Please find below our detailed responses (in blue) to comments given by Reviewers #1 and #2, where the original reviewer comments are repeated here in black for clarity and completeness.

# **REVIEWER #1**

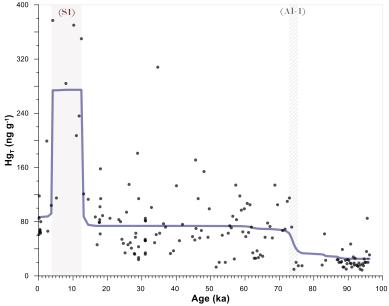
Paine and coauthors present a new Hg record in a unique sedimentological archive from Lake Bosumtwi that records West African hydroclimate over the last 96 thousand years. The study is partly honing and improving upon the Hg proxy for paleoclimate reconstructions and partly discussing hydroclimate history of the region at various timescales. The study is well executed and very well written, but the impact of the new record is a bit limited by the uncertainty in Hg driving mechanisms, but also the pre-existing reconstructions of lake level that may be more powerful for the paleoclimate framing. Please find comments, technical edits, and suggestions for improving the figures below.

We sincerely thank reviewer #1 for taking the time to provide feedback on our manuscript, and for their kind words regarding its presentation. In the response below and in our revised manuscript, we outline how we will ensure their comments and suggestions are thoroughly considered and addressed where necessary.

## Moderate comments:

The split at 73 ka should be tested with statistical tools such as change point analysis to see how significant it is, rather than a visual analysis. Much of the discussion hinges on the differences before and after this time, so it should be bolstered by significance testing.

This is a great suggestion, and so we have proceeded to carry out the change point analyses on our record. To test the significance of the shifts in Hg<sub>T</sub> we identified in the manuscript, we used Paleontological Statistics software (PAST) v.4.16 to apply a change point analysis function to the BOS04-5B Hg<sub>T</sub> data (Hammer et al., 2001). The results of this analysis are shown below, and will be incorporated into the accompanying supplementary information file.



**Figure S3**: The average changepoint model for BOS04-5B displayed as a purple curve, superimposed onto the original Hg<sub>T</sub> values. The abruptness of the curve indicates the extent to which the MCMC simulations (n = 1000000) agree on the changepoint positions, where greater smoothing indicates greater variance between simulations. Unit AI-1 is marked between 33.5 and 32.8 m depth (grey shading; Brooks et al., 2005; Scholz et al., 2007), and sapropel layer Unit S1 is marked between 3–5.5 m depth (brown shading; Shanahan et al., 2012, 2006).

We will also add the following information to our revised submission:

(1) A description of statistical methods employed by our study in section 3:

## Line 391: "3.6. Statistical analyses

Two statistical analyses were used in order to more quantitatively explore the timing, and expression of signals recorded in the BOS04-5B HgT dataset. First was a simple linear Pearson's correlation analysis, from which correlation coefficients (*r*) were calculated to indicate the direction and strength of the association between Hg (n = 157), and a suite of geochemical proxies also measured in the core. Second was a change point analysis, to determine whether distinct changes in mean values of HgT occur within the record using PAST v.4.16 software (Hammer et al. 2001). This software employs a Bayesian Markov chain Monte Carlo (MCMC) approach, which was run on default settings with a total of 1 million MCMC simulations were run for each test and a maximum of <10 changepoints. The extent to which similar processes influenced the concentration of HgT, TOC, K, and detrital matter were also explored, and correlations were subdivided based on visual examination of the HgT records (and supported by changepoint analyses). The significance of all correlations were significant at p<0.01."

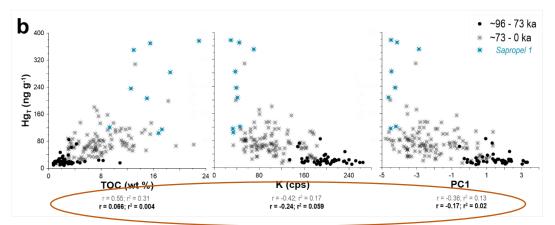
(2) Explicit reference to the changepoint analyses in section 4:

**Line 516**: "The magnitude and frequency of variability in Hg<sub>T</sub> visibly increases at ~73 ( $\pm$ 5) ka (**Fig. 4**). The quantitative significance of this shift is supported by changepoint analysis of the BOS04-5B dataset, which demonstrates a clear and step-wise shift in mean Hg<sub>T</sub> values between ~75 and 73 ka (**Fig. S3**). It also occurs in conjunction with an increase in the lake's water level (**Fig. 4b**),"

Line 542: "This unit contains clear Hg⊤ enrichments relative to the rest of the core (Fig. 2, 4a, S3)..."

3) A clear statistical test of the relationship between Hg<sub>T</sub> and the first principal component (PC1) of the BOS04-5B XRF data (proxy for lake level; McKay, 2012) both prior to, and following ~73 ka (see below).

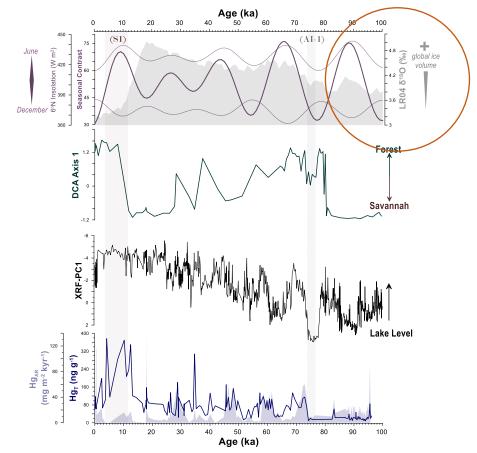
4) Explicit listing of r-values in **Figure 3b:** to further clarify the change in significance and sign of correlation of Hg, TOC, detrital matter, and lake level relationships following ~73 ka.



