



EGUsphere, referee comment RC2
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Comment on egusphere-2022-174

Anonymous Referee #4

Referee comment on "Seasonal variation of mercury concentration of ancient olive groves of Lebanon" by Nagham Tabaja et al., EGU Sphere,
<https://doi.org/10.5194/egusphere-2022-174-RC2>, 2022

This data set explores Hg uptake and cycling in olive groves – an critical agricultural tree across much of the Mediterranean basin. The experimental design seems valid to conduct an initial examination of Hg physiological cycling within olive trees and their surrounding soils. Additionally, the QA/QC of the data seems valid, which provides merit to at least the data.

Nonetheless, I cannot support publication of this manuscript in its current form for the reasons listed below. My recommendations would be to considerably scale back the scope of this manuscript and limit the discussion to the bounds of the data (extravagant speculations based on distant and unrelated data need to be removed), work on the grammar and English language of the paper, and consider the structural suggestions made below. At this point the paper may be worthy of resubmission and publication. Whether that publication is worth of publication in EGU-Sphere is a decision for the editor(s).

General comments:

- I feel like this paper would greatly benefit from combining the results and discussion. The current separated results and discussion lead to very dry reading that makes engagement somewhat difficult and there is clear repetition in the discussion section on what has already been written in the results. List then data and discuss it along in context of the story the manuscript is attempting to make. Additionally, there is repetition in the listing of the data between the text, figure 2 and table 2 (and the discussion). These data need only be listed one time.
- There is a lot of speculation and much of this speculation appears unsupported or poorly supported by the available data; the discussion oversteps the suggestions and conclusions that can be made with this dataset via the excessive speculation. **(a)** There is a whole section looking at correlations between atmospheric Hg(0) from Europe

(specifically it would seem Schauinsland, Germany; And data from 7 years prior) and foliage concentrations in these Lebanese olive groves, which are completely different ecological biomes and climatic zones. The spatiotemporal differences between these datasets is vast. **(b)** The authors jump between soils being a source of Hg to aerial parts of the plant and back to the atmosphere being the source without any definitive evidence that can clearly define which source is the actual source. Suggestions of soil sources would also contradict basically all (and a very large pool of) evidence that suggests this uptake pathway is minimal in trees. If the authors want to suggest this is a significant pathway they need strong evidence to do so and this study does not provide such evidence.

- I also believe the suggestion by another review that new foliage emergence is the main driver of the observed seasonal differences in foliar mercury is valid and needs detailed consideration.

Line 38-39: The term heavy metals is a poor descriptor and one that has been suggested multiple times to be made redundant (Duffus, 2009: <https://doi.org/10.1515/ci.2001.23.6.163>; Pourrett and Hursthouse, 2019: 10.3390/ijerph16224446). I would suggest changing the terminology throughout with a less ambiguous descriptor like "potentially toxic metals".

Lines 44-45: change to "Hg(0) is primarily transferred through the atmosphere by air mass movement and can undergo long-range transport".

Lines 45-47: This is incorrect as written. Hg(0) does not "covalently bond with organic groups to forming... MeHg". It must first be oxidized (either in the atmosphere or in terrestrial matrices after deposition), transferred to anoxic or poorly oxic conditions and it can then be methylated.

Lines 47-48: These descriptions about legacy mercury are extremely vague and need to be improved. It is also a bit out of place with the rest of the story and I think these two sentences could be deleted without effect.

Lines 48-50: Needs grammatical correction.

Lines 51: Delete "in the ecosystem".

Lines 51-63: This paragraph needs grammatical and structural (and English language) work. It is a bit disjointed and jumps from one thought to another continuously.

Lines 67-69: This ignores one of the most critical fluxes of Hg back to the atmosphere from forests: wildfires. Please add a statement on this and include references such as: McLagan et al. (2021) 10.5194/acp-638 21-5635-2021; Dastoor et al. (2022) 10.1038/s43017-022-00269-w; Friedli et al. (2009) 10.1021/es802703g.

Lines 73-76: I disagree with this statement. Tree ring Hg (dendrochronology) is predominantly used as an archiving tool for atmospheric Hg(0) (Hg(0) oxidised in leaves, transferred in phloem to bole wood, and generally considered to be stored long-term). It has been established for decades (Beauford et al., 1977: 0.1111/j.1399-3054.1977.tb01880.x; Lindberg et al. (1979) 10.2134/jeq1979.00472425000800040026x) and re-confirmed many times since that Hg in woody materials is derived from atmosphere. Please correct these statements accordingly.

