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Hydrology and Earth System Sciences Discussions

Interactive comment on "Decadal trajectories of nitrate input and output in three nested catchments along a land use gradient" *by* Sophie Ehrhardt et al.

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General response: We thank the referee for the valuable inputs and remarks. We address all comments below and hope to clarify the questions raised.

General comments: 1. The manuscript addresses the important issue of legacy stores of nutrients, which may prevent mitigation actions that reduce the inputs from having immediate effects on stream water quality. I like the date drive approach to investigate the travel times of nitrate. The paper shows that 85% of the N input is retained within the catchment. The investigation about the fate of this lost N is not very convincing and inconclusive. Based on data on inputs and outputs alone, the authors cannot proof

whether the N is retained in the soil, whether it is traveling along long flow paths, or whether it is denitrified. The authors try to give answers based on literature, but this is not very convincing. A weak point of the paper is that the entire soil and groundwater system is addressed as a black box. This is a bit strange given the focus on the paper on N –stores and travel times in soil and groundwater. Including data on e.g. groundwater heads and flow-paths and concentration depth profiles for N could provide more certainty about the fate of the lost N.

R1: We understand the reviewer remark. Our study can only hypothesize and argue about travel times (TTs) and legacy, which is unfortunately a common methodological challenge of studies on catchment scale. We treat the entire catchment including soil and groundwater system as a black box and try to understand the inherent processes by looking at the signals produced or altered by this box. By doing such a data-driven analysis, our aim is to provide observation-based evidence on the system input-output response behavior, which can then be a starting point for developing either more targeted field-based or model-based "mechanistic" studies. Groundwater measurements and soil profiles would be a great help to support the hypothesis, but those observational records are generally not available. We tried hard to overcome this lack of knowledge with a comprehensive literature review for our studied catchment and comparable study sites (see also response R4 for Referee #1).

Specific comments: 2. Title: Consider to leave out 'decadal'. I don't understand why you would only be looking at decadal trajectories

R2: Agree, we will drop "decadal" in the title.

3. Abstract: The abstract is rather long. Especially the description of the results (from "We show: : :"). Consider to start a new paragraph here to make the structure more clear. The conclusion statement is a bit weak. Management should both address longer term and short term N-loads. How does this change water quality management in practice?

R3: We will carefully review the abstract, shorten the result section and put a clear focus on consequences for management practice that results from our analysis.

4. From P3L4 until P4L22 the introduction reads like a description and a justification of the methods that you apply. It remains unclear what is not yet known from the existing scientific literature, why that is important, and what new science this paper brings.

R4: We will carefully review the Introduction part of the paper to highlight the suggested mentioned aspects: What is known? Why it is important? And what's new in our study? Note that we have partly covered on these later on at P4, L18 – L33. But we will follow your advice and make it clearer the importance and the new scientific messages conveyed from our work.

5. In P4L20 you state that "data-driven studies focused either solely on N-budgeting and legacy estimation or on TTs." What data-driven studies do you mean here? Why is this a problem / what problem do you solve by combining these? The referencing to Van Meter and Basu is quite excessive.

R5: We refer to the data-driven studies e.g. by Worral et al., 2015 and Dupas et al., 2016 (as stated in P4, L19). We need to further underline the advantage of combining the quantification of legacy and TT in one study (and from the same data base) to use the TTs to explain the legacy. We will revise the text concerning this aspect. In terms of studies cited: See also comment R10 for Referee #1 where we aim at including a greater variety of studies.

6. P2L11-12: here you state that the agricultural nitrogen input is still high since the 1980's. It did decrease in most EU member states since the '80s as a result of the introduction of manure legislation, didn't it?

R6: The reviewer is right in pointing this out that N-inputs, also from agricultural sources were significantly reduced (but they are still on a high level). We will rewrite the concerned sentence to correct this inaccuracy.

7. P2L26: "The evaluation of measures: : :" What evaluation of measures. This sentence is a bit hard to follow.

R7: Thanks - we will revise this sentence to make it clear.

8. P5L18: why is the region vulnerable to climate change?

R8: Yes, we will add the explanation on this. For details, see reply R13 to Referee #1.

9. P6L3: it's not clear where the 2 WWTP's are located. Can you add them to your map?

R9: We will add the locations to the map in Fig. 1.

10. P6L8: how much are agriculture and WWTP's (and other sources) contributing in %?

R10: This comparative N budgeting is more part of the result and discussion sections. We will add the numbers in Section 3.2 referring to the current export situation. To give an idea: currently the fraction of wastewater at the total catchment nitrate export is 14%. Note that this fraction is removed from the exported nitrate in our analysis to focus on the diffuse pathways only (see P12, L9-27).

