Soil Total Mercury Concentrations across the Greater Everglades

Matthew J. Cohen*
Sanjay Lamsal
School of Forest Resources and Conservation
Univ. of Florida
328 Newins-Ziegler Hall
PO Box 110410
Gainesville FL 32611-0410

Todd Z. Osborne
Wetland Biogeochemistry Lab.
Soil and Water Science Dep.
Univ. of Florida
106 Newell Hall
PO Box 110510
Gainesville, FL 32611-0510

Jean Claude J. Bonzongo
Environmental Engineering Sciences
Univ. of Florida
320 Black Hall
PO Box 1106450
Gainesville, FL 32611-6450

Susan Newman
Everglades Dep.
South Florida Water Management District
PO Box 24680
West Palm Beach FL 33416

K. Ramesh Reddy
Wetland Biogeochemistry Lab.
Soil and Water Science Dep.
Univ. of Florida
106 Newell Hall
PO Box 110510
Gainesville, FL 32611-0510

Elevated Hg concentrations in the Everglades pose ecological and human health risks. We mapped soil total Hg concentrations per mass (THg_M) and area (THg_A) across the Everglades, and investigated relationships with soil properties (total P [TP] and organic matter content), community type, and hydrologic compartmentalization. Samples (n = 600) from surface soils (0–10 cm) were selected from a population of 1405 sites spanning the Everglades. Overall, 168 sites had THg_M levels >0.2 mg kg^{-1}; interpolation suggests that 23% of the Greater Everglades exceeds this threshold. Hot spots (>0.4 mg kg^{-1}) were observed in eastern Water Conservation Area (WCA) 1 and west-central WCA3A; parts of WCA2A, WCA3AN, and WCA3B were locally high. Despite significant global differences in THg_M among plant communities, differences evaluated using paired proximate sites were not significant, suggesting that large spatial scale depositional gradients govern ecosystem storage. Median THg_A was 1.89 mg m^{-2} (range 0.07–12.05 mg m^{-2}), representing approximately 100 yr of atmospheric deposition at contemporary rates (~19 μg m^{-2} yr^{-1}). Correlation between TP and THg_M was positive in unimpacted areas (TP < 500 mg kg^{-1}, r = 0.69), but negative in impacted areas (TP > 500 mg kg^{-1}, r = -0.47), probably due to accelerated peat accretion rates in P-enriched areas. Moreover, while reverse correlation with distance from a canal for THg_M (0.70) and TP (–0.77) supports Hg enrichment via atmospheric deposition, THg_M hotspots in WCA3AS, WCA1, and the Holeyland and Rotenberger tracts are suggestive of local enrichment mechanisms. Finally, despite dramatic regional emissions declines, the estimated mass of Hg in surface soils across the Everglades has increased ~20% (11,000 vs. 13,100 kg) since 1996; while the statistical significance of this change is unknown, it provides a useful benchmark for future surveys.

Abbreviations: BCNP, Big Cypress National Preserve; BD, bulk density; EAA, Everglades Agricultural Area; ENP, Everglades National Park; HLRB, Holeyland and Rotenberger tracts; LOI, loss-on-ignition; MeHg, methyl-mercury; OK, ordinary kriging; THg, total mercury; THg_A, total mercury per area; THg_M, total mercury per mass; TP, total phosphorus; WCA, water conservation area.

Elevated Hg concentrations in the soils of the Everglades have been identified as one of many coincident stressors on that system that require restorative action (Stober et al., 2001). The death of an endangered Florida panther (*Puma concolor coryi*) attributed to acute Hg toxicosis (Roelke et al., 1991) exemplifies the ecological impact of Hg bioaccumulation, as do observations of Hg concentrations exceeding 1.5 mg kg^{-1} (wet mass) in sport fish from the Everglades during a 1989 statewide survey (Ware et al., 1990). These findings coincide with a fivefold increase in mean total Hg (THg) accumulation rates in Everglades sediments between recent (post-1985) and historic (pre-1900) periods (Rood et al., 1995); spatial variability in enrichment is substantial, with markedly higher enrichment rates in the northern Everglades. Similarly, Cleckner et al. (1998) observed elevated levels of methyl mercury (MeHg), a more toxic and bioavailable form, in fish and hemipterans in WCA2A (Fig. 1), while Cleckner et al. (1999) reported mediation of methylation by periphyton communities, another spatially variable ecological component. While Fleming et al. (1995) reported low levels of human exposure among those regularly consuming fish caught from the Everglades, concerns of bioaccumulation and chronic ecological effects require continuing attention to patterns and processes of enrichment.

Natural processes, such as volcanism and weathering, and human activities, notably fossil fuel burning and municipal

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*Corresponding author (mjc@ufl.edu).

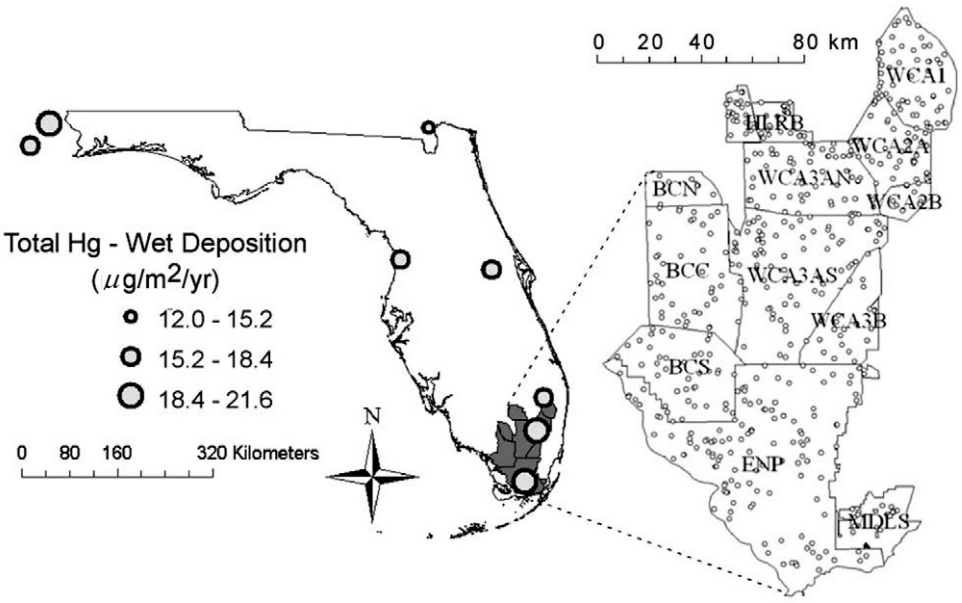

Fig. 1. Soil sampling locations across the Everglades. Shown on the Florida map are National Atmospheric Deposition Program Hg monitoring stations and recent (since 1997) annual average wet deposition rates (National Atmospheric Deposition Program, 2007). Hydrologic partitions are labeled on the Everglades map (BCN = Big Cypress National Preserve, WCA = Water Conservation Area, ENP = Everglades National Park, HLRB = Holeyland and Rotenberger tracts; MDLS = Model Lands tract)

waste incineration, release Hg, which is eventually transported to, and accumulated in, soil, sediment, and biota (Rood, 1996). Measured atmospheric deposition rates of Hg to terrestrial and aquatic systems vary widely in space and time ($\sim$0.3–30 μg m^{-2} yr^{-1}, USEPA, 1997), with geographic maxima in the eastern United States, including Florida (National Atmospheric Deposition Program, 2007). In southern Florida, the rates of Hg atmospheric deposition vary between 19 and 25 μg m^{-2} yr^{-1} (Fig. 1; Krabbenhoft et al., 1999; National Atmospheric Deposition Program, 2007), roughly 30% higher than rates in north-central Florida; no significant east–west trends have been observed (Guentzel et al., 1995), but no studies with sufficient data density to explore short-range deposition trends have been reported. Deposition rates are well monitored (five active National Atmospheric Deposition Network stations in the Florida peninsula, see nadp.sws.uiuc.edu/mdn/ [verified 12 Dec. 2008]), and despite significant interannual variability in deposition due to rainfall, trends are clearly downward. Mercury sources are estimated to be 73% local, primarily from waste incineration and fossil fuel combustion (Dvonch et al., 1999). Recent changes in waste incineration have lowered local sources by a factor of 10 (Atkeson et al., 2003), but deposition trends have not yet reflected this decline (National Atmospheric Deposition Program, 2007).

