Dear reviewer,

Thanks you so much for your constructive comments for the discussion phase. Please check our reply for all your comments point by point (followed and marked in blue). However, the revised completed manuscript will be provided in the next step of the review processes.

Best regards

<u>Responses:</u>

This manuscript presents a numerical modeling study exploring how inter- and intra-annual precipitation variability affects nitrogen (N) loads and fluxes in a catchment. The topic is timely and important for water quality and environmental management. The paper is generally well written, though some grammatical edits are needed. The methods and results are mostly clear and logically presented. The conclusions are relevant and likely to be of interest to scientists and resource managers focused on mitigating nutrient pollution.

Primary suggestions are to:

(1) Clarify and narrow the research focus: While the broad importance of precipitation variability is well established in the introduction, the final introductory paragraph should better define the unique contribution this work makes to the literature. Consider making the statement of objectives more specific and clarifying how the objectives fill gaps left by other recent studies.

Response #1:

Thanks for point that out! We have rephrased the last paragraph of the introduction part to define the unique contribution this work as " *In order to fill the gap, the present study explored the impact of rainfall variability on N dynamics and its potentially negative influence on water quality across inter-annual and intra-annual timescales. To characterize rainfall variability, a stochastic rainfall generator [Robinson & Sivapalan, 1997] was employed to generate rainfall time series with different climatic characteristics. The research was conducted in a small agricultural catchment located in Central Germany, where a hydrological model was previously established utilizing* *the fully coupled surface-subsurface numerical simulator HydroGeoSphere [Yang et al., 2018]. The framework of N dynamics was modified from the ELEMeNT approach [Exploration of Long-tErM Nutrient Trajectories; Van Meter et al., 2017]. The research is divided into two main components. First, three representative years (with high, normal, and low annual precipitation amounts, respectively) were chosen from the past two decades in Central Germany as the target scenarios. A fourth scenario with low annual precipitation amounts coupled with reduced plant uptake represents a case where vegetation is partially destroyed by extreme droughts. The statistical analyses of nitrogen loads and fluxes and a comparison across different scenarios were conducted to reveal the influence of inter-annual rainfall variability. Second, rainfall time series generated using the stochastic rainfall generator by separately altering specific parameters were used to substitute the rainfall data in the simulation period to drive the flow and nitrogen transport models. The responses of the N loads and fluxes to the parameters (e.g., the amplitudes of the seasonal variations in the storm durations and inter-storm period) were thoroughly analyzed to clarify the effect of intra-annual rainfall variability. The study will provide theoretical support to formulate fertilization strategies and protect aquatic ecosystems under climate change in the future.*"

Reference:

Robinson, J., & Sivapalan, M.: Temporal scales and hydrological regimes: Implications for flood frequency scaling, Water Resour. Res., 33(12): 2981-2999, https://doi.org/10.1029/97WR01964, 1997.

Yang J., Heidbüchel, I., Musolff, A., Reinstorf, F., and & Fleckenstein. J. H.: Exploring the dynamics of transit times and subsurface mixing in a small agricultural catchment, Water Resour. Res., 54, https://doi.org/ 10.1002/2017WR021896, 2018.

Van Meter, K. J., Basu, N. B., Van Cappellen, P.: Two centuries of nitrogen dynamics: Legacy sources and sinks in the Mississippi and susquehanna river basins, Global Biogeochem. Cycles, 31 (1), 2–23., https://doi.org/10.1002/2016GB005498, 2017.

(2) Differentiate from previous work: The study references and builds on previous work, particularly Wang et al. (2023). However, the manuscript does not always make it clear

where prior work ends and the current study begins. The manuscript also refers the reader to the previous studies for some crucial details (e.g. calibration to stream nitrate concentrations), which are not easy to find in the previous studies. I recommend adding further descriptions of the related studies at this site and delineating which analyses, model developments, and findings are new in this study.

