

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.

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## REVIEWER #2

This manuscript uses a sediment core collected that dates to 96k to link Hg to climatic events in West Africa. The manuscript is well written, and the methods and QA/QC are clearly explained.

We give earnest thanks to reviewer #2 for their kind and constructive feedback on our manuscript. In the response below and in our revised manuscript, we will endeavor to ensure the edit suggestions are considered and addressed, with alterations made where necessary.

I would remove the word new from the title.

Fair suggestion, we agree this word is superfluous and so will remove it from the main manuscript and the supplementary file. Our manuscript will subsequently be titled:

**“Evidence for millennial-scale interactions between Hg cycling and hydroclimate from Lake Bosumtwi, Ghana”**

There do not appear to be any keywords. These should not be the same as words in your title.

The following keywords will be added to the manuscript below the abstract:

**Line 36:** “**KEYWORDS:** lacustrine, mercury, geochemistry, sediment, sapropel, Lake Bosumtwi, Africa”

**Line 111:** There is a colon in this line that should not be there.

The colon will be removed so the sentence reads:

**Lines 112-115:** “*However, few terrestrial Hg records extend beyond the present interglacial (>12 ka), and even fewer come from the low-latitudes, where tropical rainforest, grassland and desert biomes are highly sensitive to millennial-scale hydroclimate variability (Bradley and Diaz, 2021; Schneider et al., 2023)*”

**Line 128:** focusses is not correct use focus

We agree the grammar of this sentence could be improved and we will revise this sentence to read:

**Lines 134-136:** “*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...*”

The section 1.3 titled West African Monsoon should be changed from this section basically introduces the reader to the research objectives. Do you have a research hypothesis?

We thank the reviewer for highlighting this discrepancy. Considering this comment, we have revisited how to best structure the introduction and made some adjustments, particularly in section headers to improve the flow and better guide the reader to our research objectives. For example, we like the sub-title suggestion made by reviewer 1 here, and so the title of this section will be edited to now read:

**Line 120:** “**1.2. Research objectives**”

Further, we will carefully revisit the introduction so the research hypothesis – **changes in hydroclimate can affect how mercury (Hg) is transported and buried in terrestrial sediments** – is clearly presented. This hypothesis was purposefully broad in order to provide a solid baseline for assessing the presence (or absence) of any Hg-climate relationships in the

Bosumtwi record using both observational and statistical methods; while also accounting for the fact that a widely accepted framework for how the terrestrial mercury (Hg) cycle responds to millennial-scale climate changes, and how these responses vary by location, is still lacking. To this end, we will also incorporate the following statement into our revised manuscript:

**Lines 130-133:** *"In light of growing evidence for a hydroclimatic influence on the terrestrial Hg cycle (e.g., Guédron et al., 2018; Nalbant et al., 2023; Paine et al., 2024), we hypothesized that humid and/or arid periods in sub-Saharan Africa would have elicited measurable changes in the Hg cycle, producing measurable signals in regional sedimentary records."*

**Line 139:** I would get rid of aims to assess and just use "assessed"

This sentence will be revised to read:

**Lines 144-146:** *"Focussing on the uppermost ~47 m of the Lake Bosumtwi sediment record, this study assesses whether major shifts in local hydroclimate produced measurable changes in how Hg has been transported to, and buried within, this system since ~96 ka"*

**Line 142:** explore should be explored. Note: I am a strong believer that what was done should be described in past tense.

Well spotted. Sentence will be amended to read:

**Lines 147-149:** *"By comparing our sedimentary Hg record with proxy data from archives across the African continent (e.g., Foerster et al., 2022; Scholz et al., 2007), we explored whether hydroclimate has exerted a measurable effect on terrestrial Hg cycling in the WAM domain in over the past ~100-kyr."*

WAM and ITCZ should be written out the first time used with the abbreviation afterwards in parenthesis.

