Response to Associate editor decision: Reconsider after major revisions.

Firstly, we wish to thank both the associate editor and the anonymous reviewer for their timely and thoughtful comments. It is our opinion that their suggestions, specifically their concerns around the applicability of the transfer function used in the previous version of the manuscript, were completely justified. In rectifying this issue in line with their concerns, we believe both the manuscript and our results have been vastly improved.

We previously used the Amesbury et al. (2016) paper for the reconstructions in this study as routine. This is one of the most widely used calibration datasets for testate amoebae, owing to its broad spatial coverage and the large number of samples included. However, we agree that given the low range of pH values in the dataset, this model was not the best choice for reconstructing pH as opposed to water table depth. Since identifying changes in pH in response to cement dust pollution, and identifying threshold values for these changes, are both central themes of the paper, it is vital that this reconstruction and our values are as accurate as possible.

In response, we have changed the transfer function model and calibration dataset used to reconstruct pH to one by Šímová et al. (2022), which uses over 250 samples from a range of sites with differing trophic conditions and a pH range of 3.5–7.9. The model also has much-improved predictive power for reconstructing pH (RMSEP100 of 0.45 pH units and an R^2 of 0.86). Doing so had the expected effect of increasing reconstructed pH values post c. 1850, relative to the previous reconstruction, while pH values for the period preceding the establishment of the factory at Kunda (ombrotrophic and poor fen phases) were similar between the two models. The direction of change was comparable between the two reconstructions. The pH values for the recent past are now substantially closer to the measured values included in this study.

Our findings now indicate that while relatively small increases in pH can drive ecosystem shifts over long time periods, much larger (~40%) increases drive more rapid, substantial changes. We have changed much of the text in the discussion and conclusions to reflect these changes in our interpretation of the results. However, despite improved comparability between measured and reconstructed values, our reconstructed pH values for the recent past remained slightly lower than measured values. We attribute this either to intra-site heterogeneity in pH (since the measurements were taken from bog pools in the NW region of the site, which may be influenced by differences in hydrology or cement dust load between areas) or to the inherent uncertainties in transfer-function-based reconstructions.

As a result, we opted to retain discussions surrounding the limitations of such models in the manuscript, despite the substantial improvements, and we have retained recommendations for future re-evaluation of our data should an improved dataset become available. It is our opinion that while future improvements may be possible, our reconstruction using this dataset is the best that can be achieved at present, within the scope of our paper. This new reconstruction provides a much more accurate estimate for critical threshold values than the previous version.

In the proceeding section, we provide a full list of changes made to the paper in this new revision to address the concerns of the associate editor and reviewer (line numbers are in reference to those in the tracked changes version of the manuscript):

Specific changes in response to testate amoeba transfer function concerns:

- New reference for Šímová et al. (2022) added to reference list.

- Line 21: 'near' added to reflect slight change in results following the addition of an improved model for pH reconstruction.

- Lines 243 – 252: We added a new section of text detailing the methodology for the updated pH reconstruction, including introducing the new calibration dataset used, model diagnostics, modern analogue test results:

New text: "We also reconstructed changes in pH using a transfer function developed by Šímová *et al.*, (2022), based on over 250 samples from east-Central Europe. The calibration dataset includes samples from peatland sites ranging from acidic bogs to calcareous fens, with a pH range of 3.5-7.9 making it suitable for reconstructing pH in sites that have likely undergone substantial changes in pH over time, such as Varudi. The transfer function was constructed using maximum likelihood calibration of Gaussian response curves (MLRC) with squared chord distance as the dissimilarity measure. The full dataset transfer function (mineral and ombrotrophic sites combined) was tested by leave-one-out cross-validation, achieving a $RMSEP_{100}$ of 0.45 pH units and an R^2 of 0.86. Both reconstructions were performed using the 'Rioja' package in R (Juggins, 2019). Comparisons between the modern calibration and fossil datasets using the 'compare.datasets' function in Rioja indicate good levels of overlap, with no non-analogue assemblages identified in the fossil data."

- Figure 10: The new testate amoebae derived transfer function is included in Figure 10

- Lines 541 – 544: We replaced values describing the results of the pH reconstruction in the results section. Mostly these changes were to the values as generally the changes seen in the new reconstruction were directionally similar as in the previous, the timings of some changes and peaks were slightly different:

Previous text: "Reconstructed pH values were relatively stable throughout much of the record (4.1 ± 0.5). Around c. 1800 cal CE, pH began to rise, reaching a maximum of 5.4 ± 0.5 by c. 1998 cal CE. By c. 2006 cal CE until the top of the core, pH returned to pre-disturbance levels".

