

Please find below our detailed responses (in blue) to comments given by Reviewer #3, where the original reviewer comments are repeated here in black for clarity and completeness.

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## Reviewer #3

<https://doi.org/10.5194/egusphere-2023-1363-RC3>

I have completed my review. I have a few suggestions for the manuscript and no basic criticisms. The main conclusion is that there is no similar pattern for the two lakes and there is not a unifying hypothesis to explain the Hg variability. Different attempts to link the Hg flux to different sources fail to be conclusive for all the periods. This can be frustrating, but it is a conclusion.

We thank reviewer #3 for taking the time to read our work, and for their constructive feedback. We agree that it is somewhat difficult to obtain a conclusive outcome in this study. However, we also feel this outcome reinforces the value of the study, as it provides valuable information on the complexity of Hg cycling through terrestrial lake systems on these timescales. Thus, we hope that it will inspire future work in this research domain.

I think the statistic applied at the data is too basic to be able to give a better understanding (if different handling of the data can produce an easier interpretation), but I'm not so expert to give some further comments on that.

Exploration of how different statistical methods may be used in analysis of Hg-based datasets would certainly be valuable in the future. We agree the level of statistics here applied is modest - this was done on purpose. While we have tested various other analyses, none of these yielded appreciable improvements or insights. For example, application of robust regression could partially account for non-linearity between Hg and host phase variability in the two datasets, yet it also required selection of one dominant host phase for each core. Distinct anomalies in both records suggest that (i) the dominant host phase did indeed change at discrete points in time and (ii) the total accumulation (availability) of Hg in the system is equally important so that normalized Hg needs to be treated with caution. From this, we concluded that more advanced data treatment is not warranted and could end up obscuring clear assessment of the processes underpinning the observed signals.

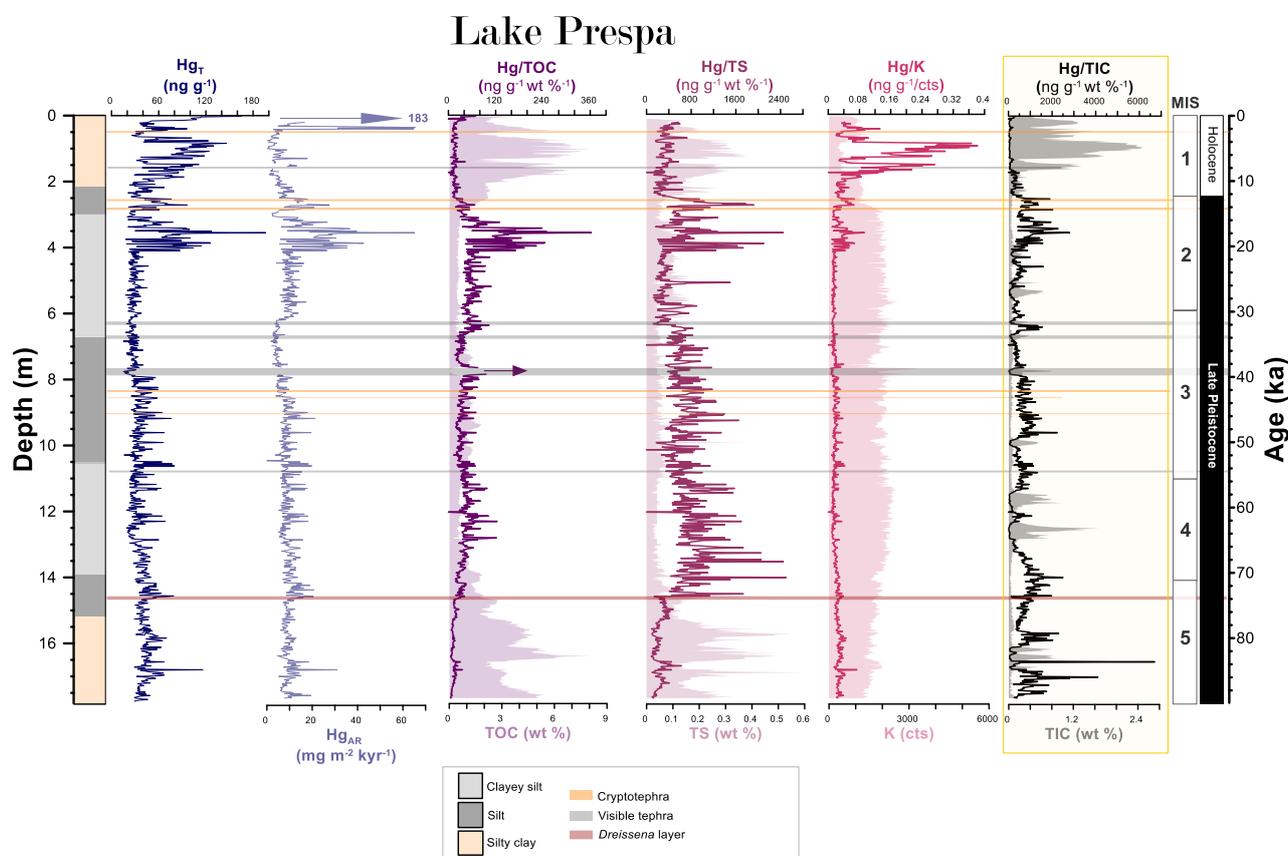
I can additionally note that during the Holocene TIC increase and it would be interesting to normalize the data also for TIC for this interval.