**Figure 3: (b)** Comparison of relationships in Lake Bosumtwi between ~96 and 73 ka (black circles), and between ~73 and 0 ka (stars). We first assess the HgT record for this lake relative to two potential host-phases: total organic carbon (TOC) values measured in this study, and detrital minerals (estimated by potassium (K)) concentrations measured by McKay (2012). We also test the relationship between HgT and first principal component (PC1) of the BOS04-5B XRF data, in which 39% of total variance is associated with terrigenous elements. PC1 was consequently interpreted as an indicator of lake level changes (McKay, 2012). R (r) and r-squared ( $r^2$ ) values for each interval are also given. The significance of all correlations were assessed using a Student's t-test, which showed that all three combinations were significant at p<0.01. Stars marked in teal correspond to deposition of sapropel unit 1 (S1) in BOS04-5B.

The pattern in Hg and pollen look a lot like ice volume...

A more explicit comparison of our record with a proxy for global ice volume would certainly add valuable context to our discussion. To do this, we will add a record of benthic foraminiferal calcite  $\delta^{18}O$  (‰) derived from the LR04 global stack to **Figure 4**, where cold glacial stages characterized by high ice volume are defined by high  $\delta^{18}O$  ratios.



**Figure 4:** Comparison of key proxy datasets. Included are (from bottom to top), total mercury (Hg<sub>T</sub>) and mercury accumulation rate (Hg<sub>AR</sub>) for Lake Bosumtwi from this study, chosen as the most appropriate proxies for Hg variability in this core (see **section 5.1**). The first principal component (PC1) of the BOS04-5B XRF data (39% of total variance) is strongly associated with terrigenous elements, and so interpreted as an indicator of lake level changes (McKay, 2012). Forest (woody) taxa abundance (presented as DCA Axis 1; Gosling et al., 2022a; Miller et al., 2016). Lack of data for woody taxa presence is assumed to imply a savannah-dominated regional landscape. Insolation at 6°N (location of Lake Bosumtwi) in June (summer) and December (winter) are calculated following the astronomical solution presented by Laskar et al. (2004) (accessed via. <u>https://vo.imccce.ft/insola/earth/online/earth/online/index.php</u>), and used to calculate seasonal insolation contrast at the Bosumtwi site since ~100 ka. Also shown is a record of benthic foraminiferal calcite  $\delta^{18}O$  (%) derived from the LR04 global stack) as a proxy for ice volume, with cold glacial stages defined by high  $\delta^{18}O$  ratios (Lisiecki and Raymo, 2005). Proxy data are all presented on the BOSMORE7 chronology. Unit Al-1 is marked between 3.5 and 32.8 m depth (grey shading; Brooks et al., 2005; Scholz et al., 2007), and sapropel layer Unit S1 is marked between 3–5.5 m depth (brown shading; Shanahan et al., 2012, 2006).

Broad coherence between data for Hg<sub>T</sub>, pollen, and ice volume is (perhaps unsurprisingly) clearest following the last glacial termination, which coincides with both the African Humid Period and transition into the Holocene interglacial. Global ice volume is intrinsically linked to North Atlantic sea-surface temperatures (SSTs), and the relationship between these temperatures and West African hydroclimate has been discussed in several studies relevant to the Lake Bosumtwi record (e.g., Gosling et al., 2022b; McKay, 2012; Shanahan et al., 2009), including those relevant to both age models that currently exist for the BOS04-5B core (e.g., Gosling et al., 2024). This work has shown that low global ice volumes typically correspond to warmer North Atlantic SSTs and subsequently to moister conditions at Lake Bosumtwi: conditions that appear to also favour higher sedimentary Hg<sub>T</sub> values. We agree that the valuable context offered by the LR04 stack should be included more explicitly in our manuscript. We will add these details to the manuscript as follows:

Line 195: "During the last glacial cycle, moisture availability in West Africa also fluctuated in conjunction with the waxing and waning of high-latitude ice sheets, and their effects on seasurface temperatures (SSTs) in the North Atlantic (deMenocal, 1995; Weldeab et al., 2007). This teleconnection exists as a function of atmospheric moisture transport and convection processes occurring in the polar and tropical regions. Low global ice volumes typically correspond to warmer North Atlantic SSTs, driving increased atmospheric moisture transport and hence more moist conditions in West Africa. Conversely, high global ice volumes generally correspond to cooler SSTs in the North Atlantic, and subsequently drier conditions in West Africa (e.g., Crocker et al., 2022; Lupien et al., 2023; Stager et al., 2011; Tjallingii et al., 2008)."