Agreed, and we will ensure we present these terms with the appropriate abbreviations the first time they are used in the manuscript:

**Line 122:** *"...the West African Monsoon (WAM)..."*

**Line 143:** *"...Intertropical Convergence Zone (ITCZ)"*

**Line 217:** which should be that, and there should be no comma preceding this

Sentence will be amended to:

**Lines 238-240:** *"The core also shows distinct co-enrichment in manganese (Mn) and iron (Fe) in certain intervals following AI-1, that are associated with manganosiderite (Mn-rich FeCO<sub>3</sub>) precipitation in the lake sediments"*

**Line 230:** which should be that. Which is used when indicating something general, while that is used when discussing something specific.

We agree this is the grammatically correct form, and so should have been used here. We will correct this sentence to read:

**Lines 254-256:** *"Our study focuses on the upper ~47 m section of a 296-m-long core extracted from deep-water (76 m) site 5 (core BOS04-5B), that extends from the present-day lake floor to the brecciated bedrock dated by <sup>40</sup>Ar/<sup>39</sup>Ar to 1.08±0.04 Ma (Jourdan et al., 2009)"*

**Line 260:** run should be analyzed

Agreed. Our wording will be corrected:

**Lines 294-295:** *"...with a known Hg value of 290 ± 9 ng g<sup>-1</sup> were analysed to calibrate the instrument before sample analysis, and then one standard for every 10 lacustrine samples"*

Please explain in the methods how the core was stored and sampled. It is very easy to contaminate samples with Hg. This must be clearly addressed.

We agree details on core handling and storage are useful to include and will add the following details to the manuscript in section **3.1. BOS04-5B**:

**Lines 262-270:** “After drilling in 2004, core BOS04-5B was shipped to the University of Rhode Island and split. The physical properties of the full ~296 m core were measured at 2-cm intervals using a Geotek® multi-sensor core logger (Koeberl et al., 2007). After logging and imaging and at 4-cm resolution, 2 cm thick slices were removed from the core half and separated into sub-samples for multi-proxy analyses, including sediment magnetic hysteresis, x-ray diffraction mineralogy, total organic and inorganic carbon content, bulk organic carbon and nitrogen isotopes, grain size, pollen, and charcoal (e.g., Gosling et al., 2022; McKay, 2012; Miller et al., 2016). Following bulk sediment analyses, the BOS04-5B core material was transferred to the Continental Scientific Drilling (CSD) Repository in Minneapolis.”

We also more clearly refer the reader to additional details related to the ICDP coring operation during which core BOS04-5B was extracted which are included in our supplementary information, under the section:

### **S1. BOS04-5B core details**

**Line 276:** which should be that. I am not going to mention any more of these and will leave this up to the authors to correct.

We thank reviewer #2 for flagging this grammatical error, and have carefully checked for other instances throughout the manuscript. The sentence in focus will be corrected to:

**Line 309:** “Both aliquots were then combusted in oxygen at 1220°C to break down the calcium carbonate and produce carbon dioxide (CO<sub>2</sub>), that was fed into a solution of barium perchlorate.”

We will also go through the full manuscript, and amend **14** instances where we identified this error was present.

**Line 364:** “broadly track” is subjective. It would help if you did some statistical analyses to make this statement more quantitative. Same with the discussion line 365.

**Line 369:** some statistical analyses would help quantify the discussion.

**Line 389:** ah here is the statistical analyses result. Please move up to the results section. Note:  $r^2$  values are meaningless without a p-value. Is it worth putting in the equation for the correlation? This might be of interest to others. Don't forget to put your statistical analyses method in the methods section.

In response to the three comments above: we agree that **section 5.1** (as-was) should be in the results section of the manuscript. Inspired by this comment, we have moved this section such that it is now:

#### **4.1. Lacustrine host phases of mercury**

With this change, the correlation matrix and its associated information will also be moved to the results section.

We also agree that statistical analyses are a powerful tool for ensuring the statements made in our discussion are based on quantitative evidence. Equally, we agree that more details of our statistical analyses need to be included in the methods section of our manuscript, and this also aligns with a comment made by reviewer #1. In light of this suggestion and to present statistical information in sufficient detail, we will make the following amendments:

- All correlation analyses in text or figures are now accompanied by  $r$ ,  $r^2$ , and p-values. 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).”