Response#2:

Thanks for the points!

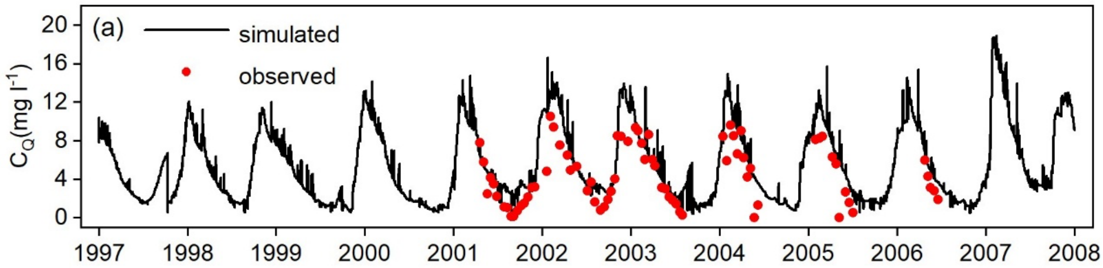
Generally, this study employed the model developed in our previous work Wang et al. [2023] for a same study site. The model setup, method for simulating the flow and N transport, parameters and the calibration are all from Wang et al. [2023]. However, new contributions are attributed to exploring the effect of rainfall variability on N dynamics. To clarify the existing work and the new contributions, we rephased the method section such that the new contributions are highlighted. Additionally, more details regarding the model calibration were added in the results section as a short review: "*The calibrated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 3a), with a Nash-Sutcliffe efficiency (NSE) value of 0.70. Subsequently, the calibrated model was validated over the entire simulation period from 1997 to 2010 (~1 day of CPU time), with a NSE value of 0.55. Due to incomplete observation and the high CPU time, the NSE is still considered to be acceptable. The calibrated best-fit values for the transport parameters are listed in Table S1 [see in Supporting Information]. More detailed calibration results refer to Wang et al. [2023].*"

(3) Clarify model-data connection: A clearer connection could be made between the numerical model and real-world observations. The use of simple abstractions in the nutrient transport component of the model is reasonable, but also requires careful consideration of (a) how well the simplified processes representations mimic actual processes, (b) the uncertainty of the parameter estimates, and (c) the accuracy of the model in terms of reproducing observations. Otherwise, there is a risk of circularity: the model is built around certain processes and parameterizations, and then used to test the importance of those same processes and parameters.

Response#3:

Thanks for the suggestions. We made the following changes to clarify model-data connection:

1. We added the observed in-stream nitrate concentrations along with the simulated ones (see the figure below) such that they can be clearly compared.



2. We added a description of the calibration results to show the accuracy of the model in terms of reproducing observations as "The calibrated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 3a), with a Nash-Sutcliffe efficiency (NSE) value of 0.70. Subsequently, the calibrated model was validated over the entire simulation period from 1997 to 2010 (~1 day of CPU time), with a NSE value of 0.55. Due to incomplete observation and the high CPU time, the NSE is still considered to be acceptable. The calibrated best-fit values for the transport parameters are listed in Table S1 [see in Supporting Information]. More detailed calibration results refer to Wang et al. [2023]".

3. We added short discussion about the uncertainty of the parameter in the section of Discussion.

Line-by-Line comments:

(4) Line 14: "performance" is wrong word. "effect"?

Response#4:

Thanks! We changed "performance" into "manifestation".

(5) 34: SON not defined at this point

Response#5:

Thanks, we rephrased as "..SON (soil organic nitrogen)…".

(6) 39 – "a small effect"

Response#6:

We corrected accordingly.

(7) 58 – not clear what "their" refers to

Response#7:

We clarified it as "the effect of rainfall variability".

(8) 59 – grammar problem

Response#8:

We rephrased the sentence into "*anthropogenic amplification of rainfall variability has been identified [Zhang et al., 2024]*".