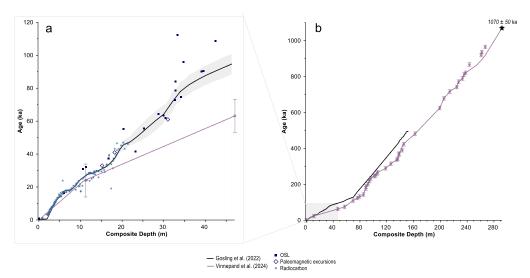
Line 521: "Furthermore, changing sedimentary TOC, terrigenous material, and pollen concentrations all corroborate a broad increase in local moisture availability, temperature, and humidity following deposition of the AI-1 unit (Fig. 4): a signal that coincides with the transition into the warmer Holocene interglacial, marked by reduced global ice volume and increased sea surface temperatures in the North Atlantic (McKay, 2012; Scholz et al., 2007; Shanahan et al., 2008b)."

...and perhaps this should be discussed in the context of the age model. The record should be presented using the newest published version of the core's age model from Vinnepand 2024. This will provide more detail throughout, and despite the lower resolution Hg data around this 73 ka time, may help pin down when this shift is, potentially in the broader context of drivers like ice volume.

While Vinnepand et al. (2024) present the most recent iteration of the BOS04-5B chronology, the resolution of tie-points used in their age-depth model in the interval of this study is very limited (n=2). The BOSMORE7 chronology used in our work utilizes a combination of radiocarbon (calibrated <sup>14</sup>C; n= 109), optically stimulated luminescence (OSL; n=22) and uranium-thorium (U/Th; n=5) dates as independent tie-points (Shanahan et al. 2013). All are concentrated within the youngest section of the core (see figure below). The application of multiple, independent, direct dating techniques to a single core succession continues to provide valuable chronological constraints for a growing number of long, Pleistocene-age sedimentary sequences (e.g., Roberts et al. 2018; Stockhecke et al. 2016). Here, this approach provides age control that is entirely independent from assumptions about past environmental conditions, and unaffected by any age uncertainty stemming from inter-site correlations; such as with the LR04 composite stack (Lisiecki and Raymo, 2005).

The lower resolution of the Vinnepand et al. (2024) chronology creates substantial discrepancies with other age-depth models in the upper ~47 m of the record. This not unsurprising given that the aforementioned study did not include <sup>14</sup>C ages in their model generation, given their aim was to generate an age-depth relationship for a much larger (and thus older) core section (~292 m) than that in our study (~47 m): "*Note that we do not plot* <sup>14</sup>C *ages (Shanahan et al., 2013) considered by Gosling et al. (2022) as these are not comparable to our study given the temporal application limits of 14C dating.*" (**Fig. 9** caption in Vinnepand et al. 2024; p. 11). However, this smoothing is also problematic. First, because it does not account for the high degree of variability in Lake Bosumtwi's sedimentation regime during the Late Pleistocene (e.g., Shanahan et al. 2013; McKay, 2012). Second, because it is offset from the absolutely dated BOSMORE7 chronology by >10-kyr for sediments >30 m depth. An offset that, in combination with ~10-kyr uncertainties for each of the three tie points in the upper 50 m of the core, substantially reduces the overall precision of the model with respect to our study interval (see **Fig. X** below).

We therefore feel the BOSMORE7 model (Gosling et al. 2022) remains more appropriate, because it explicitly includes absolute dating points and their uncertainties within the here studied interval. Whereas, the Vinnepand et al. model will be a key study for the deeper part of the record where few of those constraints are available. We have illustrated the difference between the two models in **Figure X** below.



**Figure X**: Comparison of age-depth models constructed for the BOS04-5B core by Gosling et al. (2022) (black line), and Vinnepand et al. (2024) (purple line). Panel (a) focusses on the core section relevant to this study, equating to 0 - 47 m of the composite depth. Panel (b) shows this section relative to the full, ~292 m-long BOS04-5B core succession. A black star marks the base of the crater, where  ${}^{40}$ Ar/ ${}^{39}$ Ar dating of shocked quartz yields an age of 1.08±0.04 (Jourdan et al. 2009). Data for the Vinnepand et al. (2024) model are plotted as presented in the supplementary information file presented with the source paper.

Nonetheless, although we retain the chronology as in our original work, we will add reference to the work of Vinnepand et al. (2024) in our manuscript main text:

Line 223: "The upper ~47 m of sediment corresponds to the interval ~96 ka to present, and contains a series of distinct lithological features suggesting pronounced, climate-driven changes in lake level, catchment structure, and sediment transport processes (Vinnepand et al. 2024; Gosling et al., 2022a; McKay, 2012; Miller et al., 2016)."

Line 143: "These changes all correspond to moisture-driven oscillations between a forest and grassdominated catchment in response to insolation-driven variability in WAM strength, and migration of the Intertropical Convergence Zone (ITCZ) (e.g., Vinnepand et al. 2024; Gosling et al., 2022a; Miller et al., 2016; Peck et al., 2004)"

We will also add more explicit justification for use of the BOSMORE7 chronology to our supplementary information file:

### S3. Chronology

"...In 2024, Vinnepand and colleagues used cyclicity in total natural gamma ray (NGR) data to create a cyclostratigraphic age-depth model for the full (~946-kyr) BOS04-5B core. This model will be key for study of the deeper (>200 ka) core sections where fewer absolute age markers are available (Shanahan et al. 2013). However, it is significantly lower resolution than the BOSMORE7 chronology in the upper ~47 m of the record, given that Vinnepand et al. (2024) did not include <sup>14</sup>C ages in their model generation. Not only does this limit the extent to which it can account for the high variability in Lake Bosumtwi's sedimentation regime during the Late Pleistocene (e.g., Shanahan et al. 2013; McKay, 2012), but it also creates a >10-kyr offset from the BOSMORE7 chronology for sediments >30 m depth. An offset that, in combination with ~10-kyr uncertainties for each of the three tie points in the upper 50 m of the core, substantial discrepancies. Therefore, given that the BOSMORE7 model explicitly includes absolute dating points and their uncertainties within the here studied interval, it provides age control for our data that is entirely independent from assumptions about past environmental conditions, and unaffected by any age uncertainty stemming from inter-site correlations."

