

Reply on RC2

Nagham Tabaja et al.

Author comment on "Seasonal variation of mercury concentration of ancient olive groves of Lebanon" by Nagham Tabaja et al., EGUsphere,
<https://doi.org/10.5194/egusphere-2022-174-AC2>, 2022

Your input to this study is much appreciated and of great help to this work in order to improve it and make it better for publication. Your recommendations were taken into account and the English and grammar were worked on by the help of a scientific English editor.

In this paper we did not combine the results and discussion, following the journal way of dividing the different parts of the article. The repetition that can be found in the discussion is minimal and is to make it easier for the reader to relate more directly without having to go back to the results part that is much smaller in comparison to the discussion.

The repetition between the text and figure and the table is to show the needed information in a more visual way and in a form of values for the reader to be able to be in contact with the reality of the study.

The speculations made are based on the few data found and on our own analysis and point of view on the scientific data. The comparison made with the Northern Hemisphere from data of other countries far from our region is due to the lack of data in the Eastern Mediterranean. The main suggestion of the source of the Hg concentration is the atmosphere and a minimal source from the soil that does need more study and dedication.

In this study three different generations of foliage were mixed and studied that may have affected the seasonality in foliar mercury.

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 Hursthause, 2019: 10.3390/ijerph16224446). I would suggest changing the terminology throughout with a less ambiguous descriptor like "potentially toxic metals". This phrase is corrected.

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

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. It is corrected by "This highly diffusive Hg can easily pass biological barriers (i.e. cell membranes, foliage, skin) and it must first be oxidized (either in the atmosphere or in terrestrial matrices after deposition), transferred to anoxic or poorly oxic conditions and bind covalently with organic groups forming the widespread toxic methylmercury (MeHg , CH_3Hg^+) (Clarkson and Magos 2006)."

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. These sentences were deleted.

Lines 48-50: Needs grammatical correction. It was revised by an English scientific editor.

Lines 51: Delete "in the ecosystem". This word is deleted.

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. This paragraph was checked and revised by an English scientific editor.

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. This phrase is added "An important source of Hg emission to the atmosphere is from the biomass burning, ocean currents and rivers and is also considered an essential component of the Hg global biochemical cycle (Friedli et al., 2009; McLagan et al., 2021; Dastoor et al., 2022)."

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. I used this statement from a new reference from Yanai et al. 2020.

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. The following phrase is added "Another study that used Hg stable isotopes propose a reemission and reduction of the foliage Hg can also take place of this internal leaf Hg (between 29 and 42 % of gross uptake based on the plant species studied) may occur (Yuan et al., 2018)".

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 a very minor, if not insignificant uptake pathway. This statements needs correcting. This statement is removed.

Lines 107-108: How does this compare to recommended soil guidelines? Please state this. This phrase is corrected as follows "Adding to that, soil samples collected from different areas in southern Lebanon showed values of Hg concentration ranging between 160-6480

ng/g showing a high contamination levels".

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. This acronym was made to make it easier on readers to distinguish between sites without having to spell them each time over the study knowing how hard it is to pronounce them.

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. In my opinion i believe it is better for the reader to have those sites indicated directly after the text to make it clearer concerning the locations.

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. These data were rechecked and there is no error in the values.

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. This statement was amended to the following "Collected foliage and stems were rinsed with distilled water and then dried for 48 hours in an oven at a temperature of 60°C at maximum in order to remove any dust Hg from the surface of the samples."

Line 217: The detection limit should be listed as total mass of Hg, not concentration of Hg. The system does not analyse 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. The detection limit is changed to total mass as suggested.

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. The Hg(0) is the referred to Hg concentration.

Lines 312: "the main Hg content" should be changed to "Highest Hg concentration". This phrase is corrected.

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. I agree with your argument, but as said minima Hg(0) is transferred from foliage to stems and not all comes from the roots .

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. This phrase is added : 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. I agree, but I am giving here an updated reference.

Line 336: What does nitrogen content have to do with Hg sorption and uptake in soils? Nitrogen can also be a factor affecting the Hg content in soil depending on its characteristics.

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. The author discussed dry deposition and indicated the effect of higher temperature on higher 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. I agree with your statement.

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. Another data done on the western Mediterranean showed the same results as we implicated in our study (Mastromonaco et al. 2017). Concerning this part, the problem is that there is no data related to the Eastern Mediterranean that we could find to use as a comparison in our study. But this can be a starting point to have more data collection that is closer to our studied regions.

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. These paragraphs were checked by a scientific English editor. Some data was given to support our speculations, but we tried to give our point of view in regards to the Hg cycling in stems, litter and soil system.