Changed text: “Reconstructed pH values were relatively stable throughout much of the record (4.7 ± 0.6). Around c. 1850 cal CE, pH began to rise, reaching a maximum of 7.2 ± 0.6 by c. 1985 cal CE, remaining high until the uppermost samples, where pH returned to near pre-disturbance levels (4.8 ± 0.5), this change occurring after c. 1998 cal CE”.

- Figure 14: New reconstruction added to summary figure.

- Line 725: Words added to line to show change in the rate of pH increases in line with the new reconstruction-

Previous text: “…with increasing pH…”

Changed text: “…with rapidly increasing pH…”

- Line 757: Words added to line to reflect changes in the direction, timing and magnitude of reductions in pH towards the top of the core:

Previous text: “…pH returning towards…”

Changed text: “…pH beginning to return towards…”

- Lines 855 – 895: Updated section ‘Indicators of critical thresholds to assess peatland condition and recovery due to alkaline pollution’ to reflect the new interpretations based upon the new reconstruction:

Previous text: “Around c. 1800 cal CE, the reconstructed pH for Varudi showed an increasing trend from the previous, relatively stable conditions, reaching a maximum of 5.4 ± 0.5 by c. 1990 cal CE (Figure 10). This value aligns with pH measurements for polluted sites in Estonia, supporting the reliability of our reconstruction (Paal *et al.*, 2010). We find that the transition from an ombrotrophic bog to fen-like conditions, driven by mineral soil enrichment resulting from land clearance for agriculture in the surrounding landscape, was associated with an increase of pH levels from c. pH 3.8 to pH 4.1 which represents (due to the logarithmic scale of pH) nearly a twofold increase in alkalinity. Likewise, the subsequent shift from fen-like to forested conditions corresponded with a small average increase in pH to c. 4.3, or c. 1.5 times more alkaline than the previous state and more than three times more alkaline than during the bog phase. Our results suggest that relatively small increases in pH (increases in pH of 0.2 – 0.3) may result in critical ecosystem transitions. However, we stress that the threshold values defined here are highly uncertain, owing to the limited predictive ability of the transfer function models used for our pH reconstruction and the relatively significant uncertainties associated with transfer function models in general (Amesbury *et al.,* 2016). Higher pH values measured from bog pools at the site in 1996 – 1997 of 7.6 - 8.5 are higher than our estimates for this period, and by 2022 pH had only fallen to 6.3 - 6.8 (Pärtma, 2023), suggesting that our reconstruction underestimates pH levels experienced by the site by a wide margin, possibly due to the removal of high-pH samples from Amesbury *et al.,* (2016)'s dataset, causing the range of pH measurements to be low and biased towards more acidic conditions. This may warrant future re-evaluation of our data using transfer functions specifically developed for reconstructing pH in polluted peatlands. In addition, we cannot rule out the possibility that other impacts of cement deposition (e.g., reduced photosynthesis rates due to dust deposition) and, significantly- drainage; may have also played a role in driving botanical changes".

Changed text: "Around c. 1910 cal CE, the reconstructed pH for Varudi showed an increasing trend from the previous, relatively stable conditions, reaching a maximum of 7.2 ± 0.6 by c. 1985 cal CE (Figure 10). This value aligns with previous pH measurements for polluted sites in Estonia, supporting the reliability of our reconstruction (Paal *et al.*, 2010). We find that the transition from an ombrotrophic bog to fen-like conditions during the mid 13^{th} century, driven by mineral soil enrichment resulting from land clearance for agriculture in the surrounding landscape, was associated with an increase of average pH levels from c. pH 3.9 to pH 4.1 which represents (due to the logarithmic scale of pH) more than 1.5 times increase in alkalinity. In contrast, the subsequent shift from fen-like to forested conditions corresponded with a small average increase in pH to c. 5.7, over 40 times more alkaline than the previous state and more than 60 times more alkaline than during the bog phase. Our results suggest that while substantial increases in pH can drive rapid and substantial ecosystem regime shifts, relatively small increases in pH (increases in pH of 0.2 – 0.3) may also drive critical ecosystem transitions that may be more gradual but can have equally as significant and long-lasting impacts upon peatland ecosystem functioning, such as the capacity for carbon accumulation. However, we stress that the threshold values defined here remain uncertain, owing to the relatively significant uncertainties associated with transfer function models in general (Amesbury *et al.,* 2016). Higher pH values measured from bog pools at the site in 1996 – 1997 of 7.6 - 8.5 are slightly higher than our estimates for this period (6.1 - 6.6), and by 2022 pH had only fallen to 6.3 - 6.8 (Pärtma, 2023) although our reconstruction indicates pH had fallen to 4.8 by this time, suggesting that our reconstruction, despite having a strong predictive power, underestimates pH levels experienced by the site. Alternatively, this may simply reflect local variation in pH within Varudi, as the pH measurements were taken from bog pools in the NW margin of the bog, not adjacent to the coring location. This may warrant future re-evaluation of our data using transfer functions specifically developed for reconstructing pH in polluted peatlands. In addition, we cannot rule out the possibility that other impacts of cement deposition (e.g., reduced photosynthesis rates due to dust deposition) and, significantly- drainage; may have also played a role in driving botanical changes".

