

Review of “Above- and Belowground Plant Mercury Dynamics in a Salt Marsh Estuary in Massachusetts, USA” by Wang et al. The authors carried out a systematic investigation on vegetation and soil in a salt marsh. It is important work to improve the understanding of Hg cycling in salt marsh area. However, the Hg isotopic mixing model exists critical mistakes. Firstly, some endmember isotopic signatures are lacking. The authors cited the global mean results, but I think this is not feasible. Citing results nearby the field site seems to be more convincing. Secondly, present model was built based on MDF and even-MIF. Plant uptake Hg from atmosphere or soil could cause significant negative MDF, so present model results are speculative. Thirdly, the authors cannot offer QA/QC about Hg isotopic measurements, and the error bars by measurement are lacking. In addition, the uncertainties of model estimations are not fully calculated.

ANSWER: There was a crucial misunderstanding of the reviewer in regard to the endmember mixing model, and this in part was due to a lack of clarification in our description. We DID EMPLOY both MDF and MIF fractionation in our calculations, and we DID EMPLOY the negative MDF induced by plant uptake from the atmosphere in the model. However, we failed to clarify that the negative MDF from plant uptake was used as an endmember by using the MDF signature from terrestrial foliage (with its strongly negative MDF values). Instead, the previous version suggested that directly used the atmospheric GEM signature (with positive MDF values). The clarification should resolve the concerns of the reviewer that model results are speculative. Finally, we added additional QA/QC data and an uncertainty analysis of the tertiary mixing model to the manuscript as described below.

In detail:

Introduction: I suggest the authors should add one paragraph about Hg isotope, which was used to quantify the source of plant.

ANSWER: Thank you for the suggestion. I have added a short section on the possible sources of Hg (and its isotope endmembers) between lines 62 and 64. More detailed information regarding the dominant end-member Hg sources are described in supplement document in section titled “Hg Isotope Mixing Model” (between lines 17 and 26).

Line 20-21: Here peak aboveground Hg pool is 9.0 ug m⁻², which is below that in November with 16.2 ug m⁻². What differences?

ANSWER: We clarified this, but we think that the reviewer confused the units (concentration: $\mu\text{g kg}^{-1}$ versus Hg pool: $\mu\text{g m}^{-2}$). The standing aboveground Hg pool is $9.0 \pm 3.3 \mu\text{g m}^{-2}$ and represents the total Hg mass of both live and senesced aboveground biomass in November. The value $16.2 \mu\text{g kg}^{-1}$ (not $\mu\text{g m}^{-2}$) is the Hg concentration of live aboveground biomass in November.

Line 39: largest. How much is the global input flux?

ANSWER: We added this information in lines 37 and 38 with respective citations.

Line 43-45: How about the anthropogenic pollution at present salt marshes?

ANSWER: We cannot say but revised the sentences here in lines 80 and 83. We also added a statement in the soils section that states that the small sink measured in this study is not consistent with

the large pool of Hg measured in soils in this marsh in lines 401 to 407. We discuss the potential for legacy/historical contamination from the industrial period in the region.

Line 48-53: Cite more research papers, not only review.

ANSWER: We added specific research papers here, including lines 45 and 46 (Fisher and Wolfe, 2012; Iverfeldt, 1991; Rea et al., 1996; Zhou et al., 2021) and lines 47 and 48 (Fu et al., 2019; Jiskra et al., 2018; Obrist et al., 2018; Wang et al., 2019, 2022; Zhou et al., 2021; Zhou and Obrist, 2021).

Line 53: It is overestimated.

ANSWER: We refer to the range reported by several studies, we are not quite sure why this is considered overestimated.

Line 53-59: I don't think it is related to the Hg cycling.

ANSWER: We added a sentence to clarify the linkage between NPP and Hg cycling in lines 55 and 56.

Line 102-103: Were the soil samples ground by coffee grinders? I think such operation could cause cross contamination. How much mesh were sieved with? The soil homogeneity is very important.

ANSWER: We clarified that we used a soil shatter box (8530 Shatter-Box) to mill and homogenize soils samples with particle sizes less than 2mm. Coffee grinders were used to grind and homogenize plant samples only. Both the coffee grinders and the Shatter-Box were rinsed with Milli-Q water and dried with Kimwipes between samples. We clarified these methods in lines 107 to 110.

Line 117-121: How about the Hg concentration in solution to isotopic measurement?

ANSWER: We added the requested information in lines 128 and 129.

Line 127-130: Report the isotopic results of standard samples, i.e. NIST 1515 and MESS-4. I also cannot find those values in SI. QA/QC is important for Hg isotope measurement.

