Response to the comment on EGUsphere-2022-174 by Hakan Pleijel (Referee 1) on the article "Seasonal variation of mercury concentration of ancient olive groves of Lebanon" by Tabaja et al. https://doi.org/10.5194/egusphere-2022-174-RC1, 2022

We are very thankful for your constructive comments and feedback on the manuscript submitted. We accepted all the comments provided and amended them accordingly.

Concerning the language and clarity in several statements were revised and improved. A native English speaker checked the manuscript and helped in improving the form. In regards to the phenology of leaf dynamics, it has been indicated that the collected samples were merged from three different years and merged for analysis. The phenological growth stages of olive trees described by Sanz-Cortès et al. (2002) in Spain suggest leaf development from March to November. Hence the Hg concentration measured on monthly collected leaves represents an average of Hg accumulated in young leaves (year N of collection) and older leaves (N-1 year and N-2 years) where N is equal to 2019 and 2020. According to Pleijel et al. 2021, young leave has less mercury than older leaves generations. We can expect that our Hg concentration is lower during the growth of young leaves due to the Hg dilution signal with the low Hg content of the younger leaves. In a very simple approach, we can suggest the late winter-early spring should exhibit the lowest Hg contents. However, our results show the lowest values in summer when young leaves have already grown significantly. In addition, physiology has been more integrated into the text (Line 462-469) but of course, more detailed work can be made separately to focus on this aspect.

Line 21-22: It is corrected. "This study aimed to investigate the seasonality of the mercury (Hg) concentration of olive trees foliage, an iconic tree of the Mediterranean basin."

Line 22: It is corrected. Changed was to "were".

Line 27: It is corrected by "It is noteworthy that olive fruits also have low Hg concentration (~7-11 ng/g)."

Line 30: It is corrected (Line 31-33) by "Our study draws an adequate baseline for Eastern Mediterranean and region with similar climate inventories on Hg vegetation uptake and new studies on olive trees in the Mediterranean to reconstruct regional Hg pollution concentrations in the past and present."

Line 38: It is corrected by "Mercury (Hg) is among the most widely distributed heavy metals polluting the Earth (Briffa et al. 2020)."

Line 51: It is corrected (Line 56) by "Forests are known to act as a sink of atmospheric Hg."

Line 52: It is corrected (Line 57) by "Plant foliage take up of Hg deposited on leaf surfaces through the stomata (i.e. Leaf gas exchange) and leaf cuticles".

Line 54-55: It is corrected (Line 59-60) by "where it accumulates with minimal mobility and small portions released back into the atmosphere or transferred to other plant organs"

Line 60: (Line 73-74) It is replaced from "is said" by "has been estimated".

Line 64: (Line 77) It is replaced from "earth" by "terrestrial".

Line 68: (Line 80-83) Deleted.

Line 69: (Line 82-83) Deleted.

Line 72-73: The sentence starting "Differences …" is not clear. (Line 99-109) This sentence is removed.

Line 94: It is corrected (Line 127) by "an air pollution emission area."

Line 111: "outcomes and consumed"? Not clear. (Line 152) It is replaced by "for consumption."

Lines 114-115: this sentence starting "In addition …" is not complete. (Line 155-156) This sentence is removed and objectives amended.

Line 117-119: The objectives are amended as per the feedback received. (Line 161-171) "In this study two sites, known for their century-old olive groves and located at two different altitudes in Lebanon, were selected to assess the Hg contents. In these remote areas, no direct sources of mercury contamination are reported and hence we expect very low Hg concentrations. However, due to atmospheric transport of Hg, dry or wet deposition of Hg can be expected in remote areas (Grigal, 2003). The main objectives of this study are to examine and compare Hg levels in foliage, stems, fruits, litter and soil measured in each of these two olive groves, which we monitored monthly for 18 months. The second objective is to analyze the relative importance of Hg uptake by the soil and foliage in comparison with the atmospheric Hg. Since the distribution of Hg pollution is by nature geographically widespread, and given the extent of Hg pollution in the Mediterranean and the transfer of pollution by wind and the Mediterranean Sea, long-distance contamination occurs over large areas. This study may draw an adequate baseline for Eastern Mediterranean and region of similar climates inventories on Hg vegetation uptake and new studies on olive trees in the Mediterranean to reconstruct regional Hg pollution concentrations in the past and present."