**Figure 3:** (a) 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 (\*). Grey shading marks positive correlations (light:  $>0.25$ , dark:  $>0.5$ ), and orange shading marks negative correlations (light:  $<-0.25$ , dark:  $<-0.5$ ). Unshaded boxes mark weak/negligible correlations (between 0 and 0.25, and 0 and -0.25), with values greyed-out for clarity. All remaining values are presented with black text, with those in this range related to Hg in the boldest type. (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 Hg<sub>T</sub> 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 Hg<sub>T</sub> and first principal component (PC1) of the BOS04-5B XRF data, in which 39% of total variance is strongly associated with terrigenous elements, and so interpreted as an indicator of lake level changes (McKay, 2012). **R (r) and r-squared (r<sup>2</sup>) 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 only exception to this is the correlation matrix (Fig. 3) where we supply these numbers in a data sheet (see following point).

- 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 combinations analysed. 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 3** 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 ( $p < 0.01$ ).**”

Are there any Hg analyses of the crater walls? I think it would be a good idea to add a description of the geology associated with the crater area in the site description.

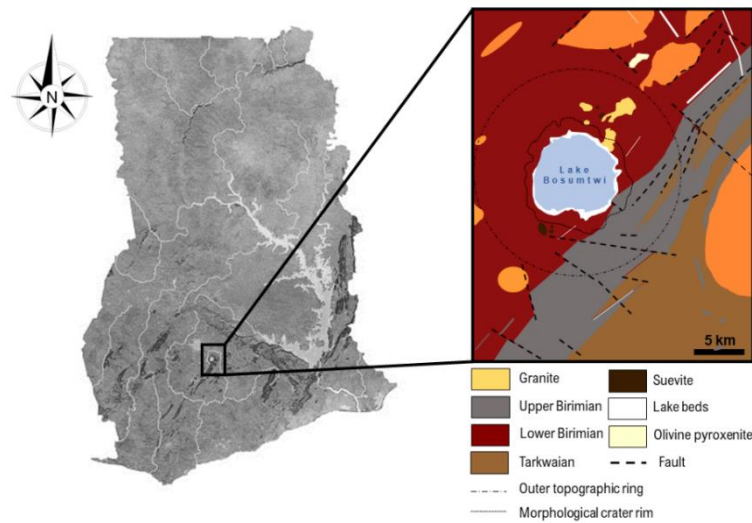
To the best of our knowledge there are unfortunately no direct Hg analyses available for the material surrounding the lake. We concur that a more detailed description of the local geological setting should have been included in our site description. Further details will be added in the main text:

**Line 153-158:** “Lake Bosumtwi is the only natural lake in Ghana, West Africa (6°30' N, 1°25' W) (Fig. 1). It occupies a  $1.08 \pm 0.04$  Ma meteorite impact crater, which is one of the youngest and best preserved impact craters on Earth (Jourdan et al., 2009). The surrounding bedrock and meteorite impact target rocks are Proterozoic metagraywackes, phyllites and metavolcanic rocks of the Birimian Supergroup (~2 Ga) (Jones et al., 1981). Lake beds, soils, and breccias constitute the most recent rock formations at the site, and are associated with evolution of the crater through time (Koerberl et al. 2007).”

Although the extent to which the geogenic Hg pool may contribute to signals recorded in Lake Bosumtwi is difficult to constrain, for example due to lack of Hg analyses on the detrital source material, we do explore plausible hypotheses related to this topic in our supplementary information file under the header:

### S12. Crater origin and geology

In this file, we also include a geological map of the Bosumtwi crater:



**Figure S6:** A schematic geological map of the Bosumtwi impact structure, Ghana. Adapted from Koeberl et al. (2007)

**Figure 3** is blurry.

We apologise for any difficulties reviewer #2 may have had in viewing our manuscript figures. Although no blur is detectable on the PDF file that was submitted to *Climate of the Past* in our original submission, we will make sure to perform additional checks on the manuscript file when uploading our revised submission.

Please identify the p-values associated with the  $r^2$  values.

