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COMMENTARY

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Pulling the rabbit out of the hat: Unravelling hidden nitrogen legacies in catchment-scale water quality models

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1 | INTRODUCTION

Human activities have significantly perturbed the global cycle of nitrogen by excess inputs of reactive nitrogen (N) to the environment (Rockström et al., 2009). The major anthropogenic sources of N to the landscape are intensive agriculture associated with the use of mineral fertilizers and manure, and atmospheric deposition resulting from the burning of fossil fuels and biomass (Duce et al., 2008). At the global scale, N inputs to the land surface are much larger than N sinks (Schlesinger, 2009) and have exceeded a critical level beyond which abrupt and severe environmental change is likely to occur (Rockström et al., 2009; Steffen et al., 2015). The consequences for the environment, climate and human life are manifold: eutrophication of inland water bodies and receiving marine systems causing toxic algal blooms, losses in biodiversity and aquatic ecosystem functioning (Diaz & Rosenberg, 2008; Rabalais, 2002), greenhouse gas emissions especially as nitrous oxide (Aguilera et al., 2021; Galloway et al., 2003), and nitrate contamination of groundwater with implications for safe drinking water production (EPA, 2007; WHO, 2016).

Several national and international efforts have been made to address the problems associated with excessive N inputs, including the Clean Water Act (EPA, 1972) in the USA, and the Nitrates Directive (EU Commission, 1991), which is part of the Water Framework Directive (EU Commission, 2000) in the EU. The Nitrates Directive has been implemented to protect surface water and groundwater across Europe against nitrate pollution and has resulted in the designation of Nitrate Vulnerable Zones (NVZs) in which action programmes that aim for balanced fertilization in agricultural areas have been applied (Bouraoui & Grizzetti, 2011; EU Commission, 2021). The European regulations have been successful in curbing net N input by reducing fertilizer usage, increasing N use efficiency, and decreasing atmospheric deposition (Bouraoui & Grizzetti, 2011). However, improvement of water quality in EU streams has been minor since the Directive's implementation: in-stream nitrate concentration has decreased by 0.02% per year in the period 1992–2018, while no appreciable changes have been noticed for groundwater (EEA, 2022). Similar observations have been made in North American landscapes (Van Meter et al., 2018; Vero et al., 2018). In summary, the problem of excess N in the environment is pervasive (EEA, 2018) and the perturbed N cycle is one of the most pressing environmental challenges (Elser, 2011; Galloway et al., 2003). Therefore, diffuse N pollution continues to be one of the largest pressures on both surface water and groundwater bodies hindering the achievement of key environmental objectives in the EU (EU Commission, 2021). This raises questions about the effectiveness of measures on the one hand (Bouraoui & Grizzetti, 2011) and challenges the state of current knowledge on reactive N transport in the landscape on the other (Vero et al., 2018).

The fate of N in the terrestrial and aquatic systems is the result of a complex interplay between hydrological transport and biogeochemical transformation processes in soil, subsurface (which includes the unsaturated zone below the root zone as well as groundwater) and surface water compartments (e.g., Böhlke & Denver, 1995; Liao et al., 2012; Lutz et al., 2020). This interplay strongly depends on the relation between hydrological transport and biogeochemical (reaction) timescales in the different compartments (Kumar et al., 2020; Ocampo et al., 2006). Both hydrological and biogeochemical processes can lead to substantial legacy effects that result in accumulation of N in the soil and subsurface, and time lags in the propagation of N input flux to N responses in water bodies (Ascott et al., 2021; Basu et al., 2022). For hydrological processes, long transport times of water and dissolved mobile species of N (in particular nitrate) are documented in soil, unsaturated zone and groundwater (e.g., Ascott et al., 2017; Kolbe et al., 2019; Weitzman et al., 2022), creating a *hydrological legacy*. Similarly, N can accumulate in immobile organic N pools in soils over long time scales, creating a *biogeochemical legacy* (e.g. Sebilo et al., 2013; Van Meter & Basu 2015). Both legacy types can thus cause a lack of immediate response in surface water and groundwater to changes in N inputs such that contemporary water quality is potentially influenced by N inputs that entered the landscape years or decades ago (e.g. Ascott et al., 2017; Bingham & Cotrufo, 2016; Van Meter et al., 2017). Hence, N legacies have substantial implications for the assessment of N-related measures and the resulting management decisions and environmental policies (Ascott et al., 2021; Basu et al., 2022; Vero et al., 2018).