## Minor comments:

I suggest adding a sentence or rephrasing the last sentence of the abstract to have a broader outlook on Hg as a proxy for hydroclimate moving forward. In the same vein, I may suggest starting the introduction more focused on the importance of reconstructing hydroclimate, rather than on Hg – and the second paragraph of the intro, too, means that this paper is focusing on the Hg proxy development, rather than understanding Bosumtwi hydroclimate. The mix of both directions in this paper is certainly a strength, but the goal of pinning down the Hg proxy is ultimately to understand hydroclimate.

We thank the reviewer for showing great interest in our work, and equally for suggesting the potential for using Hg as a hydroclimate proxy that we could explore with our record. It is an angle we certainly believe is worth pursuing in the future, given how our results show that Hg deposition may include a substantial hydrological component under the right circumstances, such as in Bosumtwi. However, it is uncertain how widely this may be applicable, to what extent Hg may directly reflect hydrology in lake sequences, and/or if hydrological signals are imprinted through certain catchment processes.

- There is a need for better characterization of the processes impacting Hg cycling in lacustrine sediments on millennial timescales, and the ways in which Hg sources, reactions, and transformations within the sedimentary environment could change in time and space (e.g., Frieling et al., 2023; Tisserand et al., 2022; Kovács et al., 2024).
- It is not yet fully clear which factors may pre-determine the sensitivity of a lake system to climate-driven perturbations in Hg cycling. Recent research has shown that geographically and/or structurally similar basins can record different sedimentary Hg signals in response to the same modes of environmental change (e.g., Paine et al., 2024), yet existing long sedimentary Hg records have typically focused on last glacial-age successions (<65 ka; e.g., Sahoo et al., 2023; Wang et al., 2024), or been limited to a single geographical domain (e.g., the Arctic; Gleason et al., 2017).

As a result, in this manuscript, we are cautious to not over interpret the data and present Hg deposition as a proxy for millennial-scale hydro climatic change. We do acknowledge that, provided the hydrological component of the Hg cycle can be isolated, it may present some opportunity to explore this as a proxy for hydrological changes in records such as that obtained from Bosumtwi. For example, measurement of Hg isotopes (e.g.,  $\delta^{200}$ Hg and  $\delta^{204}$ Hg) in sediments obtained from low-latitude and/or closed lakes could be used to quantify the contribution of Hg to the sediment from precipitation or dry deposition, relative to terrestrial remobilization of Hg stored in plants and soils (Blum et al., 2014; Yin et al., 2024). This would be promising work for a future study.

We will highlight these areas for future investigation in the manuscript in section 6:

Line 654: "Provided the hydrological component of the Hg cycle can be isolated, better characterization of the processes impacting lacustrine Hg cycling could also allow this element to be used as a proxy for hydroclimatic change in terrestrial archives. For example, measurement of sedimentary Hg isotopes in low-latitude and/or closed lakes could quantify the contribution of Hg to the sediment from precipitation or dry deposition, shedding new light on key biogeochemical reaction pathways, processes (e.g., mass-independent fractionation (MIF)), and responses to changing local hydrology across a range of timescales (e.g., Blum et al., 2014; Gao et al., 2023; Yin et al., 2024)."

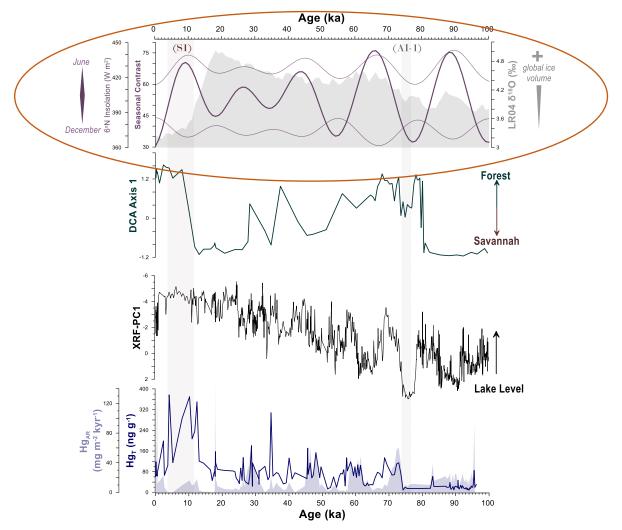
**Figure 4**: no need to show precession, and consider including key insolation curves such as 20 or 30 N and perhaps the insolation gradient (23 N - 23 S) and 65 N to test for drivers of hydroclimate throughout the late Pleistocene

We agree that precession is somewhat redundant here, and so will remove this curve from **Figure 4**. However, we also agree that insolation gradient is contextually important to our study as a key factor controlling precipitation distribution in tropical sub-Saharan Africa. With this in mind, we will add three new curves for 6°N (the latitude of Lake Bosumtwi) to **Figure 4**:

**Summer (June) insolation**  $\rightarrow$  higher values typically correspond to wetter summers in tropical sub-Saharan Africa, as higher summer insolation increases surface heating, atmospheric convection, and ITCZ strength.

