Please find below our detailed responses (in blue) to comments given by Reviewer #2, where the original reviewer comments are repeated here in black for clarity and completeness.

Reviewer #2

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This study examines long-term Hg records in sedimentary archives due to its sensitivity to centennial to millennial-scale environmental variations. Sediment analysis from two interconnected lakes, Lake Prespa and Lake Ohrid, over the past 90,000 years reveals distinct Hg patterns. Divergent Hg signals during the early and middle Holocene suggest that local factors significantly influence the Hg cycle's response to environmental changes, highlighting the role of sediment composition, lake structure, and water balance in determining the local versus global influences on Hg signals. It is a very interesting topic. This paper contains dense content and thorough analysis with well-written explanations. I am curious about whether the biota species are the same in both lakes, as this could be another factor impacting the differences in Hg records between these two bodies of water. Additionally, the layout of the paper could be improved, such as placing tables and figures at the end of the manuscript, which would enhance its readability and organization.

> We give many thanks to Reviewer #2 for their kind and constructive feedback on our manuscript and are thrilled they found it to be an interesting read. In the response below and in our revised manuscript, we will endeavor to ensure the questions raised are addressed, and alterations made where necessary.

Introduction

1. Line 35-44 In the first paragraph, I believe it would be good to emphasize the bi-directional pathway of Hg transportation. Hg can not only be emitted/released into the atmosphere but can also deposit into terrestrial and oceanic ecosystems.

   This is a good suggestion. To highlight the bi-directional nature of Hg cycling in the environment, we will add the following text to the manuscript:

   Lines 35 – 36: *Mercury (Hg) is a volatile metal released into the atmosphere, lakes, and ocean from both natural and anthropogenic sources, and actively cycled between surface reservoirs.*

   Lines 50 – 53: *Evasion back to the atmosphere, consumption by living organisms, or sequestration within aquatic sediment all represent ways in which Hg may 'leave' the terrestrial environment; the latter are known to be particularly effective sinks within the global Hg cycle (Bishop et al., 2020; Selin, 2009).*

   We believe this text would best fit into the second paragraph of the introduction (rather than the first), to ensure clarity for the reader, and ensure reviewer #2's suggestion was suitably integrated into the narrative flow.

2. Line 83, please remove this subtitle as there are no other subtitles in the Introduction section.

   Good point. This subtitle will be removed so the introduction text is presented as one single passage.

3. Line 112, it would be good to include the full name of Hg_{AR}, as this is the first instance of its mention in the manuscript.

   We thank reviewer #2 for pointing this out. This sentence will be revised to read:

   Lines 115 – 118: *… where (single) host-phase abundance or dilution cannot be easily accounted for,* ***Hg accumulation rate*** *(Hg_{AR}) may provide the most optimal assessment of Hg availability through time as long as a robust age model is available for the archive.*

4. Fig 2. Study map normally locates in "2. Site Description."

   Following this suggestion, **Figure 2** will be moved to **section 2**.

Site Description

5. Line 148 to 149, delete the dashed line.

   Good spot, this sentence will be revised to read:

   Lines 148 – 151: *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).*

6. Line 259 to 265, I recommend merging and simplifying this content with the information found between lines 207 to 211 and lines 215 and 217.

   Another good suggestion. Our initial decision to keep this information separate was in light of the fact that the secondary datasets mentioned here were not obtained as part of the same study, with different teams leading the data acquisition process. Thus, merging the data in our revised manuscript may compromise the clarity of this section. However, we do concur that this information could be presented more concisely, and so will shorten this paragraph by ~10 % through removal of superfluous wording.

7. Line 260 and 261, It appears that the same method is used to calculate TOC, but there are different references compared to lines 210 and 211. Is there a specific reason for this discrepancy?

