Plant mercury accumulation and litter input to a Northern Sedge-dominated Peatland

Ting Sun[1,2,3] and Brian A. Branfireun[4]

[1]School of Environmental Science and Engineering, Shandong University, Qingdao 266237, PR China

[2]Institute of Eco-Environmental Forensics, Shandong University, Qingdao 266237, PR China

[3]Department of Earth Sciences, University of Western Ontario, 1151 Richmond Street, London, Ontario, Canada

[4]Department of Biology, University of Western Ontario, 1151 Richmond Street, London, Ontario, Canada

Correspondence to: Ting Sun (tsun64@uwo.ca)

21   Abstract

22   Plant foliage plays an essential role in accumulating mercury (Hg) from the atmosphere and transferring it to soils in terrestrial

23   ecosystems. While many studies have focused on forested ecosystems. Hg input from plants to northern peatland peat soils has

24   not been nearly as well studied and is likely equally important from a mass balance perspective. In this study, we investigated the

25   accumulation of atmospheric Hg by the dominant plant species, few-seeded sedge [*Carex oligosperma* Michx.], wire sedge

26   [*Carex lasiocarpa* Ehrh], tussock sedge [*Carex stricta* Lamb.], and sweet gale [*Myrica gale* L.] in a boreal sedge-dominated

27   peatland. Foliar Hg concentrations decreased early in the growing season due to growth dilution. Foliar Hg concentrations were

28   subsequently positively correlated with leaf age (time). Hg concentrations were 1.4-1.7 times higher in sweet gale than in sedges.

29   A leaching experiment showed that sweet gale leached less Hg but more bioaccessible dissolved organic matter (DOM) by mass

30   than sedges. Leaching of Hg was positively related to the aromaticity of DOM in leachate, suggesting the importance of DOM

31   with higher aromaticity in controlling Hg mobility. Annual inputs of Hg through senesced leaf material to peat soils were 9.88

32   mg/ha/yr, 1.62 mg/ha/yr, and 8.29 mg/ha/yr for sweet gale, tussock sedge, and few-seeded sedge/wire sedge, respectively. Future

33   investigations into foliar Hg accumulation and input from other plant species to the sedge-dominated peatland are needed to

34   estimate the annual Hg inputs precisely.

35

36

37

38

39

40

41

1 Introduction

Mercury (Hg), especially methylmercury (MeHg), is a global concern due to its potential toxicity and ubiquitous presence in the environment (Morel et al., 1998). Hg is emitted to the atmosphere from both natural (e.g., volcanoes, wildfires, geothermal activity) and anthropogenic sources (e.g., coal combustion, artisanal gold mining, incineration) (Schroeder and Munthe, 1998; Streets et al., 2011). Atmospheric Hg exists as gaseous elemental mercury (GEM, Hg(0)), reactive gaseous mercury (RGM, Hg(II)), and particulate-bound mercury (PBM, Hg_p) with GEM as the dominant species (> 95 %) (Schroeder and Munthe, 1998). RGM and PBM have shorter atmospheric residence time ranging from hours to days, whereas GEM has a longer atmospheric residence time of several months to a year and thus is transported globally (Schroeder and Munthe, 1998). These atmospheric Hg species are eventually deposited into aquatic and terrestrial ecosystems via wet deposition (precipitation, such as rain, snow, and fog) and dry deposition (particle settling or direct partitioning to vegetation, water, and soil surface, or direct absorption by vegetation foliage) (Lindberg et al., 2007). Hg dry deposition is a larger input than wet deposition to vegetated terrestrial landscapes, contributing 70 %~85 % of total Hg deposition (dry and wet deposition) in terrestrial ecosystems (Graydon et al., 2008; Risch et al., 2017; Risch et al., 2012; St. Louis et al., 2001; Wang et al., 2016; Zhang et al., 2016), and more than 70 % of Hg dry deposition is by vegetation litterfall/incorporation into soil organic matter (SOM) (Obrist et al., 2017; Wang et al., 2016).

Forest ecosystems are important sinks of atmospheric Hg and have received widespread attention from researchers (Risch et al., 2012; St. Louis et al., 2001; Wang et al., 2016; Zhang et al., 2009); however, studies about foliar Hg accumulation in other plant types or other ecosystems such as boreal peatlands are few (see Moore et al., 1995) despite their critical role in the carbon (Gorham, 1991) and Hg cycles (Grigal, 2003). Boreal peatlands store 500 ± 100 Gt of carbon as peat (partially decomposed vegetation matter) due to slow decomposition rates in their anaerobic and acidic conditions and low temperatures (Rydin and Jeglum, 2013). In addition, boreal peatlands are sinks for inorganic Hg (St. Louis et al., 1994), and can be MeHg sources to downstream ecosystems (Branfireun et al., 1996; Mitchell et al., 2008; St. Louis et al., 1994), given their anaerobic conditions, non-limiting amounts of inorganic Hg, and often available but limited amounts of sulfate (Blodau et al., 2007; Schmalenberger et al., 2007) and bioaccessible carbon (Mitchell et al., 2008).

Little information is available about the amount of atmospheric Hg accumulated in leaves in peatlands. Moore et al. (1995) reported that Hg levels in nonvascular plants (fungi, lichens, and mosses) are almost an order of magnitude higher than those in vascular plants in wetlands, and the Hg concentrations follow the sequence: grassland herbs < trees and shrubs < aquatic

68   macrophytes < *Sphagnum* spp. mosses < lichens < fungi. However, the annual input of aboveground vegetation biomass to

69   peatland soils as senesced litter from vascular plants is greater than from bryophytes (Frolking et al., 2001) in fen-type peatlands

70   dominated by sedges and shrubs, so despite having lower Hg concentrations, the mass input may be significant. With more

71   bioaccessible litter and leachate than bryophytes, vascular plant inputs may also decompose at faster rates, releasing Hg to the

72   soil and/or facilitating net methylation (Hobbie, 1996; Lyons and Lindo, 2019).

73   Boreal peatlands are experiencing rising temperatures due to climate change (IPCC, 2018) that is likely to either increase

74   aboveground biomass and leaf area in already vascular plant-dominated peatlands or potentially shift moss-dominated peatlands

75   to more vascular plant-dominated. Changes in plant abundance and community composition may further affect Hg deposition

76   (Zhang et al., 2016) and input through litterfall, given that foliar Hg concentrations (Moore et al., 1995) and annual vegetation

77   biomass input (Frolking et al., 2001) are different among plant species. Weltzin et al. (2000) found that the productivity of

78   vascular plants in peatlands increased under higher temperatures. A full factorial greenhouse laboratory experiment of increased

79   temperature and elevated atmospheric CO_2 resulted in increased peatland graminoid productivity (both above and belowground)

80   (Tian et al., 2020). Several studies have also shown that experimental warming of northern peatlands mesocosms altered plant

81   community composition, increasing vascular plant abundance and biomass, and decreasing *Sphagnum* spp. cover (Buttler et al.,

82   2015; Dieleman et al., 2015; Weltzin et al., 2000). Changes in plant abundance and biomass in northern peatlands are

83   ecologically significant, given their role as a source of net MeHg production.

84   Foliar Hg eventually enters peat soils via litterfall and is expected to follow the sequence: (1) wash-off of aerosols, particles, and

85   gases from leaf surfaces, (2) leaching of water-soluble components, and (3) incorporation into SOM after the microbial

86   decomposition of litter. Leaching is the initial phase of litter breakdown in aquatic environments and can rapidly release up to

87   30 % dissolved matter, primarily dissolved organic matter (DOM) within 24 h after immersion of litter (Gessner et al., 1999).

88   The amount of rapidly released Hg during litter leaching is unknown and needs to be elucidated because more recently deposited

89   Hg appears to be more readily methylated than "old" Hg in peat soils (Branfireun et al., 2005; Feng et al., 2014; Hintelmann et

90   al., 2002).

91   The overall objective of this study is to link the vascular plant community (i.e., sedges and shrubs) to the peatland Hg cycle in a

92   vascular plant-dominated fen-type peatland. We use "sedge-dominated fen" instead of "vascular plant-dominated fen-type

93   peatland" hereafter, given that sedges are the primarily dominant plants in this study site (Webster and McLaughlin, 2010). The

94   specific objectives of this study are to:

95   (1) quantify the mass accumulation of atmospherically-derived Hg in leaves of dominant plant species in a sedge-dominated fen

96   over a growing season;

97   (2) estimate the Hg input from the litter of different plant species and through litter leaching to peat soils;

98   (3) clarify the role of DOM characteristics in controlling Hg leaching;

99   (4) estimate the annual areal loading of foliar Hg of different plant species to peat soils.

100   **2 Materials and methods**

101   **2.1 Study site**

102   Samples were collected from a sedge-dominated fen (10.2 ha) located in an 817 ha sub-watershed of the Lake Superior basin

103   near White River Ontario, Canada (48°21' N, 85°21' W). The growing season is roughly from June to September. The sedge-

104   dominated fen is mostly open and the vegetation community is dominated by three sedge species: few-seeded sedge [*Carex*

105   *oligosperma* Michx.]; wire sedge [*Carex lasiocarpa* Ehrh]; and tussock sedge [*Carex stricta* Lamb.] (Lyons and Lindo, 2019).

106   Sweet gale [*Myrica gale* L.] is the dominant shrub at this site (Lyons and Lindo, 2019; Palozzi and Lindo, 2017). Details of the

107   study site and the characteristics of these plants are provided in the Supporting Information (SI). In this study, few-seeded sedges

108   and wire sedges were mixed during plant sample collection as they are indistinguishable in size and form from one another when

109   not in flower/seed, and frequently co-occur.

