

Dear reviewer,

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

Best regards

Responses:

The authors present an original piece of research focusing on the catchment response of N fluxes to rainfall interannual and intra-annual variability using synthetic experiments based on the Hydrogeosphere model. I found the study clean and rigorously described, calibration method is sensible. Figures provide useful and clear illustrations of the results. The manuscript is well written. I think the discussion could be expanded and therefore I recommend a minor revision of the manuscript.

Especially I would have **2 comments on the discussion:**

1) several parts of the discussion present some reactions as simulation results when they are a direct consequence of the modelling equations. Ex.: Lines 566 to 568, the impact of temperature and wetness on mineralization is constructed mathematically in equations 1 and 2, isn't it? Same comment regarding plant uptake and denitrification. According to my opinion, the interest of the model is rather to calculate which of these mechanisms is going to dominate the response, and also it helps to consider different time scales of response, which is particularly relevant for droughts (cf. lines 97-99)

Response #1:

Thank you for the comment. We truly agree that the reaction is a direct consequence of the mechanisms that are built in the equations already. That mean, some trends / patterns can be simply known even without actually performing the simulations. This study is aimed to investigate how the rainfall variability influences the N transport and which

process is the key control of that influences. E.g, we found that the plant-uptake is the main factor caused the high in-stream concentration in extreme dry years.

In response to your comment, we have revised the discussion to clarify the dominant mechanisms governing in-stream nitrate concentration responses as “*The comparison of C_Q , N loads, and N fluxes among the four scenarios of the inter-annual rainfall variability reveals its effect on N dynamics and water quality. Mineralization, the crucial process of the transformation from SON to SIN , is highly sensitive to soil wetness. Consequently, the highest average mineralization rates, observed in WY scenario, promote the transformation of N , including plant uptake, denitrification, and leaching. In contrast, low mineralization in DY scenario leads to SON accumulation and thereby restraining overall N dynamics*”.

2) I have a general comment on the modelling choice that is N mechanisms are much more simplified than the water processes. While I am very aware of the computational challenges associated with such virtual experiments, I find intellectually disturbing to have a fully mechanistic approach to represent water combined with a representation of nitrogen very simplified in comparison. What do you think?

Response #2.1:

Thank you for this comment! We totally agree with your point that the N processes represented in the model is very simple compared with the groundwater flow. The complexities N fluxes in source zone were simplified by defining a framework describing the main pathway for $N-NO_3$ leachate with temporally constant external N input. On the one hand, this simplification neglected other processes such as the time-variant external N input, direct input of external N to inorganic N pool, the transformation from protected organic N to active organic N , and the loss of organic N via dissolution. On the other hand, this simplification allows us to focus on the main source of $N-NO_3$, rather than keeping track of the full nitrogen fluxes in the source zone of the catchment while maintaining the overall nitrogen balance using surplus as a constraint. In this sense, we think it is an effective tool to answer certain questions, for example in our study, which process controls the high nitrate concentrations in river

while dry climate.

To response to this point, we discuss the limitation of the simplified N framework additionally in section 5.6 in the revised manuscript.