(9) 72 –"a major"

Response#9:

We corrected accordingly.

(10) 59-78 – the paragraph starts with climate variability and ends with nitrate. I recommend keeping to one topic per paragraph.

Response#10:

Thanks for the suggestion. We rephrased the content accordingly into three paragraphs.

(11) 99-100 sentence fragment

Response#11:

Thanks! The sentence was modified into "*Notably, the 2018 event triggered unprecedented tree mortality across multiple species in Central European forests, accompanied by unexpectedly persistent drought legacy effects [Schuldt et al., 2020]" (Lines 100-102)."*

Reference:

Schuldt, B., Buras, A., Arend, M., Vitasse, Y., Beierkuhnlein, C., Damm, A., Gharun, M., Grams, T. E. E., Hauck, M., Hajek, P., Hartmann, H., Hiltbrunner, E., Hoch, G., Holloway-Phillips, M., Körner, C., Larysch, E., Lübbe, T., Nelson, D. B., Rammig, A., Rigling, A., Rose, L., Ruehr, N. K., Schumann, K., Weiser, F., Werner, C., Wohlgemuth, T., Zang, C. S., & Kahmen, A.: A first assessment of the impact of the extreme 2018 summer drought on Central European forests, Basic Appl. Ecol., 45, 86–103, https://doi.org/10.1016/j.baae.2020.04.003, 2020.

(12) 105 – "it is"

Response#12:

We corrected accordingly.

(13) 107 – The objectives are somewhat broad. It has been established that precipitation variability can affect N dynamics, and it would help to be more specific in this paragraph about the aspects of variability being tested and what if anything has been done to address them previously. In other words, how the specific objectives of this study relate to gaps in knowledge left by previous studies?

Response#13:

Thanks for the suggestion. The paragraph was rephrased as "*In order to fill the gap, the present study explored the impact of rainfall variability on N dynamics and its potentially negative influence on water quality across inter-annual and intra-annual timescales. To characterize rainfall variability, a stochastic rainfall generator [Robinson & Sivapalan, 1997] was employed to generate rainfall time series with different climatic characteristics. The research was conducted in a small agricultural catchment located in Central Germany, where a hydrological model was previously established utilizing the fully coupled surface-subsurface numerical simulator HydroGeoSphere [Yang et al., 2018]. The framework of N dynamics was modified from the ELEMeNT approach [Exploration of Long-tErM Nutrient Trajectories; Van Meter et al., 2017]. The research is divided into two main components. First, three representative years (with high, normal, and low annual precipitation amounts, respectively) were chosen from the past two decades in Central Germany as the target scenarios. A fourth scenario with low annual precipitation amounts coupled with reduced plant uptake represents a case where vegetation is partially destroyed by extreme droughts. The statistical analyses of nitrogen loads and fluxes and a comparison across different scenarios were conducted to reveal the influence of inter-annual rainfall variability. Second, rainfall time series generated using the stochastic rainfall generator by separately altering specific parameters were used to substitute the rainfall data in the simulation period to drive the flow and nitrogen transport* *models. The responses of the N loads and fluxes to the parameters (e.g., the amplitudes of the seasonal variations in the storm durations and inter-storm period) were thoroughly analyzed to clarify the effect of intra-annual rainfall variability. The study will provide theoretical support to formulate fertilization strategies and protect aquatic ecosystems under climate change in the future."*

(14) 163: cross section is not discernable and it is not clear what is the source of the saturation values

Response#14:

Thanks for pointing that out. We modified Figure 1 and its caption accordingly:

