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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.

Anonymous Referee #1

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This paper entitled, "Decadal trajectories of nitrate input and output in three nested catchments along a land use gradient" by Erhardt and others reports the result of a long-term nitrogen budget constrained with modeled and empirical estimates of N surplus and hydrological export. The authors found that most of the N was retained and attribute this mainly to hydrological legacy (storage within the catchmentâĂŤprimarily in the aquifer), based on a lack of observed denitrification in other studies, and the changes in the concentration-discharge relationships. The study is compelling and brings up many societally-urgent issues about preserving water quality in the Anthropocene. The paper is generally well written, though sections are uneven in their detail (either going too much into the weeds or not giving enough detail). I had a few major

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Discussion paper

questions and then provide line edits below. I believe that with a thorough revision, this article could be an important contribution to this journal. 1. How is error propagated? The authors often report four significant figures, but do not report standard deviation, confidence intervals, or some other estimate of uncertainty. Given the compound assumptions of the input chronicle models and the hydrological components, a sensitivity analysis or some kind of quantification of uncertainty seems warranted. 2. The idea of comparing biogeochemical and hydrological legacies is very compelling but it remains unclear to me how these parameters were estimated and compared. Structuring the methods around the research questions or overarching hypotheses and carrying this through the manuscript would make this flow clearer would make the results/discussion more impactful. 3. I think the discussion would be more engaging if the authors focused on the applicability of this approach to catchments generally, rather than explaining specific observations from their study. They do this effectively several times (e.g. starting on page 22 starting around line 20), but there is also quite a bit of retreatment of the results, which are specific to these sites. 4. The authors present an interesting puzzle of massive nitrogen retention/removal that cannot be attributed to typical pathways (e.g. denitrification, uptake, mineral association). The authors then conclude that N storage (the biogeochemical and hydrological legacies) account for the disconnect. However, the dismissal of denitrification seems to be based on a few studies from this area, which are not described in detail (e.g. Page 23, line 15). If these other studies are definitive and reliable, more description of their methods should be given. Another explanation is associated with point 1âĂŤcould the N removal be much lower when uncertainty in inputs and outputs are included?

Line edits Page 2 Line 5: (Elser et al. 2007) Line 6: It seems odd to say these changes were strictly terrestrial. It seems they influenced both. Line 10: Do the authors mean the natural rate of reactive N fixation has been doubled (e.g. (Vitousek et al. 1997))? Page 3 Line 2: management interventions (instead of “measures”)? Line 2: Recent study from similar agricultural and climatic context that found decadal hydrologic (Kolbe et al. 2016; Marçais et al. 2018) Line 16: I actually think there are quite a few studies, especially recently (Dupas et al. n.d.; Howden et al. 2010; Burt et al. 2011; Minaudo et al. 2015; Meter & Basu 2017; Abbott et al. 2018; Coble et al. 2018; Garnier et al. 2018; Marcé et al. 2018; Pinay et al. 2018; Fanelli et al. 2019) Line 20: How do these analyses compare with soil-surface N balance approaches that include a crop and livestock removal component (Poisvert et al. 2017; Abbott et al. 2018)? Line 30: Recent paper on concentration-discharge responses to catchment saturation (Moatar et al. 2017) Page 5 Line 18: In what dimensions is this catchment especially vulnerable to climate change? Page 8 Line 13-20: Interesting that the primary datasets do not include non-agricultural land for N deposition. Why did the authors not use one of the products that provided a consistent N deposition rate across land-use types? Perhaps this is a small portion of the overall N budget, but it would be worthwhile to specify. Page 9 Figure 2: The dissimilarity in the NO3 concentration time series is striking as are the drops to zero mg/L even at the lowest site. Consider combining Figures 2 and 3 to allow visual comparison of discharge and concentration. Page 10 Line 9: the discharge time series were used. . . Page 11 Line 8: allows increasing . . . Page 12 Line 10: Because our purpose was to balance and compare . . . Line 12: This justification seems unclear. Is it simply claiming that the longer-term trends are accurate, though the daily values are not? Page 14 Table 2: These differences in specific discharge are remarkable. Is this typical for this area or is the three-fold difference due to a known environmental or anthropogenic variable? Page 15 Line 11: Revise sentence for grammar and clarity (with implications for instead of with discussion on?) Page 16 Line 14: It is striking that the retention capacity increases 5-fold with landscape position. Is this because of shifts in soil and subsurface properties or because the retention or removal rates are dependent on substrate concentration? Page 22 Line 20: Nitrification also results in gaseous N loss via the “leaky pipe” pathway (Hart et al. 1994). Line 29: Is this referring to denitrification in the near-surface zone or throughout the whole catchment? With pyrite, sulfur, and other iron ubiquitous in the weathered and fractured zones, aquifer denitrification is likely occurring Page 23 Line 18: New methods for constraining aquifer travel time to constrain removal rates using numerical or empirical methods (Kolbe et al.

2016; Marçais et al. 2018). Page 25 Line 1: Similar to these observations, though they are on a much smaller scale (Thomas & Abbott 2018) Page 28 Line 9: were explained Line 14: catchment reaction seems like an odd description for transit time.

