Review of Manuscript preprint by Wang et al. 2023 Reviewer:
Lena Wohlgemuth

The authors present interesting findings on mercury cycling in a salt marsh estuary based on Hg flux measurements and Hg isotope fingerprinting. Studying Hg dynamics in coastal ecosystems is necessary in order to better understand Hg export to coastal oceans. The study is comprehensive and well written. I support its publication with minor comments detailed below.

**Line 25 – 34: The abstract could summarize increases in plant Hg during the growing season and the Hg mass balance in a more consolidated way. Just highlight the most relevant findings for the study. lin**

**ANSWER:** We shortened this section and removed lines 25-29 to highlight the relevant finding.

**Line 28/29: It should be made clear in the abstract, which values were not measured in this study, but taken from other studies.**

**ANSWER:** We deleted the literature values in lines 28 and 29 based on the above question.

**Line 53: …accounting for 60% to 90% of total Hg inputs to soils**

**ANSWER:** Revised. (line 49 in current version)

**Line 53 – 59: Make the link to Hg. An uninformed reader might wonder why you go into detail of plant carbon assimilation in the introduction of a Hg paper.**

**ANSWER**: Thank you for pointing this out. We added a sentence to bridge the discussion between carbon and Hg assimilation in lines 55 to 56.

**Line 63: associated; ii) strictly speaking you did not quantify the transfer of Hg from aboveground biomass to soils. I think, i) comprehensively describes your objective and that you can delete ii).**

**ANSWER**: We shortened this sentence as suggested.

**Line 71: were**

**ANSWER:** Revised. (line 67 in current version)

**Line 78/79: Please check the species names again. Are they not called Sporobolus pumilus and Spartina alterniflora?**

**ANSWER**: We changed the species names accordingly.

**Line 84: Check the title**

**ANSWER**: Thank you, we changed the title to "**Sampling and Processing of Vegetation and Soil**". (line 87 in revised version)

**Section 2.2: For readers unfamiliar with this ecosystem, it is not obvious, that sampling salt marsh vegetation means sampling of a lot of senescent plants, i.e. that senescent plant material makes up a relevant proportion of total plant biomass throughout the growing season and not only in April of the following year. I think, this should be explained at some point in the paper, e.g. in Section 2.2 to avoid confusion.**

**ANSWER**: We clarified this information in lines 95 to 97.

**Line 140: For the regression slope I assume that variability is presented by the standard error. Also, you could mention how you propagated standard deviations.**

**ANSWER**: Thanks for the suggestion. I have revised the sentence in the manuscript in lines 153 to 154.

**Line 145: …Hg concentrations (Fig. 1a)**

**Fig. 1: Where is data on Juncus gerardii? I suggest to use different colors for species and species communities.**

**ANSWER**: Thank you for pointing out the issue. I have revised the graph Figure 1a.

**Line 158 – 161: Can be moved to M&M**

**ANSWER**: We moved this sentence and simplified the description to M&M in lines 103 to 105.

**Line 165: Are numbers in brackets derived from pooled roots, rhizomes, and detritus? This should be indicated here.**

**ANSWER**: In response to the comment below, we shortened this section and removed some of these numbers to avoid confusion (detailed numbers can be seen in Table S1 and in figures).

**Line 161 – 173: I wonder if this section will improve by shortening it and highlighting only the most relevant concentration differences, that will be discussed later. All other values are already displayed in Fig. 2 or tabulated in the SI. This is a suggestion to make the concentration results section more engaging to the reader. You decide.**

**ANSWER**: Thank you for your suggestion. We shortened this section to focus on more relevant information and refer to Figure 2 and Table S1.

**Line 183: …was 39% lower than total (live and senescent) biomass…**

**ANSWER**: corrected.

**Fig. 2: The date when Hg concentrations = 0 was not determined, so don't indicate it by extending the regression line. Or did you do phenological observations to determine the beginning of the growing season?**

**ANSWER**: We clarified in the figure legend that April data was extrapolated and set to zero based on phenological observations that showed no presence of live biomass.

**Fig. 4b: Why are there two bars for April 2022? Please check.**

**ANSWER**: We clarified in the figure legend that since there was no live biomass present in April 2022, senesced biomass equals total biomass at this point.

**Fig. 4c: Why are there different standard errors for April 2022?**

**ANSWER**: This was an error, we corrected this.

**Line 195: To be more precise and align the text to Fig. 6 please define "aboveground biomass" in this section once, e.g. "aboveground biomass (live and senescent marsh leaves)".**

**ANSWER**: We clarified that aboveground biomass means live and senesced marsh leaves. We also clarified that in this section we refer to live aboveground biomass only.

