MERCURY IN BURIED LOGS OF AN HISTORIC CEDAR STAND FROM THE GREAT DISMAL SWAMP NATIONAL WILDLIFE REFUGE

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Abstract: Atmospheric deposition of mercury, a known neurotoxicant, occurs in remote locations including peatlands such as the Great Dismal Swamp. Little is known about coarse woody debris as an ephemeral mercury pool, though management actions could influence remobilization that occurs via combustion and microbial oxidation. Previously buried logs were exposed after peat burning associated with the 2008 South-One Fire and cross-sectional log samples were retrieved and analyzed. Mercury concentration increased logarithmically with increasing proximity to the nearest edge in contact with the peat. Mean mercury concentrations at the edge of the logs (0.0295 mg Hg/kg dry weight (DW) of sample) were higher than the two inner portions (0.004 and 0.003 mg Hg/kg DW sample, respectively). Mercury accumulation and retention in buried tree boles represents a significant pool of mercury that should be considered in hydrology management given that lowered water tables increase risks of remobilization.

Key Words: Mercury, Atlantic White Cedar, peatlands, Great Dismal Swamp, dendrochemistry, remobilization

INTRODUCTION

Mercury, a ubiquitous environmental toxicant of global concern, exists in several forms and the mobility and toxicity of mercury varies among these chemical species (USEPA 2009). Conversion of inorganic mercury to the more toxic form, methylmercury, is mediated by sulfate-reducing bacteria in anoxic soil conditions that are prevalent in wetlands (Branfireun et al. 1999). The addition of the methyl group makes the toxicant lipophilic, allowing it to pass readily through biological membranes into cells or tissues. Wetland microorganisms bioaccumulate methylmercury that is subsequently biomagnified (Benoit et al. 2002).

Methylmercury, acting as a neurotoxicant, can cross the fetal blood-brain barrier and lead to decreased intelligence and neurological disorders (USEPA 2009). Minamata disease was an extreme instance of methylmercury poisoning. Effluent from an acetaldehyde manufacturing industrial plant that contained mercury was released from 1932-1962, affecting Minimata Bay and the Japan Sea (Ministry of the Environment 2002, Bargagli 2005). Minamata disease was first recognized in 1956, and adults and children who consumed contaminated fish and shellfish suffered from a variety of symptoms including convulsions, numbness, and speech and walking impediments. In addition, newborns had physical and mental deficiencies and often died within a few years of birth (National Institute of Minamata Disease 2001).

There are many sources of mercury to the environment including combustion of fossil fuel from residential, power plant, and transportation activities; biomass burning; pig iron and cement manufacturing; copper, lead and zinc smelting; mercury mining; gold extraction; and caustic soda production (Streets et al. 2009). The amount of mercury emitted into the atmosphere and incorporated into the biosphere has increased since the beginning of the industrial revolution (Schuster et al. 2002). Ice cores (Schuster et al. 2002, Bargagli 2005), peat cores (Jensen and Jensen 1991, Martínez-Cortizas et al. 1999, Steinnes and Sjøbakk 2005), and sediment cores (Cooke et al. 2009) all suggest that industrialization has greatly increased mercury levels. In 2009, Streets et al. (2009) projected global emissions to increase 96% by 2050 if coal-fire power plants are not equipped with improved control technologies.

Relatively undisturbed ecosystems, distant from any point-source, are also subject to atmospheric deposition of mercury (Graydon et al. 2008). Trees incorporate elemental or inorganic mercury from the atmosphere into leaves through stomata, or direct deposition onto leaf surfaces (Ericksen et al. 2003, Witt et al. 2009a). Additional factors that can increase the total mercury load of a landscape beyond atmospheric deposition levels include fluctuating water tables in reservoirs and wetlands (Driscoll et al. 2007), forest vegetation, and soil characteristics which can cause vegetative biomass to serve as an ephemeral mercury pool (Nater et al. 1999, Friedli et al. 2009b, Witt et al. 2009a).

