



The need to integrate legacy nitrogen storage dynamics and time lags into policy and practice



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HIGHLIGHTS

- Nitrogen (N) pollution from agriculture has negative environmental impacts.
- Environmental benefits of initiatives to reduce N loads not always detectable.
- N storage dynamics and time lag invalidate steady state models often used in policy.
- Researchers should advocate for integrating N stores and time lags into policy.
- Quantifying N storage aligns with phosphorus and carbon cycling research.

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ABSTRACT

Increased fluxes of reactive nitrogen (N_r), often associated with N fertilizer use in agriculture, have resulted in negative environmental consequences, including eutrophication, which cost billions of dollars per year globally. To address this, best management practices (BMPs) to reduce N_r loading to the environment have been introduced in many locations. However, improvements in water quality associated with BMP implementation have not always been realised over expected timescales. There is now a significant body of scientific evidence showing that the dynamics of legacy N_r storage and associated time lags invalidate the assumptions of many models used by policymakers for decision making regarding N_r BMPs. Building on this evidence, we believe that the concepts of legacy N_r storage dynamics and time lags need to be included in these models. We believe the biogeochemical research community could play a more proactive role in advocating for this change through both awareness raising and direct collaboration with policymakers to develop improved datasets and models. We anticipate that this will result in more realistic expectations of timescales for water quality improvements associated with BMPs. Given the need for multi-nutrient policy responses to tackle challenges such as eutrophication, integration of N stores will have the further benefit of aligning both researchers and policymakers in the N community with the phosphorus and carbon communities, where estimation of stores is more widespread. Ultimately, we anticipate that integrating legacy N_r storage dynamics and time lags into policy frameworks will better meet the needs of human and environmental health.

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1. Introduction

Nitrogen (N) is an essential macronutrient, fundamental for growth in both plants and animals (Schlesinger, 2005). Agricultural intensification and associated N fertilizer use has underpinned the world's

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growing population, resulting in a doubling of reactive N (N_r) fluxes in the environment (Vitousek et al., 1997). Increased N_r fluxes have generated negative consequences for both human and environmental health, leading to costs associated with eutrophication and drinking water treatment alone in the billions of dollars per year (Dodds et al., 2009; House of Commons Environmental Audit Committee, 2018; Pretty et al., 2000).

In response to the ecological impacts of increased N_r fluxes, best management practices (BMPs) have been implemented to reduce N_r fluxes in catchments. Some studies have shown BMPs to reduce nutrient export at the field to plot scale (Liu et al., 2017). However, at the catchment to basin scale, in many cases, the anticipated benefits of work to reduce N_r fluxes have not been realised (Hamilton, 2012; Van Meter et al., 2018). For example, despite millions of dollars spent on implementation of best management practices (BMPs) to reduce N_r loadings from agricultural sources, the Gulf of Mexico hypoxic zone was the largest ever recorded in 2017, with the target date to reduce the size of the dead zone delayed to 2035. These observations at the catchment scale emphasise the need for the scientific community to address the apparent disconnect between action and environmental benefit in the case of N_r .

2. Disconnect between action and benefit at the catchment scale: evidence for legacy N_r storage dynamics and time lags

What is causing the apparent disconnect between actions and catchment scale benefits in the case of N_r , despite some observations of benefits at the local scale? There is now a compelling body of scientific evidence from both field and modelling research that demonstrates legacy N_r storage in different compartments of the environment. Entry and subsequent release of N_r from these stores can result in significant time lags in the environmental benefits of actions designed to reduce new N_r loads to the environment. The dynamics of legacy nitrogen storage and impacts of N_r release from stores on water quality have been shown to be significant in Europe (Ascott et al., 2016; Bell et al., 2021; Durand et al., 2011; Howden et al., 2011; Vero et al., 2018; Wang et al., 2016; Worrall et al., 2015), Asia (Jia et al., 2018; Turkeltaub et al., 2020; Wu et al., 2020; Wu et al., 2019), North America (Ator et al., 2020; Martin et al., 2021; Sprague et al., 2011; Tesoriero et al., 2013; Van Meter et al., 2016; Van Meter et al., 2018) and globally (Ascott et al., 2017; Chen et al., 2018; McCrackin et al., 2017; Xin et al., 2019). In the past delays in meeting water quality objectives due to time lags and legacy storage dynamics have been dismissed as a generic excuse (Schaure and Naus, 2010). More recently, however, policymakers are increasingly aware of the role of legacy storage in controlling the efficacy of BMPs at the catchment scale (e.g. House of Commons Environmental Audit Committee (2018); Meals et al. (2010); Stuart et al. (2016)).

