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Key Points:
- Lower, more variable water table levels and loss of Ericaceae shrubs increase total mercury and methylmercury mobility in peat
- Increased pore water mercury, methylmercury results from peat decomposition and internal regeneration of electron acceptors like sulfate
- Potential for increased mercury transport from peatlands to downstream aquatic systems as a result of climate change

Supporting Information:
- Supporting Information S1

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Mobility and transport of mercury and methylmercury in peat as a function of changes in water table regime and plant functional groups

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Abstract Climate change is likely to significantly affect the hydrology, ecology, and ecosystem function of peatlands, with potentially important but unclear impacts on mercury mobility within and transport from peatlands. Using a full-factorial mesocosm approach, we investigated the potential impacts on mercury mobility of water table regime changes (high and low) and vegetation community shifts (sedge-dominated, Ericaceae-dominated, or unmanipulated control) in peat monoliths at the PEATcosm mesocosm facility in Houghton, Michigan. Lower and more variable water table regimes and the loss of Ericaceae shrubs act significantly and independently to increase both total Hg and methylmercury concentrations in peat pore water and in spring snowmelt runoff. These differences are related to enhanced peat decomposition and internal regeneration of electron acceptors which are more strongly related to water table regime than to plant community changes. Loss of Ericaceae shrubs and an increase in sedge cover may also affect Hg concentrations and mobility via oxygen shuttling and/or the provision of labile root exudates. Altered hydrological regimes and shifting vegetation communities, as a result of global climate change, are likely to enhance Hg transport from peatlands to downstream aquatic ecosystems.

1. Introduction

Boreal peatlands store vast amounts of carbon, are critical to hydrological cycling, and are landscape scale "hot spots" for the storage and biogeochemical transformations of pollutants such as mercury (Hg) in the landscape [*Gorham*, 1991; *Branfireun et al*., 1998; *Limpens et al*., 2008]. Peatland systems act as strong sinks of atmospherically deposited inorganic Hg [*Kolka et al*., 2001; *Grigal*, 2003]. With predominantly saturated, anoxic, organic peat soils and the availability of electron acceptors such as sulfate, the hydrological and redox conditions of these systems also provide an optimal setting for the production of methylmercury (MeHg), a potent neurotoxin which bioaccumulates and biomagnifies up the food chain [*Branfireun et al*., 1996; *Scheuhammer et al*., 2007; *Tjerngren et al*., 2012]. Considerable research has focused on the influence of enhanced sulfate deposition on the process of Hg methylation in peatland ecosystems as sulfate-reducing bacteria are known to actively methylate Hg [*Gilmour et al*., 1992; *Bergman et al*., 2012; *Coleman Wasik et al*., 2015]. Recent research has confirmed that in addition to sulfate- and iron-reducing bacteria, some methanogens and syntrophic, acetogenic, and fermentative *Firmicutes* are capable of Hg methylation, including those that have been isolated from northern peatland ecosystems [*Gilmour et al*., 2013].

Peatlands, particularly those located at high latitudes, are sensitive to increasing temperatures and altered precipitation regimes resulting from global climate change [*Bridgham et al*., 1995; *Ise et al*., 2008; *Limpens et al*., 2008]. The susceptibility of peatlands to climatic change may depend upon the relative contributions of groundwater versus precipitation inputs. Bogs, wherein water table position and peat accumulation capacity are governed strongly by the balance between precipitation and evapotranspiration, are particularly vulnerable to climate change [*Winter*, 2000]. Climate-induced changes in the hydrological regimes of peatlands significantly influence microbial community structure, which may have profound effects on peat decomposition and carbon sequestration [*Nunes et al*., 2015; *Peltoniemi et al*., 2015]. Changes in bacterial community structure also have important links to methylmercury production in peatlands [*Strickman et al*., 2016]. Alterations to the carbon storage abilities of peatlands due to climate-induced changes in the

hydrology and ecology of these systems may affect the biogeochemical cycling and mobility of Hg within, and export from, these wetland systems.

With increased variability in precipitation patterns predicted to occur in the northern continental United States, prolonged periods of water table recession particularly during the summer months may have important ecological implications [*Kunkel et al*., 2003; *Groisman et al*., 2005; *Thomson et al*., 2005]. For example, these hydrological changes may result in a shift in peatland vegetation communities toward vascular-dominated functional groups [*Weltzin et al*., 2003; *Strack et al*., 2006; *Breeuwer et al*., 2009; *Dieleman et al*., 2015]. Different plant functional groups including graminoids and Ericaceae shrubs may be more suited to the hydrological conditions resulting from climate change. Although the development of shallower relative water tables as a result of peat decomposition and compression is possible, dramatic fluctuations in water levels when precipitation inputs follow prolonged dry periods may exert considerable water stress on bryophytes including *Sphagnum* mosses [*Weltzin et al*., 2000; *Whittington and Price*, 2006; *Waddington et al*., 2014]. Vascular plants may have a competitive advantage over *Sphagnum* moss species in such situations of prolonged water stress because of their ability to access deeper stores of water through their more extensive rooting systems [*Dieleman et al*., 2015]. Vascular plants can also regulate water loss through their stomata, while *Sphagnum* mosses do not possess stomata [*Breeuwer et al*., 2009]. The ecophysiological traits of the established plant functional groups may also influence the biogeochemistry of the peat and the regulation of ecosystem carbon dynamics. The rooting systems of some vascular plants such as sedges may act to shuttle oxygen to the rhizosphere, increasing the zone of peat aeration and acting as a conduit for gaseous emissions [*Weltzin et al*., 2000; *Strack et al*., 2006; *Bridgham et al*., 2008; *Waddington et al*., 2014]. Shallowly rooted ericaceous shrubs lack the adaptive structures to survive in flooded conditions [*Chapin et al*., 1996] and therefore may thrive under drier conditions. However, growth of shrubs may be hindered during periods of prolonged water stress with significant water table recession, particularly in association with increasing temperatures [*Armstrong et al*., 1991; *Weltzin et al*., 2003]. In contrast, the deep root systems of sedges may allow this plant functional group to survive and outcompete Ericaceae shrubs during prolonged drought periods [*Dieleman et al*., 2015]. The complex synergistic and antagonistic feedback mechanisms limit the ability to predict the response of peatland vegetation communities to climate change-induced water table recession. Therefore, both the sedge and Ericaceae plant functional groups should be considered, both individually and coexisting, when investigating the effects of shifting plant communities.