   Yes. Sedimentological analysis of the two cores was carried out at the same institution (University of Cologne) and the method used to calculate TOC was the same. However, TOC values for Lake Prespa and Lake Ohrid were calculated and presented in separate studies:

   Prespa → (Aufgebauer et al., 2012) (~17–0 ka), and (Damaschke et al., 2013) (~90–0 ka)

   Ohrid → (Francke et al., 2016)(~600 – 0 ka)

   Separation of the two into discrete sections of the manuscript serves to acknowledge this difference, and the corresponding text has been edited to read:

   Lines 216 – 218: *TOC was calculated as the difference between TC and TIC by Aufgebauer et al. (2012) for the upper ~3.2 m, and by Damaschke et al. (2013) for the full ~17 m succession.*

   Lines 267 – 269: *The first dataset comprises TC and TIC measured using a DIMATOC 200 (TOC calculated as the difference between TC and TIC), and TS using a Vario Micro Cube combustion CNS elemental analyser at the University of Cologne - both by Francke et al. (2016).*

Section 3.3 Mercury measurements

8. Line 292 and 293, why use different resolution to analyze Hg sediment samples from these two lakes?

   Sampling and analysis of the two cores were done as separate studies, each with key differences that influenced the sampling strategy. For example, core Co1215 from Lake Prespa was recovered in 2007 (Wagner et al., 2010): to be directly compared to core Co1202 taken from Lake Ohrid (also in 2007) (Vogel et al., 2010), and subsequently facilitate a better understanding of interactions between the two lakes during the last glacial. Given the length of this core and associated chronological interval (<100-kyr), a finer sampling strategy was chosen. Conversely, the 5045-1 core was extracted during an ICDP drilling campaign in spring 2013 (SCOPSCO - Wagner et al., 2014). Although this core is the deepest, most complete paleoenvironmental record from Lake Ohrid currently available, the sampling strategy adopted by the SCOPSCO team was intended to cover the full ~1-Myr succession (16 cm per sample). This study focusses on the upper ~100-kyr of core 5045-1, and thus explains why the Hg record from this core is lower resolution than Co1215 (Prespa – 2 cm per sample).

9. Line 293 what is the size of the powered samples, homogenize of sediment samples are really important.

A good aspect of detail that should have been included in our original submission, and so we will add the following:

> **Lines 299 – 306**: *Samples were analysed for Hg_T at a resolution of ~2 cm for Lake Prespa, and ~16 cm for 5045-1 (see sections **3.1** and **3.2**). Approximately 2 cm^3 of sediment was homogenized to fine powder for TOC (previous studies) and Hg analyses (this study). Powdered samples were weighed into glass measuring boats, with masses ranging between 35–96 mg for Co1215, and between 27–78 mg for 5045-1. For samples particularly rich in inorganic fractions (e.g., samples coinciding with tephra layers), masses needed to be greater in order to yield a sufficiently high peak area (Lumex output) for calculation of sediment mercury concentrations: justifying the range in weights for both cores*.

10. Line 296 Could you provide information relating to percent recovery for the standard material?

We were not completely sure what reviewer #2 was alluding to here, and so we provide responses to all possible interpretations of this question, which is a valuable one to ask in both respects.

1) **How much Hg was recovered from the standard material relative to the certified values** → This value is difficult to ascertain based on our analyses alone. Nonetheless, we assume for % recovery to be equal to 100 % (or very close to). Issues with Hg recovery are known to be significantly less problematic in pyrolysis-based analyses (such as we use in this study) compared to those conducted using inductively coupled plasma mass spectrometry (e.g., laser ablation)(Bin et al., 2001), which generally emerge due to several factors:
   - Hg has a very high first ionization potential, resulting in low sensitivity as only ions (not atoms) are measured by ICP-MS.
   - Hg has seven naturally occurring isotopes; all <30% abundant. Because the total element concentration is divided among many separate isotopes, the number of ions (and therefore the sensitivity) is lower for each individual isotope.

   To ensure accuracy of total Hg measurements, we also routinely check the concentration of the standard used in this study with other NIST materials, each with certified Hg contents. For example, NIST 2782 (industrial sludge, 1100 ± 190 ng g^{-1}) and NIST 1944 (New Jersey waterway sediment, 3400±500 ng g^{-1}).