Line 142: it does not become clear if Chekka town is a source of Hg emission or only of other pollutants. This sentence is cleared (Line 197-198) by "To our knowledge no direct Hg pollution is reported at Chekka and Selaata sites." (Line 221) "Here as well, we did not find indication of direct Hg pollution."

Line 143: It is corrected (Line 194) by “carbon monoxide” and “particulate material”.

Line 224: It is corrected (283-284) by “For the statistical analysis we used the R 4.1.0 program. Our data are not normally distributed, so for the effect of tissue type on Hg concentration, Wilcoxon test was used with the tissue type (foliage and stems) as the main effect.”

Line 247-249: why is not the concentrations of fruits included in these comparisons? (Line 311) The fruits are added.

Line 266-267: It is corrected (Line 329) by “A seasonal effect on foliage and stems was registered (p-value<2.2*10-16; Figure 2a,c).”

Line 282 and section 3.3: It is rearranged and corrected (Line 361-378) by

“**3.3. Inter-individual variability between trees for each site**

In the upper terrace of Bchaaleh grove, the foliage average Hg concentration of BCO4 and BCO1 varied between 42.4 ± 11.5 ng/g and 44.6 ± 13.3 ng/g respectively showing no significant difference (p-value = 0.8225). In the lower terrace of the same site, foliage average Hg concentrations of trees BCO12 and BCO9 were found to vary from 45.6 ± 12.7 ng/g to 60.7 ± 12.7 ng/g respectively (Figure 2a) exhibiting a significant difference ((p-value=0.0059). Tree BCO9 is significantly different to each of the three trees (p-value< 0.0059) while BCO1, BCO4 and BCO12 have very similar Hg contents (p-value= 0.46).

In the upper terrace of Bchaaleh grove, the stems average Hg concentration of BCO4 and BCO1 varied between 7.0 ± 2.8 ng/g and 7.1 ± 2.9 ng/g respectively showing no significant difference (p-value= 0.94). In the lower terrace, stems average Hg concentrations of BCO12 and BCO9 are 6.4 ± 2.2 ng/g and 11.2 ± 5.2 ng/g respectively showing a significant difference (p-value= 0.0054; Figure 2c). For BCO1 and BCO12 there was no significance difference (p-value= 0.5725), the same goes for BCO4 and BCO12 (p-value= 0.523).

The average concentration per tree in foliage and stems were 32.4 ± 12.2 ng/g and 8.5 ± 4.0 ng/g respectively for KWO1, 32. 8 ± 14.7 ng/g and 8.9 ± 6.0 ng/g for KWO2, 37.6 ± 14.0 ng/g and 9.3 ± 6.7 ng/g for KWO3 and 37.7 ± 13.6 ng/g and 9.6 ± 4.0 ng/g for KWO4 (Figure 2b,d). In Kawkaba grove, comparison of the foliage Hg concentration between the four studied trees shows no significant difference (0.22<p-value<1), neither for the stems (0.21<p-value<0.96).”

Line 298: It is corrected and values of foliage and stems are separated. (Line 382-390) “At first glance, seasonal variations of the Hg concentrations of the foliage of both sites suggest a covariation with climatic parameters (Precipitation amounts, Relative Humidity and Temperature) (Figure SI) and atmospheric pCO2. Foliage Hg content increased with higher precipitation and lower temperature (Autumn and Winter) while during the warmer and dryer seasons (May to mid-October), the Hg concentration of foliage decreased (Figure S). However, the Wilcoxon test for a non-normal distribution shows no significant correlation between Hg concentration of foliage and precipitation (p-value= 0.95). While temperature, relative humidity and atmospheric CO2 (pCO2) shows a significant correlation (p-value = 2.2e-16). For the stems, Hg concentration also showed no significant correlation with precipitation (p-value= 0.1147), and a significant correlation with temperature, relative humidity and pCO2 (p-value= 2.2e-16)."

Line 343: It is amended by (Line 445-452) "The litter showed higher Hg concentration than that in foliage in both Bchaaleh (62.9 ± 17.8 ng/g) and Kawkaba (75.7 ± 20.3 ng/g) (Table 1). This has been also described by Rea et al., (1996) and Zhou et al., (2021) in uncontaminated and contaminated sites where litterfall Hg contents were systematically higher than the foliage Hg contents. The bacterial and chemical decomposition of the litter decrease significantly the amount of C compared to the Hg that conversely may continue to increase due to the continued absorption of Hg from precipitation and throughfall (see Pokharel and Obrist, 2011; Zhou et al., 2021). Another possible explanation is that the leaves shed as litter are likely to mostly be the oldest leaves which have accumulated Hg during the longest period of time and thus have higher Hg concentrations than the remaining foliage have on average since they consist of both younger and older foliage (Rea et al. 1996; Pleijel et al. 2021)."