Water table recession and fluctuation, increased peat aeration, and shifting plant communities may significantly affect Hg cycling and the process of Hg methylation due to alterations in redox conditions of the peat and the internal recycling of sulfur species [*Coleman Wasik et al*., 2015]. The potential increased liberation of dissolved organic matter (DOM) may act to augment Hg methylation through the provision of labile carbon substrates needed as electron donors for methylating microbes [*Mitchell et al*., 2008a; *Graham et al*., 2013]. The establishment of deeply rooted vascular plants such as sedges may also contribute an additional labile carbon source through root exudates, stimulating methylating microbial communities [*Bridgham et al*., 1995; *Windham-Myers et al*., 2009]. Increased Hg mobilization via peat decomposition, partitioning, and/or desorption into the pore water phase as well as Hg complex formation with DOM may impact downstream aquatic ecosystems with the potential for enhanced transport and loading particularly during high-flow events such as spring snowmelt [*Bishop et al*., 1995; *Mitchell et al*., 2008c; *Haynes and Mitchell*, 2012]. Moreover, the complexation of Hg with DOM, particularly on thiol functional groups, increases Hg solubility and mobility, and possibly the bioavailability of Hg for methylation [*Skyllberg et al*., 2000; *Graham et al*., 2013]. Despite the propensity of peatland ecosystems to methylate and export Hg to downstream ecosystems, minimal research has been conducted to explore how Hg mobility and transport from peatlands may be affected by climate change.

The purpose of this study was to investigate the potential effects of climate change-induced shifts in water table regime and plant community composition on the mobility/movement of Hg within and transport from peatland ecosystems. A novel peatland mesocosm experiment known as PEATcosm (Peatland Experiment at the Houghton Mesocosm Facility) allowed for the manipulation of water tables and plant community composition to simulate some of the potential effects of climate change on peatland ecosystem functioning, including Hg cycling. Investigation at the mesocosm scale provides a process-level understanding, which may be

Table 1. PEATcosm Full-Factorial Experimental Design[a]

	Water Table (WT) Position	
Plant Functional Group	High WT	Low WT
Sedge only	High WT sedge	Low WT sedge
Ericaceae only	High WT Ericaceae	Low WT Ericaceae
Control (unmanipulated)	High WT control	Low WT control

[a] $n = 4$ mesocosm bins per crossed treatment.

applied to the landscape scale, of the influences of hydrological and vegetation changes on the movement of Hg within and export from peatlands.

2. Materials and Methods

2.1. Study Site and Experimental Design

The PEATcosm Mesocosm Facility is located in Houghton, Michigan, USA (47.11469°N, 88.54787°W), on the property of the United States Department of Agriculture (USDA) Forest Service Northern Research Station —Forestry Sciences Laboratory. The regional climate is humid continental with typical annual precipitation of approximately 870 mm at the site. Mean temperatures in this area range from −13°C in January to 24°C in July. The average growing season lasts approximately 132 days (30 year means at Houghton County Airport; NOAA National Climatic Data Center) [*Potvin et al*., 2015].

Twenty-four intact 1 m^3 (1 m × 1 m × 1 m) peat monoliths were extracted from an ombrotrophic peatland located in Meadowlands, Minnesota, USA, in May 2010. Monoliths were placed into individual mesocosm bins, transported to and subsequently installed in the PEATcosm facility. The stainless steel interior of each bin was Teflon coated to prevent direct contact and potential metal transfer between the bin and the peat. The top of each bin was open, exposing the peat to ambient climate conditions. The mesocosm bins were inserted into a climate-controlled tunnel and insulated on the sides, which allowed belowground access to one face of each of the bins as well as to facilitate a vertical temperature gradient, as would be observed in a natural peatland profile. *Potvin et al*. [2015] provides a detailed and comprehensive overview of the PEATcosm experiment including the peat harvest, experimental design, and treatment maintenance. The PEATcosm study was a full-factorial experimental design with two water table (WT) prescriptions crossed with three vascular plant functional group treatments in a randomized complete block design, with four replicates per treatment combination, resulting in a total of 24 experimental units (Table 1). The plant functional group treatments were designed to distinguish between effects of Ericaceae (nonaerenchymous shallowly rooted shrubs with enzymatically competent mycorrhizal root symbionts) and sedges (aerenchymous herbaceous graminoids with nonmycorrhizal roots that penetrate deeper into the saturated peat).