This is certainly an interesting observation. We agree that TIC variability warrants more explicit mention in the manuscript, and so will first make the following text revision:

**Lines 616 – 618:** *Between ~12 and 3 ( $\pm 0.5$ – $0.2$  ( $1\sigma$ )) ka Lake Prespa captures a series of large peaks in  $Hg_T$  and  $Hg_{AR}$ , corresponding to high TOC and TIC indicative of elevated productivity, higher rates of organic material preservation, and limited mixing (Fig. 5). Conversely,  $Hg_T$  and  $Hg_{AR}$  show a progressive decline in Lake Ohrid during MIS 1, despite coeval increases in TOC and TIC (Fig. 6).*

Below and in our revised supplementary file (Figure S5), we will show a variation on Figure 5 which includes a profile for Hg normalized to TIC (shaded in yellow). This profile appears remarkably similar to Hg/TOC, with low values shown throughout the Holocene, where clear TIC peaks are visible. This observation is not surprising, given that the TIC in Lake Prespa is generally enriched in organic-rich intervals where algal productivity is high, lake water mixing was reduced, and bottom waters were anoxic for longer periods (Leng et al., 2013). High productivity also explains the presence of endogenic calcite (TIC) during the Holocene, as it would more readily facilitate increased CO<sub>2</sub> assimilation; a process enhanced by plentiful bicarbonate supply from surface run-off into the lake, concentrated by enhanced evaporation due to warmer regional temperatures (Leng et al., 2013; Matzinger et al., 2007). Together,

this suggests that the processes underpinning the relatively strong Hg/TOC relationship during the Holocene are also linked to those associated with variability in TIC.



**Figure 5:** Total Hg ( $Hg_T$ ) and total Hg accumulation rate ( $Hg_{AR}$ ) for core Co1215 from Lake Prespa, presented as a function of depth and time, and relative to lithofacies, visible (grey shading) and cryptotephra (orange shading) layers. We include records of  $Hg_T$  (this study) normalized to records of total organic carbon (TOC) (Damaschke et al., 2013), sulphide (estimated by total sulphur (TS)) (Aufgebauer et al., 2012), detrital mineral abundance (estimated by potassium (K)) (Panagiotopoulos et al., 2014), and total inorganic carbon (TIC – highlighted in yellow) (Damaschke et al., 2013), with filled shading marking the original datasets. A distinct lake low stand based on seismic profiles and sedimentological data is marked at 14.63 - 14.58 m depth (red shading) (Wagner et al., 2014). A purple arrow marks sections where artificially high Hg/TOC values are generated by a sharp drop to near-zero TOC (<0.06 wt. %) coinciding with deposition of the Y-5 (17.1 m) tephra unit – an effect expected as background sedimentation is interrupted by volcanic ash deposition. White boxes mark the marine isotope stages defined by (Lisiecki and Raymo, 2005), and stratigraphic periods are labelled in black/white.

We suggest that changing **Figure 5** of the main text is not necessary in light of the broad similarity of the Hg/TIC and Hg/TOC profiles during the Holocene and would not constitute a meaningful addition to the manuscript. However, we will make more explicit reference to variability in TIC and it's relation to Hg during the Holocene by means of the revised text presented above, and will include the Figure above in the supplementary file.

Probably, it would be useful to mention in a more direct way tectonic activity as a potential source. The two lakes are placed in a very active tectonic zone, a present large gas emission from fault is present not far from lake Ohrid. Difficult to use this as an argument but it needs to be considered.