Line 353: It is corrected (462-463) by "accumulation in leaves after stomatal uptake."

Line 440: since the distribution of Hg pollution is by nature geographically very widespread, long-distance contamination occurs and it may be better to say "In sites without local contamination" instead of "In uncontaminated sites". Similarly on line 452 it would be appropriate to say "locally uncontaminated" rather than "non-contaminated". Line 440: It is deleted (Line 591-599). **Line 452:** It is corrected (Line 605) by "remote site".

Line 459 and 467: The conclusion is amended by (Line 605-627) "This is the first study conducted on monumental olive trees in a non-contaminated remote site of the MENA region without local contamination and followed at a monthly basis over 18 months. Findings of our study in remote sites without local contamination indicate a higher uptake of Hg in the olive foliage compared to stems, fruits items and a remarkable HgFoliage seasonal variation in both studied groves. Winter and Spring were particularly suitable for Hg accumulation in foliage in both sites. The significant correlation between our HgFoliage contents and the atmospheric Hg content and pCO2, despite the one to two months' time lag, suggests that the main source of HgFoliage is the atmospheric Hg as observed in different species and studies (conifers and hardwood). Hg is absorbed by the foliage, via the open stomata, driven by the interaction of high vegetal activity, temperature, water availability and the processes that control transpiration, which is likely to be seasonal. Hence physiological and climatic processes explain the seasonal Hg accumulation in foliage. Thus, a more intensive study taking account the phenological dynamics of olive tree foliage must be focused on. and more intensive studies on soil and litter is needed to be able to assess the source of Hg uptake in the olive trees. Further comparison and studies on the seasonal atmospheric Hg in the eastern Mediterranean basin are necessary to test our hypothesis of the reversed seasonality of Hg since contrary to the global Northern Hemisphere and western Mediteranean region vegetation, our olive groves act as a sink of Hg and CO2 when global Northern and western Mediteranean vegetation is emitting. This relationship $Hg_{Foliage}$ – Hg_{atm}-pCO_{2atm} should be further investigated along the season and locally to better understand the observed time lags. Soil surface registered the highest Hg concentration among all studied compartments due to well-known processes of litter and throughfall that incorporate Hg to the soil surface. Moreover, this study highlights significant differences between Hg_{soil} in Bchaaleh and Kawkaba groves due to differences in soil characteristics. In this study we worked on the present time samples in order to have a better understanding of the Hg cycle in the olive tree. A second step would be to reconstruct the paleo-evolution through also studying tree rings Hg concentration were we can also identify a seasonal variation. Our main contribution in this study is to see how the present-day olive trees records some elements such as Hg to better understand how the Hg in tree rings could be used for the past accumulation records."

Why are some references in capitals, e.g., lines 512-513, lines 521-523, lines 712-715. Pleijel et al and Wohlgemuth et al are no longer preprints. All references are amended.

Font size in figures should be increased to improve readability. It is amended.

Response to the comment on EGUsphere-2022-174 by Anonymous Referee on the article "Seasonal variation of mercury concentration of ancient olive groves of Lebanon" by Tabaja et al. https://doi.org/10.5194/egusphere-2022-174-RC1, 2022

Thank you very much for the pertinent revision of our manuscript. 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 revised by the help of a scientific English editor.

In this paper we did not combine the results and discussion, following the journal mostly used 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 which is much smaller in comparison to the discussion.

The repetition between the text and figure and the table have been minimized.

The speculations made previously have been revised. We removed the comparison made with data of other countries far from our region (closer Mediterranean dataset were obtained from a recent publication). In our study, the main suggestion of the source of Hg is the atmosphere via elemental gaseous Hg (Hg(0)). While soil source part is deleted since we do not have enough evidence and it was not demonstrated in previous studies.