Within ephemeral mercury pools, the proportion of mercury found in vegetation and soil depends on climate and organic matter dynamics (Friedli et al. 2009a). In most ecosystems, mercury resides in the vegetation (Friedli et al. 2009a). However, in peatlands such as the Great Dismal Swamp (GDS), which form where primary productivity exceeds decomposition (Poulter et al. 2006), most of the mercury content is in the peat (Grigal 2003, Friedli et al. 2009a, Woodruff and Cannon 2010). The amount of mercury and carbon stored in soil depends on the amount of time allowed for accumulation between fires (Friedli et al. 2009b).

The GDS may have changed from a net sink to a net source of mercury. Historic peat accumulations exceeded 6 m during the most recent 10,000 years (Oaks and Whitehead 1979) and low soil pH (3.3-3.6, Thompson et al. 2003) may have enhanced accumulation rates.

Saturated conditions are thought to have persisted longer in the past, which may have promoted wetland soil conditions favorable for peat and mercury accumulation (Woodruff and Cannon 2010). However, saturation periods began to shorten when George Washington and others conducted ditching operations in the mid 18th century (Atkinson et al. 2003a). Drier peatlands are more likely to burn and to release sequestered mercury (Witt et al. 2009b), and the GDS may now function as a net source of mercury.

Fires in the Great Dismal Swamp National Wildlife Refuge (GDSNWR) are frequently started by lightning, but the depth of burns was limited historically in the pre-ditched conditions which were associated with high water levels (Atkinson 2003b). Ditches in GDSNWR lowered water tables and in 2008, the South-One Fire burned to a depth of approximately one (1) m (USFWS 2009). The South-One Fire, one of seven fires that year, was the longest burning in Virginia's recorded fire history and cost more than $12 million to extinguish, burning 2,000 ha in 121 days (USFWS 2009).

Atlantic white cedar, *Chamaecyparis thyoides* (L.) B.S.P. (cedar), swamps are a type of temperate peatland ecosystem found within the GDSNWR and along the Atlantic Coast (Korstian 1924). Cedar swamps are composed of dense, even-aged, monotypic stands of cedar (Laderman 1989) and shade-intolerant seedlings that require an overstory-clearing perturbation event (Little 1950). These ecosystems accumulate undecomposed plant remains including buried logs. Zimmermann and Mylecraine (2003) described an unearthed population of stumps from a cedar forest that was radiocarbon dated to between the 8th and 10th centuries. An early US Geological Survey report on the GDS described the log layer in the peat, especially from cedar trees, as so dense as to be prohibitive for peat mining (Davis 1907).

Given the strong relationship between organic matter accumulation and mercury content (Friedli et al. 2007), peatlands with buried large woody debris that is resistant to decomposition have the potential to sequester additional mercury content in the buried boles, or main trunks, of trees. Given the extensive presence of buried logs in GDSNWR, the scarcity of studies that consider mercury content of buried logs (Grigal 2003) and the risk for re-emission in the event of peat fire, knowledge of mercury content of buried logs would allow development of a more accurate assessment for mercury cycling and release. The purpose of this study is to quantify mercury concentrations of buried logs in GDSNWR to enhance mercury biogeochemical modeling in peatlands, and to improve understanding of the potential for remobilization.

METHODS

Site Description

The GDSNWR is a temperate peatland located between Chesapeake and Suffolk, Virginia, and northeastern North Carolina, USA (figure 1) and was established by the Great Dismal Swamp Act of 1974 for the purpose of "*Protecting and preserving a unique and outstanding ecosystem, and perpetuating the diversity of animal and plant life therein"* (Lowie 2009). The GDSNWR encompasses over 45,000 ha of its larger, former historical acreage. Pollen analysis conducted by Whitehead (1972) and Whitehead and Oaks (1979) suggests that the GDSNWR was a Holocene landscape feature, approximately 10,000 years old, with cedar and Bald cypress (*Taxodium distichum*), appearing 6,500 years ago and becoming dominant 3,000 years ago.

Cedar trees have shallow root systems and are subject to blow-down (Little 1950) from events such as Hurricane Isabel in 2003. Following the cedar stand blow-down, salvage logging was conducted to facilitate natural regeneration through increased exposure of light to the seed bed (in areas referred to as salvage units). However, the South-One Fire (2008) in the GDSNWR burned through the peat to an estimated average depth of one (1) m and exposed previously buried logs.