Winter (December) insolation  $\rightarrow$  lower values typically correspond to drier winters, owing to weaker atmospheric convection and reduced precipitation potential.

**Seasonal contrast between June and December insolation**  $\rightarrow$  larger seasonal contrast typically indicates stronger monsoonal dynamics, as the difference in heating between



summer and winter drives the atmospheric pressure gradients that control monsoon circulation. An effect of this would be more pronounced wet seasons.

**Figure 4:** Comparison of key proxy datasets. Included are (from bottom to top), total mercury (Hg<sub>T</sub>) and mercury accumulation rate (Hg<sub>AR</sub>) for Lake Bosumtwi from this study, chosen as the most appropriate proxies for Hg variability in this core (see **section 5.1**). The first principal component (PC1) of the BOS04-5B XRF data (39% of total variance) is strongly associated with terrigenous elements, and so interpreted as an indicator of lake level changes (McKay, 2012). Forest (woody) taxa abundance (presented as DCA Axis 1; Gosling et al., 2022; Miller et al., 2016). Lack of data for woody taxa presence is assumed to imply a savannah-dominated regional landscape. Insolation at 6<sup>o</sup>N (location of Lake Bosumtwi) in June (summer) and December (winter) are calculated following the astronomical solution presented by Laskar et al. (2004) (accessed via. <u>https://vo.imcce.fr/insola/earth/online/earth/online/index.php</u>), and used to calculate seasonal insolation contrast at the Bosumtwi site since ~100 ka. Also shown is a record of benthic foraminiferal calcite  $\delta^{18}$ O (%) derived from the LR04 global stack) as a proxy for ice volume, with cold glacial stages defined by high  $\delta^{18}$ O ratios (Lisiecki and Raymo, 2005). Proxy data are all presented on the BOSMORE7 chronology. Unit Al-1 is marked between 3-5.5 m depth (brown shading; Shanahan et al., 2012, 2006).

Line 65: is it precipitation pattern? Or precipitation amount/strength? Or something else?

We agree the previous phrasing of this sentence was nonspecific. We will address this by amending the sentence as follows:

**Line 69**: "For example, changes in precipitation <u>amount</u> can influence the proportion of Hg removed from the atmosphere by wet versus dry deposition..."

Line 94: perhaps start with a sentence that is a bit more focused for this study

We agree this sentence (as was previously written) was overly broad. With this in mind, we will remove this sentence so that the paragraph starts as follows:

**Line 97**: "In the short-term, variability in hydroclimate may manifest as annual changes in rainfall intensity and seasonality, or by sub-decadal fluctuations in regional-scale climate modes (e.g., El-Nino Southern Oscillation, North Atlantic Oscillation; Hernández et al., 2020)."

#### Line 120: what is its domain?

Well spotted, this sentence does not make full sense. We will amend this to read:

**Lines 122-124**: "In Sub-Saharan Africa, the West African Monsoon (WAM) regulates precipitation amount and distribution, and drives long-term evolution of environmental characteristics and mineral-dust emissions..."

Line 122: perhaps add some more citations for orbital control of the WAM seen by leaf waxes, like from O'Mara and Kuechler

This is a good suggestion. Given the relevance of these studies to the point made here, they will be incorporated into the manuscript as follows:

**Lines 122-125:** "In sub-Saharan Africa, the West African Monsoon (WAM) regulates precipitation amount and distribution, and drives long-term evolution of environmental characteristics and mineral-dust emissions (O'Mara et al. 2022; Kaboth-Bahr et al., 2021; Kuechler et al. 2013; Weldeab et al., 2007)."

Line 124: I suggest changing drought events to arid periods, if these records are focused on orbitalscale climate variability

Good point, and we thank reviewer #1 for pointing out where different terminology is required. We will adjust this sentence to read:

Lines 125-128: "Proxy records from this domain show that orbitally-driven variations in the strength of the WAM have frequently driven distinct arid (Cohen et al., 2007; Scholz et al., 2007) and humid periods (Armstrong et al., 2023; Menviel et al., 2021) throughout the Pleistocene."

#### Line 129: over what time period?

We agree more specificity is needed here. Sentence will be amended to read:

**Lines 133-136**: "Here our focus is on sediment core BOS04-5B extracted from Lake Bosumtwi, Ghana (West Africa): a core that provides a clear and continuous record of this hydroclimate variability covering the late Pleistocene..."

**Line 161**: maybe add one sentence or phrase about how this makes the Lake Bosumtwi archive ideal for refining the Hg-paleoclimate method or generating paleoclimate reconstructions in general

We agree this is key knowledge to include and indeed discussed this specifically in our original manuscript in two dedicated paragraphs:

- (1) In **section 1.2**, where we outline our research objectives and thus highlight the suitability of Lake Bosumtwi for this purpose
- (2) In **section 2**, where we dedicate a full section that outlines why this sedimentary record is well suited to fulfil our research objectives:

## 2.1.3. Paleoclimatic significance

We will carefully revisit these during revisions to ensure all information is included, easily accessible, and add a specific reference to studying Hg behaviour. For example, the addition of the following sentence to our introduction:

**Lines 170-174**: "These properties render the lake highly sensitive to changes in atmospheric processes, but also imply that Hg inputs may originate exclusively from the atmosphere (e.g., by wet deposition). Therefore, this system is ideally suited for exploring whether specific basin characteristics (e.g., depth, nutrient status, bathymetry) could also measurably affect how Hg signals are encoded in the sedimentary record..."