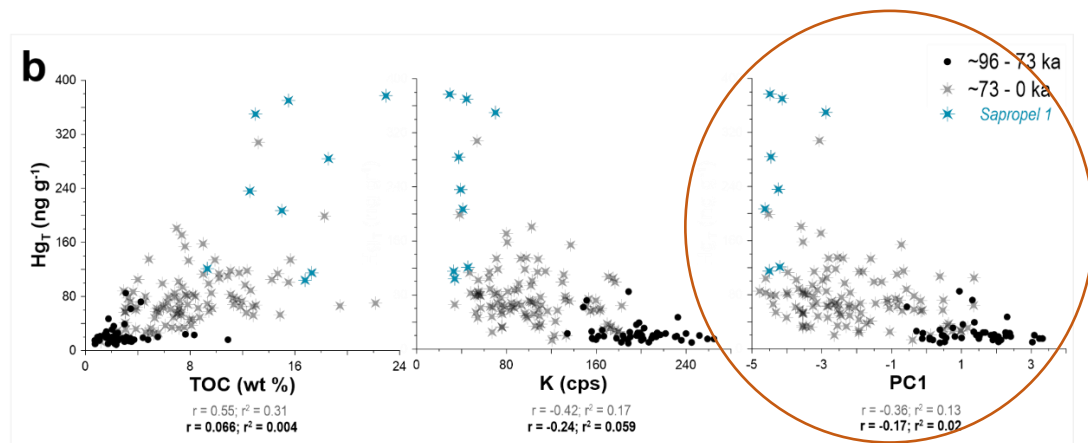
We agree this information should have been included in our original submission, and are grateful to reviewer #2 for flagging this. As mentioned above, we will make this information available in our revised submission by addition of a new sheet to our supplementary data file (titled: "**Pearson's Correlation**"), which will contain three tables: (1)  $r$  values, (2) t-squared statistics, and (3) p-values for all for all combinations analysed. We will also include a section in our supplementary file that provides details on how these p-values were calculated, and an additional statement in the caption for **Figure 3**:

**Figure 3:** "(a) 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 (\*). Grey shading marks positive correlations (light:  $>0.25$ , dark:  $>0.5$ ), and orange shading marks negative correlations (light:  $<-0.25$ , dark:  $<-0.5$ ). Unshaded boxes mark weak/negligible correlations (between 0 and 0.25, and 0 and  $-0.25$ ), with values greyed-out for clarity. All remaining values are presented with black text, with those in this range related to Hg in the boldest type. (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  $Hg_T$  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  $Hg_T$  and first principal component (PC1) of the BOS04-5B XRF data, in which 39% of total variance is strongly associated with terrigenous elements, and so 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."

Can you do some regression analyses for data presented in **Figure 4**? Would this make your discussion more robust?

This is a very good point. In **Figure 4**, we point out that a large number of elements measured in BOS04-5B by XRF are presented by means of PC1. Specifically, those that are strongly associated with the terrigenous elements Al, Si, K, Ti and Rb, whose concentrations are strongly driven by changes in the terrigenous content of core BOS04-5B, the overall sediment supply, and terrigenous drainage area: all functions of lake level (McKay, 2012). Given the importance of lake level for interpretation of our Hg record, we fully agree with reviewer #2 that a quantitative analysis of the relationship between Hg<sub>T</sub> and PC1 would be a valuable addition to our revised manuscript. Given the differences in resolution between the two datasets, PC1 values were averaged to obtain a value corresponding to the interval covered by each discrete Hg<sub>T</sub> sample (~0.5 cm). We will present the result of this analysis in **Figure 3** in our revised manuscript (see *below*), and refer explicitly to the statistical values in the discussion text. For example:

**Lines 516-521:** “The magnitude and frequency of variability in Hg<sub>T</sub> visibly increases at ~73 (±6) 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 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**), which is corroborated by a statistically-significant relationship between PC1 (lake level indicator; McKay, 2012), and Hg<sub>T</sub> in our record (**Fig. 3b** -  $r = -0.53$ ;  $r^2 = 0.29$ ).”