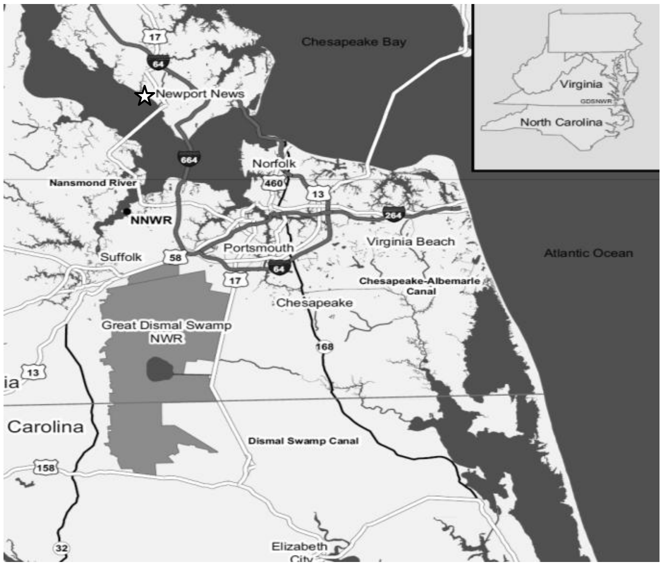


Figure 1. The Great Dismal Swamp National Wildlife Refuge location. Study site location is starred (Great Dismal Swamp National Wildlife Refuge 2006).

Sample Collection

Ground reconnaissance for exposed, previously buried logs was conducted in the summer and fall of 2009 and 2,023 ha (5,000 acres) of the GDSNWR that burned during the 2008 South-One Fire were inspected using 21.5 km-long North/South oriented transects that were spaced 143 m apart (figure 2). Buried log GPS locations were recorded for subsequent recovery. During retrieval, 32 buried log cross-sectional samples (cookies) were collected by chainsaw at the most intact cross-section location. Logs that displayed no evidence of human disturbance and remained partially buried or laid beneath a tree stump were selected (figure 3). Retrieval was suspended in fall 2009 due to high water levels and by log concealment resulting from vegetation regrowth in spring 2010. A subset of nine tree cookies, three from each of three salvage units, was selected for mercury analysis (figure 4). Selection criteria included documented orientation in the soil and a convex surface in contact with the peat.

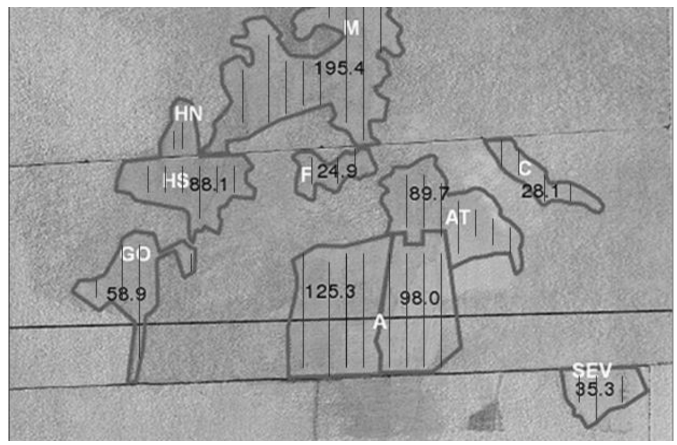
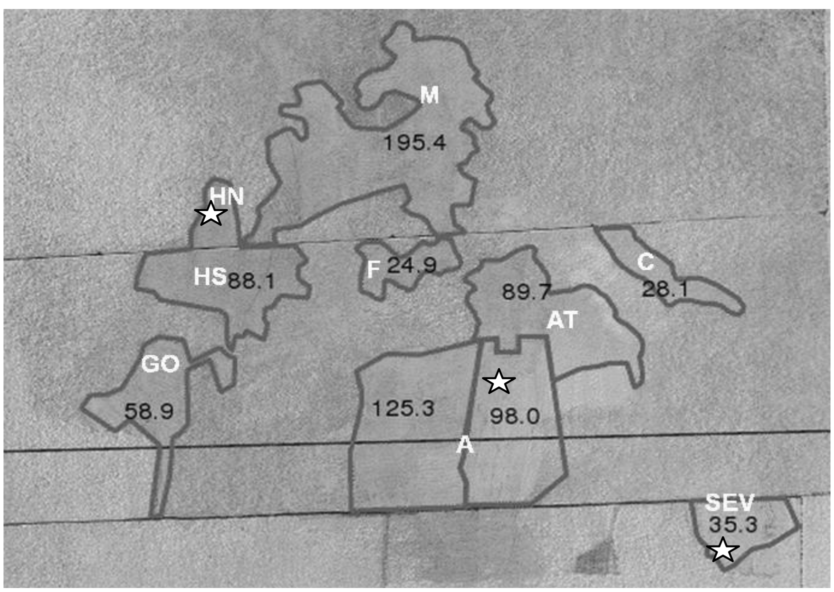