**Line 163**: perhaps just leave out this short-term definition, as only seasonal changes are described. Unless these vary on 10 year timescales? The description goes from less than 10 to over 1000 year variability.

Fair point, we will remove the short-term definition so the sentence reads:

**Line 177:** "Seasonal variability in the tropical rain belt position drives short-term hydroclimate change in West Africa."

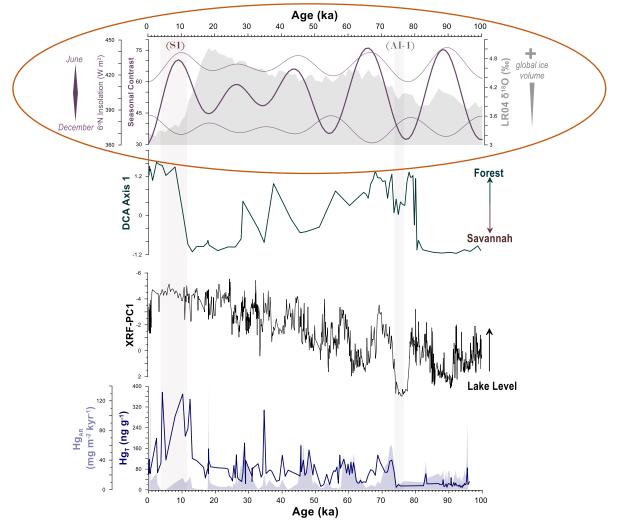
**Section 2.1.1**: is the ITCZ also expanding and contracting? Rather than just moving and strengthening/weakening? Also, there is more evidence that the insolation gradient, rather than just precession, is controlling the WAM during the Pleistocene (see O'Mara et al 2024)

Both great points, and we agree that the complexities associated with West African climate during the Pleistocene should be given more detail in this section. To better describe this complexity, we will edit the following sentences to read:

**Lines 185-187**: "Changes in axial precession produce fluctuations in seasonal insolation above the African continent, influencing the strength of the WAM, the Walker Circulation, the position and dimensions of the ITCZ, and the availability of continental moisture..."

**Lines 188-191**: "Several studies have shown weakening of the WAM and southward migration of the ITCZ in response to high precession and/or changes in insolation gradient, producing drier conditions in West Africa and subsequent reductions in terrestrial precipitation, ecosystem productivity, and recession of terrestrial forests (O'Mara et al. 2022; Menviel et al., 2021)."

Reviewer 1 also makes a great point regarding the importance of insolation, and we will directly respond to this by adding insolation curves for summer (June), winter (December), and overall seasonal contrast at 6°N (the latitude of Lake Bosumtwi) to **Figure 4** (see below). Together, these three curves will provide a more comprehensive picture of insolation variability at our study site during the Late Pleistocene, and so give us opportunity to underscore the importance of orbital forcing for variability in precipitation over long timescales in this domain.



**Figure 4:** Comparison of key proxy datasets. Included are (from bottom to top), total mercury (Hg<sub>T</sub>) and mercury accumulation rate (Hg<sub>AR</sub>) for Lake Bosumtwi from this study, chosen as the most appropriate proxies for Hg variability in this core (see **section 5.1**). The first principal component (PC1) of the BOS04-5B XRF data (39% of total variance) is strongly associated with terrigenous elements, and so interpreted as an indicator of lake level changes (McKay, 2012). Forest (woody) taxa abundance (presented as DCA Axis 1; Gosling et al., 2022; Miller et al., 2016). Lack of data for woody taxa presence is assumed to imply a savannah-dominated regional landscape. Insolation at 6<sup>o</sup>N (location of Lake Bosumtwi) in June (summer) and December (winter) are calculated following the astronomical solution presented by Laskar et al. (2004) (accessed via. <u>https://vo.imcce.fr/insola/earth/online/earth/online/index.php</u>), and used to calculate seasonal insolation contrast at the Bosumtwi site since ~100 ka. Also shown is a record of benthic foraminiferal calcite  $\delta^{18}$ O (%) derived from the LR04 global stack) as a proxy for ice volume, with cold glacial stages defined by high  $\delta^{18}$ O ratios (Lisiecki and Raymo, 2005a). Proxy data are all presented on the BOSMORE7 chronology. Unit Al-1 is marked between 3-5.5 m depth (brown shading; Shanahan et al., 2012, 2006).

**Lines 221-224**: Nobody will disagree with this statement, but an additional clause indicating why lake level alone is not a perfect measure of precipitation will lead well into the study of Hg.

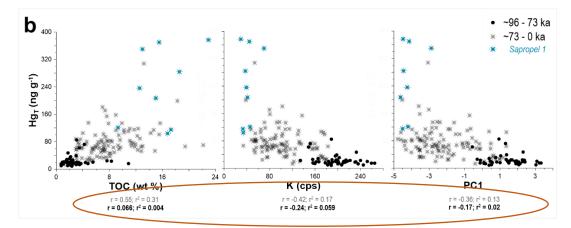
Good suggestion. We will add the suggested clause onto the end of this paragraph as follows:

Lines 244-245: "The closed hydrology of Lake Bosumtwi means that changing water levels will primarily reflect the magnitude of precipitation variability in the region, with higher lake levels typically occurring during wetter climate intervals (Russell et al., 2003; Shanahan et al., 2008b). However, lake level may have also been influenced by secondary processes such as evaporation, and, over long time-scales, sediment infill (McKay, 2012)."