110   **2.2 Sample collection and analysis**

111   Five locations several hundred meters apart were selected in the sedge-dominated fen to serve as within-site replicates to account

112   for potential local-scale variability. These five locations were roughly evenly distributed over this study area. Approximately

113   fifty whole leaves of each few-seeded sedge/wire sedge, tussock sedge, and sweet gale were collected from each location using a

114   clean blade in the middle of June, July, August, and after senescence at the beginning of October 2018 in each location, totaling

115   60 samples. For the October sampling event, the sedge leaves were still standing with the lower sections green, and although

116   senesced, shrub leaves were sampled from the branch to ensure that there was no mixing with previous years' fallen leaves.

117   Disposable nitrile gloves were worn during the sample collection. All samples were double bagged with two polyethylene bags

118   and transported to the lab using a clean cooler. Leaves of each species that were collected from each plot in October 2018 were

119   divided for foliar total Hg (THg) analyses and a foliar Hg leaching experiment. Leaves were stored frozen until they were

120   returned to the university laboratory.

121   For estimation of annual biomass of senesced leaf, seven $0.25 \ m^2$ ($0.5 \ m \times 0.5 \ m$) plots several hundred meters apart were

122   selected at the end of August 2019 during senescence and before leaf off. All aboveground biomass of few-seeded sedge/wire

123   sedge and tussock sedge and all aboveground leaf biomass of sweet gale were collected separately using a clean blade from each

124    $0.25 \ m^2$ plot. All vegetation samples were stored by species in paper bags, transported to the lab, and then oven-dried at 60 °C

125   for a minimum of 48 h. Dried leaves of each species in each plot were sorted and weighed to estimate senesced leaf biomass of

126   each species for each plot. The senesced leaf biomass of each species per hectare per year was calculated and expressed as

127   mg/ha/yr.

128   **Foliar total mercury, C content and N content.** In the laboratory, leaf samples for chemical analyses were rinsed three times

129   with deionized water (18.2 MΩ cm), freeze-dried for 48 h, ground and homogenized, and then analyzed using a Milestone™

130   DMA-80 (EPA method 7473). Leaf C content (%C; w/w) and N content (%N; w/w) before and after the foliar Hg leaching

131   experiment was analyzed using a CNSH analyzer (Vario Isotope Cube; Elementar). The ratio of leaf C content and N content

132   (C:N) were calculated. Detailed information concerning analytical methods are described in the SI, including analysis of foliar

133   total Hg, %C, and %N.

134   **Foliar mercury leaching experiment.** The foliar leaching experimental procedure followed the design of Rea et al. (2000) and

135   Del Giudice and Lindo (2017). Senesced leaves of sedges and sweet gale collected in October 2018 were rinsed twice with 100

136   mL of deionized water (18.2 MΩ cm) to quantify particulate or loosely-bound Hg and DOM that can be easily removed/leached

137   from the leaf surface. This water was reserved for subsequent analysis. After rinsing, the leaves were oven-dried at a low

138   temperature (40 °C) for 48 h, and then the leaves of each species from each location were relatively evenly separated into three

139   groups and weighed, totaling 45 groups. These oven-dried senesced leaf samples were immersed in 150 mL of deionized water

140   in clean 250 mL PETG bottles. All PETG bottles were capped, double bagged, and incubated in the dark at room temperature

141   (~21 °C) for 48 h. Senesced leaf materials were gently swirled at the beginning of the leaching experiment to ensure complete

142   wetting. Following the leaching, the leachate was vacuum filtered through a 0.45 μm glass fiber filter into clean 250 mL PETG

143   bottles. Leachate from each sample was split into two aliquots. One was preserved by acidifying to 0.5 % (vol/vol) with high-

144   purity HCl for dissolved total Hg (THg_{aq}) analysis and stored in 250 mL PETG bottles; the other was stored in the clean 60 mL

145   Amber glass bottles and analyzed within 2 d for the quantity and characteristics of DOM. All samples were stored in the dark at

146   4 °C for further analysis. Method blanks of the leaching experiment were performed at the same time following the same

147   procedure.

148   Senesced leaf material was taken out of each PETG bottle, oven-dried at 40 °C for 48 h, and re-weighed after leaching. The dry

149   leaf weight before and after the leaching process was used to calculate the mass loss. These re-dried senesced leaf samples after

150   leaching were ground and homogenized before the measurement for %C and %N as described above.

151   The dissolved total Hg (THg_{aq}) concentrations in the rinse water and leachate were analyzed using Environmental Protection

152   Agency (EPA) method 1631. Dissolved organic matter is quantified analytically as dissolved organic carbon (DOC). DOC

153   concentrations in rinse water and leachate were measured using an iTOC Aurora 1030 (OI Analytical, College Station, TX,

154   USA) using the persulfate wet oxidation method. Details on the analytical procedures and QA/QC data for concentrations of

155   THg_{aq} and DOC are provided in the SI.

156   DOM in leachate was characterized as specific ultraviolet absorbance at a wavelength of 254 nm ($SUVA_{254}$), an indicator of the

157   molecular weight (or size) and aromaticity (the content of aromatic molecules) of DOM (Weishaar et al., 2003). Higher $SUVA_{254}$

158   values suggest that DOM contains more high-molecular-weight and aromatic molecules (Weishaar et al., 2003). Fluorescence

159   excitation-emission matrices (EEMs) were also collected for calculating informative optical indices that reflect differences in

160   DOM characteristics in leachate. The reported EEMs were then converted to optical indices using R Software (R Core Team

161   2012). Three common indices were chosen in this study: the fluorescence index (FI), the humification index (HIX_{EM}), and the

162   biological index or 'freshness' index (BIX). Lower FI values (< 1.2) indicate that DOM is terrestrially derived (resulting from

163   decomposition and leaching of plant and soil organic matter) and has higher aromaticity, while higher FI values (> 1.8) indicate

164   that DOM is microbially derived (originating from processes as extracellular release and leachate of algae and bacteria) and has

165   lower aromaticity (Fellman et al., 2010; McKnight et al., 2001). High HIX_{EM} (> 1.0) values reflect the high humification of

166   DOM and DOM is composed of more highly condensed and higher molecular weight molecules (Fellman et al., 2010; Hansen et

167   al., 2016; Huguet et al., 2009; Ohno, 2002). Higher BIX values (> 1.0) reflect that more low-molecular-weight DOM was

168   recently produced by microbes (Fellman et al., 2010; Huguet et al., 2009). Details on the analytical procedures and QA/QC data

169   for $SUVA_{254}$, FI, HIX_{EM}, and BIX are provided in the SI.

170 **3 Statistical analysis**

171 Results were analyzed using IBM SPSS statistics software (IBM SPSS Inc. 24.0). The repeated-measures ANOVA was

172 performed to compare the difference in foliar THg concentrations among different plant species over the growing season and to

173 analyze the effect of leaf age on foliar Hg concentrations. Linear regressions were analyzed to examine the relationship between

174 foliar THg accumulation and leaf age. Differences in the foliage quality (%C, %N, and C:N) were analyzed using a multivariate

175 ANOVA. One-way ANOVA was used to determine the effects of plant species on concentrations of THg_{aq} and DOM quantity

176 and characteristics in leachate. The repeated-measures ANOVA, multivariate ANOVA, and one-way ANOVA were followed by

177 a *post hoc* test (Bonferroni's significant difference; honestly significant difference at the 95 % confidence interval). Weighed

178 least squares regression was used to examine the nature of the relationship between THg_{aq} concentrations and $SUVA_{254}$ in

179 leachate. Data are presented as the mean ± standard deviation (SD). Coefficient of determination (R^2) and significance p-values

180 (p) are presented for linear regression fits, and $p < 0.05$ was considered significant.

181 **4 Results and discussion**

182 **4.1 Foliar mercury accumulation in peatland plants**

183 Foliar THg concentrations were related to time/leaf age ($F_{(1.73,24.26)} = 42.75$, $p < 0.001$) and plant species ($F_{(1.23,23.38)} = 29.38$, $p <$

184 0.001) (Fig. 1). Based on *post hoc* tests, foliar THg concentrations were significantly different between plant species and

185 between the sampling months, except that there was no significant difference in foliar THg concentrations between June and

186 August. The mean foliar THg concentrations (n = 5) in June followed the sequence: few-seeded sedge/wire sedge < tussock

187 sedge < sweet gale. In July foliar THg concentrations decreased by 30 % (few-seeded sedge/wire sedge), 40 % (tussock sedge),

188 and 47 % (sweet gale), respectively. The decrease in THg concentrations is likely because of leaf growth dilution, although

189 changes in leaf biomass were not quantified as part of this study. Foliar THg concentrations were positively related to time after

190 July (few-seeded sedge/wire sedge: $F_{(1,13)} = 185.79$, $p < 0.001$, $R^2 = 0.93$; tussock sedge: $F_{(1,13)} = 200.87$, $p < 0.001$, $R^2=0.94$;

191 sweet gale: $F_{(1,13)} = 70.72$, $p < 0.001$, $R^2=0.84$). The mean foliar THg concentrations in October few-seeded sedge/wire sedge,

192 tussock sedge, and sweet gale were 1.7, 1.3, and 2.0 times higher than the initial concentrations in June. This result showed a

193 clear pattern of continuous THg accumulation from the atmosphere over time as has been shown for forests (Laacouri et al.,

194 2013; Millhollen et al., 2006b; Rea et al., 2002), given that plant roots act as a barrier of Hg transport from soils to shoots (Wang

195 et al., 2015).
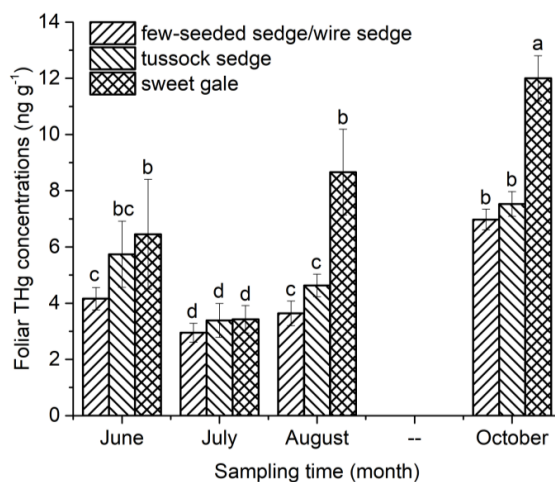
Figure 1 The intraseasonal trend in foliar total mercury (THg) concentrations (ng g⁻¹) of few-seeded sedge/wire sedge, tussock sedge, and sweet gale (ng g⁻¹). All concentrations are expressed in dry weight. Error bars are ± SD (n = 5 for each species for each time interval). The same letters above bars denote that values of foliar THg concentrations are not significantly different at the 0.05 levels.