Despite relatively high deposition rates in the southeastern United States, only a small fraction of the atmospheric load is in the methylated form (Rudd, 1995), which is the primary form of Hg found in higher trophic level organisms (Wiener and Spry, 1996). Bioaccumulation of Hg is favored by methylation of inorganic Hg species to produce MeHg (Zilloux et al., 1993), a process associated with sulfate-reducing bacteria (Gilmour et al., 1992) in anoxic sediments (Domagalski, 2001). Krabbenhoft et al. (1999) observed a significant positive association between wetland cover in a watershed and river sediment MeHg concentrations across 21 basins nationally, reinforcing previous work (Zilloux et al., 1993; Hurley et al., 1995) indicating that wetland cover strongly influences Hg dynamics. Krabbenhoft et al. (1999) specifically identified the Everglades as exhibiting high methylation efficiency (MeHg/THg in sediments >0.10), although they also observed a positive logarithmic association between THg and MeHg that suggests a reduced methylation response above THg concentrations of 1 mg kg^{-1}. Sediment methylation experiments support this observation (Rudd et al. 1983). Liu et al. (2008) reported that MeHg production derives principally from the soil pool (97 and 70% in dry and wet seasons, respectively), suggesting that knowledge of soil THg is a necessary but not sufficient predictor of ecosystem Hg dynamics.

The contemporary Everglades is subject to numerous anthropogenic stressors. Elevated nutrients, invasive exotic species, Hg bioaccumulation, and substantial changes in hydrologic dynamics have led to significant declines in ecosystem health, prompting plans for a massive and far-reaching restoration effort (Comprehensive Everglades Restoration Plan, see www.evergladesplan.org [verified 12 Dec. 2008]). Paramount among the human-induced changes are the well-documented hydrologic modifications to the South Florida ecosystem (Light and Dineen, 1994). Construction of a network of canals and dikes have drained and compartmentalized the Everglades landscape into multiple discontinuous hydrologic units (Fig. 1), including the Everglades agricultural area (EAA), Water Conservation Areas (WCAs 1, 2A, 2B, 3A, and 3B), and the Everglades National Park (ENP) and impacted Everglades ecosystem structure and function (South Florida Water Management District, 1992; Davis and Ogden, 1994). Phosphorus enrichment is attributed to agricultural activities and hydrologic partitioning of the Everglades. It remains unclear, however, if increased Hg concentrations in Everglades soils are due to oxidation of peats and release of historic Hg accumulations due to hydrologic modification (Lodenius, 1990) followed by canal transport, or are primarily from increases in atmospheric deposition.

Our principal objective in this study was to investigate the spatial pattern of Hg on both a mass and an area basis across the Everglades. Our rationale for focusing on total Hg (rather than more transient but more toxicologically relevant MeHg) follows from (i) sampling and analytical logistics and costs, (ii) the observation that, in southern Florida, THg concentrations in sediments were positively correlated with those in fish (r = 0.52, $P < 0.05$) and also with fish MeHg concentrations (r = 0.42, $P < 0.05$) (Kannan et al., 1998), and (iii) the observation that THg levels in the soil maintain spatial patterns over time

better than other ecosystem pools (Liu et al., 2008). Liu et al. (2008) further reported the dominant importance of the soil Hg pool on MeHg production, and that >80% of the annual Hg deposition is entrapped in the soil pool.

While previous studies (Rood et al., 1995; Arfstrom et al., 2000) have examined Hg distribution in Everglades soils, these were conducted in geographically limited regions (the central ENP and southern WCA3A, respectively) with relatively low observation density (n = 45 and 64, respectively). As such, they cannot provide a comprehensive assessment of spatial patterns across the system to serve as a restoration baseline. In contrast, Liu et al. (2008) obtained sufficient samples (n = 109) to develop broad spatial patterns, but reported trends along a north–south gradient only; those trends suggest a moderately significant decrease from north to south in MeHg only, but by conflating all samples at the same latitude, mask spatial patterns.

Our secondary objective was to investigate the relationships between observed Hg concentrations and potential environmental covariates. Three covariates were of particular interest: (i) the influence of proximity to canals on Hg concentrations, which indirectly provides evidence regarding sources of elevated Hg; (ii) the influence of community type on observed concentrations, which may suggest biotic factors regulating soil concentrations; and (iii) previous work (Vaithiyanathan et al., 1996) that reported a strong negative correlation between THg and TP along a nutrient enrichment gradient in WCA2A. Because the delivery mechanism for P is clear (canal inflow), quantifying the covariation of THg with TP across the entire Everglades can enhance broad-scale understanding of THg enrichment mechanisms.

Both objectives emerged from the need for performance measures for the Comprehensive Everglades Restoration Plan that can be monitored to indicate restoration effectiveness with time. Maps of Hg concentrations in surface soils at the scale of the Greater Everglades represent an essential indicator of long-term ecosystem trajectories in response to restoration and changes in atmospheric sources.

MATERIALS AND METHODS

Study Area

Our study area was the Greater Everglades, a subtropical ecosystem in South Florida (Fig 1). Nearly half the historic Everglades has been drained for agriculture and development (Davis and Ogden, 1994); the remainder has been divided by levees into hydrologic units including the EAA, ENP, Big Cypress National Preserve (BCNP), Water Conservation Areas 1, 2, and 3, and several smaller compartments (Holeyland and Rotenberger tracts [HLRB] and the Model Lands tract). For this work, we neglected coastal mangrove forests and areas of the historic Everglades that are no longer wetlands.

Soil Sampling and Laboratory Analyses

This work leveraged samples previously collected for nutrient mapping across the Greater Everglades (Bruland et al., 2006; S. Newman and K.R. Reddy, unpublished data, 2004). Soil samples from 1405 sites were collected via helicopter between May 2003 and January 2004; stratified random sampling by hydrologic partition was used to ensure broad representation of Everglades edaphic conditions. At 132 sites, field triplicates were collected to examine zero-separation-distance variability. At each site, community composition was estimated and used to classify a site into one of six categories (ridge, n = 656; slough, n = 256; wet prairie, n = 404; tree island, n = 24; sawgrass and shrub mangrove, n = 38; depressional marsh, n = 27). Geographic information for all sample locations is available at my.sfwmd.gov/dbhydroplsql/show_dbkey_info.main_menu (verified 24 Dec. 2008) and in Cohen et al. (2007).