Lines 76-77: I would suggest to add McLagan et al., (2022: <https://doi.org/10.5194/bg-2022-124>, recently accepted) to this reference on Hg dendrochemistry (using stable Hg isotopes). Some of the findings in this recently accepted study may be highly beneficial to this manuscript.

Lines 102-103: I cannot agree with this statement that roots are the primary source of Hg in contaminated areas. (1) This is unpublished work and judging from the abstract it appears they state the atmosphere as the source not the roots; (2) This is at a former Hg mine – there is MASSIVE legacy emissions of Hg(0) to the atmosphere continuing to this day at these sites, which is readily available for stomatal assimilation; (3) as previously mentioned there is countless studies during the past 50 years that show root uptake in tree ubiquitously is an very minor, if not insignificant uptake pathway. This statements needs correcting.

Lines 107-108: How does this compare to recommended soil guidelines? Please state this.

Lines 144: I really don't see the benefit of making acronyms of the sampling sites. Both a one-word towns and this just confuses readers that are not as closely linked to the study as the authors. I recommend simply writing the town names each time.

Figure 1: The climate graphs are really ancillary metadata. These are described in the method text and should be moved to the SI. Indeed, even the site map could be move to the supplementary information (SI). There are only two studies sites and again their location, climate and geography and surrounding Hg sources are described in the text. I believe this whole figure would be better served in a SI.

Line 177: "8-15 m foot circumference". There are obvious errors here. Also I highly recommend using diameter rather than circumference. It is much easier for the reader to comprehend.

Line 196-197: While I do not think this is a major problem as I believe there will be minimal Hg(0) on surfaces or within foliage and stems, it needs to be acknowledge that this heated drying method would likely eliminated any and ALL Hg(0) present in the samples.

Line 217: The detection limit should be listed as total mass of Hg, not concentration of Hg. The system does not analyze concentration (that is calculated by the mass of sample input), it is calibrated to determine the mass of Hg in any given sample. This is an important distinction.

Line 261-263: What Hg concentration is being referred to here? Hg(0) Concentration in the air? This needs to be stated. This ambiguity is exactly why the results and discussion should be combined.

Lines 312: "the main Hg content" should be changed to "Highest Hg concentration".

Lines 318-322: Once again, I disagree the main source of Hg in the stems of the trees is from the soils. What is the evidence for this? I could reference 10+ papers that have shown Hg in woody materials of trees to be almost exclusively derived from foliage and downward transport in phloem. Higher concentrations in leaves over stems is NOT evidence that Hg in stems is derived from the roots. Not all Hg taken up by foliage is transferred to woody materials, which leads to an enrichment of Hg in foliage compared to Hg in woody materials.

Lines 323-324: The downward transport of Hg in phloem eventually into roots and potential release into soils may also be contributing to Hg accumulation in soils.

Lines 333-335: This concept of soil properties driving Hg concentrations and uptake in soils was not something introduced by O'Connor et al., 2019. This is not a new idea and again has been known of for decades.

Line 336: What does nitrogen content have to do with Hg sorption and uptake in soils?

Lines 339-341: Did the authors measure wet and dry deposition of Hg? For wet deposition

to occur, there would need to be considerable Hg(II) in the atmosphere. I also see no reason as to why dry deposition would occur more in a higher temperature region. Higher temperatures favour partitioning of Hg(0) back into the gas phase, which would be suggestive of less dry deposition.

Lines 342-344: Foliage accumulates Hg(0) over time. Naturally older leaves that eventually die and senesce will be more enriched in Hg than younger leaves growing on the trees.

Lines 348-404 and Figure 3: This is far too speculative. These data are for Europe (the Authors use a site from Germany for Hg(0) in Figure 3). Lebanon is a long way from Europe and in a totally different climatic zone without typical northern temperate/boreal deciduous/conifer dominated forests. To make any sort of statement about atmospheric Hg zero concentrations this should have been measured or data taken from a long-term monitoring station in this climatic region/ecological biome. I see this whole (and very long discussion) on correlations between foliage and atmospheric Hg(0) to be too speculative to the point it is invalid. I also agree with the other reviewer that the emergence of foliage in olive trees in spring/early summer is the major driver here.

Lines 417-448: These paragraphs need grammatical and English language corrections. It is very hard to follow and from what I can derive it again seems highly speculative and to contradict the state of the science without data to support that.