This is a welcome suggestion, but as the reviewer also points out, it is difficult to directly address in the absence of quantitative data that can quantify the rate and/or intensity of Hg release from the faults present in the Prespa/Ohrid region. Although these faults were likely active during the most recent ~100-kyr of lake history, earthquake frequency nor intensity has

been reconstructed for this region on these timescales, and so discrete Hg signals cannot be linked to specific events. To acknowledge the potential for Hg emission into the catchment by seismic activity while remaining mindful of manuscript length, we will add an additional section to our attached supplementary file:

#### **Text SD4: Tectonic activity and Hg release**

Reference to this supplementary discussion will be added to the main text as follows:

**Lines 704 – 708:** *Local differences in Hg emission by neotectonic activity may have also contributed to the divergent Hg signals, owing to differences in the host rock geology, tectonic instability, and mechanical stress regimes of faults surrounding the two basins (Hoffmann et al., 2010; Lindhorst et al., 2015). However, the significance of these differences cannot be fully assessed in the absence of direct Hg emission measurements (see **Text SD4**).*

An additional argument not considered (which needs at least to be mentioned) is the dust transport. Loess belt is diffuse in the Mediterranean and there is evidence of loess deposition also in Macedonia even if not well described.

To give further credence to evidence for variable dust deposition in the Balkan region over the last glacial period, we will add the following text to the manuscript, with direct reference to the loess-based evidence that reviewer #3 rightly highlights:

**Lines 538 – 544:** *However, we see no clear evidence atmospheric dust played a major (direct) role in the local Hg cycle in our data. For example, peaks in elemental ratios typically associated with mineral dust deposits (e.g., Zr/Ti) do not correspond to peaks in Hg<sub>T</sub> and/or Hg<sub>AR</sub> (Vogel et al., 2010), nor loess-based evidence substantially higher aeolian dust fluxes over Central Europe and the Balkans during the last glacial maximum (Újvári et al., 2010; Rousseau et al., 2021). Marine sediment records also fail to capture measurable changes in dust fluxes over the Ionian and Aegean seas corresponding to pronounced Hg signals in Lake Ohrid (Ehrmann and Schmiedl, 2021).*

In the paragraph hosting this text, we further describe and assess the potential for a dust-derived Hg source to lakes Prespa and Ohrid during the last glacial. However, given the null result of this exploration, we intend to keep this section relatively short similar to our original submission.

**Page 8:** The description of the climate is not very convincing.

Although we emphasize that the primary aim of this manuscript paper is not a paleoclimate reconstruction, we agree that a more descriptive summary of the regional paleoclimate history would be useful in this section of the manuscript: to provide important context and overview of that may inform the Hg records presented herein. As such, the following changes will be made to the text:

**Lines 148 – 165:** *Major shifts in sedimentation and catchment structure of lakes Prespa and Ohrid generally correspond to the large-scale climate oscillations captured by proxy records across southern Europe throughout the last glacial-interglacial cycle (~100-kyr) (e.g., Rasmussen et al., 2014; Sanchez Goñi and Harrison, 2010; Tzedakis et al., 2006). Generally higher local temperatures and moisture availability are observed prior to 74 ka, following which conditions became distinctly colder and/or drier. This resulted in the rapid recession of forest ecosystems, intense erosion of local soils and catchments, and elevated aeolian activity. Although slightly warmer conditions were restored between ~57 and 29 ka, both moisture availability and temperature dropped again during the Last Glacial Maximum (LGM; ~29 – 12 ka) – favoring the growth and development of glaciers and (peri)glacial features in the Prespa/Ohrid catchment (Ribolini et al., 2018; Gromig et al., 2018; Ruzkiczay-Rüdiger et al., 2020), but also across the Balkan peninsula (Allard et al., 2021; Hughes and Woodward, 2017; Leontaritis et al., 2020). At ~12 ka, the Pleistocene to Holocene transition saw the rapid propagation of warmer, wetter conditions across the region (known as Termination I) with only brief excursions from this warming trend, such as episodes of transient drying and/or cooling at 8.2 ka and 4.2 ka (Bini et al., 2019; Aufgebauer et al., 2012). Anthropogenic influence on the Balkan landscape becomes increasingly clear from ~2.5 ka onwards, mainly in the form of increased erosion regimes, forest clearance, agricultural land modification, and evidence for metallurgic practices (Panagiotopoulos et al., 2013; Cvetkoska et al., 2014; Radivojević and Roberts, 2021).*

**Line 226:** Be honest is correct to quote Zanchetta et al. 2018 but also Scaillet et al. 2013 QSR 78, 147-154.

We will revise the text to include the correct two citations for this eruption date:

**Line 234:**  $Y-6$  ( $45.50 \pm 1$  ka (Zanchetta et al., 2018; Scaillet et al., 2013))

**Line 267:** Delete zirconium.

Good observation. Text will be corrected to read:

**Line 272:** Elemental intensities were obtained for K, Ti, Fe, Zr

**Lines 346-348:** This sentence is very vague. By definition Holocene is interglacial. You need to specify e.g., increase of forest and so on.

To make our definition of the Holocene clearer in this section, we will revise the text as follows:

**Lines 364 – 366:** *Widespread proxy-based evidence for warmer temperatures, forest expansion, and increased precipitation representative of interglacial climatic conditions marks the start of the Holocene epoch (~12 ka) in SE Europe...*

**Lines 414-416:** This sentence is obscure to me. Probably useless. What do you mean with “reminiscent”. I don’t see the importance of this sentence.