Mercury accumulation in leaves is affected by many factors, such as atmospheric Hg concentration, environmental conditions (e.g., solar radiation and temperature), and biological factors (e.g., leaf age, plant species, leaf area, and leaf placement) (Blackwell and Driscoll, 2015; Ericksen et al., 2003; Ericksen and Gustin, 2004; Laacouri et al., 2013; Millhollen et al., 2006a). Since all samples were collected in the same location, factors such as atmospheric Hg concentration and environmental conditions were deemed the same, leaving only biological factors as an explanation for differences.

Leaf age. Leaf age is an important biological factor in controlling foliar concentrations (Ericksen et al., 2003; Laacouri et al., 2013). The positive relationship between foliar THg concentrations and time after July suggests that leaves of all species here continued to assimilate atmospheric Hg over the growing season right up to senescence. Some studies have found that the rate of foliar Hg uptake decreased toward the end of the growing season (Ericksen et al., 2003; Laacouri et al., 2013; Poissant et al., 2008), which appears to be because of the decrease of photosynthetic activity at the end of the growing season (Koike et al., 2003). Despite the decline of foliar Hg uptake during the late growing season, foliar Hg concentrations continue to increase right up to senescence because of the immobilization of the majority of foliar Hg (Laacouri et al., 2013; Lodenius et al., 2003; Stamenkovic and Gustin, 2009).

Plant species. Plant photosynthesis, transpiration, growth rates, and leaf area are different among plant species (Antúnez et al., 2001; Laacouri et al., 2013; Millhollen et al., 2006b), and given that these are important controls on Hg accumulation, the

9

215   differences among species found in this study are not surprising. The mean foliar THg concentrations in tussock sedge were 1.2

216   times higher than that in few-seeded sedge/wire sedge, and although not measured as part of this study, tussock sedge has a

217   larger leaf area than few-seeded sedge/wire sedge (Newmaster et al., 1997). A larger leaf has more stomates and thus more leaf

218   accumulation of atmospheric Hg (Laacouri et al., 2013; Millhollen et al., 2006; Stamenkovic and Gustin, 2009). The higher

219   relative Hg concentrations in sweet gale (mean 1.7 and 1.4 times higher than few seeded/wire, and tussock sedge, respectively) is

220   likely due in part to the same leaf area relationship. In addition, Kozlowski and Pallardy (1997) reported that leaves near the top

221   of the canopy generally have higher rates of photosynthesis and stomatal conductance than those near the bottom of the canopy

222   due to light saturation. Sweet gale had potentially higher stomatal conductance due to higher incident radiation and vapor

223   pressure deficits than sedges that are lower to the saturated ground with tightly packed vertical leaves.

224   Concentrations of Hg in senesced leaves of few-seeded sedge/wire sedge, tussock sedge, and sweet gale (6.58 ng g^{-1} to 12.77 ng

225   g^{-1}) were lower than that reported in tree litter (17 ng g^{-1} – 238 ng g^{-1}) (Laacouri et al., 2013; Obrist et al., 2021; Poissant et al.,

226   2008; Rea et al., 2002; Wang et al., 2016; Zhang et al., 2009) but similar to that previously reported for sedges and shrubs in

227   Canada (10.2 ± 6.8 ng g^{-1}) (Moore et al., 1995). The foliar Hg concentrations for plant species in this study increased 1.3-2.0

228   times over the growing season, which was smaller than that (3-11 fold) reported for trees (Laacouri et al., 2013; Poissant et al.,

229   2008; Rea et al., 2002). The above results suggested that foliar Hg concentrations differ among vegetation types (Demers et al.,

230   2007; Moore et al., 1995; Obrist et al., 2012; Richardson and Friedland, 2015), which might be attributed to the larger leaf and

231   higher stomatal density/ leaf placement in trees than sedges and shrubs.

232   **Leaf carbon, nitrogen and mercury.** Leaf %C, %N, and C:N were significantly different among plant species ($F_{(6,104)} = 59.64$, p

233   < 0.001) over the growing season ($F_{(9,124)} = 45.42$, $p < 0.001$) (Fig. 2). Based on *post hoc* tests, foliar %C, %N, and C:N was

234   significantly different between sweet gale and sedges (few-seeded sedge/wire sedge and tussock sedge) but not between few-

235   seeded sedge/wire sedge and tussock sedge. Foliar %C and %N were much lower in these sedges than sweet gale, which agrees

236   well with a previous study that deciduous shrubs (i.e., sweet gale) generally have a higher foliar %C and %N than grasses

237   (Wright et al., 2004). The fixation of nitrogen in sweet gale is in part attributed to sweet gale root nodules containing symbiotic

238   nitrogen-fixing (Newmaster et al., 1997; Vitousek et al., 2002) with this greater amount of available N leading to higher

239   photosynthetic capacity (Wright et al., 2004), thus, species containing a higher foliar %N are usually accompanied with a

240   higher %C.

241
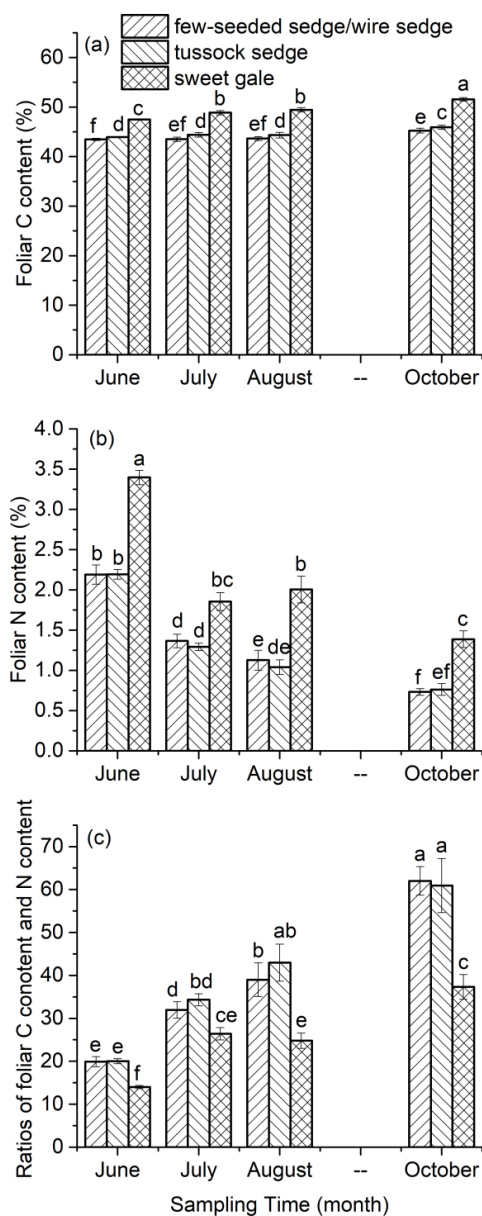
Figure 2 The carbon content (%C) (a), nitrogen content (%N) (b), and the ratio of carbon content to nitrogen content (C:N) (c) over the 2018 growing season. Vertical bars are mean ± SD (n = 5). The same letters above bars denote that values of foliar THg concentrations are not significantly different at the 0.05 levels.

There were significant increases in foliar %C (few-seeded sedge/wire sedge: $F_{(3,9)} = 25.98$, $p < 0.001$; tussock sedge: $F_{(3,9)} = 20.56$, $p < 0.001$; sweet gale: $F_{(3,9)} = 115.90$, $p < 0.001$) but sharp decreases of foliar %N (few-seeded sedge/wire sedge: $F_{(1.34,4.03)}$

11

248 $= 354.20$, $p < 0.001$; tussock sedge: $F_{(3,9)} = 252.36$, $p < 0.001$; sweet gale: $F_{(3,9)} = 170.43$, $p < 0.001$) over the growing season (Fig.

249 2). The strong decreases in foliar %N with leaf age can be attributed to the translocation of N from senescing leaves to new

250 leaves (Wang et al., 2003). A study found that approximately 77 % N, 57 % phosphorus (P), and 44 % potassium (K) were

251 translocated out of senescing leaves during mangrove leaf senescence (Wang et al., 2003). Foliar C is sequestrating continuously

252 over the growing season (Kueh et al., 2013). The element re-translocation and C sequestration in leaves may lead to the

253 foliar %C increase with time. The values of foliar C:N increased with time, which is a function of the decreases of foliar %N and

254 the increases of foliar %C.