Whilst there is now strong evidence for legacy N_r storage dynamics and increasing awareness of this amongst policymakers, a major challenge remains in how nutrient legacies are represented in models and budgets used in practice for decision making. A number of conventional modelling tools that inform policy and practice that underpins N management at the catchment scale invoke the steady state assumption (e.g. SPARROW, PolFLOW, SAGIS, SEPARATE, NEAP-N, see Chen et al. (2018) for a recent summary of approaches). These models have been used to make decisions regarding control of N_r sources in the environment in order to reduce the risk of environmental damage, alongside predicting the trajectory for recovery of the environment where impact has already occurred. Interventions made on the basis of these tools have not always been successful over predicted timescales, with time lags associated with legacy storage dynamics invalidating the steady state assumption over short (<50 year) timescales. There are also discrepancies between research and practice regarding the definition of the term 'store', with some practitioner studies (United States Environmental Protection Agency, 2011) reporting a store as flux, whilst the academic research community often deals with stores in terms of mass (Chen et al., 2018; Van Meter et al., 2016).

3. The need for policy advocacy by the biogeochemical research community

Based on the body of scientific evidence highlighted above, we argue that the biogeochemical research community could play a more proactive role in advocating for integration of legacy storage dynamics and time lags into N_r management strategies in policy and practice (Fig. 1). We envisage that this would consist of both awareness raising and direct collaboration to develop the next generation of datasets and models to support decision making regarding BMPs.

3.1. Awareness raising

Whilst there is now some understanding in the policymaking community about the importance of legacy storage dynamics, we believe that researchers should continue to raise awareness of the issue, particularly amongst practitioners working in areas where implementation of BMPs is relatively recent and rapid improvements may be desired. We envisage that researchers could have direct engagement and discussions with policymakers, contributions to government enquiries, committees (e.g. Ascott and Ward (2018)) and evidence syntheses. Engagement at the local and regional level with key stakeholders (e.g. farmers, agri-environmental community groups) may also be beneficial.

3.2. Data and model development

Beyond awareness raising, we believe researchers should collaborate directly with policymakers to develop the next generation of datasets and models to support BMP decision making. Initial requirements for such collaboration would be to ensure a consistent terminology across both research and practice regarding stores (e.g. as a mass in kg N), and sharing of existing models and datasets used in N biogeochemical research with practitioners. Historic monitoring networks have often been poorly set up to address legacy storage dynamics and associated time lags (England et al., 2008; Hamilton, 2012), and reviews of impacts of BMPs at the meso-scale have highlighted the need for long term monitoring to assess water quality changes (Melland et al., 2018). Development of co-designed monitoring networks that quantify long term fluxes to and from N_r stores and their magnitude would be beneficial. For example, this could consist of porewater profiles in the unsaturated zone and soil N storage measurements, repeated every 5–10 years. Such monitoring would quantify reductions in the magnitude of these N_r stores and provide the initial evidence that changes in management practices designed to control N_r fluxes are having the desired effect. This would provide a sentinel indicator of potential future changes in downstream components of the terrestrial water cycle. Comparing the magnitude of different N_r stores could indicate the relative impacts of anthropogenic activities on different components of the terrestrial environment such as soils, the unsaturated zone, groundwater and riparian sediments. For example, large N_r storage in the unsaturated zone suggests that future N_r concentration changes in linked receptors (i.e. groundwater and surface water) will continue to be significantly affected by release of N_r from this store, before any impacts from changes in soil N_r leaching associated with recent changes in management practices are detected in the ultimate receptor. By combining consistent terminology, sharing of existing models, and improved monitoring networks, we believe that researchers can support the development of new modelling frameworks used in policy to provide better predictions of catchment nutrient trajectories and timescales.

