Please find below our detailed responses (in blue) to comments given by Reviewer #1, where the original reviewer comments are repeated here in black for clarity and completeness.

Reviewer #1

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Paine et al. presents sedimentary Hg signals and associated processes from two lakes over the past 90ka. Overall, the data quality is high, and the manuscript is well written. It is interesting to see such different Hg signals at the same periods between two lakes just ca. 10km apart. I would like to provide some comments or seek for clarifications.

> We thank Reviewer #1 for taking the time to provide feedback on our manuscript, and for their kind words regarding its presentation and quality. In the response below and in our revised manuscript, we have sought to ensure the critiques raised are suitably considered and addressed where necessary.

Abstract:

Line 30. I would suggest adding "Hg sources" since different Hg sources also influence the local Hg cycles as discussed in the manuscript.

> Good point. We will alter this sentence to read:
>
> **Lines 29 – 31**: *The lack of coherence in Hg accumulation between the two lakes suggests that, in the absence of an exceptional perturbation, local differences in sediment composition, lake structure**, Hg sources**, and water balance all influence the local Hg cycle…*

Introduction:

The introduction section is nice! Paine et al. showed a clear summary diagram on different processes liberating/ mobilizing/ depositing Hg (i.e. Figure 1). I would still suggest adding a few lines on the sources of Hg to the lake in lines 45-53 or in other suitable paragraphs, to help readers better understand why Hg signals could be so different in lakes Prespa and Ohrid.

> We thank reviewer #1 for their kind words. We agree that this (highly relevant) contextual information could be more clearly presented in the introduction, and so will address this through revision of the following sentences:
>
> **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 59 - 62:** *Time-resolved sediment records sourced from marine and lacustrine basins are highly suitable for assessing these roles further back in time, as the Hg recorded may originate from one of several potential sources in the atmospheric (e.g., precipitation, dust), terrestrial (e.g., soils, detrital matter), aquatic, and/or lithospheric domain (**Fig. 1**).*
>
> Description of Hg sources is kept general in this section of the manuscript, with the purpose of equipping the reader with a broad overview of the state-of-the-art; prior to targeted discussion of lakes Prespa and Ohrid throughout the rest of the manuscript.

Line 86. How about changes in catchment? I would suggest adding a phrase about catchment in this sentence.

> This is an interesting suggestion, and we will add a statement including this suggestion to the previous paragraph (**lines 57 - 66**) to highlight the role local-regional scale processes may play, particularly for lacustrine records and refer directly to changes in the catchment. For the paragraph here we prefer to retain focus on the soft-sedimentary processes internal to the

lake that could influence Hg composition. To further clarify this, we will revise the following sentences:

Lines 89 – 92*: However, internal changes in bioproductivity, organic matter type and/or flux, sedimentation rate, pH, and redox conditions could all produce a distinct, local, transient, sedimentary Hg enrichment without a meaningful change in the total amount of Hg present and/or mobile in the system.*

Lines 95 – 97*: This impact is then removed, via normalization (e.g., Hg/TOC, Hg/TS), to reveal broader changes in environment Hg availability (Grasby et al., 2019; Percival et al., 2015; Shen et al., 2020; Them et al., 2019).*

Lines 97 – 99: *Such an approach is particularly beneficial for studies typically spanning >10^2-year timescales, where the goal is to isolate the effects of catchment-scale depositional and/or transport processes on Hg signals recorded in the sediment through time.*

Site Description:

Lines 143-146. Similar to the question above (i.e. line 86), how would changes of vegetation distribution in the catchment influence the Hg input to the lake sediment over different periods? Prespa lakes have a catchment of ca. 1300 km^2. I assume there is a significant Hg contribution from the catchment (e.g., through precipitation and then runoff). If not, please clarify.

This is a good question, and one that draws on a critical component of this study. The topic of catchment influences on the Hg signals recorded in the lake(s) is introduced in **section 1**, and then considered with direct reference to lakes Prespa and Ohrid in **section 4.4** of this manuscript. To define this structure more explicitly, we will add the following statement to the beginning of **section 4**:

Lines 373 – 376: *We first consider the extent to which soft sediment processes (**section 4.1**) and lithological features (**section 4.2**.) may have influenced the Hg variability observed in **Figures 5** and **6**, before adopting a catchment-scale perspective in **section 4.3** to explore the role of diverse environmental processes in Hg cycling through these two systems.*

Lines 170. Does runoff from catchment belong to the category of "direct precipitation (35%)"?