255 Senesced leaf tissue with higher foliar %C and %N had higher foliar THg concentrations (%C and Hg: $F_{(1,13)} = 191.09$, $p < 0.05$,

256 $y = 0.78x - 28.20$, $R^2 = 0.94$; %N and Hg: $F_{(1,13)} = 82.38$, $p < 0.05$, $y = 7.16x - 1.96$, $R^2 = 0.93$) (Fig. 3a and 3b). THg

257 concentrations were negatively related to foliar C:N during senescence ($F_{(1,13)} = 175.10$, $p < 0.05$, $y = 0.18x - 18.33$, $R^2 = 0.86$;

258 Fig. 3c). A previous study found soil Hg concentrations were positively related to soil organic C and N, and then given a possible

259 explanation that high C and N levels in soil reflect high vegetation productivity corresponding with high atmospheric Hg

260 deposition via litterfall (Obrist et al., 2009). Although the mechanism of these relationships between Hg concentrations and

261 contents of C and N in senesced leaves materials is still unclear, this study shows that higher C and N content in senesced leaves

262 indirectly indicates a higher input of Hg via litterfall to soils.
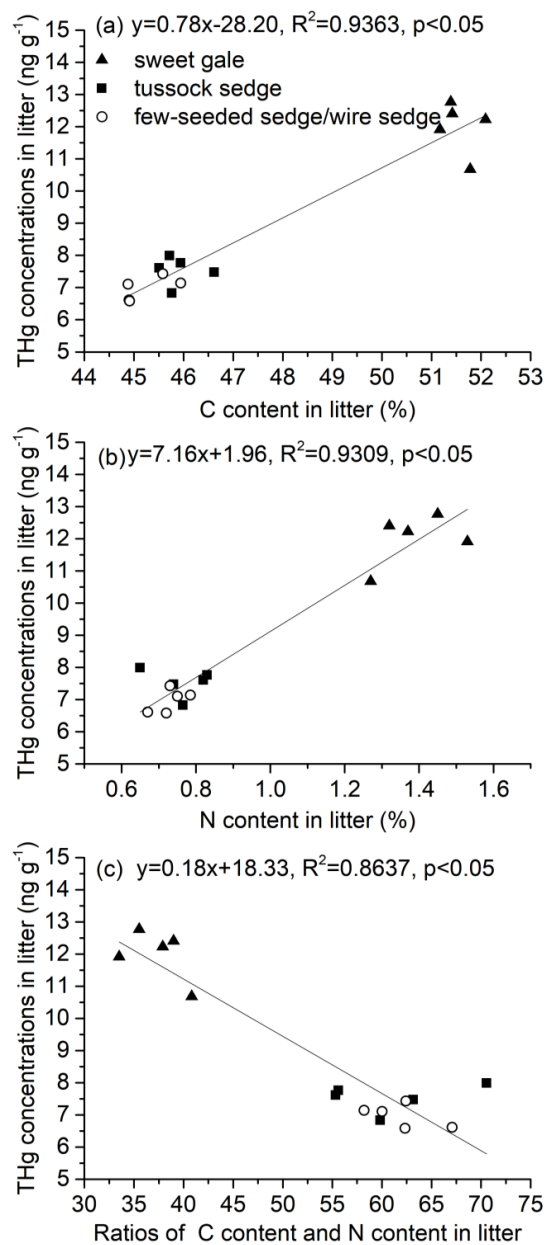
Figure 3 Correlations between (a) THg concentrations and C contents, (b) THg concentrations and N contents, and (c) THg concentrations and ratios of C content and N content (C:N) in litter. All linear correlations are statistically significant (p < 0.05).

4.2 Mercury leaching from senesced leaves

Surface-rinsable mercury. The mean mass of Hg from the surface rinse of senesced leaf material (expressed per gram of dry senesced leaf) was 0.02 ± 0.01 ng g^{-1} and 0.01 ± 0.00 ng g^{-1} (or 3.27 ± 1.68 ng L^{-1} and 1.39 ± 0.83 ng L^{-1}, expressed per liter of

269   rinse water (18.2 MΩ cm)), respectively, indicating that mass of Hg that was loosely bound on the leaf surface was small relative

270   to the total senesced leaf tissue Hg concentration (8.83 ± 2.38 ng g^{-1}) representing on average only 0.4 % Hg (tussock sedge:

271   0.6 %; few-seeded sedge/wire sedge: 0.3 %; sweet gale: 0.3 %) of total THg mass.

272   **Leachable mercury.** The mean THg$_{aq}$ mass per gram of senesced leaf had significant differences between plant species ($F_{(2,41)}$ =

273   11.55, p < 0.001; Fig. 4). Based on *post hoc* tests, there were significant differences in THg$_{aq}$ mass per gram of senesced leaf

274   between sweet gale and sedges (few-seeded sedge/wire sedge and tussock sedge) but not between few-seeded sedge/wire sedge

275   and tussock sedge. The senesced leaf of sweet gale leached the least Hg among these plant species, which is likely due to their

276   hydrophobic waxy cuticle that may both retain Hg, as well as protect the inner leaf material from leaching. Another plausible

277   explanation is that N was more easily released from sedges than C and it was the opposite for sweet gale, based on changes in

278   foliar %C and %N between before and after leaching (Table 1), whereas N groups in litter hinder the leaching of foliar Hg

279   (Obrist et al., 2009). Foliar %N of sweet gale increased after leaching, which is likely attributed to a large amount of loss of other

280   elements, such as K, Mg, and P, although they were not part of this experiment. Bessaad and Korboulewsky (2020) found that

281   60–79 % of K, 19–50 % of Mg, 22–30 % of P, and < 16 % of Ca and N were leached out from fully developed broadleaves

282   (collected in summer) during rainfall.
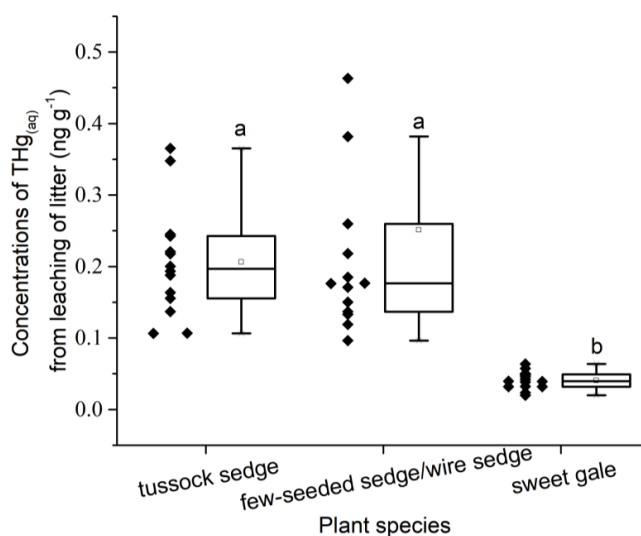
283

284   **Figure 4 Mass of mercury leached per gram of senesced leaf material (ng g^{-1}). Boxplot displays median (50th percentile; the inside line**

285   **of the box), first quartile (25th percentile; lower bound of the box), third quartile (75th percentile; upper bound of the box), whiskers**

286   **(all measures between 5th percentile and 25th percentile and between 75th percentile and 95th percentile; the straight line below and**

287   **above the box), and outliers (individual points outside of the percentile of 5th and 95th). n = 15.**

288 **Table 1 Changes of foliar carbon content (%C) and nitrogen content (%N) during leaching of litterfall. n = 15**

|	foliar %C		foliar %N	
	before leaching	after leaching	before leaching	after leaching
sweet gale	51.57 ± 0.36	51.03 ± 0.34	1.39 ± 0.10	1.50 ± 0.07
tussock sedge	45.91 ± 0.42	44.97 ± 0.54	0.76 ± 0.07	0.68 ± 0.10
few-seeded sedge/wire sedge	45.24 ± 0.49	43.83 ± 0.49	0.73 ± 0.04	0.64 ± 0.02

289

290 During experimental leaching, 3.0 %, 2.9 %, and 0.3 % of the total THg mass present in tussock sedge, few-seeded sedge/wire

291 sedge, and sweet gale senesced leaf was leached, respectively. The percentages of Hg that leached from tussock sedge, few-

292 seeded sedge/wire sedge leaves were 5.5 and 10.6 times higher than that from rinses, while the percentage of Hg that leached

293 from sweet gale senesced leaf was similar to that from rinse water (0.3 %). Rea et al. (2000) reported that surface washoff of

294 loosely bound and particulate Hg was a rapid and larger source of Hg in forest throughfall compared to continuously foliar Hg

295 leaching from live leaves. It is likely because dry leaves lack structural integrity compared to live leaves in Rea et al.'s (2000)

296 study, leading to more rapid leaching of soluble constituents (Gessner et al., 1999), including Hg, so the results of these prior

297 studies are not directly comparable to this one. Further, although Hg leached from senesced leaf material was a small (< 5 % of

298 foliar tissue Hg) but a measurable contributor to the mass balance, it is one that would be completely missed if material had been

299 collected from a litter trap that had been exposed to rainfall for any period.