I recommend reporting correlation as r, rather than r<sup>2</sup>, so we can see numerically the direction of correlation.

**Figure 3**: as mentioned above, if presented as r rather than  $r^2$ , there won't be a need for the italics and it'll be clearer.

We concur that presenting r values (in addition to  $r^2$ ) provides useful additional information regarding the relationships between the variables. To display this information more clearly, we will add r-values to the scatter plots presented in **Figure 3**, alongside the  $r^2$  values:



**Figure 3: (b)** Comparison of relationships in Lake Bosumtwi between ~96 and 73 ka (black circles), and between ~73 and 0 ka (stars). We first assess the HgT record for this lake relative to two potential host-phases: total organic carbon (TOC) values measured in this study, and detrital minerals (estimated by potassium (K)) concentrations measured by McKay (2012). We also test the relationship between HgT and first principal component (PC1) of the BOS04-5B XRF data, in which 39% of total variance is associated with terrigenous elements. PC1 was consequently interpreted as an indicator of lake level changes (McKay, 2012). R (r) and r-squared ( $r^2$ ) values for each interval are also given. The significance of all correlations were assessed using a Student's t-test, which showed that all three combinations were significant at p<0.01. Stars marked in teal correspond to deposition of sapropel unit 1 (S1) in BOS04-5B.

And when correlations are presented, please include number (n) and significance (p).

We agree this information is critical for presentation of our results, and thank the reviewer for pointing this out. This also aligns with a comment made by reviewer #2. With these comments in mind, to present statistical information in sufficient detail, and ensure our results are fully transparent, we will make the following amendments:

- Addition of p-value information to the caption and main body of Figure 3 (see above)
- Addition of both r and r<sup>2</sup> values to relevant points in the manuscript. For example:

**Lines 432-437**: "An overall positive association between  $Hg_T$  and TOC (r = 0.64;  $r^2 = 0.42$ ) suggests that Hg variability may be associated with organic carbon variability in Lake Bosumtwi. However, it is noteworthy that detrital materials (e.g., K) show negative correlations with both TOC (r = -0.73;  $r^2 = 0.53$ ) and Hg (r = -0.60;  $r^2 = 0.34$ ) so that the Hg-TOC correlation may reflect, in part, a correlation imposed by variable clay-dilution of both Hg and TOC. Moreover, these correlations are all significant at p < 0.001 (unless stated otherwise)."

**Line 447**: "A negative overall correlation between  $Hg_T$  and K is apparent throughout the record (r = -0.60;  $r^2 = 0.34$ ; **Fig. 3b**)."

- Addition of a new sheet to our supplementary data file, which will contain three tables: (1) r values, (2) t-squared statistics, and (3) p-values for all for all geochemical combinations analysed, for which it is not practical to include in the figure or text. This sheet is titled: "Pearson's Correlation"
- Addition of a new section to our supplementary file that provides a more detailed description of the correlation analyses applied to BOS04-5B:

# **S8. Correlation analyses**

- Amendment of the **Figure 3a** caption to include values for (n) and (p):

"Full-core correlation (Pearson's r) matrix for Hg, total organic carbon (TOC) (this study), and a suite

of trace elements measured in BOS04-5B by XRF (McKay, 2012). Higher r values suggest that similar processes influence the concentration of the two elements in focus). Sample size (n) was 157 for each analysis, and ~75% of the assessed trace element combinations were significant (p<0.01). The 25% that were not are marked with an asterisk (\*)."

**Line 517**: some records show a drying trend towards the LGM, particularly in eastern Africa (Garelick, Baxter, Lupien, Tierney). Please discuss how this fits into your discussion, if the focus is as transcontinental as it is now.

Interesting observation. We agree that the "*broader, regional-scale shift in hydroclimate across sub-Saharan Africa*" we mention in our discussion should have been explained in a bit more detailed context. We also thank reviewer #1 for referring us to key papers in this context. With these comments in mind, we will amend the paragraph to read:

Lines 566-578: "Proxy data generated from the BOS04-5B core suggest that progressively wetter conditions affected the catchment following ~73 ka (e.g., Gosling et al., 2022a; Shanahan et al., 2008b). Paleoclimate records based on the sediments of lakes Malawi (Tanzania), Bambili (Cameroon), Tanganyika (Tanzania/Democratic Republic of the Congo), Chew Bahir (Ethiopia) and Chala (Tanzania) (e.g., Cohen et al., 2007; Foerster et al., 2022; Lézine et al., 2019; Scholz et al., 2007), and marine sediment core material from the West African margin (Figs. 1, 5) (e.g., Kinsley et al., 2022; Skonieczny et al., 2019) also pertain to a distinct regional-scale shift in hydro climate across tropical sub-Saharan Africa at this point in time (Fig. 5). Specifically, a shift characterized by a distinct moisture gradient favouring wetter conditions in the west of the continent relative to the east, which was further amplified during the last glacial termination (e.g., Gosling et al. 2022a; Baxter et al. 2023; Lupien et al. 2022). Therefore, the coeval increase in the frequency and amplitude of Hg enrichments in Lake Bosumtwi, and associated rises in lake level, could indirectly reflect pronounced shifts in hydroclimate across tropical sub-Saharan Africa."

