

Dear Editor,

we sincerely thank the reviewers and the editor for their constructive and thoughtful comments on the revised manuscript. All additional revisions are incorporated into the updated manuscript, and changes are clearly indicated in the “track change file”. In the following, we provide our detailed responses to the major and minor comments raised. Editor and reviewer comments are reproduced in italics, followed by our responses in plain text. For clarity and transparency, each response is numbered and structured according to the order of appearance in the reviewer reports.

Reply to editor`s comments

1. Public justification

Dear authors,

Your revised manuscript has been reviewed by two referees, who had also reviewed the previous version of it. One referee has provided additional comments for you to consider, especially on the attribution of the changes observed to climate change, and I tend to agree with them. While you are comparing climatically different periods, each of them is relatively short (when we consider that climate 'normals' are established over 30-year periods), so the assessment of climate change may be perceived by some as not robust enough (even if you do state that what you see is in line with longer assessments performed by others). More importantly, to me, a range of land use and land cover changes may have occurred over the same period, including localized ones, making it more difficult to partition change drivers between climate and land cover. Personally, I think that your manuscript would be equally strong and interesting if you were “pitching” it, in the title, abstract, methods and results sections as a comparison between climatically different periods. You could then use your discussion section to state that this comparison you made may be a good proxy for what may continue to happen under climate change, assuming that the two periods you consider were good proxies for ongoing changes and changes to come... with the caveat that other changes may also occur concurrently with climate change. I invite you to consider the comments of Referee #1, as well as my short comments above, to guide the (moderate) revision of your manuscript. I look forward to receiving your revised manuscript, which I believe will be a strong contribution.

Due to the relatively short duration of the investigated periods compared to conventional 30-year climate standards, we have revised the manuscript to avoid attributing the observed changes solely to climate change. Instead, the analysis is now consistently framed as a comparison between two climatically different periods.

In line with the editor’s and referee’s comments, we now explicitly acknowledge that additional drivers may have influenced the observed changes in solute export mechanisms. In particular, we discuss potential legacy effects as well as land use changes that may have occurred over the study period. We note that historical changes in fertilization regulations or management practices within individual catchments cannot be fully excluded and may have contributed to long term solute transport dynamics through legacy effects.

At the same time, we emphasize that our analyses indicate a limited influence of land use parameters on the observed changes in SEM (solute export mechanisms) between the two climatically different periods. Specifically, arable land shows a comparatively small influence on NO₃-N export and no significant influence on NH₄-N, SRP (soluble reactive phosphorus), TP (total

phosphorus), or TOC (total organic carbon). Additional analyses of land use change, as presented in the Materials and Methods section (L. 238 to 243 in track change file) and in the Supplements (table S5), demonstrate that, despite substantial land use changes occurring between 2006 and 2018, no significant correlations were identified between changes in solute transport, expressed as differences in slope b , and changes in land use composition. This supports our interpretation that the observed changes in solute export mechanisms are predominantly associated with climatic variables.

Nevertheless, we have revised the manuscript to more explicitly emphasise the potential roles of legacy effects and land use changes as concurrent drivers. The comparison of the two climatically different periods is now framed as a useful proxy for potential future changes in solute transport mechanisms that could be caused by ongoing climate change (e.g. L. 463-465 in track changes file). At the same time, we recognise that other environmental and human-induced changes could occur simultaneously.

We believe that these revisions strengthen the manuscript and align it more closely with the editor's and referee's recommendations.

Reply to Report #2 (Anonymous referee #1)

2. Major comment:

I appreciate the authors' analysis of significant differences in temperature and evapotranspiration between the two study periods (although the duration and intensity of precipitation were not tested). However, the direct attribution of the presented results to climate change per se is, from my point of view, not scientifically warranted.

While there appears to be a correlation with the change in the climatic conditions, namely temperature and evapotranspiration (but not total precipitation), and the observed changes in solute export patterns, their causality can only be assumed rather than demonstrated. Many other things, for example, the discussed legacies of nitrate, could just as well explain the differences between the two periods. Moreover, although the observed shifts in meteorological variables are consistent with climate change, they cannot be taken as definitive evidence, as similar climatic differences could also occur in the absence of anthropogenic climate forcing. To be clear, I agree that climate change is likely to influence water quality in multiple ways, and I am happy to see that link being discussed very prominently. Nevertheless, caution is needed when inferring direct causality to climate change per se without robust attribution analysis.

Without substantially toning down the current causal claims to climate change per se and refocusing on the specific mechanisms that potentially cause the observed change in solute export patterns (for example, higher evapotranspiration or shorter, more intense rainfall events, but also other factors not related to climate), I do not believe the manuscript yet meets the standards of scientific rigor required for publication.

We acknowledge the referee's concerns regarding the strength of causal attribution to climate change per se. In response, we have substantially toned down statements implying direct or exclusive causality and removed wording that could be interpreted as definitive attribution to anthropogenic climate change. The revised manuscript now consistently frames the analysis as a comparison between two climatically different periods rather than as proof of climate change impacts.

In line with the referee's comments, we have expanded the discussion of alternative and concurrent drivers of the observed changes in solute export mechanisms. In particular, we now provide greater emphasis on potential legacy effects, acknowledging that historical changes in fertilization practices and regulatory frameworks may have influenced present-day solute transport, especially with respect to nitrate (L. 486- 493 and L. 557-571 in track changes file). Such legacy effects could contribute to delayed system responses and cannot be excluded as a factor shaping the observed differences between the two periods.