In this study three different generations of foliage were mixed and studied. We are aware that this certainly affected the seasonality in foliar mercury. This has been discussed following the rev 1 comments. We included in the text (Line 233-239) "For each olive tree, both sun exposed and shaded foliage and stems (terminal portions of 20 cm) with no evidence of pathogens were randomly taken and merged from the upper, middle, and lower canopy position of the olive trees on a monthly basis using a manual pruner. The phenological growth stages of olive trees described by Sanz-Cortès et al. (2002) in Spain suggest leaf development from March to November. Hence it should be mentioned that the Hg concentration measured on monthly collected foliage represents an average of Hg accumulated in young foliage (year N of collection where N is equal to 2019 and 2020) and older foliage (N-1 year and N-2 years). Fruits were collected in April 2019."

In addition to (Line 462-469) " This seasonal change is explained by the seasonal tree physiology variations such as the accumulation in leaves after stomatal uptake (Pleijel et al., 2021; Wohlgemuth et al., 2021). We can suggest that during winter-early spring, water is available and photosynthetic activity is not limited, hence both CO2 and Hg diffuse through opened stomata inside the foliage. As shown on figure 2, Hg in foliage is low in summer-fall and hence act as a sink of Hg. Note that despite we mixed three generations of olive leaves (year N to N-3), the most recent of which are known to be low in mercury (Pleijel et al., 2021), the seasonal signal is still very remarkable. Therefore, one can speculate that the mercury levels would have been higher if we had avoided the recently formed foliage during spring and early summer. This may also explain the large difference in Hg levels between litter and foliage."

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". This phrase is corrected (Line 38) as follows: "Mercury (Hg) is among the most widely distributed potentially toxic metals polluting the Earth (Briffa et al. 2020)".

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: (Line45-46) "It is primarily transferred through the atmosphere by air mass movement and can undergo long-range transport. Because of its high volatility and susceptibility to oxidation, elemental Hg(0) is the predominant form of Hg in the atmosphere that can be accumulated into foliage."

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 (Line 46-52) by "Because of its high volatility and susceptibility to oxidation, elemental Hg(0) is the predominant form of Hg in the atmosphere. This highly diffusive Hg can easily pass biological barriers (i.e. cell membranes, foliage, skin). Mercury has three oxidation states, namely, Hg(0) (elemental mercury), Hg(I) (mercurous), or Hg(II) (mercuric), although Hg(I) mercurous form is not stable under typical environmental conditions and, therefore, is rarely observed. It is likely that the Hg(II) high binding affinities bind covalently with organic groups to form the widespread toxic methylmercury (MeHg, CH3Hg+) (Du and Fang, 1983; Clarkson and Magos 2006; Pleijel et al., 2021)."

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 (Line 52-53).

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

(Line 53-55) “The exchange of Hg between the soil and plants is not stable and is variable dependent (e.g. cation-exchange capacity, soil pH, soil aeration, and plant species) (Patra and Sharma 2000).”

Lines 51: Delete “in the ecosystem”. This word is deleted. (Line 56) “Forests are known to act as a sink of atmospheric Hg.”

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. (Line56-64) “Forests are known to act as a sink of atmospheric Hg. Plant foliage takes up of Hg deposited on leaf surfaces through the stomata (i.e. Leaf gas exchange) and leaf cuticles (Hanson et al. 1995; Jiskra et al. 2018; Li et al. 2017; Lodenius et al. 2003; Maillard et al. 2016; Rea et al. 2002; Yanai et al. 2020) where it accumulates with minimal mobility and small portions released back into the atmosphere or transferred to other plant organs (Cavallini et al. 1999; Hanson et al. 1995; Li et al. 2017; Lodenius et al. 2003; Schwesig and Krebs 2003). All together these authors contributed to highlight the dynamic role of the foliar surfaces in terrestrial forest landscapes acting as a source or sink dependent on the magnitude of current Hg concentrations. Hanson et al. (1995) suggested a species-specific compensation concentration (or compensation points) for Hg deposition.”

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 “Though variable from year to year, Hg emission to the atmosphere from biomass burning is considered as an important driver of the global Hg biogeochemical cycle (Friedli et al., 2009; De Simone et al., 2015; 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. “Emission reduction measures adopted in Europe and North America since the 70s are corroborated by Hg dendrochemistry analysis showing a declining Hg concentration trend from the older to newer tree rings

(Yanai et al., 2020). Indeed, tree ring Hg (dendrochronology) is a powerful archiving tool for atmospheric Hg(0). After Hg(0) oxidation inside the leaves, Hg(II) bind to organic compounds and then is transported to the bole wood via the phloem (Beaufort et al., 1977; Lindberg et al., 1979). This is corroborated by the recent study of McLagan et al. (2022) showing the benefit of the stable Hg isotope analysis on dendrochemistry. "

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. We thank you for the reference. We added it as shown in the previous section.