Water quality models are indispensable tools for the assessment of management decisions and environmental policies at the catchment scale (Honti et al., 2017). Over the past decades, catchment-scale water quality models with varying complexities and parameterizations have been developed (e.g. SWAT, Arnold et al., 1998; INCA, Whitehead et al., 1998; HYPE, Lindström et al., 2010). These models mechanistically incorporate relevant processes and link N inputs to catchment exports (Rode et al., 2010; Wang et al., 2012). They are now commonly used to investigate present-state N concentrations and loads, and to predict their future trajectories in groundwater and surface water bodies under different management scenarios. While the ability of these models to capture observed N concentrations in streams is generally documented, little attention is paid to examining whether and how well these existing models represent N-legacy effects (Sarrazin et al., 2022; Van Meter & Basu, 2015). Here we (1) briefly review and show if and how N legacies are represented in current water quality models; and (2) propose stronger efforts towards proper representation and assessment of legacy components in water quality modelling. We aim at stimulating discussions in the modelling community about conceptualisation, characterization and evaluation of hidden legacy stores in catchment-scale water quality models. In the spirit of "*Getting the right answers for the right reasons*" (Kirchner, 2006), challenging the plausibility of internal model routines will immensely benefit the overall credibility of water quality assessments.

2 | REPRESENTATION OF N LEGACY IN CATCHMENT SCALE WATER QUALITY MODELS—A BRIEF OVERVIEW

Water quality models describing N transformation and transport at the catchment scale typically include a soil compartment, where N transformations (such as mineralization or denitrification), plant uptake and leaching to a subsurface compartment are quantified, and a subsurface compartment, in which transport and denitrification are elaborated to varying degrees. The soil compartment is further subdivided into a set of sub-compartments with organic (ON) and inorganic N (IN) pools (Figure 1). Most models additionally differentiate between an active (or fast) and inactive (also referred to as slow, protected or passive) ON pool (e.g. SWAT, Neitsch et al., 2011; HYPE, Lindström et al., 2010; and ELEMeNT, Van Meter et al., 2017). The active and inactive pools represent ON with fast and slow mineralization rates to dissolved inorganic N (IN), respectively. Slow mineralization of inactive ON can be caused by physical protection in soil microaggregates, association with clay and silt particles, and biochemical stabilization of the ON compound itself hampering fast decomposition (Paul, 2016; Six et al., 2002). Some models such as SWAT (Neitsch et al., 2011) and LPJmL (von Bloh et al., 2018) additionally include a fresh ON pool representing litter, crop residues and microbial biomass. It is generally assumed that IN (and possibly some dissolved ON compounds; Lindström et al., 2010) can leach from the soil to the subsurface compartment and be subsequently transported to rivers.

The explicit representation of one or several soil pools and, in particular, of immobile soil ON can enable simulation of N storage in soils over long timescales and thus of biogeochemical legacy effects of excessive N inputs (Figure 1). While this representation is comparably well established in current nutrient models, only very recently have some studies analysed the temporal dynamics of soil N storage in more detail (e.g., Ilampooranan et al., 2022; Lee et al., 2016; Sarrazin et al., 2022; Van Meter et al., 2017, 2018). Accordingly, detailed analyses of the impact of model structure and/or parameter values on the simulated build-up of soil ON have rarely been performed (but see, e.g. Ilampooranan et al., 2022; Sarrazin et al., 2022). However, as N fluxes from the soil pools propagate through the model

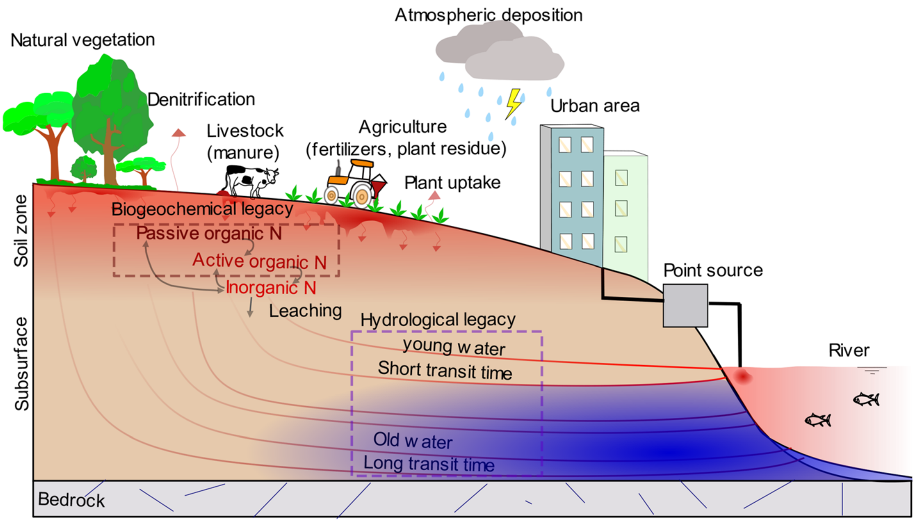

FIGURE 1 Schematic of N retention and transformation processes, with an emphasis on biogeochemical and hydrological N legacy in catchment-scale water quality models

compartments, we need to quantify and evaluate soil N legacy in water quality models to more accurately simulate N fluxes and concentrations in receiving water bodies.

