

Author's response to comments by anonymous referee #4

We would like to thank referee #4 for taking the time to carefully read our manuscript and for providing very constructive feedback. The helpful comments will improve the quality of our manuscript. Our responses to the individual comments are shown in blue below.

Summary The manuscript by Greiwe et al. describes a spatially-repeated sampling of diel variation in nitrate export along a reach in an intermediate watershed. The authors collected high-frequency diel nitrate concentrations from three stream stations, and quantified the magnitude of diel amplitude and estimated the travel times between stations. The authors used a cross-correlation approach to conclude that instream processes controlled emergent diel signals, and were minimally driven by upstream inputs. Overall, I enjoyed the paper, as it presents a means to interpret an essential ecohydrological question: which is more important, the physical or biological context, and when do these abiotic/biotic controls matter most? It is also an interesting way to use spatially-explicit data, especially that which is emerging from the application of high-frequency sensors. I found the topic highly relevant, especially as high-frequency hydrochemistry paired with discharge is becoming more widely available, and questions about source pathways and mixing have become a topic of interest of the research community.

However, there were some points of confusion that I hope the authors can clarify in a revision. I have several main comments, and some minor ones mainly focusing on improving clarity of the manuscript, that I hope the authors find insightful.

Major Comments

(1) While I am intrigued by the paper, one issue is that the authors overplayed the role of microbial processing. While this is generally assumed to be the case, this is still a “black box” situation with no microbial processing measured directly. I encourage the authors to take greater care in describing their findings and the assumptions of their interpretations, which as written are overly speculative.

Reply: We agree that some of our claims are not satisfactorily backed up with data and will tone down our conclusions. We will also delete the reference to microbial processes in the title as suggested by some of the other referees.

(2) How were tributary inputs accounted for in the authors' approach (based on Figure 1 there were some small inputs in between monitoring stations)? Part of the difficulty in parsing apart nitrate removal/production processes is the fact that there is mixing happening from multiple landscape units, which are hydrologically mixed as tributaries meet, and it was not clear how this variability in inputs was accounted for in the authors approach.

Reply: We agree that tributaries may alter diel stream nitrate patterns, and potentially even more critical, mass balances. There was only one surface input between S1 and S2. The influence of this tributary will be discussed in more detail in a revised manuscript. The streams visible outside the catchment boundaries in figure 1 in fact run parallel to the studied reach and enter the main stream downstream of S3.

(3) While the approach of using a time lag is compelling, I am curious if the authors had thought about the distributions of travel and reaction times in this study? The assumption of a mean travel time or reaction rate is to capture ‘average’ behavior and likely represents what is generally happening, but the use of a single value assumes that either transport or removal processes influencing what water/solutes make it to a point in the watershed network are occurring at a single rate. I am not encouraging the authors to use this approach, but it should likely be discussed as a

potential limitation of the study. Somewhat relatedly, why are there negative travel times in Figure 2, do you mean this to be the time lag?

Reply: We agree that our approach is a simplification. In the real world, transport will be influenced by dispersion due to non-uniform cross-sectional flow profiles, hyporheic exchange and possibly other processes. These simplification will be discussed in a revised manuscript. Negative travel times in Figure 2, indeed, result from the time lag. We will make this clear and revise this figure according to the suggestion made by referee #1.

(4) The authors could significantly shorten the discussion, as many of the processes mentioned were not directly measured and so the discussion does not need to be as nuanced as it is. Instead, presenting this as an open “call for the community” might be a more appropriate approach. Alternatively, one suggestion would be for the authors to develop a conceptual diagram of diel patterns in their watershed, indicating the open questions on the processes that the authors did not directly measure but infer as important instream drivers. Not only would this figure be useful for the community to visualize nitrate processing/transport in this system, but also likely hone the discussion around what is “known” and what is yet “unknown”.

Reply: We agree that our discussion is too elaborate which was criticized by all referees. In a revised manuscript, we will shorten its speculative parts and stick closer to what we can really show with our data. Thank you for the suggestion with the conceptual diagram. This is a nice way to illustrate the key points of our considerations and we will sketch one in our revised manuscript..

Minor Comments and Line-by-Line Suggestions

P1, Line 10: Change to “allow calculation”

Reply: Thank you, this will be adjusted.

P1, Line 15: Omit “suggested”

Reply: Thank you, “suggested” will be deleted

P2, Line 50: Please define insolation

Reply: We are referring to solar irradiance here and will clarify this in a revised manuscript.

P4, Section 2.2: Please describe in further detail how the SSC data were calibrated and turbidity-corrected.

Reply: A detailed description of the calibration procedure will be added.

P5, Section 2.3.1: Were the time lags / mean travel times estimated at the same intervals as the SSC data (i.e., did they also account for high/low Q, or are they averaged for a day)? Did you measure Q continuously at all three stations? Some additional clarity is needed here on time-scale and context for when travel times were estimated.

Reply: The cross-correlation approach yields one value per day. Although higher frequencies may be technically feasible using a floating window, lag estimates will be dominated by the lag between the strongest concentration excursions in the input signals. Higher frequencies will therefore not produce more information. Travel time estimates were based on daily means of water level recordings (15 min intervals, same as SSC probes) at the lowermost site S3 – no water level recordings are available from the other sites. We will improve clarity both in the text and also, as suggested by referee #1, by including a figure showing our raw data.

P11, Line 223: This sentence seems to come out of nowhere, I’d delete or expand on this idea before describing the Hensley & Cohen paper.

Reply: We will revise this paragraph to make it more understandable. This has also been noted by the other referees.

Figures & Tables

Generally, I thought the figure legends needed to have much greater detail. For example, in the caption for Figure 2, r should be more clearly defined. I also wouldn't put the shading for the nominal travel time on the figure, as this looks like a regression or confidence interval. In Figure 3, the letters should be defined in the figure legend and the confidence interval should be described. Additionally, in the spirit of inclusivity, I encourage the authors to check that their figures are color-blind friendly.

Reply: Thank you for pointing out the deficits in our figures. The issues with figure 2 and 3 have also been noted by the other referees and will be addressed correspondingly. We tried to act in the spirit of inclusivity by using the viridis color scheme, which was intentionally designed as color-blind friendly. However, we realize that contrasts between colors could be higher and will do our best to improve this.