300 **4.3 Quantity and characteristics of leachate dissolved organic matter**

301 **The quantity and characteristics of DOM in leachate.** The mean mass of DOC leached per gram of senesced leaf material and

302 the mass loss during senesced leaf material leaching was significantly different between plant species (leached DOC mass: $F_{(2,42)}$

303 $= 34.95$, $p < 0.001$; mass loss: $F_{(2,42)} = 11.62$, $p < 0.05$) with a same sequence following: few-seeded sedge/wire sedge < tussock

304 sedge < sweet gale (Fig. 5). The same sequence is in part because the loss of soluble carbons accounted for the majority of the

305 mass loss during litter leaching (Del Giudice and Lindo, 2017). Mass loss of sweet gale (17.7%) was significantly larger than

306 sedges (few-seeded sedge/wire sedge (8.1%) and tussock sedge (11.5%)). The released DOC accounted for $22.96 \pm 14.85\%$,

307 $23.73 \pm 12.95\%$, and $17.03 \pm 6.68\%$ of mass loss during senesced leaf material leaching for few-seeded sedge/wire sedge, tussock

308 sedge, and sweet gale, respectively. Loss of other nutrients, such as dissolved organic nitrogen (DON) and dissolved organic

309   phosphorus (DOP) (Ong et al., 2017; Liu et al., 2018; Hensgens et al., 2020) and the inorganic components and other elemental

310   organic matter (Lavery et al., 2013; Jiménez et al., 2017) also contribute to the mass loss, despite these nutrients not being
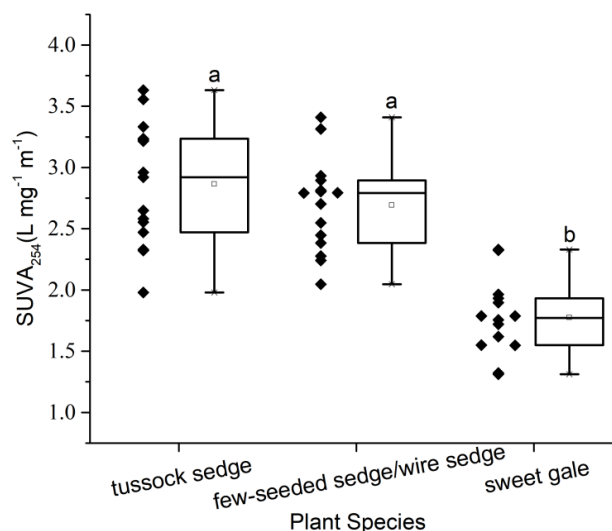
311   measured.
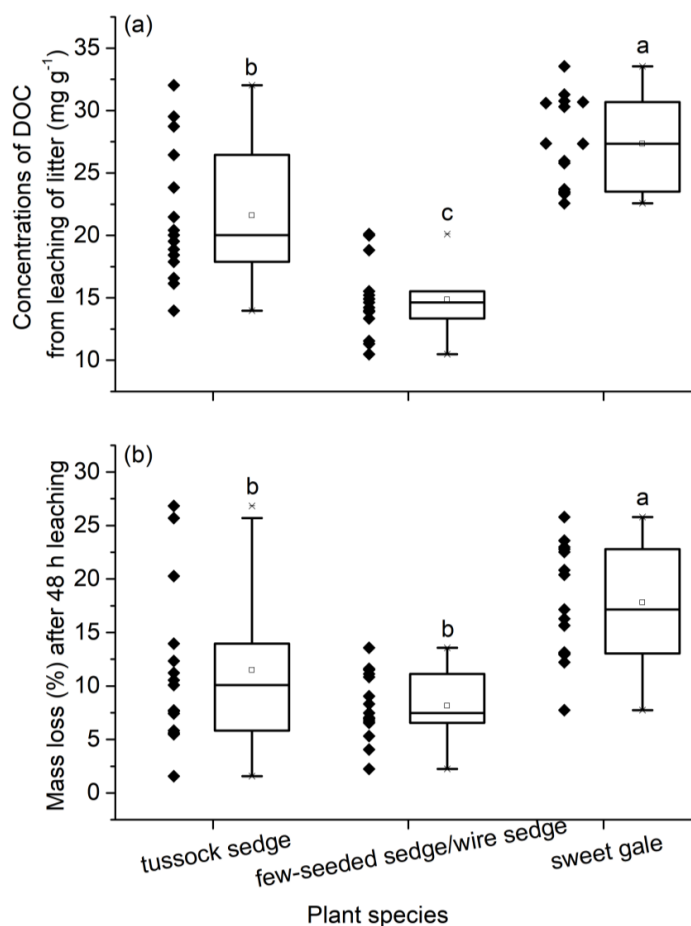
Figure 5 Mass of dissolved organic carbon (DOC) leached per gram of senesced leaf material (mg g⁻¹) (a), and mass loss after 48 h
leaching (b). Boxplot displays median (50th percentile; the inside line of the box), first quartile (25th percentile; lower bound of the
box), third quartile (75th percentile; upper bound of the box), whiskers (all measures between 5th percentile and 25th percentile and
between 75th percentile and 95th percentile; the straight line below and above the box), and outliers (individual points outside of the
percentile of 5th and 95th). n = 15.

318   Characteristics of DOM also varied among plant species (SUVA$_{254}$: $F_{(2,42)} = 24.02$, $p < 0.001$; HIX$_{EM}$:$F_{(2,42)} = 3.82$, $p < 0.05$; FI:

319    $F_{(2,42)} = 11.24$, $p < 0.001$; BIX: $F_{(2,42)} = 125.48$, $p < 0.001$) (Fig. 6 and Table 2). Based on *post hoc* tests, there were significant

320   differences in SUVA$_{254}$ between sweet gale and sedges (few-seeded sedge/wire sedge) only and in BIX among all plant species;

321   there were no significant differences in HIX$_{EM}$ among plant species.

322

Figure 6 Dissolved organic matter characteristics as measured by specific ultraviolet absorbance at the wavelength 254 nm (SUVA$_{254}$), n = 15. Boxplot displays median (50th percentile; the inside line of the box), first quartile (25th percentile; lower bound of the box), third quartile (75th percentile; upper bound of the box), whiskers (all measures between 5th percentile and 25th percentile and between 75th percentile and 95th percentile; the straight line below and above the box), and outliers (individual points outside of the percentile of 5th and 95th).

Table 2 The mean fluorescence indices of dissolved organic matter characteristics[a]

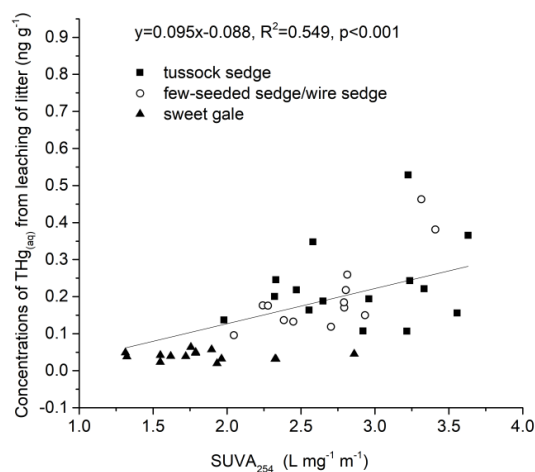| Index	Tussock sedge	Few-seeded sedge/wire sedge	Sweet gale
FI	1.19 ± 0.10	1.31 ± 0.09	1.49 ± 0.27
HIX$_{EM}$	0.16 ± 0.03	0.16 ± 0.02	0.19 ± 0.03
BIX	0.53 ± 0.05	0.63 ± 0.06	0.35 ± 0.04

329   [a]Lower values of the FI (< 1.2) suggest dissolved organic matter (DOM) has higher aromaticity and is primarily composed of high-molecular-weight DOM, while high FI values (> 1.8) indicate that DOM has lower aromaticity and is mainly composed of low-molecular-weight DOM. DOM with high HIX$_{EM}$ (> 1) values is composed of more highly condensed and higher molecular weight molecules. In contrast, higher BIX (> 1.0) values reflect that more low-molecular-weight DOM is recently produced, generally, by microbes. All indices are unitless, n = 15.

334   The mean value of $SUVA_{254}$ in leachate followed the sequence: tussock sedge > few-seeded sedge/wire sedge > sweet gale

335   leaves, respectively, indicating that leached DOM from tussock sedge and few-seeded sedge/wire sedge leaves had higher

336   aromaticity and less bioaccessible than that from the sweet gale leaves. These results are supported by indexes of FI and HIX_{EM}.

337   DOM in senesced leaf material leachate of tussock sedge and few-seeded sedge/wire sedge had lower values of FI and HIX_{EM}

338   than that of sweet gale leaves, indicative of the presence of less bioaccessible and more aromatic DOM contents in sedges than in

339   sweet gale. All BIX values (0.26–0.73) measured in this study were lower than 1.0, reflecting that DOM is mainly terrestrially

340   derived (leaching from litterfall) in this study. Although DOM leached from different litters has different characteristics, DOM

341   leaching from litters is a substantial source to surrounding ecosystems (Davis et al., 2003; Davis et al., 2006; Del Giudice and

342   Lindo, 2017). Importantly, the leached DOM (e.g., organic acids, sugars; amino acids) can provide energy and nutrients for

343   microbes (Davis et al., 2003), which will subsequently stimulate biological degradation and Hg methylation.

344   **4.4 Correlation between THg_{aq} concentrations and $SUVA_{254}$ values in leachate.**

345   The concentrations of soluble THg_{aq} were significantly related to $SUVA_{254}$ values ($F_{(1,41)}$ = 52.06, p < 0.001, y = 0.09x – 0.09, R^2

346   = 0.55; Fig. 7). Hg is tightly and readily bound to reduced sulfur groups (i.e., thiols) in DOM (Ravichandran, 2004; Xia et al.,

347   1999), especially those with higher aromaticity that have more reduced sulfur groups (Dittman et al., 2009). Mercury weakly

348   binds to carboxyl and phenol functional groups in DOM after all thiol groups are occupied at relatively high Hg concentrations

349   (Drexel et al., 2002; Graham et al., 2012), which is atypical in most natural environments in which Hg concentrations are

350   relatively low. This result agreed well with the literature indicating that DOM with higher aromaticity plays an important role in

351   controlling Hg mobility, given that the number of reduced sulfur groups far exceeds the amount of Hg in natural environments

352   (Ravichandran, 2004).

Figure 7 Correlations between the mass of mercury leached per gram of senesced leaf material (THg$_{aq}$) and the specific ultraviolet absorbance at the wavelength 254 nm (SUVA$_{254}$) in leachate.

4.5 Estimation of annual input of Hg via senesced leaves and rapid Leaching to peat soils

The annual input of leaf biomass (mg/ha/yr) of few-seeded sedge/wire sedge into peat soils was 5.55 and 1.41 fold higher than tussock sedge and sweet gale, while the annual inputs of Hg (mg/ha/yr) via sweet gale leaves were 6.29 and 1.22 fold higher than via tussock sedge and few-seeded/wire sedge leaves in the sedge-dominated fen (Table 3). Annual total Hg input through senesced leaves to peat soils were 1.56, 8.03, and 9.82 mg/ha/yr for tussock sedge, few-seeded sedge/wire sedge, and sweet gale, respectively. The input of surficial Hg and leachable Hg accounted for 0.64 % and 0.37 %, 0.31 % and 3.20 %, and 2.86 % and 0.30 % of total foliar Hg input to peat soils annually for tussock sedge, few-seeded sedge/wire sedge, and sweet gale, respectively. The majority of Hg in senesced leaves (> 96 %) was from the deposition of solid plant tissues in litter.