Samples were collected using a 10-cm-diameter, medical-grade, stainless steel corer (1 mm thick), sharpened at the bottom to minimize compaction in peat soils, with stainless steel handles (15 cm long by 2.5-cm diameter) welded 10 cm from the top of the tube. The corer was washed using site water before and after each sample. Soils sampled using separate polycarbonate core tubes were brought in contact with the steel corer to determine Hg contamination. Paired comparison of contact and noncontact soils was not significantly different from zero (P = 0.44).

Each core was sectioned into floc and 0- to 10- and 10- to 20-cm increments in the field and the sections were placed into sealed polyethylene bags and stored in coolers on ice until return to the laboratory; only the 0- to 10-cm sections were considered in this work because of (i) cost constraints, (ii) lack of a floc layer at some locations, and (iii) the upper soil profile being most representative of recent conditions. Soil samples were dried at 70°C for 3 d in plastic weigh boats and ground in 20-mL high-density polyethylene scintillation vials with acid-washed ceramic grinding balls before analysis. All samples were analyzed for organic matter (via loss-on-ignition [LOI]), TP, and bulk density (BD), using standard analytical methods at the Wetland Biogeochemistry Laboratory, University of Florida (Corstanje et al., 2006; Bruland et al., 2006; S. Newman and K.R. Reddy, unpublished data, 2004). Briefly, TP was measured using the absorbic acid procedure (Method 365.1, USEPA, 1993) using an Autoanalyzer II (Technicon, Terrytown, NY) after sample ashing at 550°C and hot acid digestion. Loss-on-ignition values were obtained by mass loss measurements following ashing. Bulk density was obtained from the dry weight of the sample divided by its corer volume.

A subset of 600 samples from the total data set was analyzed in June 2005 for THg_M (mg Hg kg^{-1} soil). These samples were selected from the population based on the Latin hypercube subsampling (LHS) protocol and using principal components axes derived from the basic soil biogeochemical factors to define the sampling space; LHS ensures end-member inclusion in the subset, but with 600 samples was not judged to introduce significant subsampling bias. The THg_M concentrations in the subset samples were determined following an acid digestion of a preweighed dry sample (~1 g) with a mixture of concentrated HCl, HNO_3, and HF in acid-cleaned and marble-capped volumetric flasks (Hossner, 1996; Donkor et al., 2005). Samples were heated overnight to a refluxing boil on a hot plate, and diluted with NANOpure water (Thermo Scientific Barnstead, Dubuque, IA) after cooling to a known final volume. Mercury concentrations in the resulting digest were analyzed by $SnCl_2$ reduction, dual Au amalgamation, and detection by cold vapor atomic fluorescence spectrometry (Bloom and Crecilius 1983). Quality assurance/quality control criteria were met by the use of reagent blanks, standard solutions, and a certified reference material (IAEA-405).

We examined THg_A by adjusting observed concentrations (in mg kg^{-1}) by bulk density (kg m^{-3}) for the 10-cm soil profiles. Other researchers working with the same data (Bruland et al., 2006) reported concentrations of soil analytes per unit mass only, principally due to relative homogeneity in bulk density across their smaller study area (WCA3A). Across the Greater Everglades, where bulk density values

vary from 0.03 to 1.88 kg m^{-3}, inference from mass alone may be problematic. While uncertainty in BD is compounded by the lack of quality assurance techniques during field sampling, we assumed that the coring device used effectively limited compaction. A lowCV (14%) between BD measurements at field triplicate sites supports this assumption. All interpolations, correlations, semivariances, and cross semivariances were computed per unit mass (THg_M, mg kg^{-1}) and per area (THg_A, mg m^{-2}).

Exploratory Analyses

Total Hg observations were summarized as a function of hydrologic partition (i.e., water conservation areas), distance from canals (as evidence of surface hydrologic delivery), and plant community type (as evidence for biotic inducement of differential enrichment); covariance with TP, BD, and LOI was also examined to better understand the local context for any observed spatial pattern. All exploratory analyses were done using Statistica 7.0 (Statsoft, Tulsa, OK).

Summary by hydrologic partition was done to examine large-scale spatial patterns. Spatial autocorrelation precludes consideration of individual points as independent, so contrast statistics (e.g., ANOVA) across large hydrologic partitions were not done.

Summary as a function of distance from a canal was motivated by an understanding of P enrichment, a primary anthropogenic influence impacting ecosystem structure and function in the Everglades (Noe et al., 2001). Because the distribution and enrichment mechanisms for P are relatively well understood (i.e., solute transport in canals; Reddy et al. 1991; Bruland et al., 2006; S. Newman and K.R. Reddy, unpublished data, 2004), associations between THg_M and TP concentrations can be used to assess any evidence for hydrologic hotspots; note that this does not assume coupled biogeochemical processes (e.g., as with S; Ullrich et al., 2001), only broad association via an enrichment mechanism. We examined global covariance (THg vs. TP, LOI, and BD) and compared concentration profiles with increasing distance from the canals. The former is based on observations (Vaithiyanathan et al., 1996) of strong negative covariance of TP and THg along a P enrichment gradient in WCA2A; evidence for similar covariance at the landscape scale is possible with these data. For the latter analysis, TP was expected to decrease with distance from canals, determined using a geographic information system buffer analysis. If THg_M followed a similar pattern, it would suggest that surface water delivery was the principal enrichment mechanism. Hydrologic flowpaths from canals to sample locations can be indirect (e.g., if a levee impedes flow), which would generally confound the relationships between distance and concentration such that the absence of correlation would be ambiguous; where significant correlation with distance is observed, however, this flowpath uncertainty is of less concern since the correlation sign is more important than the magnitude.

Summary of enrichment patterns by vegetative community type was done using a multiple comparisons Kruskal–Wallace ANOVA because assumptions of variance homogeneity were significantly violated. Sufficient sample size was available only for contrasts of three marsh communities in the Everglades (ridges: monotypic stands of *Cladium jamaicense* Crantz; sloughs: emergent, submerged, and floating leaved aquatics; and wet prairies: emergent graminoid domination). Global differences in mean value by community are potentially confounded by the spatial distribution of communities; wet prairies, for example, are generally found in ENP and BCNP, and not in deep peat areas of the central and northern Everglades (WCA3A, WCA2A, and WCA1). Moreover, spatial autocorrelation in Hg deposition and peat accretion rates may make comparison of spatially proximate sites more meaningful. Contrasts were developed using paired analyses (paired *t*-test), with pairs defined by sites of different community type separated by <5000 m. Using this criterion, we identified 160 ridge–slough pairs, 49 ridge–wet prairie pairs, and 35 slough–wet prairie pairs for comparison.

Geostatistical Analyses

Semivariance analysis and ordinary kriging (OK) were used to characterize the spatial variation and map THg_M and THg_A; both analytes were log transformed before analysis to meet the requirements of semivariance analysis, and back-transformed to produce the final maps. Spatial dependence was analyzed using semivariograms, which plot semivariance against lag spacing (Webster and Oliver, 2001). Because semivariograms are sensitive to spatial outliers, observations identified as such using Anselin's Local Moran's Index (z score less than −1.96; Anselin, 1995) were omitted from semivariance analyses (although not from exploratory analyses).

Semivariograms were developed both globally and by hydrologic partitions (e.g., WCA-3A). Interpolation cross-validation accuracy (see below) actually declined for local analysis in comparison with global analysis; as a result, we hereafter consider only global semivariograms despite the potential for nonstationary variances even after trend removal (Corstanje et al., 2008).