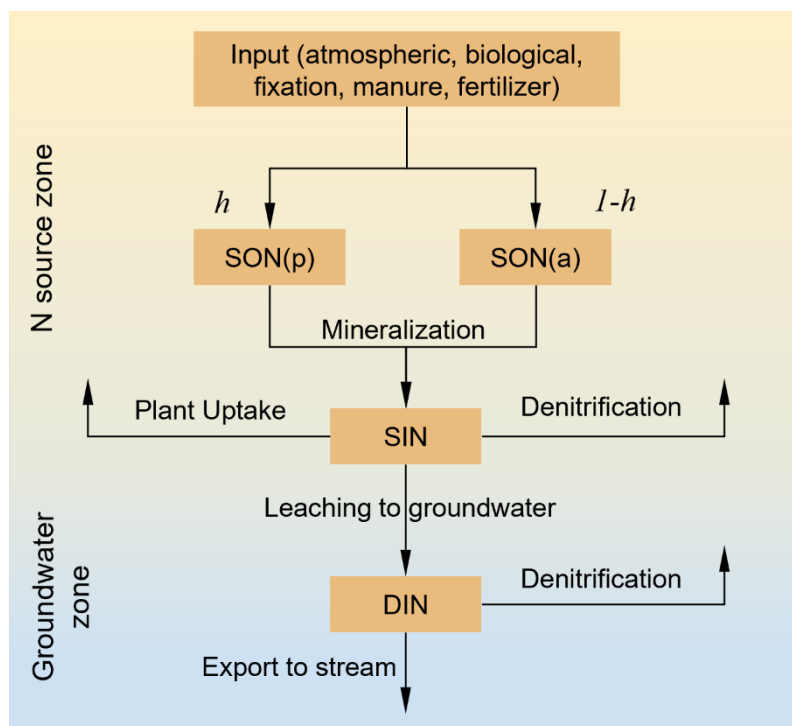


Figure 2. The framework simulating the nitrogen transport in soil and groundwater, modified based on [Yang et al., 2021].

There is no representation of plant-soil processes, and considering the results of the EDY scenario in terms of N dynamics, the response of plants growth or death to water stress seems to be a key mechanism.

Response #2.2:

Thanks for pointing that out! Yes, the plant-soil process is not represented in the model. This means, in the extreme dry scenario, the plants death was not actually simulated, but manually being assigned a lower N-uptake potential. The plant growth stages were assumed to be constant (for example, the same N uptake potential in different). This is also due to lack of representation for the plant-soil processes in the simplified N framework. N uptake process is only mathematically described using the empirical formula (Equation 6).

However, such simplification was proved to be effective in term of reproducing the

reasonable N loads and in-stream nitrate concentrations. It can still be used to identify the key effect of plant-die-off (i.e. reduced N uptake potential) on stream water quality during extreme dry climate, comparing with other processes such as mineralization and leaching.

To respond to this point, we discuss the limitation of neglecting the plant-soil processes in section 5.6 in the revised manuscript.

Also, about the fact that all external nitrogen inputs are introduced in the SON pool (lines 250, 252), I was wondering to what extent it refers to a reality? Are fertilizers mainly applied as urea?

Response #2.3:

Thanks for pointing that out!

The external N input represents atmospheric deposition, biological fixation, animal manure from the pasture area, and N fertilizer from the farmland. It does include organic and inorganic inputs. In our framework based on ELEMNT model [Van Meter et al., 2017], it is assumed that all external nitrogen inputs are introduced in the organic pool. This assumption is made based on the fact that most of the nitrate (N-NO_3) fluxes from the source zone have undergone biogeochemical transformation in the organic N pool (Haag and Kaupenjohann, 2001). The described framework simplifies complexities of different N pools and transformations via mineralization, dissolution, and denitrification within the soil zone, while preserving the main pathway for N-NO_3 leachate. In this sense, the assumption that the external N input contributes only to the SON is acceptable in the study.

To respond to this point, we discuss the limitation of the simplified N framework additionally in section 5.6 in the revised manuscript (also see our response above).

Also, I did not understand why is this framework acceptable and according to which criteria or arguments (line 258-260)?

Response #2.4:

Thanks for the comment!

Although the mechanisms of N dynamics in this framework are quite simplified, we

think it is acceptable for our study because:

- 1) It is effective in term of reproducing the reasonable N loads and in-stream nitrate concentrations in the catchment. The modeled in-stream concentrations fit well with the observations with a Nash-Sutcliffe efficiency (NSE) value up to 0.70 [Wang et al., 2023]. The simulated N mass balance was also meaningful: with an external N input of 180 kg ha/yr, the total load of SON and SIN were 468 kg ha⁻¹ and 34 kg ha⁻¹, respectively. The former accounted for 93% of the total N in the soil, being consistent with the values reported in Stevenson (1995) (organic N fraction > 90%). The mineralization rate was 179 kg ha⁻¹ yr⁻¹, which is within the range reported by Heumann et al. (2011) for study sites in Lower Saxony, Germany ([14 - 187] kg N / ha yr⁻¹). 67% of the SIN was absorbed by plants at a rate of 120 kg ha⁻¹ yr⁻¹, which is comparable to the value suggested in Nguyen et al. (2021) for the same area (around 120 kg ha⁻¹ yr⁻¹). The denitrification flux was 45 kg ha⁻¹ yr⁻¹ (4 kg ha⁻¹yr⁻¹ in the soil, 41 kg ha⁻¹ yr⁻¹ in the groundwater), which is within the range (8 - 51) kg ha⁻¹ yr⁻¹ reported in Hofstra and Bouwman (2005) for 336 agricultural areas around the world. The SIN leached into the groundwater at a rate of 55 kg ha⁻¹ yr⁻¹, which is comparable to the range (15 – 60 kg N ha⁻¹ yr⁻¹) reported in Nguyen et al. (2021). Finally, approximately 25% of the leachate was exported to the stream with the remaining 75% being removed by denitrification during the transport in the groundwater, which are comparable to the values 39% and 61% reported in Nguyen et al. (2021).
- 2) The described framework simplifies complexities of different N pools and transformations via mineralization, dissolution, and denitrification within the soil zone, while preserving the main pathway for N-NO₃ leachate. In this sense, the framework can still be used to explore the influence of rainfall on water quality via the key processes (mineralization, plant-uptake and leaching).