Figure 2. Transects surveyed for buried logs in summer and fall 2009 throughout 300 hectares (750 acres, acreage written in black, salvage unit names written in white) of the Great Dismal Swamp National Wildlife Refuge.

Figure 3. Example of buried logs exposed from a peat fire in the Great Dismal Swamp National Wildlife Refuge. Some logs were beneath a stump (left) or were partially buried (right).

Figure 4. Starred retrieval locations, within the Great Dismal Swamp National Wildlife Refuge, of the subset of nine buried logs analyzed for mercury (initials representing salvage unit names are in white and acreage is in black).



Sample Preparation

Longitudinal, radial xylem wood samples (1 cm x 0.4 cm x 0.4 cm) were collected at three locations within each cookie. Using clean techniques, samples were retrieved via 1.2-cm diameter chisel at three positions including the outermost edge of the cookie (effectively 0 cm from the edge and in contact with peat, “Edge”), near the outermost edge but not in contact with peat (1-4 cm inward from the outermost edge ,“Outer”), and at a cross section furthest from any point in which the cookie would have been exposed to either fire/atmospheric or soil influences

("Middle"). The Middle sample was always located in a portion of the bole that would be formed from when the tree was younger, although ring dating was not performed. Samples were collected with reference to distance to the nearest edge in contact with the peat, inconsistent regarding tree rings or tree age. Samples were frozen overnight and freeze-dried for 48 h.

Analysis

Mercury was analyzed among the three log positions using a Direct Mercury Analyzer-80, an atomic absorption instrument (Milestone, Shelton, CT) at the Virginia Institute of Marine Science. Analysis was performed in triplicate according to a modified USEPA Method 7473 (USEPA February 2007). Spike percent recovery was not performed; rather, standard reference materials were included once every 30 trials or at the beginning and end of each testing session and blanks were included once every five samples. Background signal variation was determined using empty sample boats (n = 23) to establish the limit of detection (LOD) and limit of quantitation (LOQ) (table 1). Two standard reference materials (SRMs) were used in this study (table 2). A curvilinear calibration curve was calculated using SRM 1575a (figure 5).

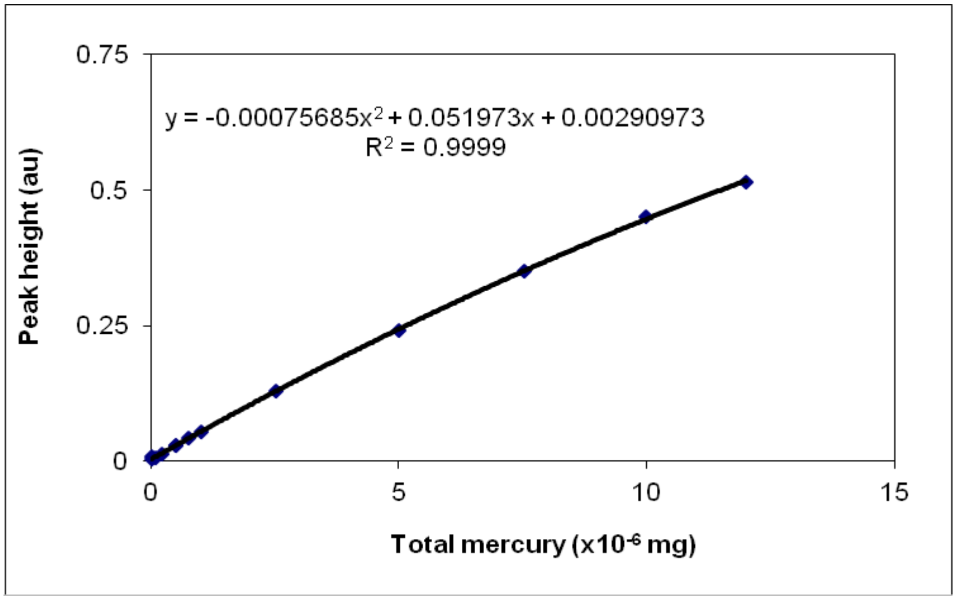
Table 1. Detection and quantitation limits determined for atomic absorption testing using empty sample boats.