To identify spatial autocorrelation structure at different scales, semivariograms were developed for lag spacings between 100 and 1000 m. A lag spacing of 500 m was selected based on semivariance stability and meeting the criterion ($n > 30$) of a minimum number of sample pairs within each lag. Model semivariograms consisted of one or two spherical models, with model parameters estimated using ordinary least squares fitting. The structural semivariance, computed as the ratio of partial sill to total sill (= nugget variance + partial sill), was used to evaluate the spatial variance explained by the semivariogram (Morris, 1999). Values approaching 1.0 indicate strong spatial structure while values near 0.0 indicate either low spatial structure or structure at spatial scales larger or smaller than those observed.

Spatial interpolation using OK provides a best linear unbiased estimator because error variance is minimized, predictions are linear combinations of available data, and the mean error is reduced to zero (Isaaks and Srivastava, 1989; Goovaerts, 1997). First-order trend removal was performed before variography to remove nonstationarity; trends were added to the interpolated surface to yield the final maps. Semivariograms were modeled in Variowin (Pannatier, 1996), and kriging was done in Geostatistical Environment Modeling Software (Remy, 2004). Final interpolation was done at a spatial resolution of 200 m.

Cross-validation was performed to estimate model error at unsampled points. Error estimation was made by interpolating using all samples except one, and comparing the predicted and observed values; iterative application of this process until all sites have been "held out" and predicted permits representation of prediction errors without sacrificing data density (Goovaerts, 1997). Prediction quality was assessed using the mean error (ME), RMSE, and r between predicted and observed values.

RESULTS

Total Mercury Subsample Properties

Selected samples were spatially distributed throughout the Everglades, with no evidence for spatial sampling bias to a particular area (Fig. 1). The average minimum distance between

Table 1. Statistical summary of total Hg per mass (THg_M) and area (THg_A) by hydrologic partition and across the Greater Everglades. Smaller hydrologic partitions (Holeyland, Rotenberger and Model Lands tracts) are not shown, but included in the overall values; BCNP = Big Cypress National Preserve, ENP = Everglades National Park, WCA = Water Conservation Area.

Statistic	BCNP	ENP	WCA3AN	WCA2B	WCA2A	WCA3B	WCA1	WCA3AS	Overall
					THg_M, mg kg^{-1}				
n	95	139	61	11	50	19	59	85	600
Mean	0.040	0.111	0.150	0.192	0.205	0.227	0.236	0.294	0.162
Min.	0.007	0.009	0.002	0.054	0.044	0.142	0.015	0.049	0.002
Max.	0.152	0.463	0.312	0.414	0.557	0.377	0.528	0.917	0.917
SD	0.027	0.089	0.064	0.104	0.122	0.066	0.119	0.199	0.141
CV, %	67.2	79.9	42.7	54.5	59.6	29.0	50.2	67.8	87.0
					THg_A, mg m^{-2}				
n	95	139	61	11	50	19	59	85	600
Mean	2.14	1.89	2.37	2.88	2.11	2.98	1.62	3.16	2.25
Min.	0.74	0.12	0.13	1.24	0.50	0.92	0.07	0.51	0.07
Max.	7.37	12.05	6.55	7.48	6.50	7.47	4.22	9.83	12.05
SD	1.07	1.66	1.13	1.82	1.23	1.69	0.84	2.23	1.59
CV, %	50.1	88.1	47.9	63.2	58.1	56.9	52.0	70.5	70.7

samples was 1902 m. In general, samples were sparser in the rocky southeastern region of the ENP and WCA3B (mean distance = 2077 and 2611 m, respectively) and in BCNP (2330 m) than in the other WCAs (WCA1, 1834 m; WCA2A, 1446 m; WCA3AN, 1767 m; WCA3AS, 1905 m). Sample densities in smaller hydrologic zones were also higher (Holeyland, 1175 m; Model Lands, 1210 m; Rotenberger, 1283 m). Overall, the sample density, with >25% of the samples within 1000 m of another site, supports exploration of lag spacings for semivariogram analysis between 100 and 1000 m. Observed means and variances were nearly identical between the selected 600 samples and the 1405 0- to 10-cm samples from the larger data set for TP (mean μ = 375 vs. 385 mg kg^{-1}, standard deviation σ = 231 vs. 237 mg kg^{-1}), total C (μ = 321 vs. 322 g kg^{-1}, σ = 147 vs. 147 g kg^{-1}), and BD (μ = 0.25 vs. 0.25 g cm^{-3}, σ = 0.25 vs. 0.24 g cm^{-3}).

Exploratory Analyses

The mean THg_M across the Everglades was 0.162 mg kg^{-1} (Table 1), with concentrations >0.2 mg kg^{-1} at 168 sites. The mean THg_A was 2.25 mg m^{-2}, approximately 100 times the estimated annual loading rate of 25 µg m^{-2} yr^{-1} given a sampling depth of 10 cm. Note that a 10-cm profile integrates over dramatically different time intervals because peat accretion varies from ~0.1 cm yr^{-1} (background) to as much as 1 cm yr^{-1} with P enrichment. Variances around both THg_A and THg_M means are large, with a CV of roughly 80% for both, and distributions for both are moderately skewed right, necessitating lognormal transformation before semivariogram analysis.

Bulk density values, which were used to compute THg_A, varied dramatically across the Everglades, with mean values in ascending order: WCA1 (BD = 0.09 g cm^{-3}) < WCA2A (0.11 g cm^{-3}), WCA3AS (0.12 g cm^{-3}) < WCA3B (0.13 g cm^{-3}) < WCA2B (0.18 g cm^{-3}), WCA3AN (0.19 g cm^{-3}) < ENP (0.24 g cm^{-3}) < BCNP (0.67 g cm^{-3}).

Variation by Hydrologic Partition

The maximum concentration (THg_M = 0.917 mg kg^{-1}) occurred in western WCA3A (Table 1); 32 sites (5%) had concentrations >0.4 mg kg^{-1}, 15 of which were in WCA3A and 12 of which were in WCA2A and WCA1. Furthermore, while no sites in the BCNP had THg_M > 0.2 mg kg^{-1}, 58% of sites in WCA1 (34 out of 59 sites) and 60% of sites in WCA3AS (51 out of 85 sites) were above that level.

Differences among hydrologic regions for THg_M were large, in part driven by differences in nominal organic matter content (Fig. 2), as expected by the binding potential of organic material vs. mineral soils (Alloway, 1990). Differences in THg_A were far less pronounced; the highest mean values were observed for WCA3AS, WCA3B, and WCA2B, while the lowest values were for BCNP, WCA2A, ENP, and WCA1 (Table 1).

Variation with Soil Properties

Overall correlations of THg_M with other measured biogeochemical parameters (TP, LOI, and BD) were significant

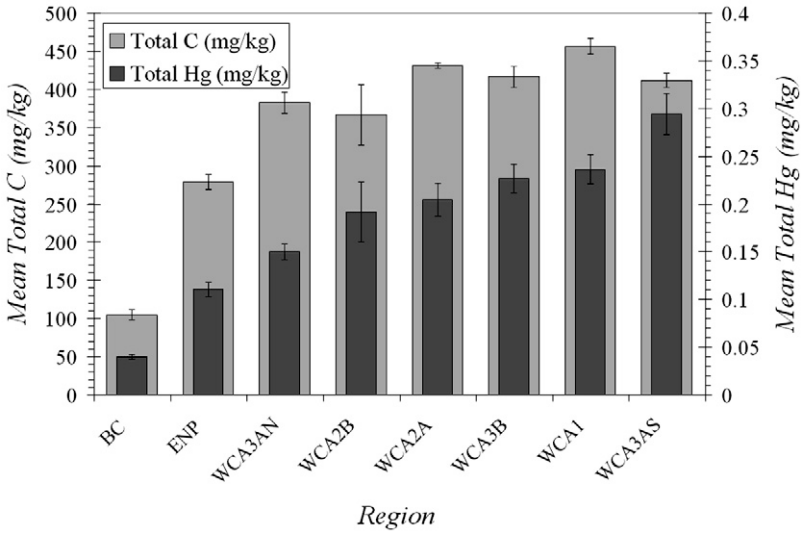

Fig. 2. Mean total C and total Hg per mass (±2 SE) by Everglades hydrologic region (BC = Big Cypress National Preserve, ENP = Everglades National Park, WCA = Water Conservation Area).

