

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.

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

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

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.

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).

Line 71: were

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

Line 84: Check the title

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.

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.

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.

Line 158 – 161: Can be moved to M&M

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

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.

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

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?

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

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

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)".

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/>).

Line 291: I redid the calculation in R (see code below) and the rounded model output was:  $f_{\text{atm}} = 0.33$ ,  $f_{\text{root}} = 0.31$ ,  $f_{\text{prep}} = 0.37$ . 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.

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.

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 ( $9.0 \mu\text{g m}^{-2} \text{yr}^{-1}$ )? 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  $1.0 \mu\text{g m}^{-2} \text{yr}^{-1}$ , but I think this merits an explanation in M&M.

Line 379: Give area in brackets

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.

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.

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  $4.1 \mu\text{g m}^{-2} \text{yr}^{-1}$  used for the mass balance, same is true for  $2.1 \mu\text{g m}^{-2} \text{yr}^{-1}$  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?

Table 1: Please make it clearer (e.g. with an asterisk), that Hg flux values (e.g. green/senescent biomass deposition of  $3.7/2.1 \mu\text{g m}^{-2} \text{yr}^{-1}$ ) 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.

Line 434: herbivory internal cycling(?)

Line 433: Avoid repetitions of the abstract

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 wash-off, though don't give negative concentration values, but leave them out.

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?

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)
```