- Lines 943 – 945: Added a section of text highlighting that substantial increases in pH can induce rapid ecosystem transitions.

New text: “while an increase of 1.5 pH units (a ~40 x increase in alkalinity) will cause rapid ecosystem transition, resulting in the loss of *Sphagnum* mosses and changes in ecosystem functioning”.

Minor comments:

- Lines 589-591: In the NPP diagram, Botryococcus braunii is missing.

Thanks to the authors comments. *Botryococcus braunii* was included in the figure but mislabelled simply as ‘Botryococcus’. We have amended this in an updated version of Figure 12.

- Lines 628-631: Based on which plant taxa you determined an ombotrophic bog phase (c.1000-1250 cal yr CE) and a poor fen phase (c. 1250-1570 cal yr CE)?

These were based upon multiple lines of evidence, including, but not limited to, the record of vegetation change from the macrofossil record (shift from *Sphagnum* to monocot/sedge dominated conditions)- this is covered in the results section and later discussion in the manuscript. However, as the reviewer previously indicated, the low taxonomic resolution of our plant macrofossil reconstruction is insufficient for drawing conclusions from alone. Instead, our conclusion is supported by the combined results of the multiple proxies. We realise that the text reads ‘botanical change’, suggesting that these conclusions are based upon this line of evidence alone- while this was not our intended meaning, we have opted to remove the words ‘of botanical change’ from L692 - 693 for clarity.

- Lines 631-634: I do not understand this sentence. Varudi bog has been impacted by drainage since 19th century (lines 155-163).

We are not entirely certain exactly where the misunderstanding arises from based upon the reviewer’s comment, but we interpret it as a concern that the phrase ‘unique, pre-disturbance context’ for understanding the effects of atmospheric pollution upon the site was unclear, given that this pollution occurred alongside other disturbances, notably drainage. If this is indeed the intended meaning, we apologise for the section not being written clearly. What we meant is that most studies of peatland responses to environmental stressors (such as those cited here) generally do not account for developmental or disturbance histories of sites (an issue highlighted by Paal et al., 2010). By contrast, our palaeoecological approach allows us to incorporate a pre-disturbance baseline into the analysis (specifically the PcA).

To address this comment, we have revised the wording of this section to better reflect our meaning (L695 – 697):

Previous text: “Unlike most studies of peatland recovery, which has typically been conducted on historically drained, mined, or agricultural sites (Roderfeld, 1993; Graf *et al.,* 2008; Wagner *et al.,* 2008; Paal *et al.,* 2010), our study presents a unique pre-disturbance context for understanding the effects of atmospheric pollution upon Estonian peatlands”.

Changed text: “Unlike most contemporary research on peatland recovery, which has typically been conducted on historically drained, mined, or agricultural sites (Roderfeld, 1993; Graf *et al.,* 2008; Wagner *et al.,* 2008; Paal *et al.,* 2010). An advantage of our palaeoecological approach is that our study presents a unique pre-disturbance context for understanding the effects of atmospheric pollution upon Estonian peatlands”.

- Lines 678–684: I agree with this interpretation (Sphagnum sect. Acutifolia are predominantly hummock-forming species); however, the sentence is too long. It would be appropriate to rephrase the sentence and, ideally, divide it into two separate sentences so that it is easily understandable for the reader. Your interpretation of wetter local environment is supported by presence of Scheuchzeria palustris pollen aroud 1500 and 1800 cal yr CE. This species tolerates overflooding rather than long-term drought

We thank the reviewer for this comment, and in response have made the following changes in line with these suggestions to improve the clarity and flow of the text, and to incorporate the presence of *Scheuchzeria palustris* into our discussion (L746 – 753), and the new reference to support this statement was added to the reference list:

Previous text: “By c. 1570 cal CE, the site had transitioned into a wetter, more minerotrophic fen, indicated by the gradual replacement of *Sphagnum* by monocots such as *Carex* sp. and *Eriophorum vaginatum* in the plant macrofossil record and an increase in *Sphagnum* section *Cuspidata* remains and subsequent declines in *Sphagnum* subsection *Acutifolia* and subsection *Sphagnum* (Figure 9), and a shift towards wetter conditions during this period indicated by the testate amoebae reconstruction, possibly indicating permanent or seasonal standing water at this time (Figure 10)”.