Table 2. Pearson correlation coefficient of total Hg per mass (THg_M) and area (THg_A) with biogeochemical properties. Impacted sites are defined as those with total P (TP) > 500 mg kg^{-1}. All variables were lognormally transformed before analysis.

Variable	THg_M	TP	Loss-on-ignition	Bulk density
		Overall (n = 600)		
THg_M	–	0.55**	0.79**	−0.74**
THg_A	0.57***	−0.01	0.06	0.13*
		Unimpacted (n = 461)		
THg_M	–	0.69**	0.81**	−0.77**
THg_A	0.58**	0.16*	0.11	0.07
		Impacted (n = 139)		
THg_M	–	−0.47**	0.63**	−0.51**
THg_A	0.76**	−0.41**	0.12	0.23*

* Correlation significant at $P < 0.05$.

** Correlation significant at $P < 0.01$.

*** Correlation significant at $P < 0.001$.

(Table 2), but correlations between those same variables and THg_A were not, with the exception of BD. The absence of strong correlation between THg_A and principle indicators of soil type and condition (with the notable exception of TP in P-enriched sites) offers some evidence for depositional uniformity in space.

Strong overall THg_M correlations were observed for LOI and BD, with a weaker correlation with TP. We observed a nonlinear relationship between TP and THg_M (Fig. 3), however, with a strong positive correlation below TP concentrations of 500 mg kg^{-1}, but moderate negative correlation above that level. Notably, soil with TP concentrations >500 mg kg^{-1} are considered P enriched (DeBusk et al., 2001). After categorizing sites using a TP > 500 mg kg^{-1} threshold, correlations remained strongly positive with LOI and strongly negative with BD, but reversed in sign for TP (0.69 in unimpacted, −0.47 in impacted sites). Also notable is that the only strongly significant ($P < 0.001$) trend observed with THg_A was a negative relationship with TP in impacted sites. This is more probably due to changes in the TP vs. BD relationship (r = −0.72

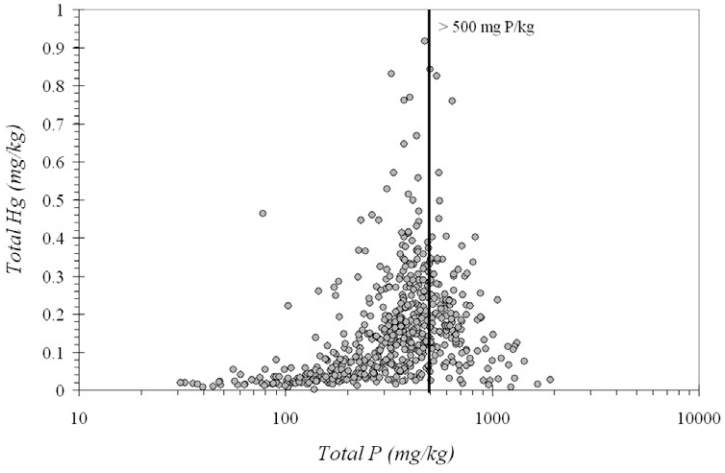

Fig. 3. Relationship between total Hg per mass and total P across the Everglades. An enrichment threshold of TP > 500 mg kg^{-1} corresponds to a value of 6.2 on the lognormally transformed x axis.

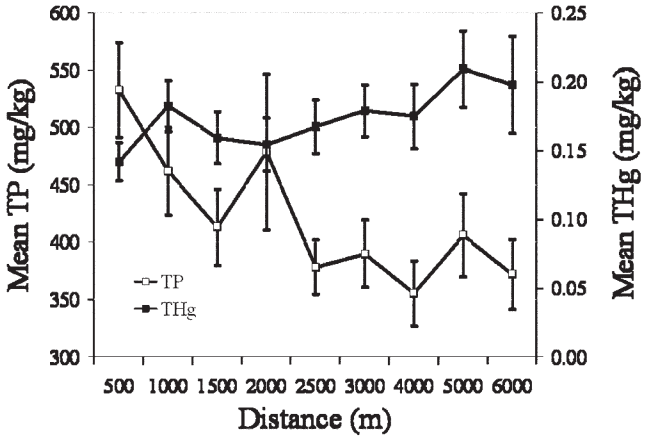

Fig. 4. Mean total P (TP) and total Hg per mass (THg) concentrations with distance from canals; error bars indicate standard errors.

and 0.16 in unimpacted and impacted sites, respectively) than to changes in areal deposition rates.

Variation with Distance from Canal

The relationship between distance from a canal and both THg_M and P suggests opposing concentration gradients (Fig. 4). Total P concentrations were negatively correlated (r = −0.73) with distance from canals, while THg_M was positively correlated (r = 0.77). The association between distance and THg_A was positive but nonsignificant (r = 0.34, P = 0.37), suggesting offsetting trends (decreasing bulk density and increasing THg_M).

Variation by Community Type

Evidence for differential enrichment, presumably by biological processes, was observed among all community types evaluated for THg_M and between slough and wet prairie for THg_A (Fig. 5). These global differences are, however, potentially confounded by larger spatial patterns in both THg loading and the distribution of different community types. Our analysis of paired proximate sites (<5000-m separation) of different community type suggests no significant differences. Paired contrast of proximate ridge and slough sites had a mean difference of 0.011 mg kg^{-1}, which was not significantly different from zero (P = 0.210). Similarly, differences between ridge and wet prairie (0.020 mg kg^{-1}, P = 0.085), and slough and wet prairie (0.016 mg kg^{-1}, P = 0.081) were not significant, although additional power from targeted sampling may be warranted.

Semivariance Analysis and Mapping

Point maps of THg_M and THg_A observations (Fig. 6A and 7A) indicate below average levels of both variables in BCNP and ENP and above average values in the WCAs. The highest THg_M and THg_A contents were found in the northwestern region of WCA3AS. Clear regional differences emerge, with important implications for management and monitoring.

Trend removal revealed systematic declines in THg_M principally from east to west, but also from north to south. Trends in THg_A were less obvious, with a weak declining trend from northwest to southeast. Semivariograms, following trend removal and exclusion of 17 observations regarded

as outliers based on Anselin's Moran *I*, show distinctly different spatial patterns for THg_M and THg_A (Fig. 6B and 7B, respectively). Comparatively low nugget variance was observed for THg_M, while the nugget for THg_A was far larger (Table 1). The range was similar between variables. We observed a clear sill in semivariance at a range of ~40 km, suggesting that a linear semivariogram model (Stober et al. 2001) is inappropriate for interpolation.