11. Figure 1: the stream is not very clear on this map. R11: We will highlight the river system.

12. P7L5 :"artificially drained" Do you mean drained by open ditches or by subsurface tube drains? How much has subsurface tube drainage?

R12: Yes, we will differentiate between "open ditches" and "tile drains" in the sentence by adding corresponding percentages. While more than half of the drains in the mid-stream sub-catchment are tube drains, the downstream sub-catchment is much more dominated by open ditches.

13. Table 1: The fraction artificially drained (last row) is much lower downstream. I

would expect more artificial drainage in the downstream part of the catchment as this is usually the wetter part of the catchment. Is there a reason why there is less artificial drainage needed in the downstream part?

R13: Thank you for this remark. We think this is related to the hydro-climatic conditions. The downstream area is significantly warmer and dryer in comparison to the colder and wetter upstream areas (see also response R20 for Referee #1 on this issue). This is also reflected in discharge behavior - the strong drop of discharge contribution from the different sub-catchments as indicated in Table 2.

14. P8L30 ": : :we do not account for wastewater fluxes at this point: : :" Why is this legitimate? Is the wastewater N flux negligible?

R14: We focused on diffuse N pathways via soil and groundwater where the legacy accumulation and time lags between input and output can potentially occur. Therefore we discounted the point contribution from both WWTPs from our N-data prior to TT analyses. See also the reply R10 above for the contribution of the WWTPs,

15. Figure 2 and 3: shouldn't these figures be presented in the results section?

R15: We understand your remark, but we still favor these figures related to data presentation in this section as it is now in the manuscript (see a similar example in Tetzlaff et al., (2014)). It's a presentation of the measured raw-data, while the results present the derived aggregated concentration and fluxes after using the WRTDS method. We will adjust the concerned section heading to "Data and methods" so to make it more clear.

16. Figure 2c: It seems like the NO3 concentration is 0 around 2007 and at the end of the graph. Please check this. There also seems to be a regime-shift in this plot just before 2000. What happened?

R16: You are right, we will correct these data points dropping to zero in Fig. 2 (see comment to that in response R15 of the Referee #1). The visible regime-shift around

2000 is related to the changing C-Q relations at the time where the dilution pattern switches to the enrichment pattern (see also Fig.7 c1 and c2). We address that in section 3.5 and in the discussion.

17. P11L6: "flow-normalized concentrations" It is not clear here why you need flow normalization. Consider to bring forward the end of the paragraph. Why would you want to take out the impact of variable flow conditions?

R17: We will drop the wording "flow-normalized" from here as the reasoning and procedure for the normalization is explained later on in this section (P12, L5-8).

18. P11L9: I don't understand how you interpolate the bi-weekly/monthly data. ": : :using a flexible statistical representation for every day of the discharge record".

R18: We will carefully revise that section to make methods more clear. The interpolation is based on a regression model using Q as a predictor, a trend component and a seasonal (sinusoidal) component. This model is fitted for every day separately utilizing a weighted regression approach that weights observation before and after that day differently based on their relevance for that specific day. Details are given in Hirsch et al. (2010). We note a mistake in the references here, and will correct this in the revised manuscript (the citation Hirsch & DeCicco in the text refers to the R-package while in the reference list the according paper Hirsch et al. is cited).

19. P13L14: "purple line"!purple dashed line

R19: Thanks - we will change that in the revised manuscript.

20. P13L21: "peaked 1980" ! peaked in 1980 R20: Thanks - we will change that in the revised manuscript.

20. Table 2: It is hard to connect the numbers for the LFS and HSF contributions in the text (<10%, 33%) with this table. It would be better not to give the cumulative contributions, so for HFS: 21, 69, 10.

R20: We will revise the table to avoid confusion between cumulative and single sub-catchment information.

21. P15L11: I don't understand ": : :besides the statistical evaluation of the time series"

R21: Thanks - we will revise this sentence to make it clear.

22: P16L6-15: During the measurement period the catchment will partly export N-inputs from before 1970/76. This could be seen as the legacy of the period before the measurement period. The missing N described here adds to the legacy from before 1970/76.

R22: Your point is right and we are aware of this discrepancy. We tried to underline this problem by stating: "overlapping time period of in- and output". A more appropriate comparison of in- and output would only be possible with the exact knowledge of TTs. In this first view of input-output-differences, we took the corresponding years for a quantitative comparison. Later on in the conclusions (P27, L22-25), we shift the input to the output ("assuming the temporal offset of peak TTs between in- and output of 12 a") and quantify the imbalance between both. With an additional sentence in the concerned section, we will underline this difference in a better way.

23. P18L6: why are these TTs for all seasons taken together not presented?

R23: We will add these lines in Fig. 6.

24. Figure7b1: The concentrations seem to drop here, before the input drops. How is this possible?

R24: Of course input changes cannot affect output earlier on. We think this drop in riverine nitrate concentration around 1985 is rather related to the sharp stop of increasing N-input at the beginning to mid of the 1970s and the following decrease of inputs.