The water table treatments were based on long-term (approximately 50 years) data from the Marcell Experimental Forest in north central Minnesota (47.51907°N, 93.45966°W), located near the peat monolith harvest site. The two target water table seasonal profiles were modeled after typical variability, average water table years ("high WT"), and typical high variability, low water table years ("low WT"). The mean difference between the high and low WT positions was approximately 20 cm throughout the experiment. These target water table profiles were maintained via a combination of artificial precipitation additions, rain exclusion covers, and regulated outflow (spring-only, from ~25 cm depth, roughly at the acrotelm-catotelm boundary) from each of the bins [*Potvin et al*., 2015]. Water table manipulations were performed only during the summer months, and water table positions were allowed to stabilize throughout the winter months. Figure 1 displays the mean water table positions for the low and high WT treatments as well as the precipitation throughout the course of this study from 2013 to 2015. The three vascular plant functional group treatments simulated anticipated climate change-induced community composition [*Chapin et al*., 1996; *Weltzin et al*., 2000; *Strack et al*., 2006] and included (1) sedge only (all Ericaceae removed), (2) Ericaceae only (all sedge removed), and (3) unmanipulated control (includes both sedge and Ericaceae) (see Figure S1 in the supporting information for photographs). The plant functional group treatments were initiated in June 2011 and maintained weekly by clipping the stems of the excluded species. The dominant sedge species present was *Carex oligosperma* Michx., while the dominant ericaceous shrubs included *Chamaedaphne calyculata* (L.) Moench., *Kalmia*

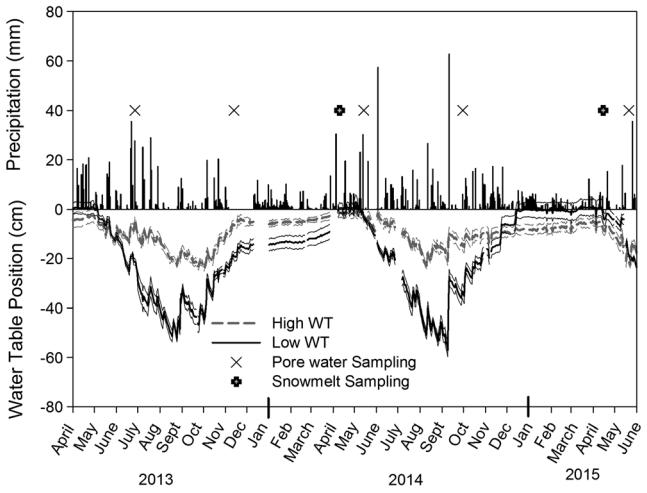

Figure 1. Mean water table positions (in cm below the peat surface) in the low and high WT treatment mesocosms and precipitation (in mm) over the course of this study from 2013 to 2015. Water table manipulations were conducted in the summer months, while water table levels were left to stabilize during the winter months. Dashed lines around the low and high WT treatment mean water table positions represent the 95% confidence interval. Pore water and snowmelt sampling events are denoted.

polifolia Wangenh., and *Vaccinium oxycoccos* L. The mosses *Sphagnum rubellum* Wilson, *S. magellanicum* Brid., *S. fuscum* (Schimp.) Klinggr., and *Polytrichum strictum* Brid. comprised the dominant bryophyte species in all of the 24 mesocosms. To a lesser extent, *P. commune* Hedw., *Eriophorum vaginatum* L., *Andromeda polifolia* L. var. glaucophylla (Link) DC., *Rhododendron groenlandicum* (Oeder) Kron and Judd, and *Drosera rotundifolia* L. were also present.

2.2. Water Sampling

Pore waters were sampled repeatedly throughout the 2013, 2014, and early 2015 growing seasons to monitor the influence of the simulated climate change effects on Hg and MeHg mobility. Pore waters were collected from a micropiezometer nest (ultrahigh-density polyethylene casing with Teflon tubing) installed in each of the 24 mesocosms at three depths—20 cm, 40 cm, and 70 cm below the peat surface (see *Romanowicz et al.* [2015] for details on piezometer construction). Sampling was conducted in June, August, and November 2013, May, July, and September 2014, and in May 2015 prior to the midsummer destructive harvest of the mesocosms at the conclusion of the experiment. Due to low midsummer water tables preventing the collection of samples for analysis from the 20 and 40 cm depths in the low WT bins, only the spring and fall data are considered throughout this analysis (see Text S1 in the supporting information for full details). Spring snowmelt runoff was also collected in 2014 and 2015 to examine impacts on the potential export of Hg from peatlands to downstream aquatic ecosystems. Outflow or runoff from each mesocosm was initiated by sufficiently high water tables sensed via a pressure transducer and controlled via an outflow line located at about 25 cm depth below the peat surface (approximate acrotelm-catotelm boundary), draining into plastic tubs in the belowground tunnel. Large rain events and spring snowmelt triggered the production of runoff. However, only runoff produced as a result of snowmelt was collected for the purposes of this study.

Pore water samples were collected from the micropiezometers using a portable peristaltic pump equipped with a Teflon line rinsed with dilute hydrochloric acid (HCl) prior to sampling. Acid-cleaned Teflon in-line filtering units (Savillex, Eden Prairie, MN) were attached to the pump line to filter all water samples to a level of 0.7 μm using ashed glass fiber filters [*Lewis and Brigham*, 2004; *Shanley et al.*, 2008]. Samples were collected in new polyethylene terephthalate glycol bottles, acidified to 0.5% by volume with trace metal grade HCl and stored at 4°C in darkness until analysis. Method blanks were collected by running deionized water through the cleaned pump line and filter units during each sampling event to ensure the cleanliness of the sampling equipment and sample handling. Mesocosm runoff was similarly collected during the snowmelt period from each bin outflow line, preserved and stored until analysis. Ultraclean trace metal techniques were followed during sample collection, laboratory handling, and analyses [*Shanley et al.*, 2008].

Pore waters from 20, 40, and 70 cm depths and mesocosm runoff were individually analyzed for both total Hg (THg) and MeHg concentrations. Ancillary chemical analyses including dissolved organic carbon (DOC), chloride, sulfate, and total phenolic concentrations were determined for pore water and runoff samples collected at the same time as those for Hg analyses.

2.3. Pretreatment Peat

Peat was collected from each of the 24 mesocosms in 2011 prior to the initiation of the water table and plant functional group prescriptions to assess any pretreatment trends in solid phase THg and MeHg

concentrations among the monoliths. Peat cores were collected with a stainless steel corer from the surface to 60 cm depth and were sectioned in 10 cm increments.