	Amount of Hg (ng)
Mean blank reading (n = 23)	0.1215866 + 0.0087235 (1 SD)
LOD (mean + 3 * SD ng)	0.1478
LOQ (mean + 10 * SD ng)	0.2088

Table 2. Mean percent recovery and standard deviation (SD) in two sets of replicates for weights of the NTIS standard dogfish tissue material (DORM-3 NIST SRM, 0.382 $\pm$ 0.007 mg Hg/kg standard).

Weight of standard	Mean percent recovery	SD	n
0.2 ng Hg	100.23	8.3	7
0.5 ng Hg	110.84	2.3	5

Figure 5. Standard curve constructed using solid, ground pine needles (NIST SRM 1575a, 0.0399 $\pm$ 0.007 mg Hg/kg standard).



This standard material is a solid made from dried ground pine needles purchased from the National Institute of Standards and Technology (NIST). It has an accepted mercury concentration of 0.0399 $\pm$ 0.007 mg Hg/kg standard. For percent recovery, DORM-3, another NTIS standard material (dogfish muscle) was purchased from the NIST and had an accepted mercury concentration of 0.382 $\pm$ 0.007 mg Hg/kg. Measurements were deemed in control if the error remained within 15% for this NIST standard reference material (SRM) 2976. Percent recovery and standard deviation were calculated at 2×10^{-7} mg Hg and 5×10^{-6} mg of mercury using DORM-3 (table 2). All instrument responses measured during testing on buried logs were greater than the limit of quantitation (0.006374 ng, table 1) and between total mercury content of DORM-3 SRM analyzed at 2×10^{-7} mg Hg and 5×10^{-6} mg Hg, at 0.0135 and 0.272 ng, respectively (table 2).

Data were analyzed in SigmaStat 9 (San Jose, CA, 2007) and plotted in Excel (Microsoft 2001, Redmond, WA). A natural logarithmic linear transformation was performed on mercury concentration and used for further analysis. The effect of distance of samples taken at the three log positions, to the nearest log edge in contact with peat on mercury concentration was examined using a Pearson Correlation Coefficient. Effect on mercury concentration of salvage units, sample position, or interaction between these two factors was established with a two-way ANOVA significance test with a post-ANOVA testing with Holm-Sidak post hoc test.

To assess if a mercury concentration varied with position in logs, the concentration gradient was calculated from positions Edge to Outer, and positions Outer to Middle (Equations 1 and 2, respectively), and gradients were compared with a t-test.

$$G_1 = (c_E - c_O) / (d_E - d_O) \qquad (1)$$

where G_1 is concentration gradient 1, c_E is the mean Edge concentration (mg Hg/kg sample), c_O is the mean Outer concentration (mg Hg/kg sample), d_E is the mean distance (cm) from peat contact to Edge sample location, and d_O is the mean distance (cm) from peat contact to Outer sample location.

$$G_2 = (c_O - c_M) / (d_O - d_M) \qquad (2)$$

where c_M is the mean Middle concentration (mg Hg/kg sample), and is the d_M = mean distance (cm) from peat contact to Middle sample location.

RESULTS

Mercury concentrations ranged from 0.0023 $\pm$ 0.0007 to 0.0355 $\pm$ 0.0143 mg Hg/kg sample (table 3). Mercury concentrations for log positions directly in contact with peat (Edge) were higher than those at other positions (Outer and Middle, $p < 0.05$)(figure 6). Mercury concentrations in Outer and Middle positions did not differ ($p > 0.05$) unless Outer and Middle positions were paired within logs (i.e. Outer position concentrations were significantly higher than for the Middle, $p < 0.05$).