3.3. An example from England (UK)

What would a proactive advocacy role for the biogeochemical research community look like in practice? Approaches to the integration of legacy N_r storage dynamics and time lags into policy would need to be informed by dialogue between researchers and

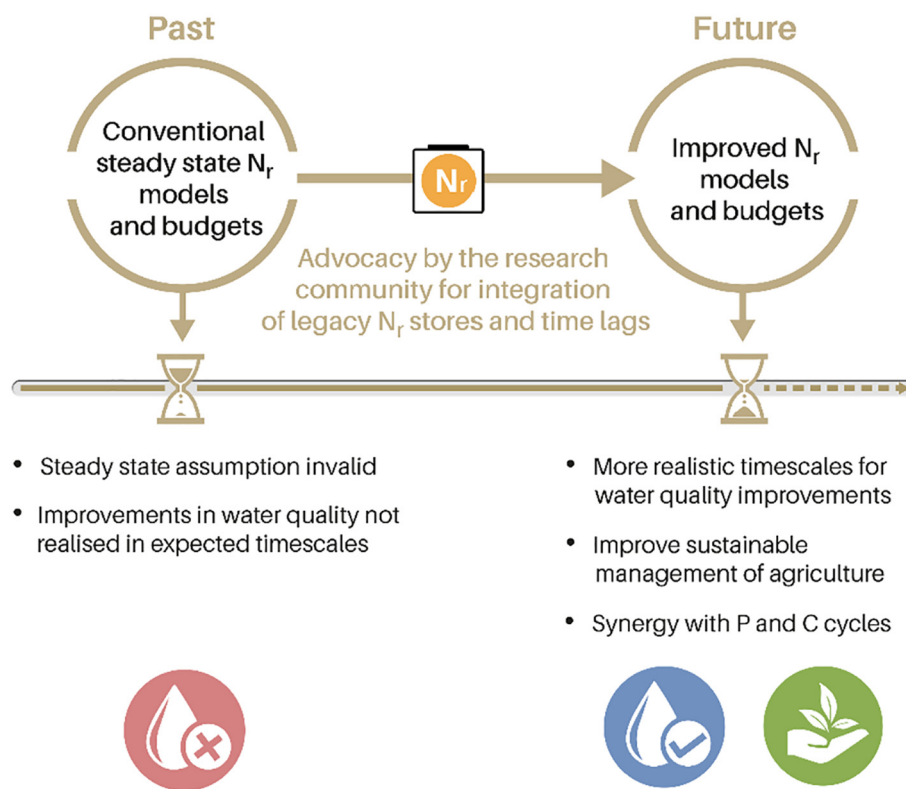


Fig. 1. Past and potential future approaches to management of legacy N_r , including the role of the research community to advocate for integration of legacy N_r stores and time lags into policy and practice

practitioners to identify discrepancies between the state of the science and models and tools used in policy within a particular setting. To illustrate the potential opportunities, here we provide an example of both awareness raising and data and model development from England (UK). In England researchers have raised awareness of the significance of legacy N storage dynamics to policymakers, national government and parliamentarians (Ascott and Ward, 2018). The methodology used to designate agricultural land in which N application may be restricted (known as Nitrate Vulnerable Zones (European Union, 1991) is reviewed every four years in England. In the latest review in 2020, time lags between nitrate leaching from the base of the soil zone and changes in nitrate concentrations in groundwater are being considered in the methodology using outputs of previous modelling of unsaturated zone travel times by Wang et al. (2012) (Hart and Kieboom, pers. comm.).

4. Synergy across macronutrient cycles

Better integration of time lags and legacy N_r stores would also align researchers and policymakers in the N community with those in the phosphorus (P) and carbon (C) communities. Successfully addressing challenges such as eutrophication requires policy responses that are co-ordinated across multiple nutrient elements (Conley et al., 2009; Harpole et al., 2011). However, different conceptual frameworks currently pervade across N, P and C communities. For example, P and C communities often more explicitly quantify the magnitude of stores compared to the N community. For P this is primarily due to issues of resource availability associated with finite resources of mineral phosphate rocks (Elser et al., 2014) and soil stores for agriculture (Haygarth et al., 2014; Sattari et al., 2012). Consequently large-scale P budgets have been developed using substance flow analysis (SFA) methods and the principles of mass balance to calculate the absolute magnitude of a number of P stores (Chen and Graedel, 2016; Yuan et al., 2018). For C