2.4. Peat Decomposition Assays

To assess peat decomposition potential for each of the crossed water table and plant functional group treatments, wooden dowels with strips of cellulose filter paper (predried at 55°C and weighed) attached at 10 cm increments using heat shrink tubing and encased in nylon mesh netting (20 × 20, 67% open area) were installed to a depth of 80 cm. Two rods were deployed in each of the peat monoliths for the duration of the 2014 growing season ($n = 48$ total). They were removed and kept frozen until processed. Peat adhering to the rods upon removal was gently rinsed off. The cellulose strips were removed, dried at 55°C, and weighed to determine the percentage mass lost in relation to the initial weight.

2.5. Analytical Methods

Total Hg concentrations of the pore water and runoff samples were determined with a Tekran Model 2600 automated Total Mercury Analyzer using cold vapor atomic fluorescence spectroscopy (CVAFS) according to U.S. EPA Method 1631 [*U.S. EPA Method 1631*, 2002]. Recovery of a THg spike was 94.5 ± 7.9% (mean ± standard deviation, $n = 33$), replication of duplicates was 1.7 ± 1.4% ($n = 32$), and the detection limit, calculated as three standard deviations of matrix blanks, was 0.25 ng L^{-1} ($n = 140$). Freeze-dried peat samples were digested in hot nitric acid, and diluted digestates were similarly analyzed by CVAFS. Methylmercury analysis was conducted by isotope dilution-gas chromatography-inductively coupled plasma mass spectrometry [*Hintelmann and Evans*, 1997] on samples, both water and solid-phase peat, that were distilled in Teflon vessels according to U.S. EPA Method 1630 [*U.S. EPA Method 1630*, 1998]. During the distillation process a trace amount of enriched stable $Me^{199}Hg$ isotope was added to each sample as an internal standard [*Hintelmann and Evans*, 1997]. Recovery of standard reference material (estuarine sediment ERM CC580) was 99.8 ± 8.6% ($n = 50$), replication of duplicates was 5.6 ± 4.9% ($n = 45$), and the MeHg detection limit was calculated ($n = 48$ matrix blanks) to be 0.04 ng L^{-1}. Field blanks were below detection limits for both THg and MeHg concentrations.

Sulfate and chloride were determined with an ICS-2000 ion chromatograph with an IonPac AS11 separator column (Dionex Corporation, Bannockburn, IL, USA). Sulfate and chloride pore water analyses were conducted only for the 2013 and 2014 growing seasons. Dissolved organic carbon was determined from piezometer samples filtered to 0.45 μm, acidified to pH 2, and analyzed with a TOC-V Analyzer (Shimadzu Scientific Instruments, Columbia, MD, USA). Pore water collection and analysis for total phenolics by microplate technique (with tannic acid standard) were as previously described in *Romanowicz et al.* [2015].

2.6. Statistical Analyses

All statistical analyses were performed using R statistical software [*R Development Core Team*, 2014] with $\alpha = 0.05$. The influence of water table regime and vascular plant functional group on THg and MeHg concentrations, as well as %MeHg over the three sampling depths of pore waters collected over the course of the experiment, was assessed using repeated measures analyses of variance (ANOVA); with water table, plant functional group and sampling depth treated as main factors and sampling event as the repeated factor. A repeated measures ANOVA was similarly performed for the snowmelt runoff Hg concentration data collected from the bins over the two sampling years. One-way ANOVAs were performed on the pretreatment solid phase peat THg and MeHg concentrations individually at 10–30 cm and 30–50 cm to assess any significant differences among assigned treatment bins prior to implementation of the treatments. These peat depths were selected for statistical analysis to coincide with the pore water sampling depths of 20 and 40 cm. Additional details regarding statistical analyses are provided in the supporting information (Text S1).

3. Results and Discussion

Both THg and MeHg concentrations in pore waters and snowmelt runoff were significantly affected by the experimental manipulation of water table (both $p < 0.0001$) and vascular plant functional group (both $p < 0.0001$), with no significant interaction ($p = 0.11$ to 0.35; Figure 2) between these factors indicating that changes in either are likely to have an impact on Hg accumulation in pore waters and subsequently, for mobility to downstream regions. Specifically, lower, more variable water table regimes significantly increased pore water and snowmelt runoff THg and MeHg concentrations, as did the change to a sedge-dominated vascular

