

General answers to the manuscript entitled: Divergent mercury sequestration dynamics in tropical dry and moist broadleaf forests

We thank the reviewers for their valuable comments, the time they dedicated to evaluating our manuscript, and their constructive suggestions, which will help us substantially improve the manuscript. Overall, the main comments concerned the use of terminology that more accurately reflects the measurements performed in this study and the need to distinguish more clearly between measured variables and inferred processes. Accordingly, terms such as “soil Hg stocks” and “litter-derived Hg inputs to soil,” which were not directly quantified with our sampling design, will be revised throughout the manuscript. We will also moderate mechanistic and causal interpretations where they cannot be fully supported by our data, and explicitly acknowledge the limitations of the study where appropriate. Our point-by-point responses are provided below.

Reviewer 2

Reviewer Comments

This study examines seasonal changes in atmospheric, foliar, forest-floor litter, and soil Hg in tropical dry and moist broadleaf forests in Costa Rica. Seasonal foliar Hg data from tropical dry forests remain limited, and the observed differences between deciduous and evergreen trees across seasons provide valuable field evidence for understanding how leaf phenology may influence Hg accumulation in tropical forests. However, the study directly measured Hg concentrations in leaves, litter, and soils, together with the standing Hg pools of forest-floor litter. These measurements represent Hg concentrations and a static litter Hg pool at the time of sampling, rather than Hg uptake rates, litterfall Hg fluxes, ecosystem Hg inputs, or net sequestration. The manuscript therefore overinterprets these metrics when describing forest Hg uptake, storage, and sequestration capacity. The authors should clearly distinguish concentrations and standing pools from fluxes and net ecosystem sequestration, and revise the mechanistic and global-scale conclusions accordingly.

Major comments

1. Lines 165–170: The authors collected forest-floor litter once using approximately 400 cm² quadrats and calculated litter mass and Hg burden per unit area. This metric represents the amount of litter present on the forest floor at the time of sampling, i.e., the forest-floor litter standing stock, rather than a litter Hg input flux over a defined period. Therefore, the terms “litter Hg input,” “litter Hg flux,” “litter-derived Hg deposition,” and “Hg transfer to soil” are not appropriate. These metrics should instead be described as “forest-floor litter mass” or “standing litter Hg burden,” and they should not be directly compared with annual Hg input fluxes measured using litterfall collectors.

Our response: *Thank you for your valuable comment. We agree, therefore, we will replace terms such as “litter Hg input,” “litter Hg flux,” “litter-derived Hg deposition,” with litter as a “Hg standing stock” throughout the manuscript. We will also clarify that our measurements should not be directly compared with annual Hg input fluxes obtained from litterfall collectors, as our sampling design was based on forest-floor litter found at a certain time rather than trap-based litter measurements.*

2. Section 2.2: The study mainly measured Hg concentrations in leaves, litter, and soils, together with the standing mass and Hg burden of forest-floor litter. These metrics do not directly represent ecosystem-scale Hg uptake rates, input fluxes, storage, or net sequestration. However, the terms “Hg sequestration,” “uptake,” “storage,” and “global Hg sink” are repeatedly used in the title, abstract, and conclusions, which may give readers the impression that these processes were directly quantified. The authors should clearly distinguish among Hg concentration, standing burden, flux, stock, and net sequestration, and restrict their interpretations to what is directly supported by the data.

Our response: *We agree, therefore, we will carefully review the terminology to avoid using specific terms that were not measured throughout the manuscript. Where such terms are used, they will be moderated where appropriate, and our interpretations will be restricted to what is directly supported by the measured data, clearly indicating the limitations of our study.*

3. Lines 170–175: The authors measured soil Hg concentrations at different depths in units of $\mu\text{g kg}^{-1}$, but did not report soil bulk density, coarse-fragment content, or other parameters required to calculate areal soil Hg stocks. Therefore, statements such as “soil Hg stocks were 2.6 times lower” in the abstract and conclusions are not supported by the available data. Unless the necessary parameters can be provided and soil Hg stocks recalculated, “soil Hg stocks” should be replaced throughout with “soil Hg concentrations.”