the quantification of the magnitude of stores is associated with climate change, with global scale budgets synthesizing fluxes and stores from a range of both observational and modelled data sources (Le Quéré et al., 2014). Whilst N_r is drawn from a large and renewable resource of atmospheric N_2 (Erismann et al., 2008), the evidence for legacy N_r in the environment highlights the need to quantify N_r stores in the terrestrial environment. Whilst fluxes from agricultural systems are the primary source of N_r to freshwater systems (Fowler et al., 2013), the same principles of time lag and stores apply to other sources (e.g. contaminated land, sewer leakage (Wakida and Lerner, 2005), mains leakage (Ascott et al., 2018).

5. Concluding remarks

Despite a strong body of scientific evidence and increasing awareness amongst stakeholders, models and budgets used by policymakers in BMP planning often do not adequately represent legacy N_r dynamics and associated time lags. Here we argue that the biogeochemical research community needs to proactively advocate for integration of time lags into future N_r management strategies through awareness raising and data and model development. This would support more realistic estimates of the trajectories of change following measures to reduce N_r loads, managing the expectations of stakeholders and supporting long term sustainable agriculture. Incorporating N_r stores and time lags into improved models and budgets used in policy and regulatory frameworks for the sustainable management of agriculture can better meet the needs of human health and the environment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