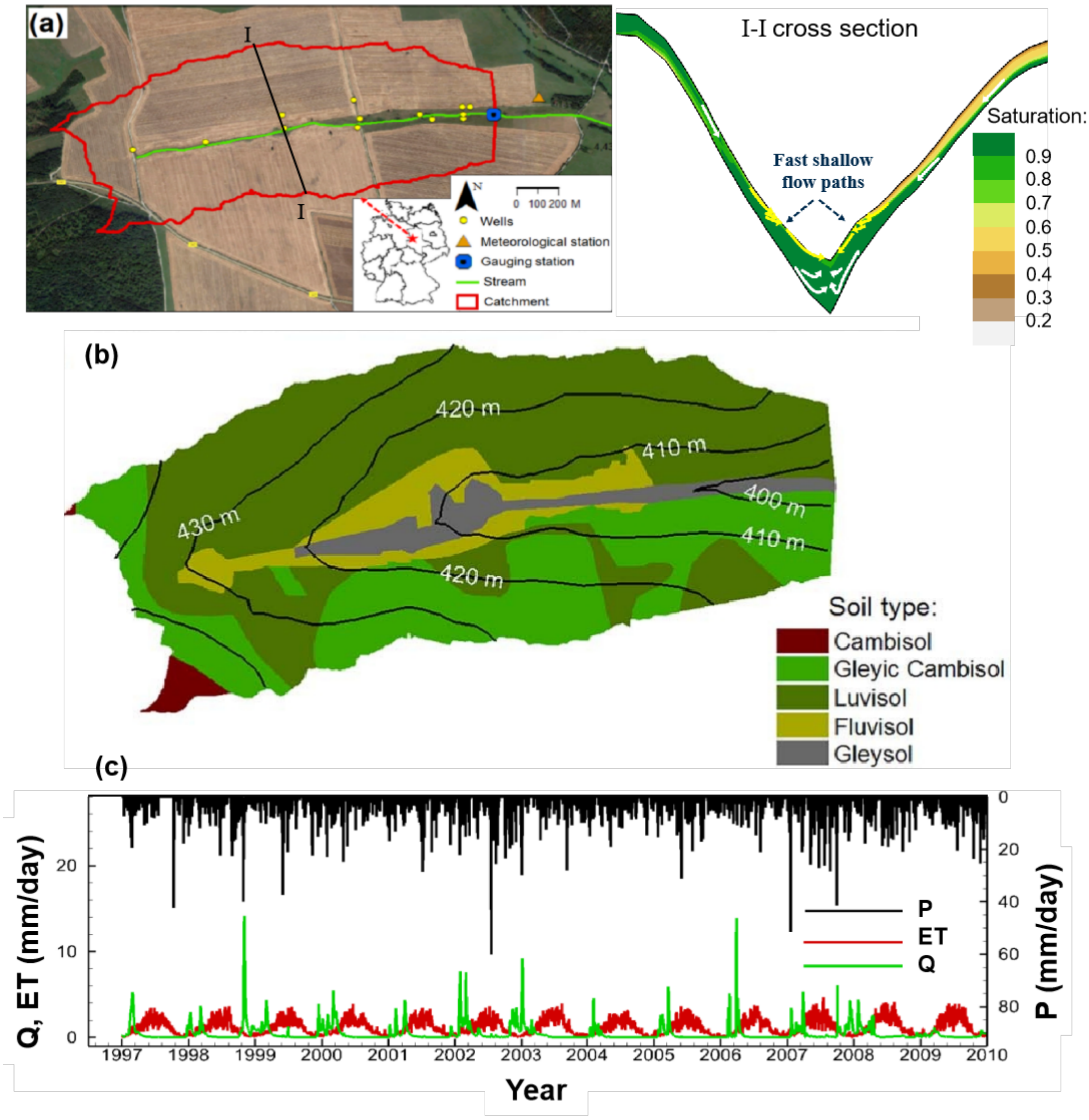


Figure 1. (a) The catchment 'Schäfertal' indicated by the red line (background image from © Google Maps), with a cross-sectional view for the flow and saturation [*Yang et*

al., 2018]. (b) The distribution of soil type in the catchment. (c) The measured daily precipitation (P), discharge (Q), and the simulated actual evapotranspiration (ET) [*Yang et al*., 2018].