Table 3. Mean mercury concentration (mg Hg/kg DW of sample) in three sections of buried logs from the GDSNWR (n = 9, SD = standard deviation).

	Edge		Outer		Middle	
Salvage Unit	Mean	SD	Mean	SD	Mean	SD
A	0.0215	0.0133	0.0029	0.0011	0.0023	0.0007
H	0.0317	0.0180	0.0039	0.0006	0.0023	0.0006
SEV	0.0355	0.0143	0.0037	0.0007	0.0035	0.0006

Sediment concentrations were not quantified in the current study, however soil Hg has been investigated in GDSNWR. Mean GDSNWR sediment concentrations reported by the USFWS (Lingenfelser 2010) and Virginia Department of Environmental Quality (DEQ 2004) were 0.09 mg Hg/kg DW of sample (standard deviation unavailable) and 0.086 $\pm$ 0.042 mg Hg/kg DW of sample, respectively (figure 6), more than 1.5 times higher than the highest concentration we found (Edge position).

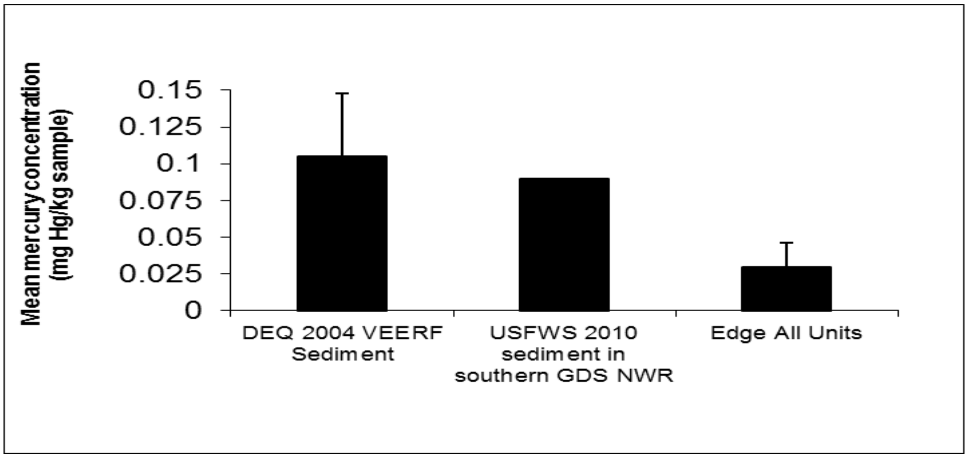


Figure 6a. Mean mercury concentrations greater than 0.01 (mg Hg/kg DW of sample) for the DEQ (2004) Sediment, USFWS (2010) Sediment and Edge of buried logs from Salvage Units A, H, and SEV in the Great Dismal Swamp National Wildlife Refuge (n = 23 for DEQ (2004), sample size unavailable for USFWS (2010)). For this study, n = 9 and error bars represent +1 SD when available.