At the same time, our analyses indicate that land use and land use change play a subordinate role as significant explanatory variables for changes in solute export mechanisms. Although substantial land use changes occurred over the study period, additional analyses reveal no significant correlations between changes in solute export metrics (slope b) and changes in land use composition. This supports our interpretation that the observed changes in solute export mechanisms are predominantly associated with climatic variables, while acknowledging that land use change and legacy effects may have contributed concurrently.

Following the guidance of the referee and the editor, we have revised the manuscript to present our analysis primarily as a comparison of climatically different periods, which may serve as a proxy for potential shifts in SEM under ongoing climatic alterations. To avoid overstating causality, terminology and definitions have been adjusted, and previous direct attributions have been replaced with more cautious formulations that emphasize plausible rather than demonstrated influences. These revisions ensure that our interpretation remains appropriately focused on the observed differences between the two periods while acknowledging uncertainties and the possible contributions of additional factors such as land-use changes and nutrient legacies.

3. Minor comments (please note that the line numbering refers to the track-changes version)

3.1 Clarification of terminology distinguishing short-term and long-term dynamics

I am not convinced by the use of the term "temporal dynamic" if it is intended to distinguish certain patterns from "long-term dynamics." The term "long-term dynamic" itself inherently refers to temporal dynamics, so the distinction as currently phrased appears conceptually inconsistent. If the authors intend to refer to short-term dynamics (e.g., seasonal or event-scale variability), this should be clearly specified to avoid confusion.

To avoid conceptual inconsistency and better distinguish short-term variability from long-term dynamics, we have replaced the term "temporal dynamic" with "seasonal dynamic" throughout the manuscript. This adjustment clarifies the intended scale of variability and ensures a clearer differentiation between seasonal patterns and long-term trends.

3.2 L204

Discharge dynamics or variability, not magnitude.

Our original wording, which referred to 'discharge magnitude', did not accurately reflect the underlying concept and may have caused ambiguity. In line with the reviewer's comment, we have revised the sentence to use the more precise term 'discharge variability' explicitly. The text has been updated accordingly in the revised manuscript.

3.3 L213

I suggest referring to Musolff et al. (2015) here as well, as they were the first to combine the CQ slope with the CV_c/CV_q in a consistent framework

With the reviewer's suggestion in mind, Musolff et al. (2015) has been included, with the manuscript revised to read as follows: "To avoid misinterpreting such near-zero b slopes as indication of chemostatic behavior, we additionally used the CV_c/CV_q ratio as proposed by Musolff et al. (2015) and Thompson et al. (2011)."

3.4 L319

It shifted to chemodynamic under conditions of high enrichment? Isn't that saying it shifted to enrichment patterns during periods of enrichment? I do not fully understanding what this is supposed to tell the reader.

We agree that the original wording could be misinterpreted and have therefore rephrased the sentence. The manuscript has been updated as follows: "Under condition of high enrichment behaviour ($b > 1$), TOC showed chemodynamic behaviour indicative of discharge-decoupled processes, whereas $b < 1$ generally reflected both chemostatic and chemodynamic patterns."

3.5 L334

"driven by (...)" belongs to the discussion.

To avoid premature interpretation, we have revised the sentence as follows: "Temporal analyses revealed significant changes in SEM across seasons (NH₄-N, SRP) and across decades (NH₄-N, SRP, TP, and TOC)."

3.6 L439

I would assume global warming is an anthropogenic impact as well?

We agree that the original phrasing was ambiguous, as global warming is itself an anthropogenic impact. To address this, the sentence has been rephrased to clearly distinguish between climatic changes and land-use influences: "Results indicate that solute concentration and SEM are influenced by climatically changing periods and land-use changes."

3.7 L625

Why except nitrate? Increased nitrate enrichment during and after drought period has for example been shown by Winter et al. (2023).

We acknowledge that nitrate enrichment can indeed increase under certain hydrological conditions, as shown by Winter et al. (2023). To avoid an overly general statement and to reflect the complexity of nitrate dynamics, we have revised the sentence accordingly. The updated text highlights that enrichment processes occur across nutrients but NO₃-N responses more moderated by legacy effects in some catchments. Revised manuscript sentence: "Accumulated nutrients are transported via surface and subsurface flow during wet periods, especially in autumn and winter, when rising water tables and increased discharge enhance hydrological connectivity (Fig. 6; Bieroza et al., 2024; Winter et al., 2023).

Both transport pathways amplify the enrichment behaviour of all nutrients. However, in the case of NO₃-N, variability can be buffered by deeper legacy sources in some catchments, which can mask enrichment patterns.”

3.8 Clarifying Structural and Thematic Differences Between Sections 4.2.3 and 4.3

What is the difference in the headlines 4.2.3 and 4.3?

The revised headlines are intended to better differentiate Sections 4.2.3 and 4.3. Section 4.2.3 (“Alteration in SEM across Climatically Changing Periods”) outlines what changed in SEM between the periods, while Section 4.3 (“Drivers and Controls Influencing Solute Export Dynamics”) explains why these changes occurred by examining the climatic and catchment-related controls involved. The new titles make this separation more explicit.