(15) 164: this figure is mostly recycled from Wang et al., 2023 but no citation is given.

Response#15:

We added the reference "Yang et al. [2018]" accordingly.

(16) 169: Please clarify if/how these data are used in the current study.

Response#16:

Thanks for the suggestion.

We clarified that by adding " The C_Q and N surplus are used to calibrate the N transport model [Wang et al., 2023]".

(17) 192 – delete "in details"

Response#17:

We corrected accordingly.

(18) 231 – what does this mean that the calibrated model was "verified" over the entire simulation period?

Response#18:

Thanks! We rephrased the sentence into "*Subsequently, the calibrated model was verified by reproducing time-variable groundwater levels for the wells over the entire simulation period [Yang et al., 2018]*."

(19) 240 – meaning unclear "delineated corresponding to the reality"

Response#19:

Thanks! We have removed the unnecessary sentences.

(20) 245-6: "in route" grammar

Response#20:

Thanks for pointing that out. The sentence was modified to "Throughout the nitrogen cycle, various forms of nitrogen undergo complex biogeochemical processes".

(21) 258-260: meaning unclear.

Response#21:

We rephrased the sentence into "The framework is able to capture the main processes of nitrogen transformation and transport in soil and groundwater [Yang et al., 2018]"

(22) 320: "validation" might not be the right word (not the same as calibration).

Response#22:

Thanks for pointing that out. The sentence was modified to "Subsequently, the calibrated model was verified by reproducing time-variable groundwater levels for the wells over the entire simulation period [Yang et al., 2018]".

(23) 323: Wang et al 2023 refers readers to Yang 2018 for more details and is not an easy source of information about the estimation of the N cycling parameters, uncertainty of those parameters or the quality of fit to the data. These are crucial aspects of the calibrated model and should be presented clearly and succinctly for the readers.

Response#23:

Thanks for the suggestion. The brief calibration results were added in the beginning of result section as "*The calibrated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 3a), with a Nash-Sutcliffe efficiency (NSE) value of 0.70. Subsequently, the calibrated model was validated over the entire simulation period from 1997 to 2010 (~1 day of CPU time), with a NSE value of 0.55. Due to incomplete observation and the high CPU time, the NSE is still considered to be acceptable. The calibrated best-fit values for the transport parameters are listed in Table S1 [see in Supporting Information]. More detailed calibration results refer to Wang et al. [2023]*".

(24) 325: "impermeable for nitrate" (and water?)

Response#24:

We rephrased the sentence as "The bedrock is treated as impermeable for water and nitrate".

(25) 333-375: probably don't need this much detail about the rainfall generator

Response#25:

Thanks for the suggestion. The section 3.3 has been simplified.

(26) 336: "a stochastic model"

Response#26:

We corrected accordingly.

(27) Table 1 – This table has too many numbers and variables for readers to easily absorb. Consider placing with a schematic, examples, or another simpler figure or table.

Response#27:

Thanks for the suggestion. We updated table 1 accordingly.

(28) 399-401 – grammar problem, meaning is lost

Response#28:

Thanks for pointing that out. The sentence has been modified to "In order to explore the effect of intra-annual rainfall variability on N dynamics, the linear regression analyses between the parameters of the stochastic rainfall generator and N loads, fluxes, as well as C_Q were conducted, in which the parameters of NY serve as a reference".

(29) 458 – Why is soil denitrification lumped with GW denitrification? Are they expected to be similar?

Response#29:

Thanks for point that out. Soil denitrification and GW denitrification are identical in mechanism, and we expected to see how much nitrogen in total is consumed by denitrification under different rainfall pattern. Therefore, they are added together in the study.

(30) 503 – These figures are confusing because the response variables (SON, SIN, LEA, Cq) are not on the z-axes.