ANSWER: We have added the quality control results for standard samples in Table S4.

Line 231-234: It is a good discussion about foliage accumulation by C3 and C4 plants. But it is not enough. I hope to get more discussion about the mechanism.

ANSWER: We cannot go into details about mechanisms here as they are not well established and is beyond the scope of this paper. For example, the main study we know on this subject is from a 1983 study. We summarized the results of this study here (lines 239 and 240).

Line 235-243: Such comparison in Hg concentration in different sites is tedious as too many factors controlled the Hg concentration.

ANSWER: We agree, but we think it is important to refer to findings of other marsh plants studies here. We shortened the section, however, to make it less tedious.

Line 259: How to identify the finer roots? <1mm or <2mm? Need more data to support it.

ANSWER: We did not measure the exact diameter of *S. patens* (*S. pumilus*) roots. Instead, we labeled them 'finer roots' in comparison to *S. alterniflora* (*S. alterniflorus*), based on our personal observations. We clarified this in brackets (at line 261 of the current paper version).

Line 277: Recent study has demonstrated the inconsistent MIF between foliage and atmosphere.

ANSWER: Many previous studies have indicated similar signatures of MIF between foliage and atmosphere. However, we do now mention that one recent study reported inconsistent MIF between foliage and the atmosphere in lines 279.

Line 281-282: The authors should compare the isotopic data with the reported grass isotopic values, rather than all the plant values. As you said, the Hg assimilation may exist great difference between C3 and C4 plant.

ANSWER: Ideally, this should be done, but to our knowledge there simply are not sufficient studies on Hg isotopes in grasses and differences between C3 and C4 plants. Hence, our best approach is to use the wider data sets from upland plants.

Line 283-285: It is speculatively. I do not agree with it.

ANSWER: We have deleted this sentence .

Line 288: How to build the model based on MDF? As we know, the plant uptake Hg could induce significant MDF. I do not believe this model results if it is based on MDF.

ANSWER: As written in our response to the first statement, there is a confusion and we indeed used the significant MDF that occur by plant uptake of atmospheric GEM. The end-member in the ternary isotope mixing model we used to calculate MDF is based on Hg isotopic signatures ***of upland foliage with negative MDF values, not atmospheric GEM as we originally wrote***. We have clarified this in the manuscript in lines 289 and 290.

Detailed information about the mixing model can be found in the supplementary document (between lines 17 and 26). In summary, the three dominant end-member Hg sources (medians) include local marsh plants roots, terrestrial upland foliage (used to represent atmospheric GEM), and precipitation. The three calculation equations are also provided in the supplementary document. Hopefully, this information proves helpful.

Line 291-292: How about the uncertainty of this estimation?

ANSWER: We now provide a detailed assessment of the uncertainties of our tertiary mixing model. Due to the low sample numbers and reported analytical uncertainty, the uncertainties are

substantial. However, two different methods to assess the uncertainties both show similar mean values of contributions. We clarified the uncertainty analysis in the section “Hg Isotope Mixing Model” in SI between lines 30 and 35.

Line 291-303: I do not believe the model results. Briefly, all the MDF values are more positive than that in measured vegetation results in Figure 6 a). I cannot think this mixing model can be solved in present pattern. Further, I suggest to add error bars (2SD) in Figure 6.

ANSWER: We added error bars to Figure 6, but their impact may not be immediately apparent since we only have two samples analyzed in duplicate with error bars. We refer to our response above about the MDF, this clarification should address the concerns of the reviewer.

Line 304-312: This paragraph is speculatively. Not all the slope of ~1 suggested the inorganic Hg photoreduction.

ANSWER: We deleted this paragraph to avoid confusion and removed the discussion points.

Line 414-422: Turnover flux is an interesting discussion. But I think such flux is overestimated.

ANSWER: We are not sure why the reviewer thinks it is an overestimate, we did our best to estimate the potential magnitude of these different component fluxes based on detailed turnover estimation of marsh ecosystem studies. As discussed, please note that the large turnover flux is largely attributed to the substantial Hg mass present in plant roots and marsh soils and as such constitutes an internal system recycling of Hg between plant-bound and soil Hg.

Line 745-749: Please cite the original research papers, not the review paper.

ANSWER: For this figure, we used and overlaid data from a figure from this review paper so citing this study here seems appropriate.

Line 752: Dry HgII deposition flux with 2.1. How to estimate it? Add the references.

ANSWER: The discussion regarding the dry HgII deposition and relevant references can be found in Section ‘4.3.1 Aboveground’ (line 346-348). Additionally, the estimated dry HgII deposition referenced is also provided in Table 1. (Engle et al., 2010, Obrist et al., 2021)