**Line 215: For citing seasonality of Hg in forest foliage, please replace Wohlgemuth et al. 2022 with Wohlgemuth et al. 2020 (https://bg.copernicus.org/articles/17/6441/2020/).**

**ANSWER**: Replaced. (line 221 of current version)

**Line 291: I redid the calculation in R (see code below) and the rounded model output was: f_atm = 0.33, f_root = 0.31, f_prep = 0.37. Almost the same values, but it might be worth checking the calculations again. Also, in the SI section on Hg isotope mixing model, please explicitly give the used endmember median values.**

**ANSWER**: Thank you for double checking. We reran the calculations with two Monte Carlo simulations and provide the best estimates as well as uncertainty ranges for these contributions. We further clarify the used endmember values in the calculation by referring to the supplementary document section "Hg Isotope Mixing Model" (Line 17-35). Additional text and a short section in SI has been provided to the Monte Carlo simulations to assess uncertainties.

**Line 290: I think a short explanation would be helpful for readers of how direct Hg uptake (translocation) from plant roots differs from precipitation Hg(II) deposition in this context, since precipitation water is also taken up by plant roots, which might confuse readers.**

**ANSWER**: We clarified that uptake of precipitation Hg includes root uptake of rainwater and possibly deposition to leaves, which we cannot distinguish in lines 63 to 64.

**Line 371: I don't quite understand, how you estimated annual throughfall deposition. Did you derive it from Hg biomass at the end of the year (9.0 µg $m^{-2}$ $yr^{-1}$)? Theoretically, wash-off Hg could be a relatively constant value at every precipitation event over the growing season in a way, that wash-off Hg is independent from stomatal GEM or Hg root uptake. In fact, values presented in SI Table 5 do not support a clear increase of wash-off Hg over the growing season. Throughfall Hg is hard to quantify and maybe I misunderstand how you calculated 1.0 µg $m^{-2}$ $yr^{-1}$, but I think this merits an explanation in M&M.**

**ANSWER**: Thanks for pointing out your confusion. We provided additional clarification in the main manuscript between lines 254 and 255. We clarified that in the absence of direct field throughfall measurements (which would be very challenging in a marsh system), the estimate is based on laboratory washing which showed on average, 11% of aboveground Hg can be washed off. We further state the limitation of this estimate and that is a very initial estimate in lines 352 to 353.

**Line 379: Give area in brackets**

**ANSWER**: The total salt marsh area of 40 $km^2$ is mentioned in section 2.1 Site Description in line 69 and Table 1 in line 761. To prevent any potential confusion, we've also included it in line 363 here.

**Line 392: Please mention the most relevant herbivores in this context (here or in Sect. 2.1). Readers are probably unfamiliar with the fauna of this ecosystem and this would help to understand why herbivory Hg is part of internal Hg cycling.**

**ANSWER**: We gave some examples of herbivores present in these ecosystems, including, marsh periwinkle and mummichog.

**Fig. 6/Fig. S3: To me, the legends could be more intuitive. Is it possible to move the two legends from inside the plot panels to the right side, such that they apply to both Fig. 6a and 6b at first**

**glance? Please note, that the isotope symbols representing organic soils and roots are almost identical by shape and color, same applies to rainfall and deep soil.**

**ANSWER**: Thanks for the suggestion. We changed the position of the legend as suggested.

**Fig. 7: This figure gives a good overview, however, I think you could improve it by clearly labelling, which arrows illustrate the fluxes used for the mass balance of this study and which arrows symbolize any possible Hg flux of the ecosystem. From my understanding, dashed deposition arrows represent Hg fluxes to aboveground biomass, non-dashed arrows represent Hg fluxes to the ground/soil pool (please define dashed/non-dashed in the caption). Therefore, I would extend the non-dashed deposition arrows of wet Hg(II), dry Hg(II), and GEM to the ground. For the deposition part of the mass balance you added up fluxes of wet and dry Hg(II) (both measured independent of aboveground biomass), throughfall Hg, and net GEM deposition determined from Hg accumulated in biomass and multiplied by the GEM percent contribution from the isotope mixing model (litterfall). So even though precipitation Hg(II) (dashed arrow) is taken up by aboveground tissues via the roots (belowground dashed arrow would be more accurate), it is not part of the 4.1 µg $m^{-2}$ $yr^{-1}$ used for the mass balance, same is true for 2.1 µg $m^{-2}$ $yr^{-1}$ dry Hg(II). For GEM, you did not determine direct GEM deposition/re-emission to/from the ground, so what do dashed/non-dashed GEM arrows mean in this context?**
**Why is there a downward dashed flux arrow labelled "Aboveground biomass Hg uptake from soil 3.1", is this a mistake?**