No, catchment-sourced run-off falls under the category of 'river runoff'. But we do agree this requires clarification, and so will adjust the wording of the preceding sentence as follows:

Lines 174 – 175*: Water input is sourced from surface runoff (56%), direct precipitation (35%), and inflow from the smaller of the two lakes.*

Chronology:

Lines 220-239. The chronologies are very well established! But I would still suggest briefly mentioning the analytical methods of ^{14}C, $^{40}Ar/^{39}Ar$, and ESR in this section or in the supplementary materials, even though some relevant references are already cited. This can provide some pedagogical information to readers on age reconstruction using different techniques.

We agree that full disclosure of chronological details is important in this study. However, full description of these details in the main text may add unnecessary 'bulk' to the manuscript. We will follow the reviewer's suggestion to include a bit more detail on this in the SI text, and two short statements will be added to refer the reader to these methods in the SI:

Lines 242 – 243: All twenty-seven tie-points and accompanying chronological details are presented in **Text SI3** and **Table S3**

Lines 293 – 294: *Full description of the 5045-1 chronology and associated methods are presented in **Supplementary Text SI4.***

We will also make the following additions to the supplementary file:

(1) Three new sub-sections to our accompanying supplementary file:

SI3.1. Existing data & methods

SI3.2. A new Co1215 chronology

SI4.1. Existing data & methods

(2) Two new tables to our accompanying supplementary file, which present important information related to ESR dating of the *Dreissena* shell layer in core Co1215 (Prespa) as first presented in (Damaschke et al., 2013):

Table S1: Radionuclide contents of bulk sediment samples presented in (Damaschke et al., 2013). All analysis was done by high-resolution gamma-ray spectrometry with samples K-5835 and K-5836 (2011) measured at the Cologne lab, and sample K-5800 (2009) analysed at the VKTA lab, Dresden. All errors represent the 1σ level. For dose rate calculation the mean values were used.

Lab Code	U (ppm)	Th (ppm)	K (%)
K-5835	3.59 ± 0.19	17.57 ± 1.02	2.37 ± 0.09
K-5836	3.53 ± 0.19	17.99 ± 1.03	2.38 ± 0.09
K-5800	3.80 ± 0.40	17.30 ± 0.60	2.30 ± 0.07

Table S2: Parameters for dose rate calculation, total dose rates, equivalent dose values and ESR ages presented in (Damaschke et al., 2013). All errors represent the 1σ level.

Sample	Depth (m)	U (ppm) [shells, ICP-MS]	Dose Rate (Gy kyr^{-1}*)	Equivalent Dose (Gy)	ESR Age (ka)
K-5835	14.70 – 14.88	0.06 ± 0.01	1.36 ± 0.10	93.71 ± 2.03	68.9 ± 5.1
K-5836	14.58 – 14.70	0.06 ± 0.01	1.36 ± 0.10	114.41 ± 6.73	84.1 ± 7.8
K-5800	14.58 – 14.63	0.08 ± 0.01	1.36 ± 0.10	100.2 ± 11.2	73.9 ± 9.9

* Calculation of dose rates includes the following parameters and assumptions: alpha-efficiency value 0.10±0.02, thickness before and after surface etching 0.82/0.75 mm, an average water content of 47±4.7% (weight water/wet sediment), calculation of the cosmic dose contribution is based on the actual sampling depth including additional shielding through a water column.

The chronologies for both cores also do integrate age estimations for discrete, often independently dated tephra layers. In **Table S6,** we already provide references for the ages given for each of these layers, to ensure the reader can easily access the source publications and their corresponding associated analytical set-ups. This data is also provided in an accompanying spreadsheet.

Mercury Accumulation:

Lines 316-318. Are the methods to calculate sedimentation rates and dry bulk density for Lake Ohrid the same as the ones for Lake Prespa?

Sedimentation rate (SR) → Yes, the only difference was that two different research teams did these calculations. For Lake Prespa, SR was calculated and presented as mm yr^{-1}, whereas for Lake Ohrid SR values were presented as cm yr^{-1}. This meant that different calculations were required to convert these values to m kyr^{-1} for use in this study.