Our response: *Thank you for your valuable comment. We agree that soil Hg stocks were not directly quantified because the parameters required for stock calculations were not measured. Therefore, we will replace “soil Hg stocks” with “soil Hg concentrations” throughout the manuscript where appropriate, as requested.*

4. In the TDBF, leaves collected during the dry season were mainly newly flushed after rainfall, whereas those collected during the wet season were mature leaves. The higher Hg concentrations in wet-season mature leaves may therefore primarily reflect greater leaf age and longer exposure to atmospheric Hg, rather than higher water availability, greater stomatal opening, or stronger transpiration during the wet season. Because leaf age, season, and water availability are confounded in the current sampling design, the present results do not demonstrate that water availability is the main driver of foliar Hg variation.

Our response: *We agree that leaf age, season, and water availability are partly confounded in our sampling design and therefore cannot be fully disentangled. However, in the TDBF, deciduous trees shed their leaves during the dry season and leaf flushing generally occurs following the onset of rainfall, meaning that water availability is closely linked to leaf phenology, as explained in the manuscript. Moreover, as also stated in the manuscript, Borchert (1994) described evergreen trees in the same area of our TDBF study as facultative evergreens, with leaf phenology primarily driven by water status. In this regard, rainfall promotes leaf growth, while drought causes leaves to fall. Without leaves, there is no foliar Hg sequestration. To further support this hypothesis, in situ ecohydrological research carried out at our exact study site (Estación Experimental Forestal Horizontes, Costa Rica) by Kühnhammer et al. (2022) demonstrated that trees from TDBF exhibit a highly dynamic, individual-level coupling between water availability, foliage phenology, and tree physiological*

activity. These findings provide direct, site-specific empirical evidence that water availability fundamentally drives foliage dynamics and individual transpiration rates, which are the physiological mechanisms that control stomatal conductance and gas exchange (and thus foliar Hg sequestration). On the other hand, TMBF maintained similar values of foliar Hg in both dry and wet seasons. The rainforest, as explained in the Materials and Methods, is influenced by constant precipitation even during the short dry season. Therefore, we consider that water drives plant physiology and that this is linked to Hg sequestration dynamics. In this regard, water availability is a key driver of foliar Hg variation, especially in seasonal forests such as the TDBF.

5. Lines 640–650: The authors compare plots G1–G2 with G3–G6 in the TMBF and attribute the lower soil Hg concentrations in G1–G2 to historical grazing, cacao cultivation, and lower tree-species diversity. They further suggest that recovery of soil Hg stocks requires a long time. However, this comparison lacks an undisturbed primary-forest reference and pre-disturbance soil Hg data. In addition, the study measured soil Hg concentrations rather than Hg stocks. Therefore, the current evidence is insufficient to demonstrate that anthropogenic disturbance caused a 2.6-fold loss of soil Hg stocks or to determine the time required for recovery. The conclusion that maintaining species diversity enhances Hg sequestration also extends beyond what the data can support.

Our response: *We agree that, because the deforested/reforested site was not spatially replicated and plots G3–G6 are also secondary forests containing remnants of primary forest, these results should be interpreted as a case study rather than as definitive evidence of a deforestation effect. We will also clarify this limitation in the reviewed manuscript. In particular, there was not suitable undisturbed primary-forest close to our study area that could serve as a direct reference and pre-disturbance soil Hg are not available. Therefore, our study cannot quantify the magnitude of Hg loss attributable to past disturbance or determine the time required for Hg recovery. Nevertheless, the comparison remains relevant because we compared the deforested/reforested site with a secondary tropical forest that has been regenerating for more than 30 years. Although this forest cannot be considered a pristine primary-forest reference, it provides an independent, older secondary-forest contexts against which Hg concentrations and patterns can be evaluated.*

Regarding soil Hg stocks, because bulk density was not measured, we will replace the term “soil Hg stocks” with “soil Hg concentrations” where appropriate. We will also moderate the statement linking higher tree-species diversity with enhanced Hg sequestration, as this relationship cannot be demonstrated from our data.