366 **Table 3 Annual input of senesced leaves, and senesced leaf Hg, surficial Hg, and leached Hg during leaching into peat**

367 **soils per hectare and per year in the sedge-dominated fen (mg/ha/yr).**

Species	Senesced leaf biomass (mg/ha/yr)	Litter total Hg input (mg/ha/yr)	Washoff Hg input (mg/ha/yr)	Leachate Hg input (mg/ha/yr)
Tussock sedge	2.07×10^8	1.56	0.01	0.05
Few-seeded sedge/wire sedge	1.15×10^9	8.03	0.03	0.23
Sweet gale	8.18×10^8	9.82	0.03	0.03
Total	2.17×10^9	19.41	0.07	0.31

368

369 Based on the data from the study growing season, the annual input of Hg in total via senesced leaves (19.40 mg/ha/yr) was 5-

370 22 % of litterfall in forest ecosystems (e.g., jack pine/black spruce/balsam fir forest, red maple/birch forest, Norway spruce

371 forest; 86-372 mg/ha/yr) (St Louis et al., 2001; Graydon et al.,2008; Shanley and Bishop, 2012), which can be attributed to those

372 forest ecosystems having both higher mean foliar Hg concentrations (21-51 ng g^{-1}) (Zhou and Obrist, 2021) and much greater

373 aboveground biomass and litterfall inputs (2000-3488 kg/ha/yr) (Graydon et al., 2008) than plants in this study. The overall

374 annual Hg inputs via these senesced leaves to peat soils in this sedge-dominated fen were 59 % of that via wet deposition using

375 the mean precipitation Hg input estimates from the Experimental Lakes Area (33 mg/ha/yr) for the years 2001-2010, which is in

376 the same general geographic region of Ontario (St Louis et al., 2019).

377 **5 Conclusions**

378 This study shows that the widely-observed pattern of foliage accumulation of Hg from the atmosphere and changes in foliar Hg

379 concentrations over time are the same in peatland vascular plants as they are for forest trees and that the patterns are related to

380 time/leaf age and plant species. Although THg concentrations in litterfall in this study are relatively lower than that in the forest

381 litterfall, Hg input through litterfall to peatland soils cannot be neglected, given that peatlands are "hotspots" of MeHg

382 production (Mitchell et al., 2008). Foliar leaching of lower molecular weight DOM from peatland shrubs such as sweet gale

383 provides energy for bacteria (including Hg methylators) and can enhance microbial metabolism. Hg released from ubiquitous

384   sedge litter during leaching is relatively more quickly than the much slower release of tissue-associated Hg through the

385   decomposition of plant tissues. Thus, the supply of inorganic Hg to sites of methylation in peatlands has both fast and slow

386   pathways that may shift under climate change, given that peatland plant species composition and biomass will certainly change

387   under climate change.

388

389   ■DATA AVAILABILITY

390   All data generated or analysed during this study are included in this published article and its supplementary information files.

391   ■SUPPLEMENT

392   The supplement related to this article is available online.

393   ■AUTHOR CONTRIBUTION

394   Ting Sun carried the project out, collected all samples, performed the leaching experiment, analyzed samples and data, and wrote

395   the manuscript. Brian A. Branfireun designed the experiments, provided supervision, and edited the manuscript.

396   ■COMPETING INTERESTS

397   The authors declare that they have no conflict of interest.

398   ■DISCLAIMER

399   Publisher's note: Copernicus Publications remains neutral with regard to jurisdictional claims in published maps and institutional

400   affiliations.

401   ■ACKNOWLEDGMENTS

405 ■ABBREVIATIONS

406 Hg, mercury; MeHg, methylmercury; GEM, gaseous elemental mercury; RGM, reactive gaseous mercury; PBM, particulate-

407 bound mercury; THg, total mercury; THg_{aq}, dissolved total mercury; SRB, sulfate-reducing bacteria; %C, carbon content; %N,

408 nitrogen content; C:N, the ratio of leaf C content and N content; dissolved organic matter (DOM); DOC, dissolved organic

409 carbon; $SUVA_{254}$, specific ultraviolet absorbance at a wavelength of 254 nm; EEMs, fluorescence excitation-emission matrices;

410 FI, fluorescence index; HIX_{EM}, humification index; BIX, biological index; soil organic matter (SOM); CRM, certified reference

411 material; RSD, relative standard deviation.

412

413

414

415

416

417

418

419

420

421

422

423

424

References

Antúnez, I., Retamosa, E. C., and Villar, R.: Relative growth rate in phylogenetically related deciduous and evergreen woody species, Oecologia, 128, 172-180, DOI:10.1007/s004420100645, 2001.

Bessaad, A. and Korboulewsky, N.: How much does leaf leaching matter during the pre-drying period in a whole-tree harvesting system? Forest Ecol. Manag., 477, 118492, DOI:10.1016/j.foreco.2020.118492, 2020.

Blackwell, B. D. and Driscoll, C. T.: Deposition of mercury in forests along a montane elevation gradient. Environ. Sci. Technol., 49, 5363-5370, DOI:10.1021/es505928w, 2015.

Blodau, C., Mayer, B., Peiffer, S., Moore, T. R.: Support for an anaerobic sulfur cycle in two Canadian peatland soils, J. Geophy. Res-Biogeo., 112, G02004, DOI:10.1029/2006JG000364, 2007.

Branfireun, B. A., Heyes, A., and Roulet, N. T.: The hydrology and methylmercury dynamics of a Precambrian Shield headwater peatland, Water Resour. Res., 32, 1785-1794, DOI:10.1029/96WR00790, 1996.

Branfireun, B. A., Krabbenhoft, D. P., Hintelmann, H., Hunt, R. J., Hurley, J. P., Rudd, J. W. M.: Speciation and transport of newly deposited mercury in a boreal forest wetland: A stable mercury isotope approach. Water Resour. Res., 41, W06016, DOI:10.1029/2004WR003219, 2005.

Buttler, A., Robroek, B. J., Laggoun-Défarge, F., Jassey, V. E., Pochelon, C., Bernard, G., Delarue, F., Gogo, S., Mariotte, P., Mitchell, E. A. D.: Experimental warming interacts with soil moisture to discriminate plant responses in an ombrotrophic peatland. J. Veg. Sci., 26, 964-974, DOI:10.1111/jvs.12296, 2015.

Ong Demers, J. D., Driscoll, C. T., Fahey, T. J., Yavitt, J. B.: Mercury cycling in litter and soil in different forest types in the Adirondack region, New York, USA, Ecol. Appl., 17, 1341-1351, DOI:10.1890/06-1697.1, 2007.

Davis, S. E., Corronado-Molina, C., Childers, D. L., Day Jr, J. W.: Temporally dependent C, N, and P dynamics associated with the decay of *Rhizophora mangle* L. leaf litter in oligotrophic mangrove wetlands of the Southern Everglades, Aquat. Bot., 75, 199-215, DOI:10.1016/S0304-3770(02)00176-6, 2003.

Davis, S. E., Childers, D. L., and Noe, G. B. J. H.: The contribution of leaching to the rapid release of nutrients and carbon in the early decay of wetland vegetation. Hydrobiologia **569**, 87-97, DOI:10.1007/s10750-006-0124-1, 2006.

Del Giudice, R. and Lindo, Z.: Short-term leaching dynamics of three peatland plant species reveals how shifts in plant communities may affect decomposition processes, Geoderma, 285, 110-116, DOI:10.1016/j.geoderma.2016.09.028, 2017.

Dieleman, C. M., Branfireun, B. A., McLaughlin, J. W., Lindo, Z.: Climate change drives a shift in peatland ecosystem plant community: implications for ecosystem function and stability, Glob. Change Biol., 21, 388-395, DOI:10.1111/gcb.12643, 2015.

Dittman, J. A., Shanley, J. B., Driscoll, C. T., Aiken, G. R., Chalmers, A. T., Towse, J. E.: Ultraviolet absorbance as a proxy for total dissolved mercury in streams, Environ. Pollut., 157, 1953-1956, DOI:10.1016/j.envpol.2009.01.031, 2009.

Drexel, R. T., Haitzer, M., Ryan, J. N., Aiken, G. R., Nagy, K. L.: Mercury (II) sorption to two Florida Everglades peats: evidence for strong and weak binding and competition by dissolved organic matter released from the peat, Environ. Sci. Technol., 36, 4058-4064, DOI:10.1021/es0114005, 2002.

Ericksen, J., Gustin, M., Schorran, D., Johnson, D., Lindberg, S., Coleman, J.: Accumulation of atmospheric mercury in forest foliage, Atmos. Environ., 37, 1613-1622, DOI:10.1016/S1352-2310(03)00008-6, 2003.

Ericksen, J. A. and Gustin, M.: Foliar exchange of mercury as a function of soil and air mercury concentrations, Sci. Total Environ., 324, 271-279, DOI10.1016/j.scitotenv.2003.10.034, 2004.

Fellman, J. B., Hood, E., and Spencer, R. G.: Fluorescence spectroscopy opens new windows into dissolved organic matter dynamics in freshwater ecosystems: A review, Limnol. Oceanogr., 55, 2452-2462, DOI:10.4319/lo.2010.55.6.2452, 2010.

