Dear Editor,

Please find our response to the two external reviewers comments on the above manuscript. Both reviews were very helpful in clarifying some ideas and interpretations. We have addressed all of the reviewers comments below, to what we feel is a complete and thorough way. We are grateful for the opportunity to resubmit this manuscript for publication in Biogeosciences.

Sincerely, Ting Sun and Brian A. Branfireun

Reviewer 1

Sun et al. "Plant mercury accumulation and litter input to a Northern Sedge-dominated Peatland" investigated the foliar Hg concentration and flux via Sedge plant in peatland. In addition, they also carried out a leaching experiment to explore the Hg behavior in leaching process driven by DOM. I think this MS is important research to understand the Hg biogeochemical cycle and well written. However, I think this discussion is not enough in this MS, especially in relationship between DOM and Hg. I hope more effective discussion should be added in the revised MS. Moreover, the figures should be revised fully.

Reply: *We are grateful for the reviewer's deeply informed and insightful comments. We have addressed the detailed comments that were made on the PDF copy of the earlier version of the manuscript. We think figures in black and white have complied with the journal's guidelines for figures. Please find our replies to your comments and suggestions below.*

Introduction: It is too divergent. The author should focus on the Hg specific behavior in Hg cycles at peatland, highlighting the importance in global Hg cycles and differences from forest ecosystems. Add more recent references.

Reply: *We agree with this suggestion. We have modified the introduction based on the reviewer's suggestion. The revised text (now on Page3-5, Line 43-108) reads as follows "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, Hgp) 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).*

Vegetation is generally considered a sink for atmospheric Hg, with the majority of Hg in vegetation leaves accumulated from the atmosphere (Jiskra et al., 2018; Obrist et al., 2017). Plant leaves accumulate Hg from the atmosphere mainly through stomatal uptake (Lindberg et al., 1992). Stamenkovic and Gustin (2009) suggested that the non-stomatal pathway of Hg deposition to the leaf cuticle and subsequently retention and incorporation into leaf tissue also plays an important role in accumulating atmospheric Hg. Plant roots are thought to generally act as a barrier of Hg transport from soils to shoots (Wang et al., 2015), and it has been shown that less than 10% of Hg in roots is transported to the aboveground portion of plants (Ericksen et al., 2003; Mao et al., 2013). Some studies have found that a great proportion of foliar Hg in halophytes in salt marshes was translocated from the root (Canário et al., 2017; Cabrita et al., 2019; Weis and Weis 2004). The plausible reason is that plants in the hydroponic growth system have fewer apoplastic barriers (i.e. Casparian bands and suberin lamellae) in root architecture than plants grown in contaminated soils (Redjala et al., 2011).

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 in boreal peatlands ecosystems are few (see Moore et al., 1995) despite their critical role in the carbon (Gorham, 1991) and Hg cycles (Grigal, 2003). Boreal peatlands are a type of wetland that stores large amounts of Hg (Grigal 2003) and can be major 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 facilitating net MeHg production (Mitchell et al., 2008). Elucidation of foliar Hg input from the dominant plant types to boreal peatlands is important to further estimate the supply of bioavailable Hg(II) for net MeHg production.

Previous studies have found that the majority of Hg in plant leaves in wetlands was from the atmosphere (Brahmstedt et al., 2021; Enrico et al., 2016; Fay and Gustin 2007) and nonvascular plants (e.g., fungi, lichens, and mosses) had higher foliar Hg concentrations than vascular plants (Moore et al., 1995; Pech et al., 2022). Although foliar Hg concentration is lower in vascular plants than in nonvascular plants, Hg mass input to peatlands may be substantial, given the greater litter input from vascular plants than from nonvascular plants (Frolking et al., 2001). With more bioaccessible litter and leachate than bryophytes (Hobbie, 1996; Lyons and Lindo, 2019), vascular plant inputs also have a faster initial decomposition rate (0.2 y^{-1}) than bryophytes (0.05-0.08 y^{-1}) (Frolking et al., 2001), leading to a rapid Hg release to the soil Boreal peatlands are experiencing rising temperatures due to climate change (IPCC, 2018) that is likely to both increase aboveground biomass in vascular plant-dominated peatlands (Tian et al., 2020) and *promote a shift from moss-dominated to more vascular plant-dominated plant communities (Buttler et al., 2015; Dieleman et al., 2015; Weltzin et al., 2000) further affecting Hg deposition (Zhang et al., 2016). To date, the amount of atmospheric Hg accumulated in dominant plants in the vascular plant-dominated (i.e., graminoid plants and shrubs) peatlands, an important type of boreal wetlands (Rydin and Jeglum, 2013), is unknown.*