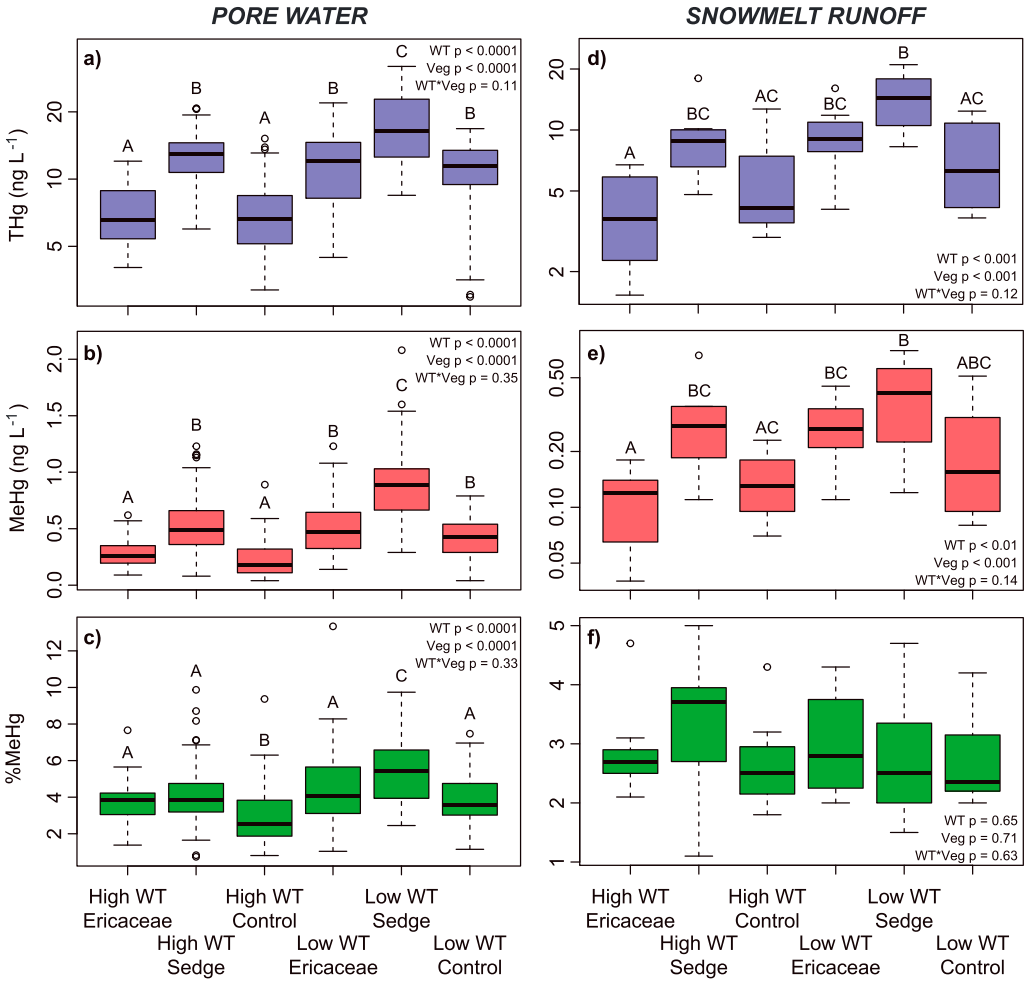

Figure 2. Total Hg and MeHg concentrations and %MeHg in pore water and snowmelt runoff in relation to water table and vascular plant functional group manipulations (treatment means of all depths and sampling events). (a) pore water THg, (b) pore water MeHg, (c) pore water %MeHg, (d) snowmelt runoff THg, (e) snowmelt runoff MeHg, and (f) %MeHg in snowmelt runoff. Letters denote statistically similar groups based on transformed data. No significant differences were observed in snowmelt runoff %MeHg across treatments. Significance of treatment effects both individual (water table "WT" and plant functional group "Veg") and interactive ("WT*Veg") for THg, MeHg, and %MeHg for both pore waters and snowmelt runoff are noted. $n = 318$ pore water samples total ($n = 50$–56 per treatment).

plant community composition with no Ericaceae shrubs present. The combined low WT-sedge only treatment resulted in the highest pore water and runoff concentrations of both THg and MeHg (Figure 2). One-time mass transfer loads were determined for the 2014 spring snowmelt period by multiplying the THg and MeHg concentrations of the runoff by the amount of water drained from the mesocosms' acrotelm-catotelm boundaries (Table S1). The trends in THg and MeHg loads among the six crossed WT and vascular plant functional group treatments (Figure S2) were consistent with those observed for THg and MeHg pore water and runoff concentrations (Figure 2). No significant, systematic differences were observed in solid phase peat THg or MeHg concentrations at either the 10–30 cm or 30–50 cm depth intervals as these values were quite variable among the six assigned experimental treatment combinations prior to the experiment (Table 2). The differences observed in pore water and runoff THg and MeHg concentrations can therefore be confidently attributed to the experimental manipulation.

For both THg and MeHg pore water concentrations, the effect of water table significantly interacted with both the depth ($p = 0.0002$ to 0.008) and the timing of the sampling event ($p < 0.0001$ to 0.002). Total Hg and MeHg concentrations decreased from the 20 cm to the 70 cm sampling depths in the low WT bins, whereas THg and MeHg concentrations at the three sampling depths were not significantly different from one another in the high WT bins. Greater fluctuations in water table position in the low WT bins (mean

Table 2. Mean ± Standard Deviation THg and MeHg Concentrations (in ng g^{-1}) in Solid-Phase Peat Prior to Experimental Manipulations ($n = 8$ Samples Per Treatment and Depth Increment)

	THg Concentration (ng g^{-1})		MeHg Concentration (ng g^{-1})	
	10–30 cm	30–50 cm	10–30 cm	30–50 cm
High WT Ericaceae	91.8 ± 63.4	87.8 ± 38.4	5.4 ± 6.6	5.9 ± 5.7
High WT sedge	82.8 ± 27.0	104.7 ± 40.5	5.0 ± 3.6	8.1 ± 6.7
High WT control	80.9 ± 33.3	91.3 ± 21.3	4.5 ± 6.0	5.8 ± 4.1
Low WT Ericaceae	65.3 ± 15.5	103.3 ± 28.2	1.0 ± 0.7	6.6 ± 6.4
Low WT sedge	92.0 ± 33.3	83.2 ± 22.6	5.6 ± 3.9	3.5 ± 1.8
Low WT control	75.9 ± 23.8	100.2 ± 28.9	2.8 ± 3.0	6.8 ± 5.9