- Ascott, M.J., Ward, R.S., 2018. Written evidence submitted by the British Geological Survey. <http://data.parliament.uk/writtenevidence/committeeevidence.svc/evidencedocument/environmental-audit-committee/nitrate/written/77020.html>.
- Ascott, M.J., Wang, L., Stuart, M.E., Ward, R.S., Hart, A., 2016. Quantification of nitrate storage in the vadose (unsaturated) zone: a missing component of terrestrial N budgets. *Hydrol. Process.* 30 (12), 1903–1915. <https://doi.org/10.1002/hyp.10748>.
- Ascott, M.J., Goody, D.C., Surridge, B.W.J., 2018. Global patterns of nitrate storage in the vadose zone. *Nat. Commun.* 8 (1), 1416. <https://doi.org/10.1038/s41467-017-01321-w>.
- Ascott, M.J., Goody, D.C., Surridge, B.W.J., 2018. Public water supply is responsible for significant fluxes of inorganic nitrogen in the environment. *Environmental Science & Technology* 52 (24), 14050–14060. <https://doi.org/10.1021/acs.est.8b03204>.
- Astor, S.W., Blomquist, J.D., Webber, J.S., Chanat, J.G., 2020. Factors driving nutrient trends in streams of the Chesapeake Bay watershed. *J. Environ. Qual.* 49 (4), 812–834. <https://doi.org/10.1002/jeq2.20101>.
- Bell, V.A., Naden, P.S., Tipping, E., Davies, H.N., Carnell, E., Davies, J.A.C., Dore, A.J., Dragosits, U., Lapworth, D.J., Muhammed, S.E., Quinton, J.N., Stuart, M., Tomlinson, S., Wang, L., Whitmore, A.P., Wu, L., 2021. Long term simulations of macronutrients (C, N and P) in UK freshwaters. *Sci. Total Environ.* 776, 145813. <https://doi.org/10.1016/j.scitotenv.2021.145813>.
- Chen, M., Graedel, T.E., 2016. A half-century of global phosphorus flows, stocks, production, consumption, recycling, and environmental impacts. *Glob. Environ. Chang.* 36, 139–152. <https://doi.org/10.1016/j.gloenvcha.2015.12.005>.
- Chen, D., Shen, H., Hu, M., Wang, J., Zhang, Y., Dahlgren, R.A., 2018. Chapter five - legacy nutrient dynamics at the watershed scale: principles, modeling, and implications. In: Sparks, D.L. (Ed.), *Advances in Agronomy*. Academic Press, pp. 237–313 <https://doi.org/10.1016/bs.agron.2018.01.005>.
- Conley, D.J., Paerl, H.W., Howarth, R.W., Boesch, D.F., Seitzinger, S.P., Havens, K.E., Lancelot, C., Likens, G.E., 2009. Controlling eutrophication: nitrogen and phosphorus. *Science* 323 (5917), 1014–1015.
- Dodds, W.K., Bouska, W.W., Eitzmann, J.L., Pilger, T.J., Pitts, K.L., Riley, A.J., Schloesser, J.T., Thornbrugh, D.J., 2009. Eutrophication of U.S. freshwaters: analysis of potential economic damages. *Environ. Sci. Technol.* 43 (1), 12–19. <https://doi.org/10.1021/es801217q>.
- Durand, P., Breuer, L., Johnes, P., 2011. Nitrogen processes in aquatic ecosystems. In: Sutton, M.A., et al. (Eds.), *The European Nitrogen Assessment*. Cambridge University Press, Cambridge.
- Elser, J.J., Elser, T.J., Carpenter, S.R., Brock, W.A., 2014. Regime shift in fertilizer commodities indicates more turbulence ahead for food security. *PLoS One* 9 (5).
- England, J., Skinner, K.S., Carter, M.G., 2008. Monitoring, river restoration and the Water Framework Directive. *Water Environ. J.* 22 (4), 227–234. <https://doi.org/10.1111/j.1747-6593.2007.00090.x>.
- Erisman, J.W., Sutton, M.A., Galloway, J., Klimont, Z., Winiwarter, W., 2008. How a century of ammonia synthesis changed the world. *Nat. Geosci.* 1, 636. <https://doi.org/10.1038/ngeo325>.
- European Union, 1991. Council Directive 91/676/EEC of 12 December 1991 Concerning the Protection of Waters Against Pollution Caused by Nitrates From Agricultural Sources. European Union, Brussels.
- Fowler, D., Coyle, M., Skiba, U., Sutton, M.A., Cape, J.N., Reis, S., Sheppard, L.J., Jenkins, A., Grizzetti, B., Galloway, J.N., 2013. The global nitrogen cycle in the twenty-first century. *Philos. Trans. R. Soc. B* 368 (1621), 20130164.
- Hamilton, S.K., 2012. Biogeochemical time lags may delay responses of streams to ecological restoration. *Freshw. Biol.* 57 (s1), 43–57. <https://doi.org/10.1111/j.1365-2427.2011.02685.x>.
- Harpole, W.S., Ngai, J.T., Cleland, E.E., Seabloom, E.W., Borer, E.T., Bracken, M.E.S., Elser, J.J., Gruner, D.S., Hillebrand, H., Shurin, J.B., Smith, J.E., 2011. Nutrient co-limitation of primary producer communities. *Ecol. Lett.* 14 (9), 852–862. <https://doi.org/10.1111/j.1461-0248.2011.01651.x>.
- Haygarth, P.M., Jarvie, H.P., Powers, S.M., Sharpley, A.N., Elser, J.J., Shen, J., Peterson, H.M., Chan, N.-I., Howden, N.J., Burt, T., 2014. Sustainable phosphorus management and the need for a long-term perspective: the legacy hypothesis. *Environ. Sci. Technol.* 48 (15), 8417–8419.
- House of Commons Environmental Audit Committee, 2018. *UK Progress on Reducing Nitrate Pollution*, London, UK.
- Howden, N.J.K., Burt, T.P., Worrall, F., Mathias, S., Whelan, M.J., 2011. Nitrate pollution in intensively farmed regions: what are the prospects for sustaining high-quality groundwater? *Water Resour. Res.* 47 (6), W00L02. <https://doi.org/10.1029/2011WR010843>.
- Jia, X., Zhu, Y., Huang, L., Wei, X., Fang, Y., Wu, L., Binley, A., Shao, M., 2018. Mineral N stock and nitrate accumulation in the 50 to 200m profile on the Loess Plateau. *Sci. Total Environ.* 633, 999–1006. <https://doi.org/10.1016/j.scitotenv.2018.03.249>.
