–584.
- Gabriel, M.C., Axelrad, D.M., Lange, T., Dirk, L., 2010. Chapter 3B: Mercury and Sulfur Monitoring, Research and Environmental Assessment in South Florida. South Florida Environmental Report. South Florida Water Management District, West Palm Beach, FL.
- Gawlik, D.E., Rocque, D.A., 1998. Avian communities in bayheads, willowheads, and sawgrass marshes of the central Everglades. *Wilson Bull.* 1, 45–55.
- Guentzel, J.L., Landing, W.M., Gill, G.A., Pollman, C.D., 1998. Mercury and major ions in rainfall, throughfall, and foliage from the Florida Everglades. *Sci. Total Environ.* 213, 43–51.
- Herring, G., Gawlik, D.E., Rumbold, D.G., 2009. Feather mercury concentrations and physiological condition of great egret and white ibis nestlings in the Florida Everglades. *Sci. Total Environ.* 407, 2641–2649.
- Hofmockel, K., Richardson, C., Halpin, P., 2008. Effects of Hydrologic Management Decisions on Everglades Tree Islands, Everglades Experiments. Springer, New York, pp. 191–214.
- Liu, G., Cai, Y., Kalla, P., Scheidt, D., Richards, J., Scinto, L.J., Gaiser, E., Appleby, C., 2008a. Mercury mass budget estimates and cycling seasonality in the Florida Everglades. *Environ. Sci. Technol.* 42, 1954–1960.
- Liu, G., Cai, Y., Mao, Y., Scheidt, D., Kalla, P., Richards, J., Scinto, L.J., Tachiev, G., Roelant, D., Appleby, C., 2009. Spatial variability in mercury cycling and relevant biogeochemical controls in the Florida Everglades. *Environ. Sci. Technol.* 43, 4361–4366.
- Liu, G., Cai, Y., Philippi, T., Kalla, P., Scheidt, D., Richards, J., Scinto, L., Appleby, C., 2008b. Distribution of total and methylmercury in different ecosystem compartments in the Everglades: implications for mercury bioaccumulation. *Environ. Pollut.* 153, 257–265.
- Liu, G., Naja, G.M., Kalla, P., Scheidt, D., Gaiser, E., Cai, Y., 2010. Legacy and fate of mercury and methylmercury in the Florida Everglades. *Environ. Sci. Technol.* 45, 496–501.
- Mason, D.H., Valk, A.V.d., 2002. Vegetation, peat elevation, and peat depth on two tree islands in water conservation area 3-A. In: Sklar, F.H., Valk, A.V.d. (Eds.), *Tree Islands of the Everglades*. Kluwer Academic, Netherlands, pp. 337–356.
- NADP, 2007. National Atmospheric Deposition Program.
- Orem, W.H., Willard, D.A., Lerch, H.E., Bates, A.L., Boylan, A., Comm, M., 2003. Nutrient geochemistry of sediments from two tree islands in water conservation area 3Bm the Everglades, Florida. In: Sklar, F.H., Valk, A.V.d. (Eds.), *Tree Islands of the Everglades*. Kluwer Academic Publishers, pp. 153–186.
- Post, D.M., 2002. Using stable isotopes to estimate trophic position: models, methods, and assumptions. *Ecology* 83, 703–718.
- Rumbold, D.G., Howard, N., Matson, F., Atkins, S., Jacques, J.J., Nicholas, K., Owens, C., Warner, K.S.a.B., 2007. Appendix 3B-1: Annual Permit Compliance Monitoring Report for Mercury in Downstream Receiving Waters of the Everglades Protection Area. South Florida Environmental Report. South Florida Water Management District, West Palm Beach, FL.
- Sah, J.P., 2004. Vegetation structure and composition in relation to the hydrological and soil environments in Shark Slough. In: Ross, M.S., Jones, D.T. (Eds.), *Tree Islands in the Shark Slough Landscape: Interactions of Vegetation, Hydrology, and Soils*. Everglades National Park, U. S. Department of the Interior.
- Sklar, F., Dreschel, T., Warren, K., 2011. Chapter 6: Ecology of the Everglades Protection Area. South Florida Environmental Report. South Florida Water Management District, West Palm Beach, FL.
- Stober, Q.J., T., K., Jones, R., Richards, J., Ivey, C., W., R., Madde, M., Trexler, J., Gaiser, E., Scheidt, D., R. S., 2001. South Florida Ecosystem Assessment: Phase I/II (Technical Report) –Everglades Stressor Interactions: Hydropatterns, Eutrophication, Habitat Alteration, and Mercury Contamination. USEPA Region 4, Athens, GA.
- Wetzel, P.R., 2003. Tree island ecosystems of the world. In: Sklar, F.H., Valk, A.V.d. (Eds.), *Tree Islands of the Everglades*. Kluwer Academic Publishers, pp. 19–69.
- Wetzel, P.R., Valk, A.G.v.d., Newman, S., Gawlik, D.E., Gann, T.T., Coronado-Molina, C.A., Childers, D.L., Sklar, F.H., 2005. Maintaining tree islands in the Florida Everglades: nutrient redistribution is the key. *Front. Ecol. Environ.* 3, 370–376.
- Wetzel, P.R., Sklar, F.H., Coronado, C.A., Troxler, T.G., Krupa, S.L., Sullivan, P.L., Ewe, S., Price, R.M., Newman, S., Orem, W.H., 2011. Biogeochemical processes on tree islands in the greater Everglades: initiating a new paradigm. *Crit. Rev. Environ. Sci. Technol.* 41, 670–701.