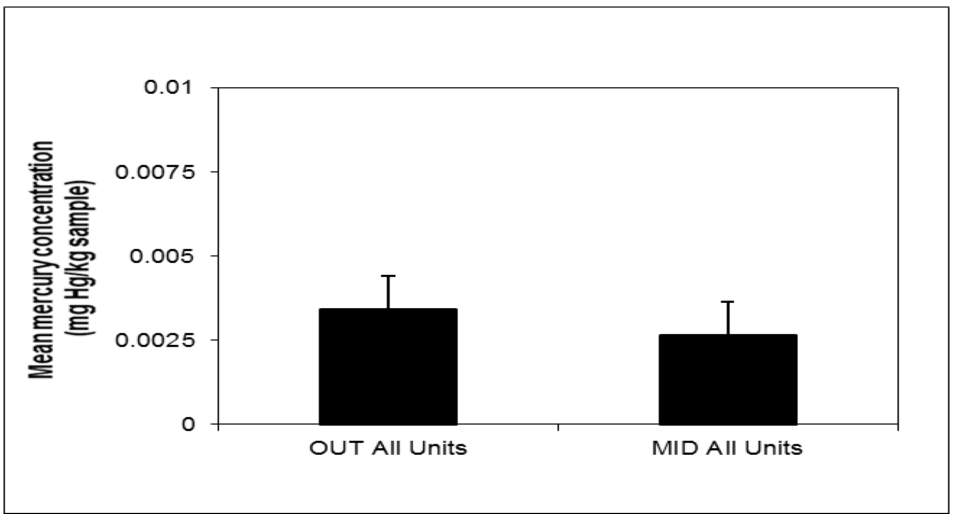


Figure 6b. Mean mercury concentrations less than 0.01 (mg Hg/kg sample) for the Outer, Middle, of buried logs from Salvage Units A, H, and SEV in the Great Dismal Swamp National Wildlife Refuge (n = 9, error bars represent +1 SD).

Natural logarithm of mercury concentrations were significantly and negatively related to distance to nearest edge in contact with peat ($r^2 = 0.416$, $p < 0.001$, $n = 26$)(figure 7). The magnitude of the gradient of mercury concentration change from the Edge to the Outer samples was -0.0196 $\pm$ 0.0150 mg Hg/kg DW of sample/cm and was higher than from the Outer to Middle samples (-0.000144 $\pm$ 0.000192 mg Hg/kg DW of sample/cm, $p < 0.05$)(figure 7).

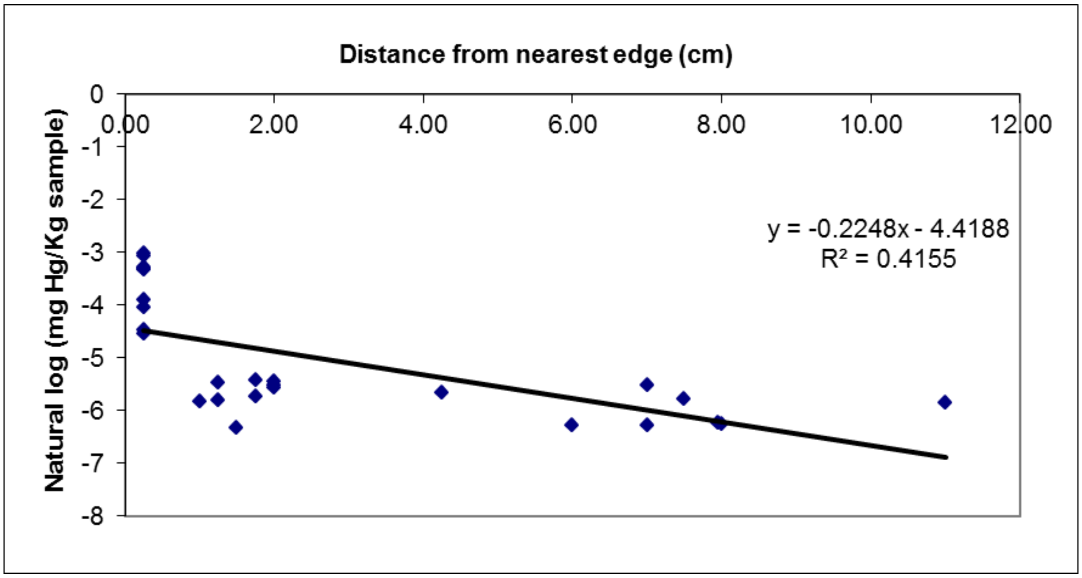


Figure 7. Natural logarithm of mercury concentration and distance to nearest edge of buried logs found in the Great Dismal Swamp National Wildlife Refuge ($n = 26$, $r^2 = 0.416$, $p < 0.001$).

DISCUSSION

The findings in this study are consistent with others in that mercury deposition rates were increasing in the past two centuries (Biester et al. 2007) and that remote places are subject to mercury deposition (Graydon et al. 2008). A review of mercury concentrations in wood by Grigal (2003) reported that 60% of observations from the north central United States were between 0.001 to 0.004 mg Hg/kg per sample with a mean of 0.0025 mg Hg/kg per sample. This concentration is similar to those measured in the Middle and Outer sections of the buried log cookies analyzed in this study, but an order of magnitude lower than the Edge sections that were analyzed in the current study.