To response this point, we briefly describe the simulated N loads and in-stream nitrate concentrations, by comparing with observations or other literatures (which has been done in our previous Wang et al., 2023) in the revised manuscript.

I have also a few minor comments below:

3) line 41: I suspect that fertilization strategies will evolve with the changes in timing of plant growth stages with warming

Response #3:

Thanks! This is a very good point. We agree that the fertilization strategies may evolve with warming. However, in this study, the plant growth stages were assumed to be the same for different years (for example, the same sowing date in either wet year or dry year). This is due to lack of representation for the plant-soil processes in the model. Such assumption was proved to be effective in term of reproducing the reasonable N loads and in-stream nitrate concentrations [Yang et al., 2021], however, may leads to uncertainties in the simulated N fluxes.

4) introduction should maybe be restricted to context elements that are directly linked to the study, lines 45 to 61: it could go more directly to the point of rainfall variability skipping the details on extremes. Lines 79 to 82: I missed the link with the present study here.

Response #4:

Thank you for your suggestions. We have revised the first paragraph of the Introduction as “*The hydrological processes are susceptible to meteorological conditions on various spatial and temporal scales [Ionita et al., 2017; Laaha et al., 2017; Zhang et al., 2021]. In the past decades, extreme climate events intensified by human-induced climate change have frequently occurred globally [Pall et al., 2011; Min et al., 2011; Williams et al., 2015; Hari et al., 2020], most of which caused water scarcity and poor water quality at regional scales [Zwolsman and van Bokhoven, 2007; Delpla et al., 2009; Whitehead et al., 2009; Stahl et al., 2016; Ballard et al., 2019; Bauwe et al., 2020; Geris et al., 2022]. Heavy rainstorms and severe droughts being the predominant extreme climate events around the globe share the common characteristic of rainfall variability [Trenberth et al., 2011; Pendergrass et al., 2017; Hanel et al., 2018]. In the context of global warming scenarios, rainfall variability has been amplified*

anthropogenic [Zhang et al., 2024]. Thus, the effect of rainfall variability on water resources has attracted much attention around the world” (Line 47-59)”.

We also deleted the following sentences describing historical extreme events, which are not directly relevant to this study *“Extreme rainfall events in 2002, 2013, and 2021 caused significant threats to human safety and substantial damage to the environment, economy, and infrastructure [Ulbrich et al., 2003; Thielen et al., 2016; Voit & Heistermann, 2024]. Three notable summer droughts occurred in 2003, 2015, and 2018–2019 (consecutive), driven by precipitation deficits combined with temperature anomalies during the growing season [Fink et al., 2004; Schär et al., 2004; Ciais et al., 2005; Orth et al., 2016; Hanel et al., 2018; Hari et al., 2020; Camenisch et al., 2020].”*

5) lines 145-146: at which frequency are concentration measured?

Response #5:

Thanks! We clarified this by adding “..at 14-days to monthly intervals [Dupas et al., 2017], covering the period 2001–2010.”

6) line 339-340 what is a "good performance under Climate change" for the stochastic generator?

Response #6:

Thank you for raising this point.

Wilusz et al. [2017] coupled the rainfall generator [Robinson & Sivapalan, 1997], a rainfall-runoff model [Kirchner, 2009], and the rank StorAge Selection (rSAS) transit time model [Harman, 2015] to decompose the relationship between rainfall variability and the time-varying fraction of young water (<90 days old) in streams (FYW). The rainfall generator was specifically employed to synthesize daily rainfall time series with statistical properties that are identical with or systematically different from historic rainfall. Crucially, the generator is capable of representing individual storm events, between-storm variability, within-storm variability, and seasonality [Robinson &

Sivapalan, 1997]. Thus, based on these capabilities, the sentence was modified into “*It outputs rainfall series representing different rainfall patterns under climate change*”.