Dry bulk density (DBD) → No, but only because analyses were conducted by different studies/research teams. For Lake Ohrid, density data were directly measured by Francke et al. (2016), whereas for Lake Prespa we calculated DBD using TOC and water content data obtained by Aufgebauer et al. (2012) and Damaschke et al. (2013). For our calculations, we assumed an average wet density of 1 g cm^{-3} for wet sediments, and 2.6 g cm^{-3} for dry sediments.

To clarify these methodological details in the manuscript text, we will make the following addition to section 3.4:

> **Lines 323 – 324**: *Sedimentation rate (SR) values for both Prespa and Ohrid were calculated by combining stratigraphic and lithological observations with the age-depth relationship ascertained for each core, respectively.*
>
> **Lines 326 – 328:** *DBD values were calculated on the basis of sedimentological data available for each core. For the Lake Ohrid dataset, DBD values were already available following the analyses of Francke et al. (2016). To acquire these values for Lake Prespa, we employed the formula…*
>
> **Lines 332 – 333**: …assuming an average wet density of 1 g cm^{-3} for wet sediments, and 2.6 g cm^{-3} for dry sediments.

Results and Discussion:

Lines 380-381 and Figure 4. Hg_T concentration appears consistently high during MIS2 in both Lake Prespa and Ohrid. Where does the Hg come from?

Good observation, and one we highlight as a key defining feature of the presented records. This section focusses solely on the soft-sediment processes that may be influencing the Hg concentration of the two records; after the Hg has reached the lake. So, rather than changing the wording of this sentence to introduce different Hg sources (and potentially compromise overall clarity), in our revised manuscript we will better guide the reader through this narrative flow by adding the following statement to **section 4**:

> **Lines 373 – 376***: We first consider the extent to which soft sediment processes (**section 4.1**) and lithological features (**section 4.2**.) may have influenced the Hg variability observed in Figures 5 and 6, before adopting a catchment-scale perspective in **Section 4.3** to explore the role of diverse environmental sources and processes in Hg cycling through these two systems.*

Lines 393-397. This explanation is quite superficial, even though it makes sense. I would suggest going deeper to find evidence to explain it a bit more. For example, (1) how did the catchment shift regarding vegetation? (2) what can shift the rates of Hg emissions and/or exchange between surface reservoirs? Hg loss by reduction of Hg^{2+} in lake ecosystems can be very important (e.g., by photoreduction, Jiskra et al., 2021. https://doi.org/10.1021/acsearthspacechem.1c00304)

Omission of information/discussion related to catchment-scale processes from this section is deliberately done in order to maintain a clear narrative structure. We hope that the inclusion of the statement referenced above clarifies this reasoning, which we agree could have been more clearly provided in our original submission. The reviewer also raises some very valid points regarding the strength of our interpretation as written, and we agree that this could benefit from additional supporting evidence. In light of this comment, we will edit the passage in focus as follows:

> ***Lines 416 – 429****: … 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* Hg_T*, and so could imply more effective Hg burial by autochthonous organic material compared to allochthonous (Leng et al., 2013; Damaschke et al., 2013). However, in the presence of abundant availability of binding ligands such as for the Lake Prespa record, maximum Hg burial is limited principally by supply regardless of productivity, and so changing Hg signals in Lake Prespa more likely reflect changes in environmental Hg availability; resulting from externally-driven oscillations in Hg emission and/or exchange between (local) surface reservoirs such as forests, water courses, and soils (Bishop et al., 2020; Obrist et al., 2018)). This interpretation is supported by the lack of a close statistical correspondence between Hg, organic matter, sulphur, or detrital mineral content, source, or composition (**Fig. 4**), which suggests that Hg burial efficiency is only weakly associated with host phase availability in this system.*

Regarding point (1), the influence of vegetation changes will be discussed in greater detail in section 4.4. For example:

> **Lines 489 – 493***: High organic and low clastic material concentrations point to warmer climate conditions during this interval, in which both catchments experienced an increase in moisture availability, pronounced*

forest expansion, and plant diversification – collectively acting to stabilize hillslopes and reduce deep soil erosion (Francke et al., 2019; Panagiotopoulos et al., 2014; Sadori et al., 2016, 2016).