Foliar Hg eventually enters peat soils via litterfall and is expected to follow the sequence: (1) wash-off of aerosols, particles, and gases from leaf surfaces, (2) leaching of water-soluble components, and (3) incorporation into SOM after the microbial decomposition of litter. Leaching is the initial phase of litter breakdown in aquatic environments and can rapidly release up to 30 % dissolved matter, primarily dissolved organic matter (DOM) within 24 h after immersion of litter (Gessner et al., 1999). It has been established that dissolved organic matter (DOM) is closely related to Hg mobility in terrestrial and aquatic ecosystems (Haitzer et al., 2002; Ravichandran, 2004; Kneer et al., 2020), given the strong affinity between Hg and reduced sulfur groups (i.e., thiols) in DOM (Xia et al., 1999). DOM with higher aromaticity has more thiols ligands and has a stronger correlation with Hg (Dittman et al., 2009). The rapid and abundant leaching of DOM, especially those with higher aromaticity from litterfall may lead to large amounts of Hg leaching. The amount of rapidly released Hg during litter leaching is unknown and needs to be elucidated because more recently deposited Hg appears to be more readily methylated than "old" Hg in peat soils (Branfireun et al., 2005; Feng et al., 2014; Hintelmann et al., 2002). Despite previous studies showing that Hg mass in live leaf leachate is insignificant compared to that on leaf surfaces and in SOM (Rea et al., 2001; Rea et al., 2000), litterfall generally lacks structural integrity and likely leaches more Hg compared to live leaves.

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 vascular plant-dominated fen-type peatland. We use "sedge-dominated fen" instead of "vascular plant-dominated fen-type peatland" hereafter, given that sedges are the primarily dominant plants in this study site (Webster and McLaughlin, 2010). The specific objectives of this study are to:

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

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

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

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

Line 23: While should be revised.

Reply:*We agree with this suggestion.We have revised this in the manuscript. The revised text (now*

Page 2, Line23) as "Plant foliage plays an essential role in accumulating mercury (Hg) from the atmosphere and transferring it to soils in terrestrial ecosystems, while many studies have focused on forested ecosystems."

Line 27: It is inconsistent.

Reply:*We agree with this comment. The clarified text (Page 2, Line 27) reads as follows "Foliar Hg concentrations decreased early in the growing season due to growth dilution, and after that were subsequently positively correlated with leaf age (time)".*

Line 70: It is confused. Further explain it.

Reply:*We agree with this suggestion. The clarified text (Page 4, Line 77) reads as follows "Although foliar Hg concentration is lower in vascular plants than in nonvascular plants, Hg mass input to peatland might be important, given the more amount of litter input from vascular plants than from nonvascular plants (Frolking et al., 2001)."*

Line 71-72: Plant decomposition turnover time should be mentioned as you talked about the Hg biogeochemical cycles.

Reply:*We agree with this suggestion. We have incorporated additional content in this section. The revised text (Page 4, Line 80-81)reads as follows "With more bioaccessible litter and leachate than bryophytes (Hobbie, 1996; Lyons and Lindo, 2019), vascular plant inputs also have a faster initial decompose rate (0.2 y^{-1}) than bryophytes (0.05-0.08 y^{-1}) (Frolking et al., 2001), leading to a rapid Hg release to the soil and/or facilitating net methylation.".*

Line 73-83: I cannot get the significance of this section. I agreed with those points, but global warming has nothing to do with this research. Only one sentence is enough to highlight the importance of peatlands.