7) Figure 4 a: it would be useful to have measured C_Q in the plot too. The legend refers to "acceptable simulations" but so far as I understand it is more the variability associated to the generator in each scenarios (n=100) isn't it?

Response #7:

Thank you for the suggestion!

We have added the measured in-stream nitrate concentration data to Figure 3a. Indeed, gray areas in Figure 4(b1)~(b4) are formed by the 100 realizations. We changed the caption of Figure 4 to clarify this as “*The grey areas are formed by the realizations of each scenario*”.

8) lines 577-578: are preferential flowpaths represented in the model?

Response #8:

Thank you! Preferential flow paths on top of the aquifer (fast shallow flow paths) has been identified and discussed in our previous study [Yang et al., 2018], plotted in Figure 1a. Such fast shallow flow paths established a hydraulic connection between channel and the slope top in the wet seasons. We added “(I-I cross section, Figure 1a)” to refer to the flow paths.

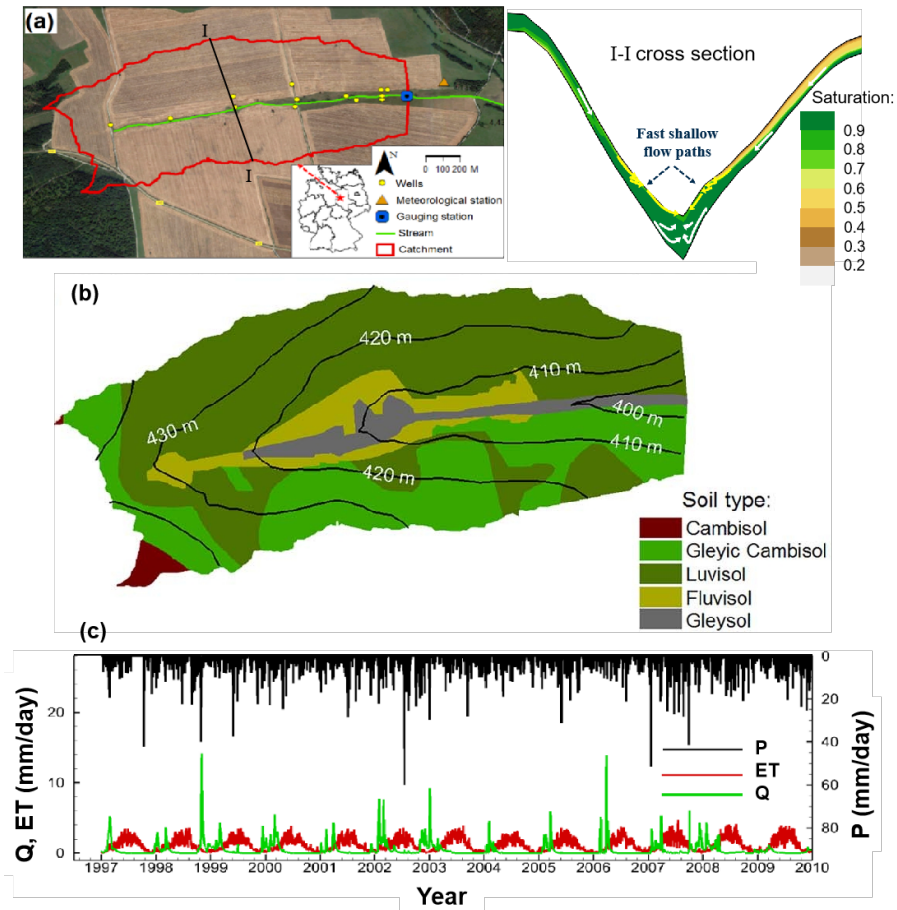


Figure 1. (a) The catchment ‘Schäfertal’ indicated by the red line (background image from © Google Maps), with a cross-sectional view for the flow and saturation [Yang *et al.*, 2018]. (b) The distribution of soil type in the catchment. (c) The measured daily precipitation (P), discharge (Q), and the simulated actual evapotranspiration (ET) [Yang *et al.*, 2018].

9) it would be useful to see the effect on water fluxes as well: especially recharge flux for groundwater and Actual ET in Figure 10 or Figure S2 (lines 653 to 656)

Response #9:

Thank you for your suggestion. We added the recharge fluxes (Rainfall-actual ET) and the actual ET in Figure S2 of the supporting information.