Changed text: “By c. 1570 cal CE, the site had transitioned into a wetter, more minerotrophic fen, indicated by the gradual replacement of *Sphagnum* by monocots such as *Carex* sp. and *Eriophorum vaginatum*, increases in *Sphagnum* subsection *Cuspidata* and subsequent declines in *Sphagnum* subsections *Acutifolia* and *Sphagnum* in the plant macrofossil record (Figure 9). This shift towards wetter conditions is also supported by the testate amoebae reconstruction, possibly indicating permanent or seasonal standing water at this time (Figure 10), and the presence of *Scheuchzeria palustris* pollen between c. 1500 – 1800 cal CE (Figure 11), a flood-tolerant species (Tallis and Birks, 1965)”.

- Lines 679-680: In the macrofossil diagram, Eriophorum vaginatum is missing. Is it included as Eriophorum sp. (spindles)?

We apologise for the oversight, *Eriophorum* sp. was meant here, text changed to reflect this (L747).

- Lines 726-734: It would be appropriate to rephrase the sentence and, ideally, divide it into two separate sentences so that it is easily understandable for the reader.

We agree with the reviewer and have changed the structuring of the sentence to improve clarity and flow as suggested:

Previous text: "We find no evidence for botanical succession toward pre-disturbance conditions in the plant macrofossil record (Figure 9), while the presence of calciphilous *Tomentypnum nitens* at the core surface reflects the legacy of alkaline conditions at the site (Malmer et al., 1992; Hájek et al., 2006; Apolinarska et al., 2024), likely resulting from the movement of mineral-rich water, enriched by the buried cement dust as it is transported upwards via capillary rise, indicating that these cement deposits continue to influence surface water chemistry and vegetation communities at the site, although *Sphagnum* has returned to the site since its disappearance, indicating a degree of recovery. Identification and monitoring of the species present on the site could provide additional insight into the extent of this recovery".

Changed text: "We find no evidence of botanical succession toward pre-disturbance conditions in the plant macrofossil record (Figure 9). The presence of the calciphilous moss *Tomentypnum nitens* at the core surface likely reflects the legacy of alkaline pollution at the site (Malmer et al., 1992; Hájek et al., 2006; Apolinarska et al., 2024). These cement deposits may still influence surface water chemistry and vegetation through the upward movement of water enriched by the buried dust. Nonetheless, *Sphagnum* has re-established at the site since its disappearance in the 1960s, suggesting a degree of recovery. Identification and monitoring of the present-day flora could provide additional insights into the extent of this recovery".

- Lines 820-823: Why do you compare the reconstructed pH with pH measurements from other polluted sites in Estonia? Why do you not compare your reconstructed pH with the pH measurements made directly at Varudi (lines 834-839)? I would assume that these values would better show whether your pH reconstruction is reliable or not.

We do compare our reconstruction with measurements later in this section of the discussion (L945 – 951) with previously unpublished data from Varudi, although such data are relatively scarce, and we intended to support our findings through incorporation of the wider literature. However, considering our new reconstruction and its better agreement with our pH measurements from the past c. 16 years, this line seems slightly redundant. We have replaced it with a different statement, from another study (same first author), and have added the reference to the citations. Some changes in the text and values reflect the new reconstruction.

Previous text: "Around c. 1800 cal CE, the reconstructed pH for Varudi showed an increasing trend from the previous, relatively stable conditions, reaching a maximum of 5.4 ± 0.5 by c. 1990s cal CE (Figure 10). This value aligns with pH measurements for polluted sites in Estonia, supporting the reliability of our reconstruction (Paal *et al.*, 2010)".

Changed text: "Around c. 1910 cal CE, the reconstructed pH for Varudi showed an increasing trend from the previous, relatively stable conditions, reaching a maximum of 7.2 ± 0.6 by c. 1985 cal CE (Figure 10). This value aligns with previous pH measurements for forest litter in the zone surrounding the Kunda cement factory of 7.1 – 7.4, supporting the reliability of our reconstruction (Paal *et al.*, 2015)".

- Lines 870-871: The pH reconstruction is decreasing since ca 2000, not increasing.

We apologise for the error and have changed the text to decreasing in L994 in agreement with the suggested change.

Other, non-requested changes made:

- Line 719- Typo fixed, **B**ritish.
- Figure 11- small cosmetic changes made to figure, full stops removed from '...type' in taxa names.
- Supplementary data updated to include new model details.
- Figure and table of supplement numbered as Figure S1 and Table S1. New text added (L415: "Model diagnostics are shown in Figure S1").
- Figshare temporary link changed to DOI (L966).
- Word 'for regulating' added to L987 to improve clarity.