position approximately 35 cm below the peat surface with a standard deviation of 12 cm) likely resulted in enhanced leaching and mobility of Hg into the pore waters due to greater aerobic decomposition of the surface peat layers and may account for the observed differences in Hg concentrations with depth. These differences are not the result of evaporative concentration of Hg under low water table conditions as there was no significant difference in chloride pore water concentrations between the two water table treatments ($p = 0.76$). In contrast, the comparatively minimal water table variability in the high WT bins (mean position approximately 14 cm below the peat surface with a standard deviation of 5 cm) reduced this mobilization, likely as a result of reduced leaching, yielding both lower and more temporally consistent pore water THg, MeHg, and DOC concentrations at all sampling depths. In association with the effect of WT position, THg and MeHg concentrations were slightly higher during the early growing season sampling events as compared to the fall samplings. This may be due to considerable water table variability as a result of draining at the beginning of each growing season to reestablish the water table treatments following spring snowmelt. Despite this seasonal variability, a consistent trend in mean THg and MeHg concentrations was observed among the six crossed WT and vascular plant functional group treatments in each of the five sampling events from 2013 to 2015 (Figures S3 and S4). A marginally significant interaction ($p = 0.02$) was observed for THg concentrations only between the vascular plant functional group and the sampling depth. The influence of vegetation on Hg mobility appears to be more evident nearer to the rooting zone, particularly with the removal of Ericaceae shrubs.

Water table position ($p < 0.0001$) and vascular plant functional group ($p < 0.0001$) also significantly affected the percentage of THg found as MeHg in pore waters (Figure 2c). The observed differences in %MeHg may be the result of the greater solubility of MeHg in water as compared to inorganic Hg [*Morel et al.*, 1998]. However, the significant differences in pore water %MeHg among treatments suggest that differences in Hg methylation may also be caused by the water table and plant functional group treatments. In contrast to the pore waters, no significant effect of either water table ($p = 0.62$) or vascular plant functional group treatment ($p = 0.77$) was observed for the %MeHg in mesocosm runoff collected during the spring snowmelt periods. This trend was also consistent over the two sampling periods as no significant direct or interactive influence of time was observed. There were no significant differences in runoff %MeHg between any of the six crossed treatments combinations ($p = 0.87$) (Figure 2f). This suggests that a similar process is responsible for the transport of both THg and MeHg to runoff during spring snowmelt.

One contributing factor controlling the mobilization and accumulation of THg and MeHg into peat pore waters as well as transport in snowmelt runoff may be the degradation of peat through decomposition. Pore water THg and MeHg concentrations were both positively and significantly but weakly correlated with DOC concentrations ($R^2 = 0.10$ for THg, $R^2 = 0.09$ for MeHg; both $p < 0.0001$) (Figure S5), which would be expected to similarly increase as peat decomposes. Pore water DOC concentrations significantly increased in the low WT treatment bins throughout the sampling period ($p < 0.001$), while those in the high WT treatments did not significantly change over time. Mean THg and MeHg concentrations as well as %MeHg in the shallow pore waters (20 and 40 cm below the peat surface) were positively correlated with the amount of cellulose decomposition (expressed as percent loss) in the top 40 cm of peat, near the mean water table position in the low WT mesocosms (Figure 3). These relationships between pore water THg and MeHg concentrations and the percentage of mass lost from the cellulose decomposition assays are significant when all pore water sampling events are considered (Figure S6). Although the decomposition data were only from 2014, the

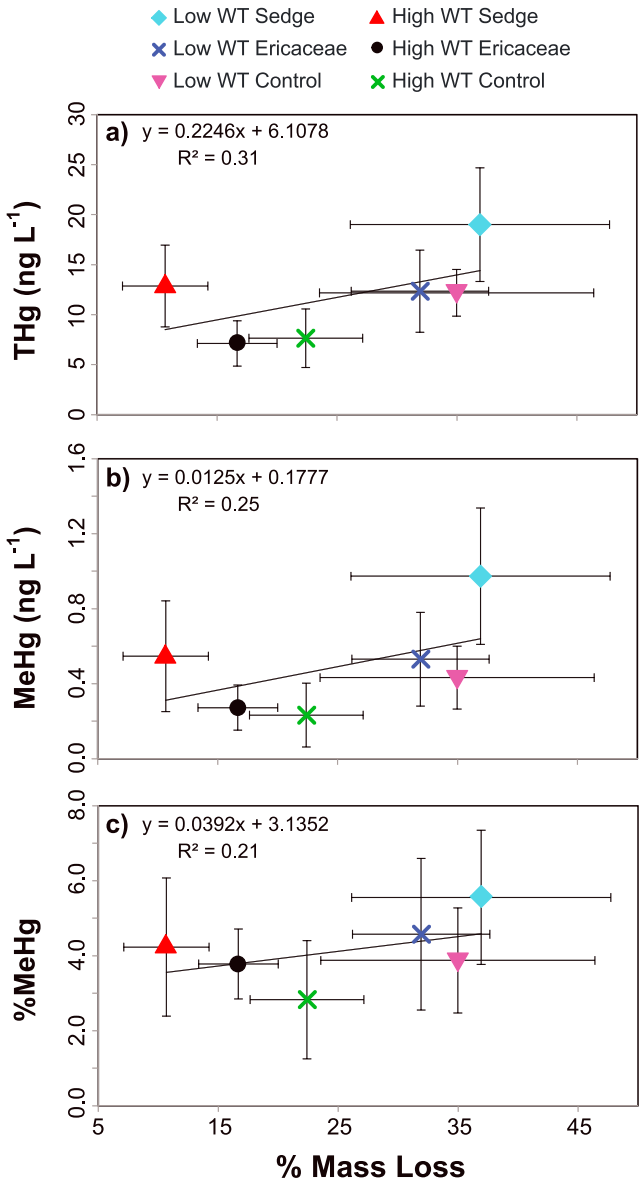

Figure 3. Relationships between shallow pore water (20 and 40 cm below the peat surface) Hg and mean % mass loss of cellulose decomposition assays in the top 40 cm of the peat. (a) THg concentrations, (b) MeHg concentrations, (c) %MeHg. Error bars represent standard deviation. Decomposition assays were harvested in 2014 and represent potential peat decomposition among the treatments. Pore water Hg data were averaged by treatment from all five sampling events.

percentage of mass lost during this time should still be representative of potential peat decomposition as a result of the water table and plant functional group treatment effects. The low WT-sedge only treatment which yielded the highest concentrations of THg and MeHg in pore waters also exhibited the greatest potential for peat decomposition through the cellulose decomposition assays.