Semivariance analysis indicated a strong spatial structure for THg_M, with a relative structure parameter indicating that >80% of the spatial variance is explained by the model semivariogram. The semivariogram for THg_A explained less of the total semivariance (56%), suggesting that spatial structuring was absent or occurring at different scales than our observations. There was no evidence of significant anisotropy in either variable.

Interpolation (Fig. 6C and 7C) shows significant hotspots and strong differences among subregions. The prediction of THg_M (Fig. 6C) shows hotspots in western WCA3AS, northern Holeyland (HL), and southern WCA2A and WCA1. Enrichment zones in the Shark River Slough and coastal marshes in the western ENP were also observed. The hotspot in western WCA3AS had eight soil samples with THg_M concentrations ranging from 0.646 to 0.917 mg kg^{-1}; notably, those same locations had relatively low TP concentrations (325.41–638.96 mg kg^{-1}, average 439.51 mg kg^{-1}), and THg_M and TP levels in these sites were weakly correlated ($r = 0.16$; $P = 0.58$). The map of THg_A (Fig. 7C) shows hotspots in WCA3AS and HL, plus a moderate hotspot in WCA3B; other hotspots observed for THg_M (i.e., WCA1 and WCA2A) are absent.

From the prediction maps, the spatial extent of THg_M and THg_A in quantized ranges (Table 3) indicates that >77% of the area had THg_M values <0.2 mg kg^{-1}, and <2% had concentrations more than double that threshold. This result, combined with our exploratory analyses that suggested that 5% of observations exceeded 0.4 mg kg^{-1}, illustrates the localized nature of severe Hg enrichment. That is, large areas had low THg_M levels, and hotspots were geographically constrained. For THg_A, nearly 60% of the landscape had levels in the upper 10 cm exceeding 100 yr of deposition at contemporary rates (>2 mg m^{-2}).

Cross-validation efficiency for both THg_M and THg_A (Table 4) showed strong agreement between predicted and observed THg_M, but reduced agreement for THg_A. The ME is close to 0 for both, substantiating that OK predictions were unbiased. The RMSE for THg_M is 0.098 mg kg^{-1}; this relatively high error is principally due to six validation sites, two of which were underpredicted and four that were highly overpredicted. A high correlation value ($r = 0.70$) illustrates relatively strong prediction efficiency for most sites. The RMSE

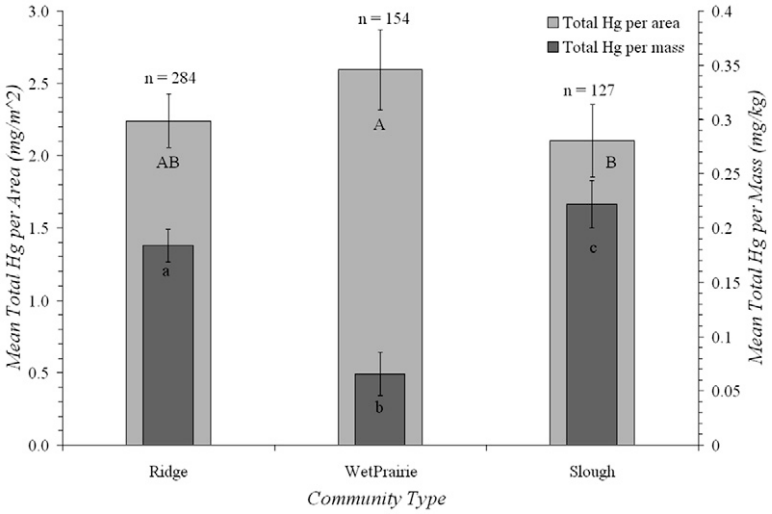

Fig. 5. Mean (±95% confidence intervals) total Hg per mass and per area levels by community type across the Everglades; the sample size for each community is also shown. Different letters denote significant differences ($P < 0.05$) based on a Kruskal–Wallace ANOVA.

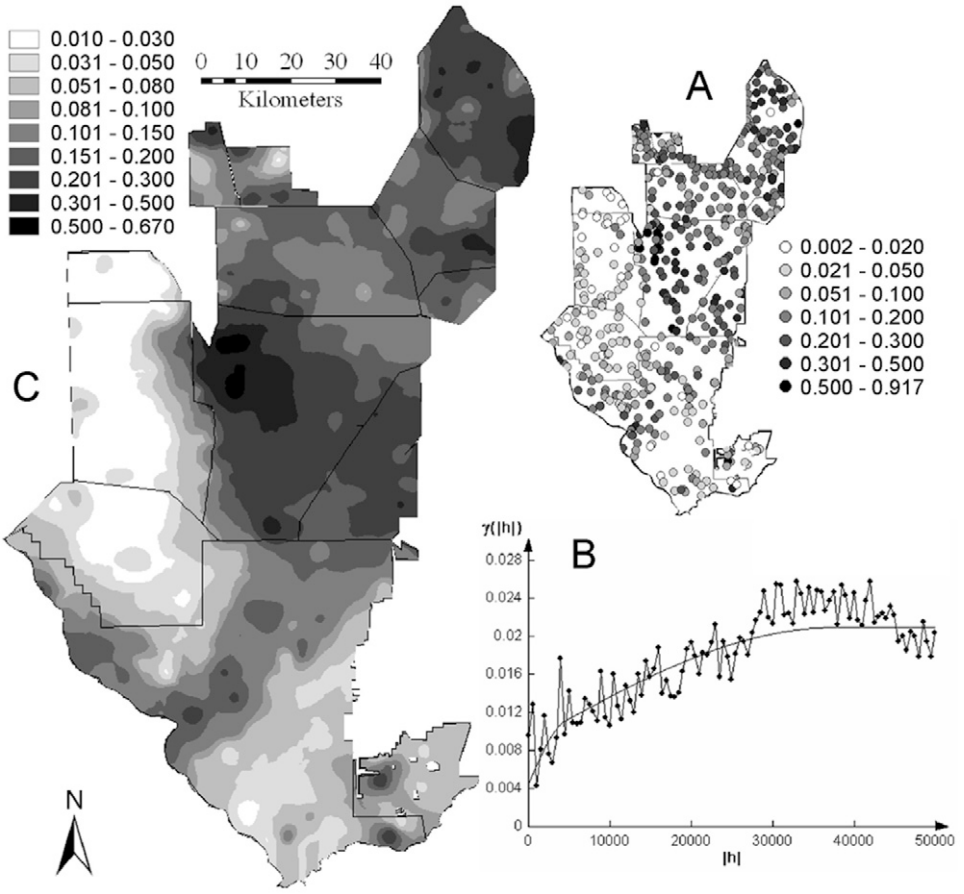

Fig. 6. Total Hg per mass (THg_M, mg kg^{-1}) showing (A) point observations, (B) modeled global semivariograms (500-m lags), and (C) an ordinary kriging prediction map.