25. Figuret7c1: The higher concentrations in summer and fall during the peak around

1990 are surprising. This would indicate that the concentrations in deep groundwater with long travel times to surface water are higher than the concentrations in shallow groundwater with short travel times. Is this groundwater N that infiltrated in the midstream catchment and seeps up in the downstream catchment?

R25: We can understand your confusion, but as explained in Section 4.2, the higher concentrations downstream in summer and fall are result of different nitrate source contribution during LFS and HFS. HFS-signals downstream are dominated by contributions from the wetter midstream sub-catchment with higher discharge per area and generally lower concentrations (see also Table 2 and similarity of midstream and downstream high flow concentrations shown in Fig. 7), whereas the low flow concentrations are dominated by the groundwater discharging from the downstream sub-catchment with much lower groundwater recharge and likely higher groundwater nitrate concentrations.

26. Figure 7a2-c2: add a legend. R26: We will add the color gradient to Fig. 7.

27. P20L9: refer to figure 7a2. R27: We will add the suggested reference.

28.P20L7-P21L17: This text in combination with figure 7 is quite a hard puzzle.

R28: Thank you for that comment. We will carefully revise this section and pay close attention to the information needed for the discussion later on.

29. P22L1: "was difficult" ! "was impossible" R29: Thanks, we will revise that sentence.

30. P22L1-2: Degradation of organic matter may play a role.

R30: You are right. We add this aspect in the sentence.

31. P22L17-20: I don't understand why "steeper terrain suggests a deeper infiltration" and "leaching of NO3 from a wider depth range than flat terrains". I would expect the opposite; deeper infiltration and leaching from a wider depth range in flat terrains. Of course, this depends on the geology.

R31: We would like to refer the paper by Jasechko et al. (2016) at this point: “Conversely, the reduced prevalence of young streamflow in steeper terrain suggests that steeper landscapes tend to favor deeper vertical infiltration rather than shallow lateral flow. A tendency for greater infiltration in mountainous watersheds may seem counter-intuitive, but is consistent with conceptual models of runoff generation and groundwater flow that suggest that topographic roughness drives long groundwater flow pathways that bypass first-order streams.”

32. P22L26:”to for an” ! ”for an”

R32: Thanks, we will change this as suggested.

33. P23L9-10: “Hence,: : :output” I think that this conclusion that denitrification is weakly supported by the previous text. Groundwater quality measurements would be very useful here.

R33: We hope to improve the overall argumentation through a support by findings in another study Hannappel et al. (2018) who analyzed groundwater and an enhanced discussion of the isotope evidence by Müller et al. (2018) – see more information in response R4 for Referee #1.

34. P23L16: why did Kuhr et al exclude denitrification?

R34: We would drop this “grey” citation (this is a report, not peer reviewed) in our manuscript and refer to Hannappel et al (2018) – see the previous comment R33.

35. P24L4-9: from this paragraph and especially the last 2 sentences it seems like it is not important whether the legacy store is growing or the denitrification capacity is used, however on P22L23-25 you stated that this difference is important.

R35: We will make our point clearer in the revised text. With the long-term data collection, we can only hypothesize whether the missing N is stored or denitrified, although it would be important for management. Beside management advices, we can show that the catchment N-input is unsustainable high, either due to the ongoing build-up of an

even bigger legacy or due to relying on a denitrification capacity which is unlikely to be infinite.

36. Figure 8: This figure does not make any sense to me.

R36: We will revise this figure to make our conceptual understanding of the catchment more clear.

37. P25L3-5: I don’t think that you can make this assumption; the flow contributions from a certain depth can vary a lot due to interannual variability

R37: We don’t think that there is evidence of a long-term change of flow paths in the catchment. Hydroclimatic conditions did not change; land use, topography and river network are stable over the long observation period. We will add these aspects here to better justify our assumption.

38. P26L3: You can also argue that groundwater seeping up is more important in the downstream catchment. This would mean more discharge of relatively old water.

R38: The TTs in the downstream part are shorter than those in Midstream, and not the other way round. Our argumentation is based on the greater prevalence of young streamflow in flatter terrain as shown also by Jasechko et al. (2016). See also response R31.

References uses (that are not in the main manuscript) âĂć Hannappel, S., Kopp, C. and Bach, T. (2018) Characterization of the denitrification potential of aquifers in Saxony-Anhalt. Grundwasser 23(4), 311-321. âĂć Tetzlaff, D., Birkel, C., Dick, J., Geris, J. and Soulsby, C. (2014) Storage dynamics in hydropedological units control hillslope connectivity, runoff generation, and the evolution of catchment transit time distributions. Water Resources Research 50(2), 969-985.

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