The prolonged periods of water table drawdown sustained in the low WT treatments likely caused the enhanced observed peat decomposition by increasing the zone of aerated peat, resulting in greater aerobic respiration as compared to the relatively high water table position in the high WT treatments [*Whittington and Price*, 2006; *Ise et al.*, 2008; *Waddington et al.*, 2014]. The degradation of soil organic matter leads to increased DOC mobilization and accumulation in pore waters as reflected by higher concentrations in the mesocosm pore waters, with repeated fluctuations in water table position resulting from precipitation inputs likely leaching carbon from the peat into the pore waters following periods of drawdown [*Fenner et al.*, 2007]. This may also account for the greater mobilization of THg and MeHg at the upper depths of the peat

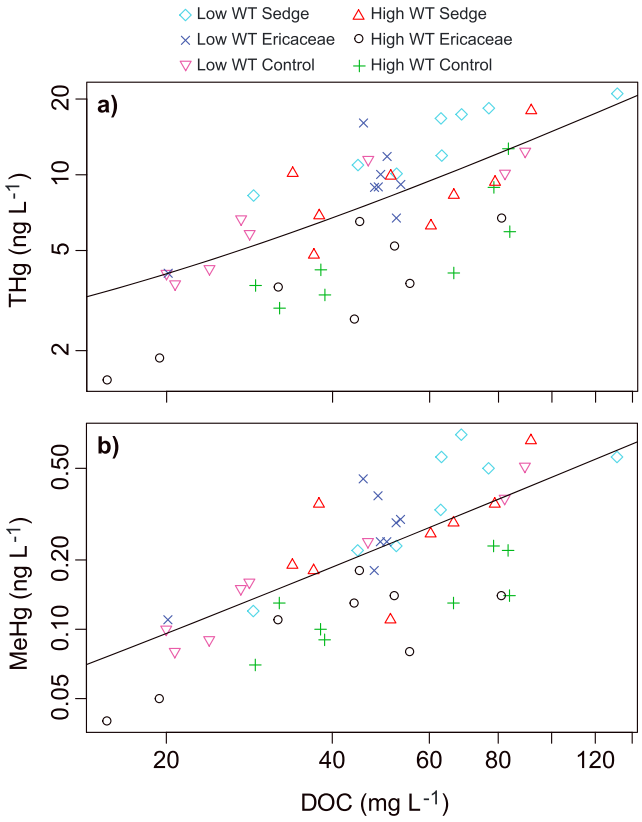

Figure 4. Hg-DOC relationships in 2014 and 2015 snowmelt runoff for all 24 mesocosms ($n = 48$ total). (a) THg in relation to DOC concentrations and (b) MeHg in relation to DOC concentrations. Data are plotted on log-transformed axes.

monoliths, with higher pore water THg and MeHg concentrations in the 20 and 40 cm samples within the zone of fluctuation of the low WT treatments. Significant, but relatively weak, positive correlations with total phenolics in pore water were observed for MeHg ($R^2 = 0.12$; $p < 0.0001$) and THg ($R^2 = 0.10$; $p < 0.0001$) (Figure S7). *Romanowicz et al.* [2015] found a positive correlation between pore water total phenolics and peroxidase enzyme activity. Oxidative enzymes including peroxidase and phenol oxidase contribute to the degradation of lignin and polymeric complexes [*Romanowicz et al.*, 2015]. As pore water concentrations of THg and MeHg increased with total phenolics, which were positively correlated with peroxidase enzyme activity in the PEATcosm pore waters [*Romanowicz et al.*, 2015], organic matter decomposition was likely the source of increased pore water Hg mobility (Figure S7). Mercury forms complexes with organic matter [*Driscoll et al.*, 1995] predominantly with thiol moieties [*Graham et al.*, 2012]. Therefore, enhanced peat decomposition and degradation are likely responsible for much of the observed increase of both THg and MeHg, as well as DOC accumulation in the mobile pore water phase. Given that solid phase peat THg and MeHg concentrations among bins were not significantly different prior to initiation of the experiment, and THg concentrations in particular are not expected to change significantly over time, the observed pore water THg and MeHg trends are a clear indication that Hg mobility has increased.

In addition to the peat decomposition effect on Hg and MeHg concentrations in pore water, the water table manipulations may also act to enhance MeHg production in the peat. Prolonged periods of water table drawdown followed by rewetting of the peat through precipitation input may act to reset the oxidation-reduction conditions in the zone of aeration [*Coleman Wasik et al.*, 2015]. Mercury methylation is a redox-sensitive process, facilitated by a diverse set of anaerobic bacteria and Archaea [*Gilmour et al.*, 2013]. The periodic oxidation of peat through water table variability may stimulate MeHg production via regeneration of the oxidized forms of terminal electron acceptors, such as the oxidation of sulfide to regenerate sulfate [*Coleman Wasik et al.*, 2015]. The highest sulfate concentrations in pore waters were indeed observed in the low WT treatment mesocosms (Figure S8).