Reply:*We agree that one sentence is enough for this section. The clarified text (Page 4, Line 81-87) reads as follows "Importantly, boreal peatlands are experiencing rising temperatures due to climate change (IPCC, 2018) that is likely to either increase aboveground biomass in vascular* *plant-dominated peatlands (Tian et al., 2020) or promote a potential shift from moss-dominated peatlands to more vascular plant-dominated (Buttler et al., 2015; Dieleman et al., 2015; Weltzin et al., 2000). Changes in plant abundance and community composition may further affect Hg deposition (Zhang et al., 2016) in boreal peatlands. So far, the amount of atmospheric Hg accumulated in dominant plants in the vascular plant-dominated (i.e., graminoid plants and shrubs) peatlands, an important type of boreal wetlands (Rydin and Jeglum, 2013), is unknown."*

Line 105: Offer the percentage of each dominate species.

Reply:*We agree with this suggestion. We have incorporated the species percent cover of each dominant species in this section. The revised text (Page 5, Line 117-118)reads as follows "The species percent cover of few-seeded sedge; wire sedge; tussock sedge, and sweet gale was 35.0 ± 21.79%, 0.3 ± 0.12%, 73.0 ± 18.81%, 44.8 ± 10.63%, respectively (Palozzi and Lindo 2017)."*

Line 130 and SI 32-37: Why did you choose the DORM-4 (Fish protein certified reference material) as the standard sample, not the plant standard samples?

Reply:*Thank you for the reviewer's comment. In this study, DORM-4 was used to validate instrument (Milestone™ DMA-80) recovery and stability not for the method validation step (accuracy, precision, recovery of samples). Milestone™ DMA-80 is very stable and reliable. A single calibration suits all wide variety of samples with concentrations ranging from ppm to ppt. The DMA-80 calibration provides long-term reliability due to the stability of the system and the long lifetime of the catalyst tube and gold amalgamator. These features allow us to eliminate the daily calibrations often required by conventional instrumentation. In this study, the concentration range of the regular calibration was from 0 to 1 mg/kg. Total Hg concentration in DORM-4 is 0.410 ± 0.055 mg/kg, which is in the middle of the calibration concentration range and thus can validate instrument recovery and stability very well. There is one CRM "IAEA-140/TM Trace elements and methylmercury in seaweed"for total mercury concentration, but the Hg concentration of CRM is 0.038 (0.032-0.044 mg/kg), which is not suitable to validate instrument recovery and stability.*

Line 183: Explain the subscript in F(1.73,24.26).

Reply:*Thank you for the reviewer's comment. For the repeated-measures ANOVA, we get the results F(x,y) and p. F is the statistics of the repeated-measures ANOVA, p-value indicates that if the mean difference among all groups was statistically significant. x and y is calculated based on the degree of freedom between subjects and the degree of freedom within subjects. Based on Mauchly's Test of Sphericity, if the results did not follow Mauchly's Test of Sphericity, the results needed to be modified with epsilon (e). We double-checked our analysis throughout the manuscript and made the necessary corrections in the manuscript. The revised text (Page 8, Line 192) reads as follows "Foliar THg concentrations were related to time/leaf age ($F_{(3,36)} = 108.86$, $p < 0.001$) and plant species ($F_{(2,12)} = 51.85$, $p < 0.001$) (Fig. 1)".*

Line 208-209: In references, the Hg concentration in foliage showed the linear increasing, inconsistent with the decreased uptake rate as description of the sentence.

Reply:*Thank you for this comment. The rate of leaf Hg uptake means the flux rate of gaseous Hg to plant, which is different from foliar Hg concentration. Despite the decrease in foliar Hg uptake rate, the leaf still continuously uptake Hg from the atmosphere and the majority of Hg was incorporated into leaves, thus, the Hg concentration in foliage still showed a linear increase.*

Line 218: The larger leaf also caused the bigger biomass, offsets the stomates effect. How to explain it?