2) **The average % deviation of standard concentrations from their accepted value** → Across all analytical runs, over 95 % of standards yielded concentrations that were within ±20% of the expected value (here being 290 ng g^{-1}), and 68 % were within ±10 %. For both cores, standards with >20 % deviation (11 out of 184 total standard runs) were not used in calibration of the instrument. Details of standard measurements for each record are included as a supplementary Excel file (BGs) SUPPLEMENT_standard runs). In this spreadsheet, we include details of the total number of standards run, standard sample masses, measured Hg concentrations, the peak area derived from varying masses of standard, and the percentage deviation of calculated Hg concentrations from the expected value.

3) **Percentage core material recovered during drilling** → At the DEEP site of Lake Ohrid, six parallel holes yielded 1526m of sediment in total. Accounting for sediment–core overlap, the total composite field recovery amounts to > 95% (545 m). Full details are presented in Wagner et al. (2014), and this information will be added to the manuscript:

   > **Lines 253 – 255**: *Sediments below 1.5 m depth were recovered from six closely-spaced drill holes at the site in 2013 (5045-1A to 5045-1F), with a total composite field recovery amounting to > 95% (545 m); accounting for sediment–core overlap (Wagner et al., 2014).*

   Core recovery from Lake Prespa had not been published at the time of writing, and so we cannot provide a % value. However, we assume this was also high (>95 %) given the lack of any major gaps in sedimentation and/or disturbance of the core samples; likely due (at least in part) the undisturbed sedimentation at the Co1215 site inferred from hydroacoustic surveillance (Wagner et al., 2010).

11. Line 299 Could you please specify the exact table or figure that indicates the calibration results here?

    As above, details of standard runs for each record are included as a supplementary Excel file ((BGs) SUPPLEMENT_standard runs), and reference to this information will also be incorporated into the revised manuscript as the following statement:

    Line 314: *Details of standard runs for each core are included as a supplementary file.*

12. Line 303 I recommend removing the subtitle 3.3.1 since there are no other subtitles in this section.

    This is a fair suggestion and can understand why it was made here. However, we believe this subtitle serves an important purpose of guiding the reader through this part of the manuscript – as new formulae, data, and methods were introduced as part of our Hg_{AR} calculations. Hence, separating this information into a sub-section helps to guide the reader through our workflow more clearly.

13. Fig 4. The legend for MIS 3-5 in the figure is not easy to identify. It's up to your discretion whether to consider using different colors to improve clarity.

    We agree that the accessibility and clarity of this figure needed improvement; specifically, the presentation of data for MIS 3-5. We include details of our proposed changes, and a copy of the revised **Figure 4** below:

    MIS 5 – changed from circles to plus symbols. Colour changed to lilac.
    MIS 4 – changed from circles to triangles, with reduced transparency. Colour changed to light blue.
    MIS 3 – changed from circles to squares. Colour changed to navy blue.
    MIS 2 – colour changed to black.