Response#30:

Thanks for the suggestion! Figure 8 and Figure 9 are presented in the same way. We will use Figure 8 as an example. In the stochastic rainfall generator, the distributions of the average storm duration and the average inter-storm period over the course of a year are depicted by Sine functions (S3 & S4 in the Supporting Information), whose characteristics are determined by the amplitudes of the seasonal variations in the average storm duration and the average inter-storm period (α_γ and α_δ) reflecting climate change, when other parameters keep invariable. As for a certain α_γ, there is a specific distribution of the average storm duration over the course of a year, which is presented in the x-z plane. The color of the panels represents the value of α_γ. With α_γ decreasing, the distribution of the average storm duration transitions and the year shifts from wet to dry. When the year becomes drier, the annual SON load increases, while the SIN load, leaching flux, and in-stream nitrate concentration decreases on the y-axes. Therefore, the transformation and transport of N are subject to retardation in a dry year when storms with shorter duration occur mid-year.

(31) 564: 5.1 section title: consider being more specific about what increased rainfall does to the N dynamics

Response#31:

Thanks for the suggestion. We changed section tile of 5.1 into " N transformation and transport upgrade in wet years ".

(32) 608: It seems notable that the high flows during the 2018 to 2019 drought are as high as the high flows from 2014-2018, and the main difference during 2018-2019 seems to be in the low flow periods.

Response#32:

Thanks for pointing that out. Yes, as you observed, droughts mainly occurred in the middle of the year, resulting in extreme low flow (Figure 12). During the low-flow periods, nitrogen absorbed by vegetation in the growth stage is significantly reduced and nitrogen (SON & SIN) starts to accumulate in the soil. The accumulated nitrogen was subsequently flushed out during the rewetting period [Winter et al., 2023].

Reference:

Winter, C., Nguyen, T., Musolff, A., Lutz, S., Rode, M., Kumar, R., Fleckenstein, J.: Droughts can reduce the nitrogen retention capacity of catchments, Hydrol. Earth Syst. Sci., 27(1):303-318, https://doi.org/10.5194/hess-27-303-2023, 2023.

(33) 670: 5.6. Consider discussing: Data limitations, uncertainty of parameters, model process representations

Response#33:

Thanks for the suggestions.

We added the discussion to address the limitation in data, model representation and parameter uncertainty as "*In addition, the spatial variation of denitrification rate coefficient may lead to uncertainty in the simulated results. Thus, the effect of temperature on denitrification rate should be investigated in detail and depicted in the framework of N dynamics. Due to the lack of groundwater nitrate concentration, the nitrate transport model was calibrated using only in-stream nitrate concentration data [Wang et al., 2023]. More nitrate concentration data in groundwater and river will contribute to establish a better nitrate transport model.*"

(34) Table S1 – van Meter reference is missing date; bibliography is not included in this document

Response#34:

Thanks for pointing that out. We have corrected the table S1:

Table S1. The parameters for the N source zone. The parameters were adjustable and calibrated (referring to Yang et al. [2022]).

Parameter	Process	Adjustable range	Reference	Value of best fit
k_a	Mineralization	0~0.7	Van Meter et al. [2017]	0.0109 day^{-1}
k_p	Mineralization	0~0.7	Van Meter et al. [2017]	0.0008 day^{-1}
$p1$	Plant uptake	60~160	Van Meter et al. [2017]	160 kg·ha^{-1}
$p2$	Plant uptake	1~10		10 kg·ha^{-1}
$p3$	Plant uptake	1~60		24 day
λ_s	Denitrification in soil	0~0.7	Nguyen et al. [2021]	0.0003 day^{-1}
k_l	Leaching	1~1000		3.3906 day^{-1}
q_{ref}	Leaching	$1e^{-4}$~1		0.0654 m·day^{-1}
λ	Denitrification in water	$1e^{5}$~$1e^{-1}$	Nguyen et al. [2021]	0.0088 day^{-1}