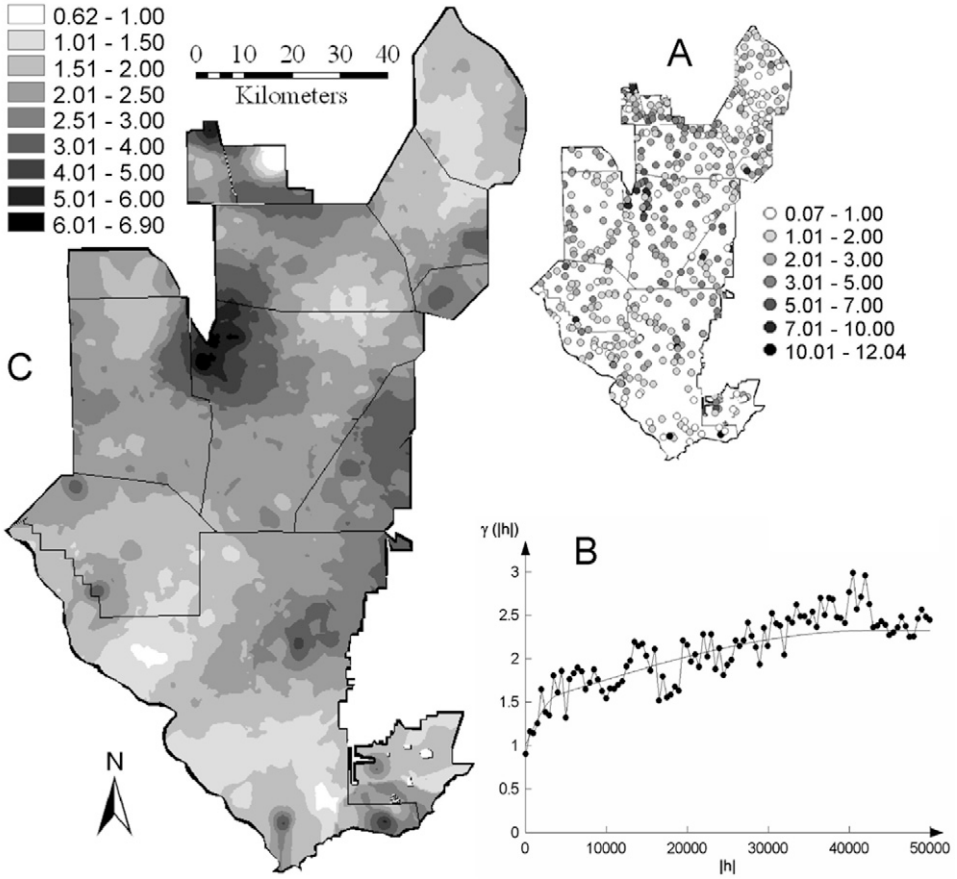

Fig. 7. Total Hg per area (THg_A, mg m^{-2}) showing (A) point observations, (B) modeled global semivariograms (500-m lags), and (C) an ordinary kriging prediction map.

for THg_A was high (1.46 mg m^{-2}) with only a fair correlation (r = 0.45), suggesting less predictable spatial structuring.

DISCUSSION

Most studies of Hg in the Everglades have articulated the processes and effects of methylation and bioaccumulation (e.g., Guentzel et al., 1995; Beyer et al., 1997; Cleckner et al., 1998; Cai et al., 1999; Sepulveda et al., 1999). Studies of spatial enrichment patterns have generally been highly localized (Rood et al., 1995; Arfstrom et al., 2000) with insufficient data to provide regional hotspot information critical to risk management and restoration monitoring. One important exception (Stober et al., 2001, conducted in 1996) examined regional Hg spatial patterns in the soil as part of a multistressor assessment and found significant variability across space and, more importantly, time. While that study made simplifying assumptions about the character of spatial structure and offered no estimate of prediction errors for direct comparison, the spatial patterns observed were generally consistent with our findings. The most notable difference was the range of observed values; Stober et al. (2001) observed value between 2.4 and 330 μg kg^{-1}, while we observed values between 2 and 917 μg kg^{-1} for approximately the same number of samples (n = 731 vs. 600, respectively). Despite these differences in concentration, coarse regional patterns (i.e., averages by hydrologic partition) followed the same trends, with low values in the ENP and markedly higher values in WCA1 and 2. Lower THg_M values in the ENP are possibly due to the dominance of calcitic soils, which have low metal cation affinity, rather than peat (Rood et al., 1995), which has very high affinity (Drexel et al., 2002). Furthermore, there is strong agreement in the concentration frequency distribution, with median values of 0.120 and 0.126 mg kg^{-1} in the previous study and our study, respectively. The 1996 and current study disagree to the maximum extent in WCA3AS, where we identified a major hotspot that was previously present but substantially less severe, and in WCA3AN, which we observed to be moderately enriched (mean THg_M = 0.150 mg kg^{-1}) and was observed to be unenriched previously (mean THg_M = 0.093 mg kg^{-1}). Another recent study to examine Hg spatial patterns in more detail (Liu et al., 2008) was principally focused on mass budget work; while they reported a weak declining trend in soil THg_M from north to south, the absence of east–west information makes comparison between their data and the current work difficult.

Table 3. Spatial coverage of ordinary kriging predictions of total Hg per mass (THg_M) and area (THg_A) across the Everglades.

THg_M		THg_A	
Range	Coverage	Range	Coverage
mg kg^{-1}	%	mg m^{-2}	%
0.01–0.02	3.2	0.62–1.00	1.7
0.02–0.10	37.7	1.00–1.50	17.1
0.1–0.20	36.4	1.50–2.00	22.5
0.2–0.30	18.7	2.00–3.00	40.4
0.3–0.40	2.7	3.00–4.00	13.6
0.4–0.50	0.8	4.00–5.00	3.9
0.5–0.67	0.5	5.00–6.90	0.9

One potential limitation of our analysis is our inclusion of only the top 10 cm of soil. The annual mass storage in other pools is small, and Hg burial rates below the top 10 cm are <8% of the annual deposition mass (Liu et al., 2008). Arfstrom et al. (2000) reported concentrations (ng cm^{-3}) with depth that strongly support our assumption that recent atmospheric Hg enrichment remains in the upper soils (typically <5 cm). We also elected not to analyze floc, in part because it is not present at all sites; excluding this component, which is strongly seasonal and highly labile, is supported by the observation in Liu et al. (2008) that a comparatively small fraction (3.5% in the wet season, 0.7% in the dry season) of deposited Hg goes to this pool.

Nominal levels of THg_A are also comparable with previous work. Stober et al. (2001) observed mean THg_A of roughly

1.8 mg m^{-2}; mean values were high in the ENP (Shark Slough, ~2.5 mg m^{-2}; Taylor Slough, 2.2 mg m^{-2}) and WCA3A (2 mg m^{-2}) and low in WCA1 (1.4 mg m^{-2}) and WCA2 (1.7 mg m^{-2}). Values in our study were generally higher (2.27 mg m^{-2}), with uneven agreement in spatial pattern; higher values were observed in WCA3A (3.05 mg m^{-2}), WCA2B (2.88 mg m^{-2}), and Rotenberger (2.81 mg m^{-2}), with low values in WCA1 (1.62 mg m^{-2}), the ENP (1.88 mg m^{-2}), and BCNP (2.14 mg m^{-2}).

Table 4. Semivariogram parameters for total Hg per mass (THg_M) and area (THg_A) and ordinary kriging cross-validation error statistics.

Variable	Semivariogram parameters						Error statistics		
	Nugget	Model 1†		Model 2‡		Q§	ME¶	RMSE	r
		Partial sill	Range	Partial sill	Range				
			m		m	%			
THg_M	0.004	0.005	4500	0.012	37,500	81	0.0006	0.098	0.70
THg_A	1.009	0.426	4000	0.858	43,000	56	0.0024	1.461	0.45

† Parameters for first spherical model.

‡ Parameters for second spherical model.

§ Relative structure (partial sill/total sill).

¶ ME, mean error (Schloeder et al., 2001).