6. Lines 680–685: Based on mature-leaf Hg concentrations at the two study sites being higher than the global average, the authors conclude that TDBF and TMBF are important global Hg sinks and recommend including TDBF in global Hg budgets. However, high foliar Hg concentrations do not necessarily indicate a strong ecosystem-scale Hg sink. Forest Hg sequestration also depends on foliar biomass, leaf age, leaf turnover, leaf area index, and Hg re-emission, among other factors. The current results are therefore insufficient to demonstrate that these forest types function as major global Hg sinks, and the relevant statements should be substantially weakened.

Our response: We agree that high foliar or soil Hg concentrations alone are insufficient to demonstrate ecosystem-scale Hg sink strength. However, previous studies cited in our manuscript, including modelling and flux-based approaches, have identified tropical forests as major global Hg sinks. Our results are consistent with this broader understanding, particularly given the relatively high Hg concentrations observed in foliage and topsoil as described in the manuscript. However, because we did not quantify key ecosystem-scale parameters such as foliar biomass, leaf turnover, leaf area index, Hg re-emission, or net Hg fluxes, our data cannot independently demonstrate the magnitude of Hg sequestration by these forests. We will therefore substantially moderate the corresponding statements and refer to TDBF and TMBF as important Hg sequestrators rather than as major global Hg sinks, as requested.

Minor comments

1. The Methods state that four soil cores were collected from each plot. With 12 plots, the total should therefore be 48 cores, yet the manuscript reports $n=24$ $n=24$ $n=24$ cores. In addition, 192 samples across four depth intervals correspond to 48 cores. Please verify and correct the number of soil cores and soil samples throughout the manuscript.

Our response: Thank you for your valuable comment. We confirm that 48 soil cores were collected in each forest type across the 12 plots (6 on each forest, and with 4 replicates per plot). As each core was divided into four depth intervals, this resulted in 192 soil samples. We will therefore revise the statement from “Four soil profile samples were collected per plot from both forest types ($n = 24$ cores)” to “Four soil cores were collected per plot in each forest type ($n = 48$), resulting in 192 soil samples across four depth intervals.” We will also verify and correct these numbers throughout the manuscript.

2. The Methods report a total of 391 leaf samples, whereas the sample sizes shown in Fig. 3 sum to 355. Please verify the sample numbers and explain the discrepancy.

Our response: Thank you for your careful reading. We will correct the number of samples. We apologise for the mistake.

3. The manuscript treats water availability and seasonality as key mechanisms explaining foliar Hg variation, but does not report actual precipitation, air temperature, relative humidity, or soil moisture during the three sampling campaigns or the two MerPAS deployment periods. Please provide site-specific measurements or data from nearby meteorological stations to better support these interpretations.

Our response: We will retrieve and compile available site-specific data on precipitation, air temperature, relative humidity, and, if available, soil moisture from nearby meteorological monitoring stations for the corresponding sampling campaigns and MerPAS deployment periods. These data will be included in the Supplementary Information to better support our interpretation of seasonal and water-availability effects.

4. Section 2.6 is numbered twice. The Statistical Analysis section should be renumbered as Section 2.7.

Our response: Thank you for your careful reading. It will be corrected; we apologise for the mistake.

5. The phrase “A significance level of significance” in line 214 is redundant and should be revised.

Our response: Thank you for your careful reading. It will be corrected; we apologise for the mistake.

6. “Specific leaf area index” in line 402 should be corrected to “specific leaf area.”

Our response: Thank you for your careful reading. The section “Foliar functional traits and their relationship with Hg accumulation in tropical forests” will be deleted, as we did not get strong correlations, but it will be shortly explained in the revised manuscript.

7. The unit “20–40 $\mu\text{g } \mu\text{g kg}^{-1}$ ” in line 418 contains a duplication and should be corrected.

Our response: Thank you for your careful reading. It will be corrected; we apologise for the mistake.

References:

Kühnhammer, K., Dahlmann, A., Iraheta, A., Gerchow, M., Birkel, C., Marshall, J. D., and Beyer, M.: Continuous in situ measurements of water stable isotopes in soils, tree trunk and root xylem: Field approval, Rapid Commun. Mass Spectrom., 36, e9232, <https://doi.org/10.1002/rcm.9232>, 2022.