Reply: *We agree with the reviewer's point that "bigger leaf not only has more stomatal openings and more surface but also has more biomass.". The more biomass may offset effects of stomates on atmospheric Hg accumulation by leaves to a certain degree. Leaf biomass may do not proportionally increase with leaf area and stomates, leading to a higher absolute Hg concentration in tussock sedge leaves than in few-seeded sedge/wire sedge leaves.The clarified text (Page 10, Line227-232) reads as follows "A larger leaf has a higher density of stomata and thus more leaf accumulation of atmospheric Hg (Laacouri et al., 2013; Millhollen et al., 2006; Stamenkovic and Gustin, 2009). A larger leaf area may also provide more adsorption sites for non-stomatal Hg uptake. Increased biomass corresponding with a bigger leaf area can offset the effects of stomate number on atmospheric Hg accumulation by leaves to a certain degree. A plausible explanation is that leaf biomass does not proportionally increase with leaf area and stomata, leading to a higher absolute Hg concentration in tussock sedge leaves than in few-seeded segde/wire sedge leaves.".*

Line 229-231: It is insipid. Further explain is needed to clarify the reason why the Hg concentration in peatland vegetation is lower than that in tree litter.

Reply:*We agree with this suggestion. We have incorporated additional content in this section. The revised text (Page 10-11, Line 236-252)reads as follows "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 g-1) were lower than that reported in tree litter (21 ng g^{-1}–78 ng g^{-1}) in North-America and Europe (Laacouri et al., 2013; Obrist et al., 2021; Poissant et al., 2008; Rea et al., 2002; Wang et al.,* *2016) but similar to that previously reported for grasses and herbaceous plants (~10 ng g^{-1}) (Moore et al., 1995; Olson et al., 2019). The foliar Hg concentrations for plant species in this study increased 1.3-2.0 times over the growing season, which was smaller than that (3-11 fold) reported for trees (Laacouri et al., 2013; Poissant et al., 2008; Rea et al., 2002). The above results further confirm that foliar Hg concentrations differ among vegetation types (Demers et al., 2007; Moore et al., 1995; Obrist et al., 2012; Richardson and Friedland, 2015). It has been suggested that Hg previously retained in leaves can be photo reduced to Hg0 that is re-emitted to the atmosphere, and consistent Hg0 re-emission from the foliage is positively related to photosynthetically active radiation (PAR) (Yuan et al., 2019). The plants in open boreal peatlands lacking a tree overstorey like that in this study would receive very high exposure to ultraviolet (UV), which may result in a greater photoreduction of Hg previously retained in leaves and then Hg loss than tree leaves that are more often shaded. Moreover, despite angiosperms having higher stomatal conductance due to fewer stomata but more numbers (de Boer et al., 2016; Jordan et al., 2015), stomatal opening in dark-adapted leaves after light exposure was generally faster in gymnosperms than in angiosperms but stomatal closing upon the darkness of light-adapted leaves was faster in angiosperms than in gymnosperms (Xiong et al., 2018). This phenomenon may lead to a higher Hg concentration in trees (a type of gymnosperms) than in sedges and sweet gales (two types of angiosperms). More studies are needed to elucidate this mechanism of foliar Hg accumulation by different plant types.".*

Line 334-342: This discussion is not enough. What does the SUA represent? I hope not only the amount of aromaticity. You should explain more about each factor. For example, "indicating that leached DOM from tussock sedge and few-seeded sedge/wire sedge leaves had higher aromaticity and less bioaccessible than that from the sweet gale leaves", the higher aromaticity and less biaccessible, so what? This discussion is meaningless.

Reply: *We agree with this suggestion. The explanation of $SUVA_{254}$,FI and BIX was given in page 7-8. We have incorporated additional content in this section. The revised text (Page 17-19, Line 337-374)reads as follows "Characteristics of DOM also varied among plant species ($SUVA_{254}$: $F_{(2,42)} = 24.02$, $p < 0.001$; FI: $F_{(2,42)} = 11.24$, $p < 0.001$; BIX: $F_{(2,42)} = 125.48$, $p < 0.001$) (Fig. 6 and Table 2). Based on post hoc tests, there were significant differences in SUVA254 and FI between sweet gale and sedges (few-seeded sedge/wire sedge) only and BIX among all plant species. FI (1.2-1.8) and BIX (<1.0) reflected that DOM in leachate was generally of plant origin, suggesting that the microbially-derived OM was a smaller component. The mean value of $SUVA_{254}$ in leachate followed the sequence: tussock sedge ≈ few-seeded sedge/wire sedge > sweet gale leaves, respectively, indicating that leached DOM from tussock sedge and few-seeded sedge/wire sedge leaves had higher aromaticity and higher molecular weights than that from the sweet gale leaves. $SUVA_{254}$ was negatively related to DOM concentrations ($F_{(1,43)} = 48.37$, $p < 0.001$, $y = –0.69x + 3.93$, $R^2 = 0.53$) when all plant species were considered, suggesting that sweet gale prefers to release more amount of lower aromatic DOM.*