Mercury concentrations were not consistent throughout the radius of the buried logs. Mercury concentrations in the Outer to Middle positions ranged from 0.0023 to 0.0039 mg Hg/kg DW of sample and the Edge position concentrations ranged from 0.0215 to 0.0355 mg Hg/kg DW of sample. Mercury concentration decreased as distance from the nearest edge increased. The trend in concentration gradients supports a potential for diffusion from relatively high concentrations in the soil toward lower concentrations in the interior of buried logs. The concentration gradient was greater in magnitude from the Edge to Outer (-0.0196 mg Hg/kg DW of sample/cm) compared to the Outer to Middle (-0.000144 mg Hg/kg DW of sample/cm) positions, suggesting that mercury accumulates slowly in the logs, increasing with the steepest gradient towards the Edge. GDSNWR watershed DEQ stations and USFWS mean sediment mercury concentrations were approximately 0.09 ppm (DEQ 2004, Lingenfelser 2010) which are more than double the mean concentration in Edge samples that were in contact with the peat (0.0295 mg Hg/kg DW of sample), which are an order of magnitude greater than the means of the two inner sections of the buried logs (Outer and Middle, 0.00353 and 0.002686 mg Hg/kg DW of sample, respectively).

Abreu et al. (2008) described living trees as sentinel recorders in which mercury concentrations within annual rings correspond to environmental concentrations, but several factors may affect Edge concentrations and confound interpretation of our buried logs. The perimeter of our buried logs was incomplete and thus resulted in a non-uniform tree-ring age along the Edge. Fire can also make the charred post-burn soil substrate act similarly to activated charcoal and enhance mercury absorption (Burke et al. 2010). Translocation from ray parenchyma cells, as described by Nabais et al. (1999), may also confound age-related concentrations, but studies have not yet been performed for cedar.

Studies have estimated mercury concentration in wildlife in the GDSNWR to be much greater than concentrations in buried logs reported here. However, it is important to recognize that the unique properties of cedar, namely its resistance to decomposition in peatlands, result in a vast volume and mass of buried logs that remains sequestered unless mobilized by decomposition or fire.

Fire can indirectly mobilize mercury by conversion to more mobile or soluble species and through burning soil-stabilizing vegetation (Gimeno-Garcia et al. 2001). Bound mercury can be converted directly into gaseous forms or through particulates as a part of ash (Brinkley and Christensen 1991). As peatlands are altered and drained, they are more likely to burn and volatilize sequestered mercury (Witt et al. 2009b), releasing it into surrounding watersheds (Simola and Lodenius 1982). Once these forms of mercury arrive in wetland and aquatic ecosystems, they may be transformed into methylmercury and pose human health risks (USEPA 2009).

CONCLUSION AND MANAGEMENT IMPLICATIONS

Existing models estimating remobilization of terrestrial mercury pools through fire emissions do not account for undecomposed buried bolewood in peatland soil (Turetsky et al. 2006, Wiedinmyer and Friedli 2007, Friedli et al. 2009b). The GDSNWR has historic peat accumulations greater than 6 m in depth (Oaks and Whitehead 1979) and contains extensive accumulations of buried logs (Davis 1907). DeBerry et al. (2003) report that in cedar stands, total above-ground tree biomass accounted for more than 99% of total above-ground biomass in both the GDSNWR, yet bolewood is an understudied component of mercury cycling (Grigal 2003). This study finds that buried logs from a peatland historically supporting cedar stands to have between 0.0295 mg Hg/kg DW of sample and 0.004 mg Hg/kg DW of sample available for remobilization of mercury that is not accounted for in models.

Models predicting the likelihood of mercury mobilization from buried logs should consider both the extensive buried log population of cedar stands and the increased risk of ignition for the Edge position of those logs. Water level management strategies that protect peat from burning may have the added benefit of reducing peat and buried log oxidation that result from decomposition and fire. Therefore, the same peatland management strategies may favor both carbon sequestration and mercury immobilization ecosystem services.

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