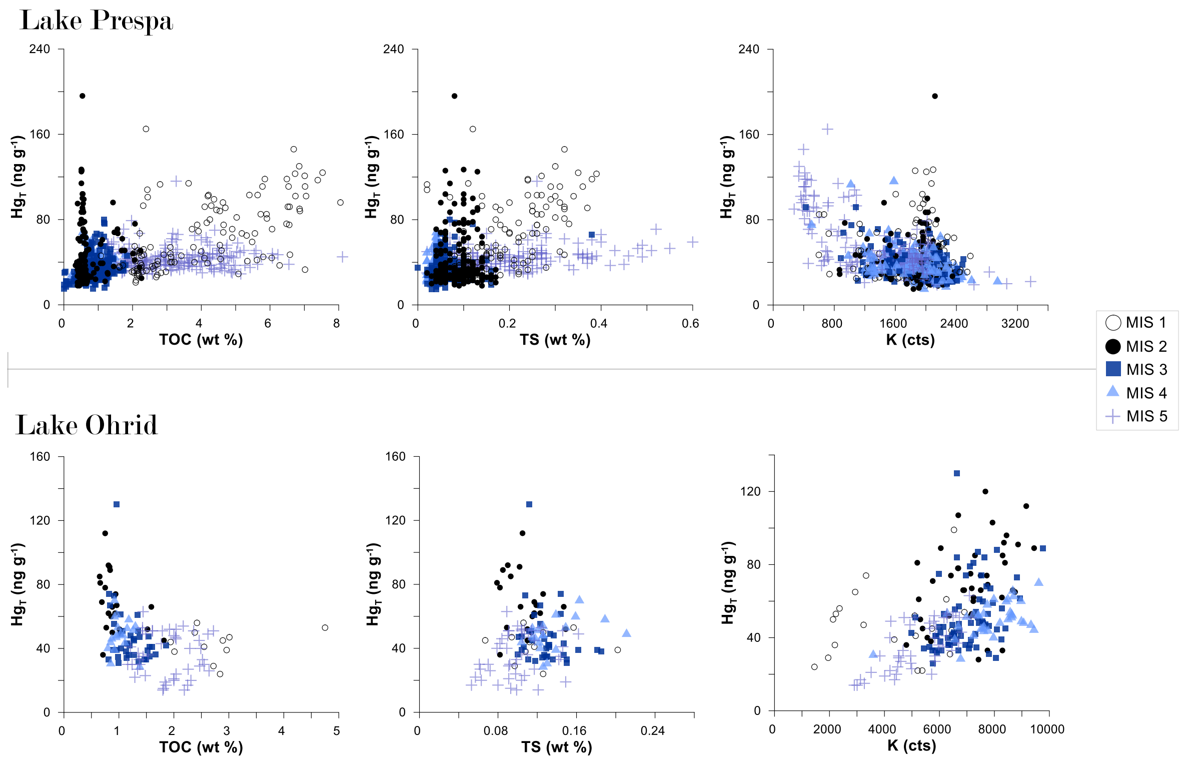

Figure 4: A comparison of host-phase relationships between lakes Prespa and Ohrid. Points are coded relative to stratigraphic period: the Holocene (12–0 ka, transparent circles), and the late Pleistocene (90–12 ka, filled symbols). We

compare Hg_T records for both lakes relative to total organic carbon (TOC), sulphide (estimated by total sulphur (TS)), and detrital minerals (estimated by potassium (K) concentrations) – note that aluminium (Al) data are more commonly used as an indicator of detrital mineral abundance, but these are currently unavailable for 5045-1.

14. Line 390, what is p-value for the relationship between Hg_T and TS for MIS 1?

The p value for the Hg_T/TS relationship in Lake Prespa for MIS 1 is 0.8534. To make p-values for both cores readily accessible to the reader but without adding more quantitative data to the main text, we propose adding a column into supplementary **Table S4**:

Table S4: Comparison of host-phase relationships (presented as the r-squared (r^2) value) between Lake Prespa and Lake Ohrid. r^2 values marked in bold/italic signal that the linear relationships observed between Hg_T, and each compound examined was negative.

Lake	Host	MIS	r^2 value	p-value
Prespa	Hg_T/TOC	1	0.3375	<0.0001
		2	0.0105	0.7858
		3	0.1053	<0.0001
		4	0.1381	<0.0001
		5	*0.0002*	*0.8559*
	Hg_T/TS	1	0.2511	<0.0001
		2	0.0007	0.8534
		3	0.056	0.0002
		4	0.0431	0.0132
		5	0.0751	0.0001
	Hg_T/K	1	*0.4418*	*0.7580*
		2	*0.0184*	*0.3531*
		3	*0.0024*	*0.4390*
		4	*0.031*	*0.0362*
		5	0.0109	0.1530
Ohrid	Hg_T/TOC	1	0.019	0.7580
		2	*0.1495*	*0.0006*
		3	*0.1477*	*0.0021*
		4	*0.0293*	*0.3256*
		5	*0.0004*	*0.8976*
	Hg_T/TS	1	*0.007*	*0.1277*
		2	*0.0367*	*0.2530*
		3	*0.0074*	*0.3750*
		4	0.1197	0.0417
		5	0.0805	0.0560
	Hg_T/K	1	0.0287	<0.0001
		2	0.1574	0.0005
		3	0.1403	0.0068
		4	0.3248	0.0004
		5	0.5239	<0.0001

15. I am wondering if biota species are the same between these two lakes Hg pool/accumulation, you have compared the hydrology, sedimentation regime, and geochemistry of them.