Lines 526-527: this is a narrow statement – see a nice explainer of fuel-limited and moisture-limited by Karp 2023

We agree this statement is overly simple. We will re-write this sentence as follows and include the recommended citation:

**Lines 579-589**: "Hydroclimate was a key driver of changes in fire activity in tropical sub-Saharan Africa during the late Pleistocene. However, although wetter climatic conditions may be broadly associated with heightened fire activity due to associated increases in terrestrial biomass, recent work has shown that discrete changes in precipitation can elicit notably different fire responses between sites (e.g., Karp et al. 2023; Gosling et al., 2021; Moore et al., 2022). The influence of biomass burning on the Hg record presented here appears similarly complex; despite being a well-constrained factor in the Bosumtwi catchment, and evidence that wildfires are also a significant source of Hg, accounting for ~13% of natural Hg (re-)emissions to the modern atmosphere (Francisco López et al., 2022). Given that no clear relation is visible between Hg<sub>T</sub>, Hg<sub>AR</sub>, and two discrete macro- (Kiely, 2023) and micro- (Miller et al., 2016) charcoal profiles generated from the BOS04-5B core, we suggest that the effects of Hg emitted during wildfires did not leave a clear imprint on Hg variability in this record (**Fig. S9**)."

### Technical:

### Lines 63-64: remove one of the 'direct's

These two sentences will be edited to read:

**Line 67**: "The transport and transformation of Hg at the Earth's surface is linked to the hydrological cycle (Bishop et al., 2020; Selin, 2009). Water plays a direct role in the efficiency of both Hg deposition and reemission."

#### Line 148: change Ma to Myr-old, or "dated to 1.08 Ma"

Sentence will be edited to read:

**Line 154**: "It occupies a meteorite impact crater dated to  $1.08 \pm 0.04$  Ma, which is one of the youngest and best preserved impact craters on Earth..."

### Line 230: cite the map in Fig 1

Good suggestion. Reference to Figure 1 will be added as follows:

Line 254: "Our study focuses on the upper ~47 m section of a 296-m-long core extracted from deepwater (76 m) site 5 (core BOS04-5B; **Fig. 1**)..."

Line 240: changed generated to described or similar

We will edit wording of this sentence to read:

**Line 273**: "Age control for the ~47 m of sediment analysed in this study is provided by the BOSMORE7 model, presented by Gosling et al. (2022)."

Line 256: might help to include the maximum spacing between samples too

Agreed. This sentence will be amended to:

**Lines 288-290**: "For this study, we analysed 165 samples spanning the composite depth interval 47.7 to 0 m, with an average temporal resolution of ~0.6 ka between each sample (range: 0.01 to 5.85 kyr)."

Line 261-262: can simply cite the supplemental data, rather than including a sentence

The full sentence will be deleted, and we will amend the previous sentence as follows:

Line 295: "and then one standard for every 10 lacustrine samples (supplementary data)."

**Lines 383-385**: instead of talking directly about Figure 2, try removing the first part of this sentence and then citing Figure 2 at the end.

We agree this longer reference to Figure 2 adds unnecessary bulk to the sentence. From this, the sentence will be edited as follows:

Line 429: "Two mechanisms emerge as plausible drivers of Hg variability in Lake Bosumtwi (Fig. 2)."

**Line 386**: I don't think this last sentence of the paragraph is necessary – there are a lot of these statements throughout that are a bit redundant that can be cut down

We agree with this point, and so will amend this paragraph so that the sentence in question is removed:

Lines 426-430: "Studying time-resolved changes in lake sediment Hg concentration provides a valuable opportunity to study changes in the pre-industrial Hg cycle, how these changes translate to measurable sedimentary signals, and their links to local and regional-scale environmental variability (Cooke et al., 2020). Two mechanisms emerge as plausible drivers of Hg variability in Lake Bosumtwi (**Fig. 2**). First is organic matter (host) availability, and second is external change in net Hg input to the system."

We will also carefully revisit the entire text to eliminate any other superfluous statements.

Line 516-517: "these records" needs clarification aside from the figure citation.

The section being referenced here will be amended so to read:

Lines 568-573: "Paleoclimate records produced from the sediments of lakes Malawi (Tanzania), Bambili (Cameroon), Tanganyika (Tanzania/Democratic Republic of the Congo), Chew Bahir (Ethiopia) and Chala (Tanzania) (e.g., Cohen et al., 2007; Foerster et al., 2022; Lézine et al., 2019; Scholz et al., 2007), and marine cores extracted from the West African margin (**Figs. 1, 5)** (e.g., Kinsley et al., 2022; Skonieczny et al., 2019) also pertain to a distinct regional-scale shift in hydro climate across sub-Saharan Africa at this point in time (**Fig. 5**)."

# Line 526: citation

We will make the following amendment to this opening statement:

**Lines 579-580**: "Hydroclimate was a key driver of changes in fire activity in sub-Saharan Africa during the late Pleistocene (Moore et al. 2022)."

Sections 5.2.1 and 5.2.2: I suggest renaming these sections into more descriptive titles

We agree that the original section headers are overly vague, and would benefit from being more descriptive. With this in mind, we will adjust the section headers to read:

# 5.1.1. Arid conditions (~96 to 73 ka)

# 5.1.2. Humid conditions (~73 to 0 ka)

Line 539: change ka to kyr

The text will be corrected to read:

Line 594: "The resolution of the BOS04-5B record (~0.6 kyr per sample)..."