466 Frolking, S., Roulet, N. T., Moore, T. R., Richard, P. J., Lavoie, M., Muller, S. D.: Modeling northern peatland decomposition
467   and peat accumulation, Ecosystems 4, 479-498, DOI10.1007/s10021-001-0105-1, 2001.
468 Gessner, M. O., Chauvet, E., and Dobson, M.: A perspective on leaf litter breakdown in streams, Oikos 85, 377-384,
469   DOI10.2307/3546505, 1999.
470 Gorham, E.: Northern peatlands: Role in the carbon cycle and probable responses to climatic warming, Ecol. Appl., 1, 182-195,
471   DOI:10.2307/1941811, 1991.
472 Graham, A. M., Aiken, G. R., and Gilmour, C. C.: Dissolved organic matter enhances microbial mercury methylation under
473   sulfidic conditions, Environ. Sci. Technol., 46, 2715-2723, DOI:10.1021/es203658f, 2012.
474 Graydon, J. A., St. Louis, V. L., Hintelmann, H., Lindberg, S. E., Sandilands, K. A., Rudd, J. W., Kelly, C. A., Hall, B. D.,
475   Mowat, L. D.: Long-term wet and dry deposition of total and methyl mercury in the remote boreal ecoregion of Canada,
476   Environ. Sci. Technol., 42, 8345-8351, DOI:10.1021/es801056j, 2008.
477 Haitzer, M., Aiken, G. R., and Ryan, J. N.: Binding of mercury (II) to dissolved organic matter: The role of the mercury-to-DOM
478   concentration ratio, Environ. Sci. Technol., 36, 3564-3570, DOI:10.1021/es025699i, 2002.
479 Hansen, A. M., Kraus, T. E., Pellerin, B. A., Fleck, J. A., Downing, B. D., Bergamaschi, B. A.: Optical properties of dissolved
480   organic matter (DOM): Effects of biological and photolytic degradation, Limnol. Oceanogr., 61, 1015-1032,
481   DOI:10.1002/lno.10270, 2016.
482 Hensgens, G., Laudon, H., Peichl, M., Gil, I. A., Zhou, Q., Berggren, M.: The role of the understory in litter DOC and nutrient
483   leaching in boreal forests, Biogeochemistry, 149, 87-103, DOI:10.1007/s10533-020-00668-5, 2020.
484 Hintelmann, H., Harris, R., Heyes, A., Hurley, J. P., Kelly, C. A., Krabbenhoft, D. P., Lindberg, S., Rudd, J. W., Scott, K. J., St.
485   Louis, V. L.: Reactivity and mobility of new and old mercury deposition in a boreal forest ecosystem during the first year
486   of the METAALICUS study, Environ. Sci. Technol., 36, 5034-5040, DOI:10.1021/es025572t, 2002.
487 Hobbie, S. E.: Temperature and plant species control over litter decomposition in Alaskan tundra, Ecol. Monogr., 66, 503-522,
488   DOI:10.2307/2963492, 1996.
489 Huguet, A., Vacher, L., Relexans, S., Saubusse, S., Froidefond, J. M., Parlanti, E.: Properties of fluorescent dissolved organic
490   matter in the Gironde Estuary, Org. Geochem., 40, 706-719, DOI:10.1016/j.orggeochem.2009.03.002, 2009.
491 Jiménez, M. A., Beltran, R., Traveset, A., Calleja, M. L., Delgado-Huertas, A., Marba, N.: Aeolian transport of seagrass
492   (Posidonia oceanica) beach-cast to terrestrial systems, Estuar. Coast. Shelf S., 196, 31-44,
493   DOI:10.1016/j.ecss.2017.06.035, 2017.
494 Jiskra, M., Sonke, J. E., Obrist, D., Bieser, J., Ebinghaus, R., Myhre, C. L., Pfaffhuber, K. A., Wängberg, I., Kyllönen, K.,
495   Worthy, D.: A vegetation control on seasonal variations in global atmospheric mercury concentrations, Nat. Geosc., 11,
496   244-250, DOI:10.1038/s41561-018-0078-8, 2018.
497 Koike, T., Kitaoka, S., Ichie, T., Lei, T., Kitao, M.: Photosynthetic characteristics of mixed deciduous-broadleaf forests from leaf
498   to stand, Terra, Publishing, Tokyo, Japan. 2003.
499 Kueh, J. H. R., Ab Majid, N. M., Seca, G., Ahmed, O. H.: Above ground biomass-carbon partitioning, storage and sequestration
500   in a Rehabilitated forest, Bintulu, Sarawak, Malaysia, Sains Malays., 42, 1041-1050, 2013.
501 Laacouri, A., Nater, E. A., and Kolka, R. K.: Distribution and uptake dynamics of mercury in leaves of common deciduous tree
502   species in Minnesota, USA, Environ. Sci.Technol., 47, 10462-10470, DOI:10.1021/es401357z, 2013.
503 Lavery, P. S., McMahon, K., Weyers, J., Boyce, M. C., Oldham, C. E.: Release of dissolved organic carbon from seagrass wrack
504   and its implications for trophic connectivity, Mar. Ecol-Prog. Ser., 494, 121-133, DOI:10.3354/meps10554, 2013.
505 Lindberg, S., Bullock, R., Ebinghaus, R., Engstrom, D., Feng, X., Fitzgerald, W., Pirrone, N., Prestbo, E., Seigneur, C.: A
506   synthesis of progress and uncertainties in attributing the sources of mercury in deposition, AMBIO, 36, 19-33,
507   DOI:10.1579/0044-7447(2007)36[19:ASOPAU]2.0.CO;2, 2007.

508 Liu, S. L., Jiang, Z. J., Zhou, C. Y., Wu, Y. C., Arbi, I., Zhang, J. P., Huang, X. P., Trevathan-Tackett, S. M.: Leaching of
509   dissolved organic matter from seagrass leaf litter and its biogeochemical implications, Acta Oceanol. Sin., 37, 84-90,
510   DOI:10.1007/s13131-018-1233-1, 2018.
511 Lodenius, M., Tulisalo, E., and Soltanpour-Gargari, A.: Exchange of mercury between atmosphere and vegetation under
512   contaminated conditions, Sci. Total Environ., 304, 169-174, DOI:10.1016/S0048-9697(02)00566-1, 2003.
513 Lyons, C. L. and Lindon, Z.: Above-and belowground community linkages in boreal peatlands, Plant Ecol., 221, 615-632,
514   DOI:10.1007/s11258-020-01037-w, 2019.
515 McKnight, D. M., Boyer, E. W., Westerhoff, P. K., Doran, P. T., Kulbe, T., Andersen, D. T.: Spectrofluorometric characterization
516   of dissolved organic matter for indication of precursor organic material and aromaticity. Limnol. Oceanogr., 46, 38-48,
517   DOI:10.4319/lo.2001.46.1.0038, 2001.
518 Millhollen, A., Obrist, D., and Gustin, M.: Mercury accumulation in grass and forb species as a function of atmospheric carbon
519   dioxide concentrations and mercury exposures in air and soil, Chemosphere, 65, 889-897,
520   DOI:10.1016/j.chemosphere.2006.03.008, 2006a.
521 Millhollen, A. G., Gustin, M. S., and Obrist, D.: Foliar mercury accumulation and exchange for three tree species, Environ. Sci.
522   Technol., 40, 6001-6006, DOI:10.1021/es0609194, 2006b.
523 Mitchell, C. P. J., Branfireun, B. A., and Kolka, R. K.: Spatial characteristics of net methylmercury production hot spots in
524   peatlands, Environ. Sci. Technol., 42, 1010-1016, DOI:10.1021/es0704986, 2008.
525 Moore, T., Bubier, J., Heyes, A., Flett, R.: Methyl and total mercury in boreal wetland plants, Experimental Lakes Area,
526   Northwestern Ontario, J. Environ. Qual., 24, 845-850, DOI:10.2134/jeq1995.00472425002400050007x, 1995.
527 Morel, F. M., Kraepiel, A. M., and Amyot, M.: The chemical cycle and bioaccumulation of mercury, Annu. Rev. Ecol. Syst., 29,
528   543-566, DOI:10.1146/annurev.ecolsys.29.1.543, 1998.
529 Newmaster, S. G., Harris, A. G., and Kershaw, L. J.: Wetland plants of Ontario. Lone Pine Publishing, Edmonton, Canada, 1997.
530 Obrist, D., Agnan, Y., Jiskra, M., Olson, C., Colegrove, D., Hueber, J., Moore, C., Sonke, J., Helmig, D.: Tundra uptake of
531   atmospheric elemental mercury drives Arctic mercury pollution, Nature, 547, 201–204, DOI:10.1038/nature22997, 2017.
532 Obrist, D., Johnson, D., and Lindberg, S.: Mercury concentrations and pools in four Sierra Nevada forest sites, and relationships
533   to organic carbon and nitrogen, Biogeosciences, 6, 765-777, DOI:10.5194/bg-6-765-2009, 2009.
534 Obrist, D., Johnson, D. W., and Edmonds, R. L.: Effects of vegetation type on mercury concentrations and pools in two adjacent
535   coniferous and deciduous forests, J. Plant Nutr. Soil Sc., 175, 68-77, DOI:10.1002/jpln.201000415, 2012.
536 Obrist, D., Roy, E. M., Harrison, J. L., Kwong, C. F., Munger, J. W., Moosmuller, H., Romero, C. D., Sun, S.W., Zhou, J.,
537   Commane, R.: Previously unaccounted atmospheric mercury deposition in a midlatitude deciduous forest, P. Natl. Acad.
538   Sci. USA., 118, DOI:10.1073/pnas.2105477118, 2021.
539 Ohno, T.: Fluorescence inner-filtering correction for determining the humification index of dissolved organic matter, Environ.
540   Sci. Technol., 36, 742-746, DOI:10.1021/es0155276, 2002.
541 Ong, C. S. P., Joon, C. J., and Yule, C. M.: The contribution of leaching to nutrient release from leaf litter of two emergent tree
542   species in a Malaysian tropical peat swamp forest, Hydrobiologia, 794, 125-137, DOI:10.1007/s10750-017-3086-6, 2017.
543 Palozzi, J. E. and Lindo, Z.: Boreal peat properties link to plant functional traits of ecosystem engineers, Plant Soil, 418, 277-
544   291, DOI:10.1007/s11104-017-3291-0, 2017.
545 Poissant, L., Pilote, M., Yumvihoze, E., Lean, D.: Mercury concentrations and foliage/atmosphere fluxes in a maple forest
546   ecosystem in Quebec, Canada, J. Geophys. Res-Atmos., 113, DOI:10.1029/2007JD009510, 2008.
547 Ravichandran, M.: Interactions between mercury and dissolved organic matter: A review, Chemosphere, 55, 319-331,
548   DOI:10.1016/j.chemosphere.2003.11.011, 2004.
549 Rea, A., Lindberg, S., Scherbatskoy, T., Keeler, G.J.: Mercury accumulation in foliage over time in two northern mixed-

550   hardwood forests, Water Air Soil Poll., 133, 49-67, DOI:10.1023/A:1012919731598, 2002.