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. (Line 133-137) This statement is removed.

Lines 107-108: How does this compare to recommended soil guidelines? Please state this. This phrase is corrected as follows (Line 147-150) "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 (Borjac et al. 2020) as indicated by World reference Senesi et al. 1999, Kabata-Pendias 2001 ".

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. I changed the acronyms to town names.

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. On the behalf of all the authors, we believe it is better for the reader to have those sites indicated directly after the text to make it clearer concerning the locations. Concerning the climatic data, it is removed since it is available in the text. The climatic data was placed in the supplementary information as Figure S1.

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. This sentence has been changed to (Line 228-229): "For the Hg concentration analysis, four olive trees (3 to 5 m diameter and an average height of the trees 4-6m) were sampled in each of the two groves from February 2019 to September 2020."

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 (Line 250-252) "Collected foliage and stems were rinsed with distilled water and then dried for 48 hours in an oven at a temperature of 50°C at maximum (Demers et al. 2013; Li et al. 2017; Pleijel et al. 2021). This procedure likely eliminate any 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 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. (Line 272-275) "The absolute detection limit (ADL) of the analytical technique (AMA 254) was estimated at 0.04 ng Hg. As a consequence, the method detection limit (MDL) for samples analyzed were 0.7 ng Hg/g for soil, litter and foliage and 0.4 ng Hg/g for stem and wood. These MDL were much lower than the measured Hg concentration in the various samples."

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 concentration referred to here is the Hg concentration in the foliage and stems (Plant tissues), litter and soil. (Line 323) "Seasonal variation of Hg concentration in plant tissues, litter and soil"

Lines 312: "the main Hg content" should be changed to "Highest Hg concentration". This phrase is corrected as follows: (Line 397-399) "Our data corroborates previous studies (Bargagli 1995; Higueras et al. 2016) showing that olive foliage has the main highest Hg content concentration among of plant tissues."

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. This statement is deleted (Line 403-406).

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: (Line 414-418) "Inside the leaves the oxidized Hg(II) has high affinities to bind covalently with organic groups (Du and Fang, 1983; Clarkson and Magos 2006; Pleijel et al., 2021). The Hg can be translocated by phloem transport to the stems and eventually into roots and potential release into soils may also be contributing to Hg accumulation in soils (Giesler et al., 2017; Schaefer et al., 2020)."

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. The following references are added (Line 432-433) transport (Richardson et al., 2013; Chen et al., 2016; O'Connor et al., 2019).

Line 336: What does nitrogen content have to do with Hg sorption and uptake in soils? (Line 433-437) "Nitrogen can also be a factor affecting the Hg content in soil depending on its characteristics. Nitrogen increase can change the equilibrium of soil solution and the morphology of roots, causing a possible increase in Hg availability in soil and increases the Hg uptake by the plant (Alloway, 1995, Barber, 1995, Carrasco-Gil et al. 2012). The increase in Hg availability in the soil is due to the organic Nitrogen that provides a high absorption capacity, retaining the atmospheric Hg deposition (Obrist et al. 2009). Nitrogen supply prevents oxidative stress in roots, but also can improve root development and increase the uptake of Hg from the soil (Carrasco-Gil et al. 2012)."

These associations are likely the result of a high sorption capacity provided by organic C and N groups which efficiently retain atmospheric Hg deposition, and by high C and N pools reflecting a high ecosystem productivity which also leads to elevated atmospheric deposition inputs via leaf litterfall and plant senescence. Soil N proved to be a particularly reliable predictor for soil Hg levels explaining over 90% of the variance in both concentrations and pool sizes across all measured sites and soil horizons.

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 authors measured litter that represent a large portion of Hg dry deposition to forested soils in terrestrial ecosystems. This statement is deleted (Line 441-443).

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. (Line 449-452) "Another possible explanation is that the leaves shed as litter are likely to mostly be the oldest leaves which have accumulated Hg during the longest period of time and thus have higher Hg concentrations than the remaining foliage have on average since they consist of both younger and older foliage (Rea et al. 1996; Pleijel et al. 2021)."

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.