Previous studies have found that characteristics of DOM controlled Hg mobility and methylation (Cui et al., 2022; Jiang et al., 2018; Ravichandran 2004; Xin et al., 2022; Wang et al., 2022). Hg *is tightly and readily bound to reduced sulfur groups (i.e., thiols) in DOM (Ravichandran, 2004; Xia et al., 1999). Mercury weakly binds to carboxyl and phenol functional groups in DOM after all thiol groups are occupied at relatively high Hg concentrations (Drexel et al., 2002; Graham et al., 2012), which is atypical in most natural environments in which Hg concentrations are relatively low. Higher terrestrial (plant-derived) DOM had a greater DOM-Hg affinity (Wang et al., 2022). Additionally, DOM with higher aromaticity and molecular weight strongly bonded with Hg2+, potentially because these DOM provide more sulfidic groups such as thiols (Dittman et al., 2009; Wang et al., 2022). Therefore, terrestrial DOM and/or DOM with higher aromaticity and molecular weight may transport more Hg into peat soils during the litter leaching phase." and "The concentrations of soluble THgaq were significantly related to* $SUVA_{254}$ *values* ($F_{(1,41)}$ = 52.06, $p < 0.001$, $y = 0.09x – 0.09$, $R^2 = 0.55$; *Fig. 7). This result suggested that DOM with higher aromaticity plays an important role in controlling Hg mobility (Ravichandran, 2004). The value of* R^2 *was only 0.55, which can be attributed that the number of reduced sulfur groups in DOM far exceeds the amount of Hg in natural environments and other factors, such as pH and sulfide may affect the binding between DOM and Hg (Ravichandran, 2004). In this study, DOM with higher aromaticity may transport more Hg from litter to soils, and senesced leaves of sedges had a higher potential in leaching Hg into peatland soils than the senesced leaves of sweet gales in this study.".*

Line 342-343: I can find the evidence to support that stimulate biological degradation and Hg methylation in this study.

Reply: *We agree with this suggestion. There are many papers about the bioaccessible DOM (e.g., organic acids, sugars, amino acids) can stimulate biological degradation (Ganjegunte et al., 2006; Zhang et al., 2019) and Hg methylation (Mitchell et al., 2008; Schaefer et al., 2011; Leclerc et al., 2015). (See the below references). However, we removed this information from the manuscript to make the section focusing on Hg cycling in boreal peatlands.*

Ganjegunte, G. K., Condron, L. M., Clinton, P. W., Davis, M. R., Mahieu, N.: Effects of the addition of forest floor extracts on soil carbon dioxide efflux.Biology and Fertility of Soils, 43, 199-207, DOI10.1007/s00374-006-0093-6, 2006.

Zhang, Z. Y., Wang, W. F., Qi, J. X., Zhang, H. Y., Tao, F., Zhang, R. D.: Priming effect of soil organic matter decomposition with addition of different carbon substrates. Journal of Soils and Sediments, 19, 1171-1178. DOI10.1007/s11368-018-2103-3. 2019

Line 381-382: In this MS, I cannot find any MeHg data. But the authors always highlighted the MeHg production. This is confused.

Reply: *We agree with this suggestion. The clarified text (Page 21, Line) reads as follows "The THg concentrations in senesced leaves in this study are relatively lower than that in the forest litterfall".*

Line 383: This conclusion is not available in this MS.

Reply: *We agree with this suggestion. We have removed this sentence from the manuscript.*