551   Rea, A. W., Lindberg, S. E., and Keeler, G. J.: Assessment of dry deposition and foliar leaching of mercury and selected trace

552   elements based on washed foliar and surrogate surfaces, Environ. Sci. Technol., 34, 2418-2425, DOI:10.1021/es991305k,

553   2000.

554   Richardson, J. and Friedland, A.: Mercury in coniferous and deciduous upland forests in northern New England, USA:

555   implications of climate change, Biogeosciences, 12, 6737-6749, DOI:10.5194/bg-12-6737-2015, 2015.

556   Risch, M. R., DeWild, J. F., Gay, D. A., Zhang, L., Boyer, E. W., Krabbenhoft, D. P.: Atmospheric mercury deposition to forests

557   in the eastern USA, Environ. pollut., 228, 8-18, DOI:10.1016/j.envpol.2017.05.004, 2017.

558   Risch, M. R., DeWild, J. F., Krabbenhoft, D. P., Kolka, R. K., Zhang, L. M.: Litterfall mercury dry deposition in the eastern

559   USA, Environ. Pollut., 161, 284-290, DOI:10.1016/j.envpol.2011.06.005, 2012.

560   Rydin, H. and Jeglum, J. K.: The biology of peatlands. (2nd ed.), Oxford, United Kingdom, Oxford University Press. 2013.

561   Schaefer, J. K. and Morel, F. M.: High methylation rates of mercury bound to cysteine by *Geobacter sulfurreducens*, Nat.

562   Geosci., 2, 123, DOI:10.1038/NGEO412, 2009.

563   Schaefer, J. K., Rocks, S. S., Zheng, W., Liang, L., Gu, B., Morel, F. M.: Active transport, substrate specificity, and methylation

564   of Hg (II) in anaerobic bacteria, P. Natl. Acad. Sci. USA, 108, 8714-8719, DOI:10.1073/pnas.1105781108, 2011.

565   Schmalenberger, A., Drake, H. L., and Küsel, K.: High unique diversity of sulfate-reducing prokaryotes characterized in a depth

566   gradient in an acidic fen, Environ. Microbiol., 9, 1317-1328, DOI:10.1111/j.1462-2920.2007.01251.x, 2007.

567   Schroeder, W. H. and Munthe, J.: Atmospheric mercury: An overview, Atmos. Environ., 32, 809-822, DOI:10.1016/S1352-

568   2310(97)00293-8, 1998.

569   Shanley, J. B. and Bishop, K.: Mercury cycling in terrestrial watersheds. In: Banks, M. S. (Ed.), Mercury in the Environment:

570   pattern and process, pp. 119-141. 2012.

571   Skyllberg, U., Xia, K., Bloom, P. R., Nater, E. A., Bleam, W. F.: Binding of mercury (II) to reduced sulfur in soil organic matter

572   along upland-peat soil transects, J. Environ. Qual., 29, 855-865, DOI:10.2134/jeq2000.00472425002900030022x, 2000.

573   St. Louis, V. L., Graydon, J. A., Lehnherr, I., Amos, H. M., Sunderland, E. M., St. Pierre, K. A., Emmerton, C. A., Sandilands, K.,

574   Tate, M., Steffen, A., Humphreys, E. R.: Atmospheric concentrations and wet/dry loadings of mercury at the remote

575   Experimental Lakes Area, Northwestern Ontario, Canada, Environ. Sci. Technol., 53, 8017-8026,

576   DOI:10.1021/acs.est.9b01338, 2019.

577   St. Louis, V. L., Rudd, J. W., Kelly, C. A., Beaty, K. G., Bloom, N. S., Flett, R. J.: Importance of wetlands as sources of methyl

578   mercury to boreal forest ecosystems, Can. J. Fish. Aquat. Sci., 51, 1065-1076, DOI:10.1139/f94-106, 1994.

579   St. Louis, V. L., Rudd, J. W., Kelly, C. A., Hall, B. D., Rolfhus, K. R., Scott, K. J., Lindberg, S. E., Dong, W.: Importance of the

580   forest canopy to fluxes of methyl mercury and total mercury to boreal ecosystems, Environ. Sci. Technol., 35, 3089-3098,

581   DOI:10.1021/es001924p, 2001.

582   Stamenkovic, J. and Gustin, M. S.: Nonstomatal versus stomatal uptake of atmospheric mercury, Environ. Sci. Technol., 43,

583   1367-1372, DOI:10.1021/es801583a, 2009.

584   Streets, D. G., Devane, M. K., Lu, Z., Bond, T. C., Sunderland, E. M., Jacob, D. J.: All-time releases of mercury to the

585   atmosphere from human activities, Environ. Sci. Technol., 45, 10485-10491, DOI:10.1021/es202765m, 2011.

586   Ullrich, S. M., Tanton, T. W., and Abdrashitova, S. A.: Mercury in the aquatic environment: A review of factors affecting

587   methylation, Crit. Rev. Env. Sci. Tec., 31, 241-293, DOI:10.1080/20016491089226, 2001.

588   Vitousek, P. M., Cassman, K., Cleveland, C., Crews, T., Field, C. B., Grimm, N. B., Howarth, R. W., Marino, R., Martinelli, L.,

589   Rastetter, E. B., Sprent, J. I.: Towards an ecological understanding of biological nitrogen fixation, Biogeochemistry, 57,

590   1-45, DOI:10.1023/A:1015798428743, 2002.

591   Wang, W. Q., Wang, M., and Lin, P.: Seasonal changes in element contents in mangrove element retranslocation during leaf
592   senescene, Plant Soil, 252, 187-193, DOI:10.1023/A:1024704204037, 2003.
593   Wang, X., Bao, Z., Lin, C. J., Yuan, W., Feng, X.: Assessment of global mercury deposition through litterfall, Environ. Sci.
594   Technol., 50, 8548-8557, DOI:10.1021/acs.est.5b06351, 2016.
595   Wang, X., Tam, N. F., He, H. D., Ye, Z. H.: The role of root anatomy, organic acids and iron plaque on mercury accumulation in
596   rice, Plant Soil, 394, 301-313, DOI:10.1007/s11104-015-2537-y, 2015.
597   Webster, K. L. and McLaughlin, J. W.: Importance of the water table in controlling dissolved carbon along a fen nutrient
598   gradient, Soil Sci. Soc. Am. J., 74, 2254-2266, DOI:10.2136/sssaj2009.0111, 2010.
599   Weishaar, J. L., Aiken, G. R., Bergamaschi, B. A., Fram, M. S., Fujii, R., Mopper, K.: Evaluation of specific ultraviolet
600   absorbance as an indicator of the chemical composition and reactivity of dissolved organic carbon, Environ. Sci.
601   Technol., 37, 4702-4708, DOI:10.1021/es030360x, 2003.
602   Weltzin, J. F., Pastor, J., Harth, C., Bridgham, S. D., Updegraff, K., Chapin, C. T.: Response of bog and fen plant communities to
603   warming and water-table manipulations, Ecology, 81, 3464-3478, DOI:10.1890/0012-
604   9658(2000)081[3464:ROBAFP]2.0.CO;2, 2000.
605   Wright, I. J., Reich, P. B., Westoby, M., Ackerly, D. D., Baruch, Z., Bongers, F., Cavender-Bares, J., Chapin, T., Cornelissen, J.
606   H. C., Diemer, M., Flexas, J., Garnier, E., Groom, P. K., Gulias, J., Hikosaka, K., Lamont, B. B., Lee, T., Lee, W., Lusk,
607   C., Midgley, J. J., Navas, M. L., Niinemets, U., Oleksyn, J., Osada, N., Poorter, H., Poot, P., Prior, L., Pyankov, V. I.,
608   Roumet, C., Thomas, S. C., Tjoelker, M. G., Veneklaas, E. J., Villar, R.: The worldwide leaf economics spectrum, Nature,
609   428, 821-827, DOI:10.1038/nature02403, 2004.
610   Xia, K., Skyllberg, U., Bleam, W., Bloom, P., Nater, E., Helmke, P.: X-ray absorption spectroscopic evidence for the
611   complexation of Hg (II) by reduced sulfur in soil humic substances, Environ. Sci. Technol., 33, 257-261,
612   DOI:10.1021/es980433q, 1999.
613   Zhang, H., Holmes, C., and Wu, S.: Impacts of changes in climate, land use and land cover on atmospheric mercury, Atmos.
614   Environ., 141, 230-244, DOI:10.1016/j.atmosenv.2016.06.056, 2016.
615   Zhang, L., Wright, L. P., and Blanchard, P.: A review of current knowledge concerning dry deposition of atmospheric mercury,
616   Atmos. Environ., 43, 5853-5864, DOI:10.1016/j.atmosenv.2009.08.019, 2009.
617