Concerning point (2), we agree that evasion also should have been given more sufficient credence in our original submission. In light of this suggestion, we will integrate more explicit mention of this process at several points in our revised manuscript. For example:

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).*

Lines 457 – 461: *In the absence of a pronounced host-phase influence, retention of a measurable Hg signal requires that the net influx of Hg into the lake (e.g., surface runoff, wet/dry deposition) exceeds the amount leaving the system due to processes such as runoff or evasion. Therefore, we surmise that the Hg_T and Hg_{AR} signals recorded in Lake Prespa and Lake Ohrid are records of net Hg input to the two lakes rather than the efficiency of sedimentary drawdown.*

It is possible that photoreduction did influence the Hg composition of the Prespa and/or Ohrid sediments. For example, the photoreduction of divalent Hg to gaseous Hg (more susceptible to evasion) is controlled mainly by dissolved organic matter (DOM) in aquatic settings (Luo et al., 2017; O'Driscoll et al., 2018), and several Holocene-age lake and peat cores show evidence for increased evasion of Hg corresponding to sediments enriched in organic matter and/or containing evidence for heightened productivity. Collectively, this could point to more effective Hg photoreduction under warm climatic conditions (Jiskra et al., 2015; Schaefer et al., 2020; Biester et al., 2018; Hermanns et al., 2013), but potentially also more in Lake Prespa where Hg variability is more closely correlated to TOC variability. However, we are limited in our ability to constrain this flux for either of the two lakes, and further speculation on climate-driven differences in Hg loss by photoreduction may not be a valuable/useful addition to our revised manuscript:

(1) **The extent to which photoreduction has influenced the Hg composition of a sediment record is best distinguished through use of stable isotopes, which were not measured in this study**. Isotope analysis can provide crucial indications of whether photoreduction of divalent Hg to gaseous Hg has occurred, most commonly by identification of odd-mass Hg stable isotope anomalies (e.g., Δ^{199} and Δ^{201} Hg), (Kurz et al., 2019; Jiskra et al., 2022). Given this data currently are not available for lakes Prespa and Ohrid, we cannot confidently distinguish the mechanisms by which Hg is removed from these systems beyond simple speculation; nor the extent to which these mechanisms influenced the observed Hg composition.

(2) **Studies in which isotopic methods have been applied are nearly all limited to the Holocene (<11 ka)**. The most robust estimates of terrestrial Hg evasion are mainly based on enriched isotope tracing studies of recently (<10^2-year) deposited sediments (Obrist et al., 2018); many of which are focused primarily on the marine environment (Soerensen et al., 2016; Jiskra et al., 2021; Horowitz et al., 2017). This means that there are currently few studies that seek to quantify the influence that climate shifts associated with glacial-interglacial conditions may have exerted on Hg evasion and/or photoreduction. Even fewer explore this in the terrestrial realm; limiting the applicability of present understanding to the records presented here.

There remains much to learn about the ways in which Hg loss may affect the signals retained in lake sediments on multi-millennial timescales. However, despite the aforementioned uncertainties, we do fully concur with reviewer #1's point that this component of the Hg cycle is important and well worth further study; especially in older, pre-Holocene records such as those from lakes Prespa and Ohrid. Directly inspired by this comment, we will also add reference to the potential of Hg isotope analysis in this research domain in the closing **section 4.4**:

Lines 743 – 746: *Intra-basin heterogeneity in Hg sources, reactions, and transformations could also be examined through measurement of stable Hg isotopes; particularly in millennia-scale sedimentary records* *where the nature of these processes may change through time (Blum et al., 2014; Jiskra et al., 2022; Kurz et al., 2019).*

Lines 535-554. It is not clear to me why Hg accumulation profile in Lake Prespa spanning 10 ka from 33 to 23 ka is much flatter than the one in Lake Ohrid. Why isn't Hg accumulation elevated in Lake Prespa as the one in Lake Ohrid during this period? Does it link to the shallow characteristic of Lake Prespa or limited Hg input?