The total mass of Hg in the upper 10 cm previously computed (Stober et al., 2001) across the study area (5500 km^2) was approximately 11,000 kg. Our work, which includes the Model Lands, Holeyland and Rotenberger tracts and BCNP (total area = 8221 km^2) indicated a storage of 18,184 kg of Hg in the upper 10 cm; across the same area as the previous study, we estimate 13,097 kg of Hg. While evidence of a mass increase may be in part due to sampling or interpolation errors, this result suggests, at least, that recent declines in deposition rates have yet to be expressed in the soil pool. Since soil stores >99% of Hg in the system, it would be expected to respond slowly, particularly given relatively small mass loss rates via evasion and surface outflow (Liu et al. 2008).

The THg_A observations lack strong spatial structure, with local exceptions; the prediction efficiency of semivariogram models for THg_A underscore this conclusion and suggest that deposition is relatively uniform in space. What drives stronger spatial structuring in the THg_M observations, therefore, is strong spatial patterns in soil C (production and storage), which in turn reflects hydroperiod and nutrient enrichment. This inference is supported by observations of strong covariance between THg_M and LOI globally.

Short-range spatial variability was comparatively high (~20%), indicating measurement error, or processes at the pedon or community scale that lead to local variation. Working across a smaller extent and grain, Delfino et al. (1994) reported a wide range of Hg concentrations (0.01–0.275 mg kg^{-1}) in soil cores collected from 36-m^2 grids in WCA3A. Spatial variability [the difference between paired samples: $([Hg]_a - [Hg]_b)/[Hg]_{mean}$] for separation distances of 0.1, 1, 2, 3, and 4 m was 17, 20, 42, 50, and 34%, respectively (Delfino et al., 1994).

One possible explanation for such high local variability is that different vegetative communities, which form a mosaic across short ranges (~50 m) due to differences in soil elevation, accumulate Hg in different ways. Based on our data, however, there is little evidence to support this explanation. While there are strong differences evident between communities at the regional scale, when geographically proximate samples are paired for comparison, differences are nonsignificant. Without locally strong community effects, any global effects are more likely a function of regional deposition or peat oxidation and accretion conditions than indicators of a differential risk of Hg enrichment among ecotypes. We also suggest, however, that more detailed sampling of paired sites would be necessary to reasonably reject the hypothesis of community effects on enrichment. The most probable trend emerging from the analysis of paired data suggests that sawgrass ridges have higher concentrations than wet prairies but lower than sloughs, which is difficult to explain given the intermediate hydroperiods of wet prairies vis-à-vis ridges and sloughs. Global differences in THg_A between communities are also significant; what are particularly notable about these differences are the comparatively high levels in ridges vs. both sloughs and wet prairies.

Assuming that atmospheric deposition rates are relatively uniform, hotspots that were observed in THg_A must be due to other processes. One possibility is the delivery of Hg in canal water from the EAA. Phosphorus enrichment due to agricultural runoff (from the EAA) in the northern Everglades is well documented, with soil concentrations in WCA2A and WCA3AN as much as five times higher than in unenriched locations (Reddy et al., 1991; Craft and Richardson, 1993; DeBusk et al., 2001). Previous work (Vaithiyanathan et al., 1996) showed a strong negative correlation, however, between TP and THg_M along a P enrichment gradient in WCA2A; our data confirm their findings at the regional scale. In particular, TP decreases with distance from canals, indicating that canals are the principal P source, while THg_M increases with distance. This effect probably occurs because of stimulatory effects of TP on soil accretion; given relatively even Hg deposition rates, the increased accretion will reduce the mass of Hg per mass of soil. Furthermore, we observed a nonsignificant but positive correlation (r = 0.34) between THg_A and distance from canals throughout the Everglades, and corroborated that observation locally in western WCA3AS. These contrasting trends for P and Hg suggest that canal transport is unlikely to be source, a conclusion that reinforces the findings of Arfstrom et al. (2000). Measurements of Hg in EAA canals (Atkeson et al., 2003) support the supposition that a negligible fraction (~2%) of Hg arrives from canal runoff.

Covariance between THg_M and TP is nonlinear. At both TP concentration extremes, we observed low THg_M values, with a marked peak at intermediate TP values. At TP values <500 mg kg^{-1}, strong positive covariance between TP and THg_M (r = 0.69) was observed, a trend that reversed above that threshold (r = –0.47). Furthermore, our data (Fig. 3) suggest low variance in THg_M at both high (>1000 mg kg^{-1}) and low (<300 mg kg^{-1}) TP levels, and markedly higher variance at concentrations around 500 mg kg^{-1}. This confirms previously observed asymmetric variances at the local scale in

northern WCA2A (Vaithiyanathan et al., 1996, Fig. 3), where the TP–THg_M correlation was strongest at concentrations exceeding 1000 mg kg^{-1}.

An alternative source of the relative enrichment of THg_A in parts of the ENP, WCA3AS, WCA3AN, WCA3B, Holeyland, and WCA2A is the loss of peat due to subsidence or oxidation. Stober et al. (2001, Fig. 6.38) reported changes in soil elevation between 1946 and 1996. Concordance between areas of substantial soil loss (>30-cm decline in elevation, principally in northern WCA3A, eastern WCA2A, southern WCA3B, the northeastern ENP, and HLRB) and the regions of elevated Hg, especially THg_A, is compelling; in particular, regions of high current THg_A are within or downstream of areas where major subsidence has been documented, although with a few important exceptions (e.g., eastern WCA1). This may suggest that Hg enrichment ensues from peat oxidation and subsequent incorporation into the surface soils. As such, considerations of internal load, particularly under modified hydrologic conditions that change the wetland C balance, are of considerable research importance.

Mercury emissions rates in South Florida have declined from 3000 kg yr^{-1} in 1991 to an estimated 250 kg yr^{-1} in 2000, primarily due to the reduction in municipal solid waste combustion and medical waste incineration (Atkeson et al., 2003); Liu et al. (2008) estimated an annual deposition of <140 kg yr^{-1} across the Everglades. Despite these improvements, we observed little evidence to suggest that total Hg concentrations or mass are declining. Indeed, our findings suggest that THg_M remains significantly elevated in areas, but unevenly distributed throughout the Everglades. The average THg_M value was 0.156 mg kg^{-1}, and all samples fell toward the lower end of the range of concentrations (0.01–4 mg kg^{-1}) for organic soils (Alloway, 1990). These findings are consistent with those reported by Rood et al. (1995), where surveyed soil cores from the Everglades showed the highest surface Hg concentrations in WCA1 and parts of WCA2 and southern WCA3A, and with Arfstrom et al. (2000), who reported spatial patterns of THg for lower WCA3AS.

As important as the total and average mass estimates could be for future monitoring of restoration progress, the spatial pattern of enrichment is relevant to contemporary management. While the principal concern for Hg bioaccumulation and toxicity arises primarily from its occurrence in natural systems as MeHg, there is evidence for relatively strong covariance between regional THg_M hotspots identified in this study and MeHg levels in periphyton and mosquito fish (*Gambusia affinis*) observed previously (Stober et al., 2001). Furthermore, path analyses suggest that, in some areas, soil THg_M concentrations are the best predictor of soil MeHg (Stober et al., 2001). If mineralization of organic-bound Hg following hydrologic modification has played an important role in the Hg budget in recent decades, hydrologic restoration coupled with reduced atmospheric Hg inputs should begin to lower surface soil concentrations and reduce eco-toxicological risks.

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