**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 Hg<sub>T</sub> 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 Hg<sub>T</sub> and first principal component (PC1) of the BOS04-5B XRF data, in which 39% of total variance is strongly associated with terrigenous elements, and so interpreted as an indicator of lake level changes (McKay, 2012). R (r) and r-squared (r<sup>2</sup>) 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.

This same approach could not be taken to apply regression analyses of relationships between the BOS04-5B Hg and forest (woody) taxa abundance (presented as DCA Axis 1), due to differences in sampling position, which would result in significant interpolation. Although the pollen and Hg concentrations were measured in the same core, they were each measured on different sample sets at a similar resolution; meaning that values are offset from each other by ~20 cm (equating to >100 years). This implies interpolation is particularly risky when considering the effects of short-lived events such as wildfires on Hg concentrations, charcoal and pollen, and thus we cannot appropriately test for a relation between these proxies.

Despite these limitations, we fully agree with reviewer #2’s comment that exploration of Hg-vegetation-catchment interactions is an important area for future study. Namely, that studies of this nature could provide crucial context for understanding which mechanisms are most significant for transport, accumulation, and cycling of Hg in different ecosystems, and comparing similarities and differences in Hg signals between lakes with comparable or

opposing structures (e.g., closed versus open), composition (e.g., grassland versus forest), and sensitivity to hydroclimate.

**Line 501:** please include the Outridge reference since he was the first to state this.

We agree this reference is important to include here, and so will amend the citation to read:

**Lines 548-551:** “Scavenging of Hg from the water column by algae is also a process now recognised as an important driver of Hg export to lacustrine sediments; particularly in systems where primary productivity, organic matter production, and burial capacity is high (Outridge et al. 2007; Biester et al., 2018; Schütze et al., 2021)”

**Line 508:** can you provide information on documented fire events in this area that would directly impact water in the crater?

We are grateful to reviewer #2 for highlighting this, as it underscores how we have (wrongly) omitted to mention the existing literature that is relevant here. Namely, studies that have analysed micro charcoal concentrations in the BOS04-5B core, and subsequently used this data to provide millennial scale fire histories for this region. For example:

Miller, C.S., Gosling, W.D. (2014) Quaternary forest associations in lowland tropical West Africa. *Quaternary Science Reviews* **84**: 7–25

Miller, C.S., et al. (2016) Drivers of ecosystem and climate change in tropical West Africa over the past ~540000 years. *Journal of Quaternary Science* **31**: 671–677

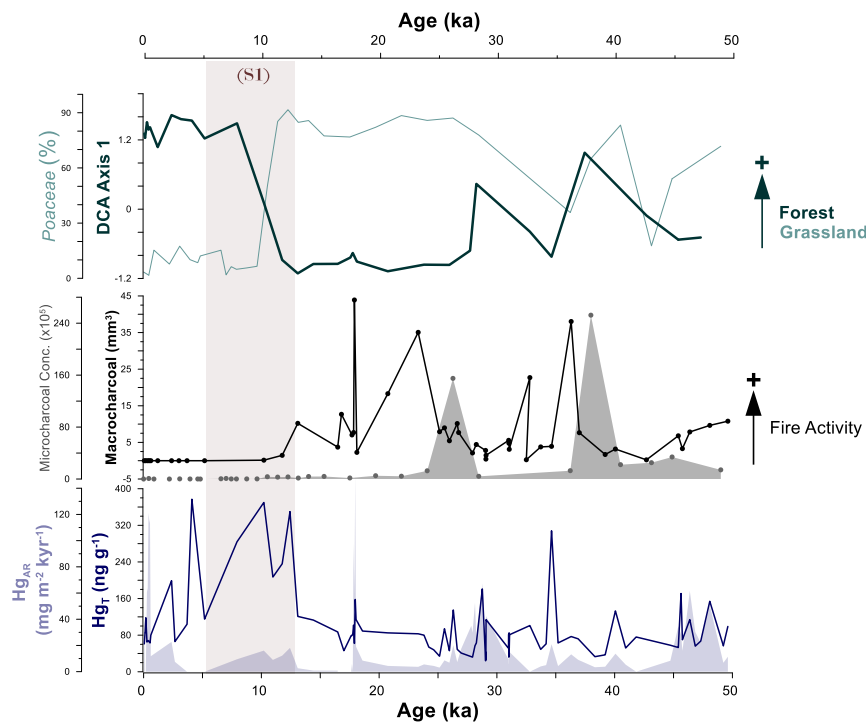
Gosling, W.D. et al. (2021) Preliminary evidence for green, brown and black worlds in tropical western Africa during the Middle and Late Pleistocene. *Palaeoecology of Africa* **35**: 13–25