We have split out this comment to answer the two specific queries (see also below). This certainly warrants further clarification both here and in our revised manuscript. At the core of our interpretation are the differences that exist between the two lakes in terms of bathymetric structure, catchment characteristics, and the extent to which both of these were affected by glaciation. Independent evidence has shown Lake Prespa to be significantly more sensitive to transient climate change than Lake Ohrid (Jovanovska et al., 2016), and so **section 4.3** seeks to explore how this differing sensitivity could also influence Hg cycling. Reviewer #1 rightly points out that we have omitted to mention a key (and intriguing) feature of the two profiles: the distinct lack of a Hg_{AR} signal during the LGM in Lake Prespa. First, we will include a more direct reference to this feature:

Lines 566 – 570: *Lakes Ohrid and Prespa show another two striking differences in Hg composition between 35–12 ka (**Fig. 7**). First, Lake Prespa does not record a distinct Hg_T or Hg_{AR} signal during the LGM, and second, Lake Ohrid does not record a distinct Hg_T or Hg_{AR} signal corresponding to deglaciation. Given their close proximity and environmental similarity, both lakes could be expected to record similar overall signals if the climate-driven processes influencing Hg_{AR} were broadly similar.*

This will be followed by a more explicit discussion of the potential reasons underpinning the observed signals; discussion directly inspired by the questions raised in this review:

Why isn't Hg accumulation elevated in Lake Prespa as the one in Lake Ohrid during this period?

Lines 570 – 580: *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…It is possible that the heightened presence of ice at the peak of glaciation served as a natural barrier between the surface and the sediments of Lake Prespa, effectively slowing the net flux of Hg into delivery of solutes to the basin. A simultaneous lack of ice cover on Lake Ohrid could also justify why Hg_{AR} remained high in this lake during the LGM, as the Hg influx pathway would be unaffected by ice formation (**Fig. 7**).*

Lines 581 – 594: *Volume changes may have also influenced the hydrological connection between lakes Ohrid and Prespa during deglaciation (Cvetkoska et al., 2016; Jovanovska et al., 2016; Leng et al., 2010)…For Lake Prespa, a measurable change in lake volume would reduce the number (and pressure) of active sinkholes, and subsequently the outflow of water and solutes (e.g., Hg) into Lake Ohrid(Wagner et al., 2014) – increasing both Hg_T and Hg_{AR}.*

Does it link to the shallow characteristic of Lake Prespa or limited Hg input?

Lines 594 – 600: *Together, the collective impact of disproportionately large, climate-driven reductions in water level could explain why rates of Hg accumulation were significantly higher in Lake Prespa during deglaciation compared to the LGM. Glacial meltwaters would elevate the net Hg input compared to the LGM, and reduced ice cover would permit a more direct pathway for Hg to be delivered into the basin; both processes becoming effective while underground permafrost continued to limit the intra-basin exchange of water and solutes.*

Key differences and implications

The whole section is overall well written, but it is lack of some interpretation on Hg loss from my perspective. Hg loss can be very different between these two lakes, therefore affecting the net Hg signals in the sediments. I would suggest adding a few lines on this information to make your interpretation more convincing.

We thank the reviewer for their kind critique, and equally for giving constructive pointers from which to improve this section of the manuscript. As discussed above, we are cautious to put precise constraints on the extent to which different processes may have influenced Hg loss from the two lakes (e.g., evasion, photoreduction). Nonetheless, we concur that Hg loss is a vital component of lacustrine Hg cycling that warrants mention in this section, and so we will make the following text additions:

> **Lines 692 – 697:** *Increased distance from lake margin to core site means distribution of material over a greater total area, and thus increased potential for net Hg loss either by evasion from the water surface (Cooke et al., 2020), removal of water (and suspended material) via riverine outlets (Bishop et al., 2020), or processes taking place within the water column (Frieling et al., 2023). Therefore, preservation of a measurable Hg signal in a deep lake (e.g., Lake Ohrid) would require a sufficiently large influx of Hg…*

This will add to the interpretations made in the preceding section (**4.4**), where we already speculate on the potential for loss of Hg from Lake Prespa as a result of transport through the karst system underlying the two lakes:

> **Lines 587 – 589**:*…decreases in the reconstructed $\delta^{18}O$ of lake water and TIC in both lakes during the last glaciation point to a reduction in the contribution of karst-fed waters to Lake Ohrid (Lacey et al., 2016; Leng et al., 2013).*

> **Lines 592 – 594**: *For Lake Prespa, a measurable change in lake volume would reduce the number (and pressure) of active sinkholes, and subsequently the outflow of water and solutes (e.g., Hg) into Lake Ohrid (Wagner et al., 2014) – increasing both Hg_T and Hg_{AR}.*

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