- Le Quéré, C., Peters, G.P., Andres, R.J., Andrew, R.M., Boden, T.A., Ciais, P., Friedlingstein, P., Houghton, R.A., Marland, G., Moriarty, R., Sitch, S., Tans, P., Arneeth, A., Arvanitis, A., Bakker, D.C.E., Bopp, L., Canadell, J.G., Chini, L.P., Doney, S.C., Harper, A., Harris, I., House, J.I., Jain, A.K., Jones, S.D., Kato, E., Keeling, R.F., Klein Goldewijk, K., Körtzinger, A., Koven, C., Lefèvre, N., Maignan, F., Omar, A., Ono, T., Park, G.H., Pfeil, B., Poulter, B., Raupach, M.R., Regnier, P., Rödenbeck, C., Saito, S., Schwinger, J., Segsneider, J., Stocker, B.D., Takahashi, T., Tilbrook, B., van Heuven, S., Viovy, N., Wanninkhof, R., Wiltshire, A., Zaehle, S., 2014. Global carbon budget 2013. *Earth Syst. Sci. Data* 6 (1), 235–263. <https://doi.org/10.5194/essd-6-235-2014>.
- Liu, Y., Engel, B.A., Flanagan, D.C., Gitau, M.W., McMillan, S.K., Chaubey, I., 2017. A review on effectiveness of best management practices in improving hydrology and water quality: needs and opportunities. *Sci. Total Environ.* 601–602, 580–593. <https://doi.org/10.1016/j.scitotenv.2017.05.212>.
- Martin, S.L., Hamlin, Q.F., Kendall, A.D., Wan, L., Hyndman, D.W., 2021. The land use legacy effect: looking back to see a path forward to improve management. *Environ. Res. Lett.* 16 (3), 035005. <https://doi.org/10.1088/1748-9326/abe14c>.
- McCrackin, M.L., Jones, H.P., Jones, P.C., Moreno-Mateos, D., 2017. Recovery of lakes and coastal marine ecosystems from eutrophication: a global meta-analysis. *Limnol. Oceanogr.* 62 (2), 507–518. <https://doi.org/10.1002/lno.10441>.
- Meals, D.W., Dressing, S.A., Davenport, T.E., 2010. Lag time in water quality response to best management practices: a review. *J. Environ. Qual.* 39 (1), 85–96. <https://doi.org/10.2134/jeq2009.0108>.
- Melland, A.R., Fenton, O., Jordan, P., 2018. Effects of agricultural land management changes on surface water quality: a review of meso-scale catchment research. *Environ. Sci. Pol.* 84, 19–25. <https://doi.org/10.1016/j.envsci.2018.02.011>.
- Pretty, J.N., Brett, C., Gee, D., Hine, R., Mason, C., Morison, J., Raven, H., Rayment, M., Van der Bijl, G., 2000. An assessment of the total external costs of UK agriculture. *Agric. Syst.* 65 (2), 113–136.
- Sattari, S.Z., Bouwman, A.F., Giller, K.E., van Ittersum, M.K., 2012. Residual soil phosphorus as the missing piece in the global phosphorus crisis puzzle. *Proc. Natl. Acad. Sci.* 109 (16), 6348–6353. <https://doi.org/10.1073/pnas.1113675109>.
- Schaure, S., Naus, J., 2010. 10 Years of the Water Framework Directive: a toothless tiger? A Snapshot Assessment of EU Environmental Ambitions. European Environmental Bureau, Brussels.
- Schlesinger, W.H., 2005. *Biogeochemistry*. Elsevier.
- Sprague, L.A., Hirsch, R.M., Aulenbach, B.T., 2011. Nitrate in the Mississippi River and its tributaries, 1980 to 2008: are we making progress? *Environmental Science & Technology* 45 (17), 7209–7216. <https://doi.org/10.1021/es201221s>.
- Stuart, M.E., Ward, R.S., Ascott, M.J., Hart, A., 2016. *Regulatory Practice and Transport Modelling for Nitrate Pollution in Groundwater*. British Geological Survey, Keyworth, UK.
- Tesoriero, A.J., Duff, J.H., Saad, D.A., Spahr, N.E., Wolock, D.M., 2013. Vulnerability of streams to legacy nitrate sources. *Environmental Science & Technology* 47 (8), 3623–3629. <https://doi.org/10.1021/es305026x>.
- Turkeltaub, T., Ascott, M.J., Goody, D.C., Jia, X., Shao, M.-A., Binley, A., 2020. Prediction of regional-scale groundwater recharge and nitrate storage in the vadose zone: a comparison between a global model and a regional model. *Hydrol. Process.* 34 (15), 3347–3357. <https://doi.org/10.1002/hyp.13834>.
- United States Environmental Protection Agency, 2011. *Reactive Nitrogen in the United States: An Analysis of Inputs, Flows, Consequences, and Management Options*, Washington DC, USA.
- Van Meter, K.J., Basu, N.B., Veenstra, J.J., Burras, C.L., 2016. The nitrogen legacy: emerging evidence of nitrogen accumulation in anthropogenic landscapes. *Environ. Res. Lett.* 11 (3), 035014.
- Van Meter, K.J., Van Cappellen, P., Basu, N.B., 2018. Legacy nitrogen may prevent achievement of water quality goals in the Gulf of Mexico. *Science* <https://doi.org/10.1126/science.aar4462>.
- Vero, S.E., Basu, N.B., Van Meter, K., Richards, K.C., Mellander, P.-E., Healy, M.G., Fenton, O., 2018. Review: the environmental status and implications of the nitrate time lag in Europe and North America. *Hydrogeol. J.* 26 (1), 7–22. <https://doi.org/10.1007/s10040-017-1650-9>.
- Vitousek, P.M., Aber, J.D., Howarth, R.W., Likens, G.E., Matson, P.A., Schindler, D.W., Schlesinger, W.H., Tilman, D.G., 1997. Human alteration of the global nitrogen cycle: sources and consequences. *Ecol. Appl.* 7 (3), 737–750.
- Wakida, F.T., Lerner, D.N., 2005. Non-agricultural sources of groundwater nitrate: a review and case study. *Water Res.* 39 (1), 3–16.
- Wang, L., Stuart, M.E., Bloomfield, J.P., Butcher, A.S., Goody, D.C., McKenzie, A.A., Lewis, M.A., Williams, A.T., 2012. Prediction of the arrival of peak nitrate concentrations at the water table at the regional scale in Great Britain. *Hydrol. Process.* 26 (2), 226–239. <https://doi.org/10.1002/hyp.8164>.
- Wang, L., Stuart, M.E., Lewis, M.A., Ward, R.S., Skirvin, D., Naden, P.S., Collins, A.L., Ascott, M.J., 2016. The changing trend in nitrate concentrations in major aquifers due to historical nitrate loading from agricultural land across England and Wales from 1925 to 2150. *Sci. Total Environ.* 542, 694–705. <https://doi.org/10.1016/j.scitotenv.2015.10.127>.
- Worrall, F., Howden, N.J.K., Burt, T.P., 2015. Evidence for nitrogen accumulation: the total nitrogen budget of the terrestrial biosphere of a lowland agricultural catchment. *Biogeochemistry* 123 (3), 411–428. <https://doi.org/10.1007/s10533-015-0074-7>.