To acknowledge these studies, we will amend this section of our manuscript to read:

**Lines 558-562:** “For Lake Bosumtwi these direct inputs may have come from precipitation, and/or from increased flux of charcoal (and associated released of Hg from vegetation during wildfires) into the lake following local wildfire events; the latter documented by variations in the micro charcoal concentration of BOS04-5B (Gosling et al., 2021; Miller et al., 2016; Miller and Gosling, 2014).”

However, it is important to note is that, similar to the pollen data, we could not conduct regression analyses of relationships between the BOS04-5B Hg and charcoal abundance due to differences in sampling position. The BOS04-5B has been assessed for both macro and microcharcoal abundance (Kiely et al., 2025; Miller et al., 2016), however, almost all samples assessed for this purpose were measured on different sample sets at a similar resolution. This creates a ~20 cm (>100 years) offset, and hence reduces the suitability of interpolation for alignment of these datasets; particularly when assessing the effects of short-lived events such as wildfires on Hg concentrations. We cannot entirely discount the possibility that wildfires did affect Hg fluxes into Lake Bosumtwi, although other drivers were likely more influential; namely moisture-driven changes in sedimentation, productivity, and detrital material supply. Details of this analysis will be provided in an accompanying supplementary information file:

#### **S14. Fire activity**



**Figure S9:** 50-kyr records of total mercury ( $Hg_T$ ) and mercury accumulation rate ( $Hg_{AR}$ ) for Lake Bosumtwi from this study, with proxy datasets from prior studies of the same lake. These include forest (woody) taxa abundance represented by detrended correspondence analysis (DCA) axis 1 (Gosling et al., 2022a; Miller et al., 2016), percentage abundance of Poaceae (grass) pollen (Miller et al., 2016), microcharcoal concentrations (Miller et al., 2016), and macrocharcoal volume (Kiely, 2023). A distinct lake low stand (LS) based on seismic profiles and sedimentological data 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). 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).

Again, would it be worth doing some correlation analyses for data show in **Figure 5** to make your paper more quantitative?

This is an interesting suggestion. On one hand, we do agree that there may be some value in performing analyses of this nature on the data presented in **Figure 5**. However, this would require (non-trivial) interpolation of nearly all datasets involved. Assessing whether false positive or negative results occur would be especially challenging given various differences in how age models were constructed and whether they fully account for any errors that may exist. Aside from the challenge of properly taking these chronological uncertainties into consideration, any correlations produced are in any case likely to highlight the correspondence (or lack thereof) with the most obvious changes in each record, and so would give limited additional insight beyond what is visible to the eye. Thus, we believe that the potential benefits of performing further statistical analyses on (interpolated versions of) the data presented in **Figure 5** do not sufficiently outweigh the risks of potential misinterpretation.

**Line 537:** seeks? How about this? This work combined geochemical data obtained for a sediment core collected from Lake Bosumtwi, Ghana, with climate data in order to explain measured trends in Hg concentrations.

This statement does a great job at summarizing our study rationale. Inspired by this suggestion, we will amend the first line of our concluding section to read:



**Lines 592-594:** *“This study combines new sedimentary Hg data from Lake Bosumtwi, Ghana, with proxy data from archives across the African continent to explore whether hydroclimate has exerted a measurable effect on regional Hg cycling over the past ~96-kyr.”*

I like the schematic, **Figure 6**.

We are delighted that reviewer #2 found this figure a valuable addition to the manuscript, and are grateful for the positive feedback.