This is a great point. We posit that the link between Hg and biota in lakes Prespa and Ohrid exists as a function of their respective differences in bathymetric structure. In summary:

Lake Prespa → shallower (~14 m) waters host a dominantly mesotrophic (nutrient-rich) system where benthic and planktonic diatom species are present in equal abundance and allude to moderate/high biological productivity. We hypothesize that elevated productivity (inferred from the presence of these species) would typically favor more effective Hg scavenging by organic particles in Lake Prespa, and so could explain why the Hg/TOC relationship is notably stronger in this record compared to lake Ohrid.

Lake Ohrid → deep (~240 m) waters of Lake Ohrid host a highly oligotrophic (nutrient poor) environment characterized by low levels of biological productivity, and a high abundance of planktonic diatom species. These conditions would be less favorable for algal scavenging of Hg, and so could explain: (1) why the Hg in Lake Ohrid is inversely correlated to organic matter availability, and (2) why Hg signals better correspond to low-amplitude climate variability rather than transient disturbances.

The role of biotic processes as they relate to Hg could certainly have been described more explicitly within the manuscript; particularly **section 4.4**. Directly inspired by reviewer #2, we will add a paragraph detailing the aforementioned differences to **section 4.4**, with the concluding statement:

> **Lines 719 – 723**: *While the overall signal will remain dominated by Hg availability, broad-scale differences in productivity between lakes Prespa and Ohrid through time could provide an additional explanation for the disparate expression of recorded Hg signals (**section 4.1**); with notably higher productivity in the shallower Lake Prespa further increasing its sensitivity to changes in nutrient status, erosion, and hydrology.*

We will also supplement our interpretation of the Hg profiles with additional references to biological data earlier in the manuscript. For example:

- **Section 4.1,** where we consider the role of changing organic processes in creation and/or preservation of the Hg signals we observe in the two lakes:

  > **Lines 416 – 421***: On one hand, Hg signals could reflect changes in the dominant sources of organic and detrital materials deposited in the lake. For example, combined isotopic and sedimentological data record episodes of stronger algal blooms during MIS 1 and 5 (Leng et al., 2013), supported by coeval abundance of freshwater diatom genera such as Cyclotella and Aulacoseira (Cvetkoska et al., 2015). All correspond to elevated HgT, and so could imply more effective Hg burial by autochthonous organic material compared to allochthonous (Leng et al., 2013; Damaschke et al., 2013).*

- **Section 4.3**, where we draw upon biotic evidence for lower water levels in Lake Prespa during the LGM to propose why this lake records distinctly different glacial/interglacial signals:

  > **Lines 570 – 576***: One plausible explanation could be a disproportionately large change in Lake Prespa's total volume compared to Lake Ohrid, with implications for seasonal ice cover. Increased abundance of small Fragilariaceae and benthic Eolimna submuralis diatom species point to generally low temperatures and lake levels during MIS 2 (Cvetkoska et al., 2015), and are reinforced by elevated concentrations of coarse sand and gravel grains (IRD) alluding to persistent ice formation on the lake surface (Damaschke et al., 2013; Wagner et al., 2010).*

A full presentation of the biological character of each lake is beyond the scope of this study, although detailed descriptions of this nature are presented in the following publications - all of which are cited in our manuscript:

Cvetkoska, A., et al. (2018) Spatial patterns of diatom diversity and community structure in ancient Lake Ohrid. *Hydrobiologia* **819**, 197–215.

Cvetkoska, A., et al. (2016) Ecosystem regimes and responses in a coupled ancient lake system from MIS5b to present: the diatom record of lakes Ohrid and Prespa. *Biogeosciences* **13**, 3147–3162

Cvetkoska, A., et al. (2021) Drivers of phytoplankton community structure change with ecosystem ontogeny during the Quaternary. *Quaternary Science Reviews* **265**, 107046, https://doi.org/10.1016/j.quascirev.2021.107046

Jovanovska, E., et al. (2022) Environmental filtering drives assembly of diatom communities over evolutionary timescales. *Global Ecology and Biogeography* **31**, 954–967

Leng, M. J., et al. (2013) Understanding past climatic and hydrological variability in the Mediterranean from Lake Prespa sediment isotope and geochemical record over the last glacial cycle, *Quaternary Science Reviews* **66**, 123–136