Citations: Abbott, B.W., Moatar, F., Gauthier, O., Fovet, O., Antoine, V. & Ragueneau, O. (2018). Trends and seasonality of river nutrients in agricultural catchments: 18 years of weekly citizen science in France. Science of The Total Environment, 624, 845–858. Burt, T.P., Howden, N.J.K., Worrall, F. & McDonnell, J.J. (2011). On the value of long-term, low-frequency water quality sampling: avoiding throwing the baby out with the bathwater. Hydrological Processes, 25, 828–830. Coble, A.A., Wymore, A.S., Shattuck, M.D., Potter, J.D. & McDowell, W.H. (2018). Multiyear Trends in Solute Concentrations and Fluxes From a Suburban Watershed: Evaluating Effects of 100-Year Flood Events. Journal of Geophysical Research: Biogeosciences, 123, 3072–3087. Dupas, R., Minaudo, C., Gruau, G., Ruiz, L. & GascuelâĂŘOdoux, C. (n.d.). Multidecadal Trajectory of Riverine Nitrogen and Phosphorus Dynamics in Rural Catchments. Water Resources Research, 0. Elser, J.J., Bracken, M.E.S., Cleland, E.E., Gruner, D.S., Harpole, W.S., Hillebrand, H., et al. (2007). Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. Ecology Letters, 10, 1135–1142. Fanelli, R.M., Blomquist, J.D. & Hirsch, R.M. (2019). Point sources and agricultural practices control spatial-temporal patterns of orthophosphate in tributaries to Chesapeake Bay. Science of The Total Environment, 652, 422–433. Garnier, J., Ramarson, A., Billen, G., Théry, S., Thiéry, D., Thieu, V., et al. (2018). Nutrient inputs and hydrology together determine biogeochemical status of the Loire River (France): Current situation and possible future scenarios. Science of The Total Environment, 637–638, 609–624. Hart, S.C., Stark, J.M., Davidson, E.A. & Firestone, M.K. (1994). Nitrogen mineralization, immobilization, and nitrification. Methods of Soil Analysis: Part 2âĂŤMicrobiological and Biochemical Properties, 985–1018. Howden, N.J.K., Burt, T.P., Worrall, F., Whelan, M.J. & Bieroza, M. (2010). Nitrate concentrations and fluxes in the River Thames over 140 years (1868–2008): are increases irreversible? Hydrological Processes, 24, 2657–2662. Kolbe, T., Marçais, J., Thomas, Z., Abbott, B.W., de Dreuzy, J.-R., Rousseau-Gueutin, P., et al. (2016). Coupling 3D groundwater modeling with CFC-based age dating to classify local groundwater circulation in an unconfined crystalline aquifer. Journal of Hydrology, 543, Part A, 31–46. Marçais, J., Gauvain, A., Labasque, T., Abbott, B.W., Pinay, G., Aquilina, L., et al. (2018). Dating groundwater with dissolved silica and CFC concentrations in crystalline aquifers. Science of The Total Environment, 636, 260–272. Marcé, R., Schiller, D. von, Aguilera, R., Martí, E. & Bernal, S. (2018). Contribution of Hydrologic Opportunity and Biogeochemical Reactivity to the Variability of Nutrient Retention in River Networks. Global Biogeochemical Cycles, 32, 376–388. Meter, K.J.V. & Basu, N.B. (2017). Time lags in watershed-scale nutrient transport: an exploration of dominant controls. Environ. Res. Lett. Minaudo, C., Meybeck, M., Moatar, F., Gassama, N. & Curie, F. (2015). Eutrophication mitigation in rivers: 30 years of trends in spatial and seasonal patterns of biogeochemistry of the Loire River (1980–2012). Biogeosciences, 12, 2549–2563. Moatar, F., Abbott, B.W., Minaudo, C., Curie, F. & Pinay, G. (2017). Elemental properties, hydrology, and biology interact to shape concentration-discharge curves for carbon, nutrients, sediment, and major ions. Water Resour. Res., 53, 1270–1287. Pinay, G., Bernal, S., Abbott, B.W., Lupon, A., Marti, E., Sabater, F., et al. (2018). Riparian Corridors: A New Conceptual Framework for Assessing Nitrogen Buffering Across Biomes. Front. Environ. Sci., 6. Poisvert, C., Curie, F. & Moatar, F. (2017). Annual agricultural N surplus in France over a 70-year period. Nutrient Cycling in Agroecosystems, 107, 63–78. Thomas, Z. & Abbott, B.W. (2018). Hedgerows reduce nitrate flux at hillslope and catchment scales via root uptake and secondary effects. Journal of Contaminant Hydrology, 215, 51–61. Vitousek, P.M., Aber, J.D., Howarth, R.W., Likens, G.E., Matson, P.A., Schindler, D.W., et al. (1997). Technical Report: Human Alteration of the Global Nitrogen Cycle: Sources and Consequences. Ecological Applications, 7, 737.

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