- Wu, H., Song, X., Zhao, X., Peng, X., Zhou, H., Hallett, P.D., Hodson, M.E., Zhang, G.-L., 2019. Accumulation of nitrate and dissolved organic nitrogen at depth in a red soil Critical Zone. *Geoderma* 337, 1175–1185. <https://doi.org/10.1016/j.geoderma.2018.11.019>.
- Wu, H., Song, X., Liu, F., Zhao, X., Zhang, G.-L., 2020. Regolith property controls on nitrate accumulation in a typical vadose zone in subtropical China. *CATENA* 192, 104589. <https://doi.org/10.1016/j.catena.2020.104589>.
- Xin, J., Liu, Y., Chen, F., Duan, Y., Wei, G., Zheng, X., Li, M., 2019. The missing nitrogen pieces: a critical review on the distribution, transformation, and budget of nitrogen in the vadose zone-groundwater system. *Water Res.* 165, 114977. <https://doi.org/10.1016/j.watres.2019.114977>.
- Yuan, Z., Jiang, S., Sheng, H., Liu, X., Hua, H., Liu, X., Zhang, Y., 2018. Human perturbation of the global phosphorus cycle: changes and consequences. *Environmental Science & Technology* 52 (5), 2438–2450. <https://doi.org/10.1021/acs.est.7b03910>.