In addition to the clear influence of the hydrological treatments, the dominant vascular plant functional group also significantly affects the accumulation of Hg in pore waters. The correlations between Hg concentrations and decomposition demonstrate the likely influence of vascular plant functional groups affecting Hg mobility by mechanisms other than decomposition. This is exemplified by the increased explanation of variability in the relationships when the sedge-only treatments for both water table prescriptions are removed ($R^2 = 0.93$ for THg, $R^2 = 0.69$ for MeHg, and $R^2 = 0.24$ for %MeHg). The removal of Ericaceae and the establishment of sedges may lead to greater Hg methylation and the amount of Hg mobilized in pore waters, as sedge roots exude labile carbon compounds and oxygen in the rhizosphere [*Crow and Wieder*, 2005; *Bridgham et al.*, 2013]. Under both water table treatments, the sedge-dominated peat monoliths had the highest

concentrations of THg and MeHg in pore water (Figures 2a and 2b), higher than may be explained by potential peat decomposition and leaching into pore waters (Figure 3). Sedges increase peat aeration and subsequent oxidation via their aerenchyma tissues [*Bridgham et al*., 2008; *Waddington et al*., 2014] and may, similarly to water table fluctuation, prime Hg methylation by stimulating the regeneration of the terminal electron acceptors required for methylating microbial communities [*Coleman Wasik et al*., 2015]. The leakage of oxygen from the roots of salt marsh sedges has been observed to stimulate aerobic respiration by acting as a terminal electron acceptor as well as facilitating anaerobic respiration by regenerating alternative electron acceptors including nitrate, ferric iron, and sulfate [*Mueller et al*., 2016]. Pore water sulfate concentrations were slightly higher for the sedge-dominated treatments subjected to both water table prescriptions (Figure S8). Thus, in addition to supplying the necessary terminal electron acceptors for soil organic matter decomposition [*Mueller et al*., 2016], sedges may prime MeHg production through the provision of oxygen and labile organic compounds via root exudation to the rhizosphere, which are required for methylation to occur [*Mitchell et al*., 2008a]. Conversely, the presence of Ericaceae shrub roots in the Ericaceae-only and unmanipulated control treatment bins (both of which had lower Hg concentrations than the sedge treatment) may compete with the heterotrophs for available oxygen [*Romanowicz et al*., 2015], thereby reducing sulfate regeneration (slightly, although not significantly; Figure S8) and suppressing MeHg production. Ericaceous shrubs form a symbiotic relationship with mycorrhizal fungi, which may mediate changes in soil microbial communities, compete for available oxygen thereby limiting terminal electron acceptor regeneration, and decrease peat decomposition through the suppression of heterotrophs. This has been demonstrated in the PEATcosm experimental monoliths wherein a negative correlation was observed between Ericaceae biomass and peroxidase as well as β-glucosidase enzyme activity [*Romanowicz et al*., 2015]. Inhibition of MeHg production in the upper rooting zone depths of the Ericaceae-only treatments, as well as the unmanipulated control mesocosms as compared to the sedge-only monoliths, may contribute to the peat pore water Hg concentration differences observed.

The trends in THg and MeHg concentrations in snowmelt runoff are similar to those observed in the pore waters (Figure 2). Snowmelt runoff THg and MeHg concentrations both exhibit a significant ($p < 0.001$), positive correlation with DOC concentrations ($R^2 = 0.45$ for THg, $R^2 = 0.42$ for MeHg; Figure 4). Increased Hg export in association with organic carbon has similarly been observed in other systems [*St. Louis et al*., 1994; *Driscoll et al*., 1995]. The statistically similar %MeHg across the treatments along with the positive correlation with DOC concentrations provides further support for peat degradation being the principal driving factor controlling not only the accumulation of Hg in pore waters but also its transport within and potential export from peatlands. The concentrations of THg and MeHg observed in snowmelt are likely the result of peat decomposition liberating Hg in association with organic matter, which has accumulated in pore waters throughout the growing season and flushed from the peat during the spring snowmelt period.

4. Conclusions

Climate-induced changes in the hydrological regime and vegetation communities of peatland systems have the potential to significantly enhance both THg and MeHg accumulation in peat pore water and transport downstream during spring snowmelt. Although peatlands are strong sinks of inorganic Hg in the landscape, peatlands are also well-documented sources of MeHg [*St. Louis et al*., 1994]. By potentially increasing the amount of MeHg exported to aquatic ecosystems for uptake into the food chain, as well as the amount of inorganic Hg available for in-lake methylation, wildlife and human populations which rely upon fish consumption for subsistence may be exposed to increased Hg levels [*Scheuhammer et al*., 2007; *Mergler et al*., 2007]. Given that peatlands cover approximately 3% of the global land area and are located predominantly in the subarctic and boreal regions which are vulnerable to the effects of climatic change [*Bridgham et al*., 2008], significant stores of Hg sequestered with vast stocks of carbon [*Grigal*, 2003] may be liberated from either impacts on water table regime, plant community changes, or a combination of both.

The small scale of the peat monoliths in this study facilitated a direct assessment of the influence of water table position and vascular plant functional groups on Hg mobility by removing potential confounding factors such as heterogeneities in microtopography and hydrological flow paths and sources. At larger scales, spatial heterogeneities in Hg methylation and accumulation have been observed, including MeHg hot spots at the outer edges of peatlands, which tend to increase MeHg export from peatland systems [*Mitchell et al*.,

2008b]. The results of this study are most applicable to the predominant interior peatland area and suggest that higher MeHg concentrations observed at the outer edges of peatlands may be even further enhanced by climate change-driven increases in Hg methylation and/or mobility from the peatland interior. Certainly, results from this experimental work may be scaled to larger peatland ecosystems and may aid in developing modeling approaches to forecast the response of peatlands under future climate change scenarios. Overall, the results of this study suggest that deeper, more fluctuating water tables and shifts toward sedge-dominated plant functional groups will measurably enhance Hg and MeHg concentrations in peatland pore waters and likely lead to greater Hg and MeHg export to downstream ecosystems during spring snowmelt.

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