**ANSWER**: That you for your suggestions. We clarified that dashed arrows represent Hg fluxes related to the aboveground biomass dynamics and clarified this in the text. We changed the solid arrow for aboveground tissue deposition (including green, senesced, and throughfall) to a dashed line, and renamed this to "Aboveground tissue Hg uptake"). We also shifted the numbers for wet and deposition (4.1 and 2.1 µg $m^{-2}$ $yr^{-1}$) next to the solid lines to show these are non-vegetation depositions. Finally, we put a question mark next to the direct soil GEM emission/deposition since this was not measured here.

We intentionally didn't extend the non-dashed deposition arrows for wet Hg(II), dry Hg(II), and GEM via vegetation to the ground, because we don't need to partition respective deposition sources in this figure plus it would lead to difficult interpretations (as discussed by the reviewer, e.g., that we don't know the fraction of precipitation Hg take up directly on leaves versus transported from soil water). Finally, we removed the downward arrow of aboveground biomass Hg uptake, the reviewer is correct that this deposition is already represented in the aboveground tissue Hg uptake/turnover numbers and included in the total plant-derived deposition.

**Table 1: Please make it clearer (e.g. with an asterisk), that Hg flux values (e.g. green/senescent biomass deposition of 3.7/2.1 µg $m^{-2}$ $yr^{-1}$) represent calculated percentages of measured values. I think, that this is not intuitive from the percent of Hg sources given in the next column for a reader, who only looks at this table without reading the text. The asterisk indicating the vegetated salt marsh area should only apply to the last column.**

**ANSWER**: Thanks for your suggestion. We have removed the asterisks in the "Item" column. In the "Percent of Hg sources" column of Table 1, we have labeled their calculated percentages. Furthermore, the relevant information has been provided in the manuscript, specifically between lines 349 and 353.

**Line 434: herbivory internal cycling(?)**

**ANSWER**: Yes, the herbivory belongs to internal Hg cycling. In order to avoid confusion, I revised the sentence between lines 420 and 421.

**Line 433: Avoid repetitions of the abstract**

**ANSWER**: Thanks for your suggestion. We have revised the sentence between lines 418 and 421.

**Table S5: Please check the average value of estimated throughfall of S. alterniflora, it seems wrong. It is possible, that washed biomass samples (S. alterniflora in Oct-21 and Nov-21) are higher than respective unwashed samples due to measurement uncertainties and low Hg concentrations in washoff, though don't give negative concentration values, but leave them out.**

**ANSWER**: Thank you for bringing up this issue. We removed the "Estimated throughfall" values for individual months and now only provide an average value through the growing season to address this concern.

**Section Summary and conclusion: You give all relevant fluxes, sources, and pools of the study, which is good. I wonder, if you could go a step further and bring this study in line with other studies on Hg input to coastal oceans, e.g. how this sink compares to other coastal sinks or input fluxes. Can you derive any implications from your findings for the ecosystem, e.g. in the introduction you mention, that the salt marsh is a Hg hotspot?**

**ANSWER**: Thank you for your suggestion. We have added more discussion accordingly in lines 401 to 407.

R code for checking Hg isotope mixing model

```
library(matlib)

# Isotope compositions:
d202Hg_GEM <- -2.84
D200Hg_GEM <- -0.02
D199Hg_GEM <- -0.37
d202Hg_root <- -0.69
D200Hg_root <- 0.03 D199Hg_root
<- 0.17 d202Hg_prep <- -0.3
D200Hg_prep <- 0.17
D199Hg_prep <- 0.4
D200Hg_veg <- median(c(0.11, 0.06, 0.07, 0.04)) d202Hg_veg
<- median(c(-1.07, -1.61, -1.21, -1.29))
D199Hg_veg <- median(c(0.20, 0.43, 0.42, 0.32))

# ternary isotope mixing model
A <- matrix(c(D200Hg_GEM, d202Hg_GEM, 1, D200Hg_root,
d202Hg_root, 1, D200Hg_prep, d202Hg_prep, 1), 3, 3) b <-
c(D200Hg_veg, d202Hg_veg, 1) showEqn(A, b)

Solve(A, b)
```