We feel that comparison of the Lake Ohrid Hg record with another ancient lake succession does add value to this passage, as it introduces the argument that Hg availability ( $Hg_{AR}$ ) is limited by the Hg fluxes to the catchment and that, even in presence of abundant host-phases, it will not correlate and/or may get diluted instead. However, we agree this sentence could be written more clearly. Thus, we will revise the text to improve the readability of the point being made:

**Lines 444 – 447:** *the relationship between  $Hg_T$  and organic matter in Lake Ohrid also shows an inverse correlation (Fig. 4), similar to the trend observed in the uppermost sediments of a ~5 Ma succession from Lake Baikal (Russia) (Gelety et al., 2007). These trends may be explained by a scenario where the Hg flux to Ohrid from direct deposition and/or surrounding catchment is typically the limiting factor, rather than availability of potential host phases.*

**Lines 475:** I don’t think MIS 3 is considered anymore an interglacial (for a while). I think this sentence should be deleted.

We agree that much of the text here is probably superfluous. While remaining mindful that comparing MIS 3 to the preceding stages serves as a useful frame on which to present our results, but also acknowledging the validity of the point reviewer #3 raises here, the passage will be edited to instead read:

**Lines 507 – 509:** *During MIS 3, proxy records suggest that conditions in the Prespa/Ohrid region were milder than MIS 4, but cooler and drier than MIS 5 (Fig. 7) (Panagiotopoulos et al., 2014; Sadori et al., 2016; Wagner et al., 2019)*

**Line 565:** Delete interglacial.

Sentence will be corrected to read:

**Lines 615 – 616:** *The timing and amplitude of  $Hg_T$  and  $Hg_{AR}$  signals recorded in Lake Prespa and Lake Ohrid sediments are noticeably different during the Holocene (MIS 1).*

**Lines 560-563:** This seems speculative. Do you have evidence of this in the records?

The reviewer is right in assuming this statement is speculative. It is unclear from our data why the transient Oldest (17.5-14.5 ka) and Younger (12.9-11.7 ka) Dryas climate events did not appear to leave measurable Hg signals in the sediments, and so here we sought to provide a potential explanation. However, something we agreeably did not sufficiently provide in our original submission, are details of how both Lake Prespa and Ohrid record clear indications of these cold events:

**Lake Prespa** → Core Co1215 records a clear, transient change in tree pollen concentrations corresponding to the Younger Dryas. Specifically, studies have revealed an increase in the abundance of cold-resistant open steppe vegetation, which was likely due to a net reduction in winter temperatures and moisture availability in the catchment (Panagiotopoulos et al., 2013; Aufgebauer et al., 2012). A transient shift in diatom flora also support this interpretation, reflecting a nutrient pulse linked to enhanced catchment erosion, lake-level reduction, and wind stress (Cvetkoska et al., 2014).

**Lake Ohrid** → Evidence for Younger Dryas-associated cooling has been identified in core 5045-1 in the form of low tree pollen percentages, low TOC concentrations, and high ( $^{234}U/^{238}U$ ) activity ratios –indicative of cold and dry local conditions, as well as deep hillslope erosion owing in a more open vegetation-type catchment structure (Francke et al., 2019). Short core JO2004-1 extracted from the southern part of the Ohrid basin also records a transient drop in calcite precipitation corresponding to the Younger Dryas, alluding to distinctly colder local temperatures (Lézine et al., 2010).

Inspired by reviewer #3's question, we will better integrate this evidence into the manuscript text as follows:

**Lines 602 – 612:** *Both lakes contain clear evidence for an abrupt return to glacial conditions during this time. Lake Prespa sediments record shifts in tree pollen and diatom assemblages alluding to a net reduction in local winter temperatures and moisture availability (Aufgebauer et al., 2012; Panagiotopoulos et al., 2013; Cvetkoska et al., 2014), and high uranium ( $^{234}U/^{238}U$ ) activity ratios, low tree pollen percentages, and low TIC concentrations in Lake Ohrid also pertain to intense hillslope erosion owing to a more open catchment structure (Francke et al., 2019; Lézine et al., 2010). Geomorphological evidence also pertains to local glacier stabilization (Gromig et al., 2018; Ribolini et al., 2018; Ruszkiczay-Rüdiger et al., 2020) (Fig. 7). Nonetheless, we suggest these events may have been too (a) short-lived, and/or (b) climatically mild to produce a similarly distinct response in the terrestrial Hg cycle as the processes operating during, and immediately following, the LGM; potentially explaining the lack of an associated sedimentary Hg signal.*