In the process of the reply to the review we rechecked the papers available on the atmospheric mercury closer to our region. We were not able to find any information in the region on atmospheric mercury data, but we passed across an interesting newly published paper by Martino et al. 2022 that is the only atmospheric gaseous elemental mercury (GEM) measurement near an olive site that is comparable to our study. Martino et al. 2022 publication shows the factors affecting the GEM values are mainly the factories, sea emissions, fires, and vegetation. He also observed the normalized difference vegetation index values (NDVI) with GEM concentration along different seasons from 2018 to 2020, show that the higher vegetative uptake explains the GEM depletion in certain seasons and years. Even though this paper is not in the same area as that of our sites, we take profit to compare with same years studied (2019-2020). We used their data from table 1 representing the monthly median GEM values and compared it to our data. Our values of the foliage in Bchaaleh and Kawkaba show higher values of Hg concentration in winter and lower in summer, where seasonality is clear as that of Martino et al. 2022. This seasonality can change due to many factors such as wind, fires, industrial surroundings and other factors. At the same time, we decided to remove the northern Hemisphere data upon your feedback.

Other data was done on Total Gaseous Mercury (TGM) in the French coastal Mediterranean site which showed higher values in winter/spring and lower values in summer in the Mediterranean region. This studies also showed seasonality with the atmospheric mercury due to dispersion of pollutants in the troposphere and high TGM values that are due to air masses from regional and local sources (Marusczak et al. 2015).

Mastromonaco et al. 2017 Showed also that the main source of mercury occurs from the Mediterranean Sea water and is affected by wind speed with higher values of GEM concentration registered in autumn in comparison to summer, with higher evasion rates found in summer in comparison to the autumn season. Another study that includes countries closer to Lebanon by Kotnik et al. 2014 found a seasonal variation in Hg in the Mediterranean region near Spain, Italy, Turkey and Israel (1998-1999) with the highest Hg average concentration during winter and the lowest during autumn.

Concerning the emergence of foliage in olive trees in spring/early summer being a major driver, this is shown through the seasonality registered during the different seasons due to the merging of three different years of the foliage in our study.

The following was amended in the discussion: (Line 474-485, 511-512). “Year 2020 has higher foliage Hg values than 2019. This can be related to a higher vegetation uptake in 2020 in comparison to 2019.

Martino et al., (2022) showed seasonal variability of Gaseous Elemental Mercury (GEM) in the atmosphere in southern Italy (Figure 3a). They also related this seasonality to high energy use for heat during winter thus higher emissions due to coal combustion (Weigelt et al., 2015) and the effect of air mass trajectory on the GEM seasonality where the winter peak is connected to the Hg re-emission from sea surface and with long-range transport, in addition to the GEM oxidation rate in warmer months (Horowitz et al., 2017; Martino et al., 2022). Martino et al. (2022) comparing the normalized difference vegetation index values (NDVI) with GEM concentration of the same seasons over the different years (2018 to 2020), show that the higher vegetation uptake explains the GEM depletion in certain seasons and years. This was also collaborating Jiskra et al., (2018) for the Northern Hemisphere site. Since no data of atmospheric mercury in Lebanon or surrounding countries are available we used the atmospheric Hg time series data of Martino et al. (2022) (Figure 3a). We observed opposite trends between foliage Hg concentration and air Hg (negative covariation in 2019 and positve covariation in 2020) (Figure a,d,e)." " Alternatively other studies reported a positive correlation between atmospheric Hg and crops (Niu et al. 2011) as observed in our study between the Hgfoliage and the atmospheric Hg in 2020 (Figure 3a,d,e)."

Line 486-510 was deleted.

Figure 3 is also amended (Line 556).

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 scientific point of view and references are given. (Line 579-587) "We can suggest the following Hg cycling in the system of the olive grove/soil. In winter-early spring the highest concentrations in foliage continuously feed the litter and can explain the following maximal spring Hg content of the litter. The decomposition of the litter organic matter during the wettest conditions likely liberate Hg in the Hg(0) or Hg(II) forms or MeHg either towards the atmosphere or the surface soil (see table 2) respectively (Gworek et al. 2020). A fraction of the degraded organic matter is transferred through gaseous evaporative processes towards the atmosphere while another fraction of the Hg is leaching towards the deeper soil in addition to dry Hg deposition during dry season (Teixeira et al. 2017). We can also speculate that the small Hg decrease observed in soil 0-30, 30-60 cm during the winter season in Bchaaleh can be due to the minimal absorption of total Hg and MeHg through the roots and xylem sap to the above ground tissues (Johnson and Lindberg 1995)."