Most well-established water quality models thus have routines for *biogeochemical* legacies, but they are often not evaluated or checked against field observations. Considering the second type of legacy that is, *hydrological* legacy, there is not only a lack of validation, but generally also a lack of model routines (or only theoretical routines) to represent N legacy caused by long transport times in the subsurface (e.g. Lindström et al., 2010; Neitsch et al., 2011). Fortunately, there have been recent developments exploring the representation of hydrological N legacy in water quality models following two main approaches. The first is based on implementation of two storage compartments, that is, a passive and an active hydrological storage, and is thus comparable to the implementation of an active and inactive ON pool in the soil compartment (Shafii et al., 2019; Yang et al., 2018). The main challenge of this approach lies in how to parameterise the size of the passive storage and the exchange fluxes between active and passive storages (Hrachowitz et al., 2016). Moreover, biogeochemical N processes (e.g. denitrification) within the hydrologically passive storage are usually neglected (Shafii et al., 2019; Yang et al., 2018). The second approach for modelling hydrological legacy of N is based on the concept of transit times, which essentially characterize the journey of water and dissolved solutes from inflow (infiltration) to outflow via discharge or evapotranspiration (e.g. Ilampooranan et al., 2019; van der Velde et al., 2010; Wade et al., 2002). The range of transit times caused by a large number of different flow paths in a catchment is specified by the transit time distribution (TTD; Botter et al., 2010; Harman, 2015; Rinaldo et al., 2011; van der Velde et al., 2010). The TTD can be used to describe reactive transport of dissolved IN along these flow paths to the stream network (Figure 1). Therefore, it also allows representing long lag times between mobilization of IN (N leaching) and its export to streams (Ilampooranan et al., 2019; Nguyen et al., 2021; Van Meter et al., 2017).

3 | HOW CAN WE BETTER REPRESENT N LEGACIES IN MODELS?

Most of the current water quality models represent *biogeochemical legacy* via simulation of N storage in soils. In contrast, efforts to explore *hydrological legacy* for N are scarce and restricted to modelling for scientific purposes, while established water quality models used by practitioners typically do not consider this legacy component. One reason for the latter is that the modelling focuses mainly on the representation of in-stream concentrations and loads, which can be adequately reproduced by current catchment-scale water quality models via model calibration—despite the missing description of hydrological legacy. However, as understanding and quantifying legacy stores is of paramount importance to mitigate the negative effects of excessive nutrient loads, water quality models need to represent both biogeochemical and hydrological N legacy. Hence, new synthesis approaches are needed in water quality models to better reflect and analyse the joint effects of hydrological and biogeochemical legacies and the timescales in which they occur.

Water quality models that do implement hydrological N legacy generally use TTDs that are constant in time (but see, e.g., van der Velde et al., 2010; Ilampooranan et al., 2019; Nguyen et al., 2021). While steady-state TTDs might be adequate to predict annual N export, prediction of finer-scale N export dynamics and the role of legacy in modifying these dynamics can benefit from recent advancements by the hydrological modelling community in the description of dynamic TTDs and storage selection (SAS) functions (Benettin et al., 2017; Harman, 2015; Rinaldo et al., 2015; van der Velde et al., 2012). Dynamic TTD descriptions have been increasingly used to simulate the effect of hydrological conditions on reactive solute transport (e.g., Benettin et al., 2015; Kumar et al 2020; Lutz et al., 2017). They enable us to represent and account for time-varying hydrological dynamics more realistically as they allow incorporating knowledge on catchment functioning under varying environmental conditions (Harman, 2015; Rinaldo et al., 2011). Hence, we believe that N transport descriptions with time-variant TTDs should be the preferred approach for characterising hydrological N legacy, particularly at the sub-annual or seasonal scale.

In the rainfall-runoff (water quantity) modelling community, there is a growing tendency to report and evaluate model-internal components such as simulated soil moisture, evapotranspiration, groundwater dynamics, and total water storage anomalies, to prevent the model from giving good results for the wrong reasons (Bouaziz et al., 2021; Clark et al., 2011; Rakovec et al., 2016). We argue that such model-internal plausibility checks should also become a routine procedure in water *quality* modelling. In particular, the effect of biogeochemical legacy can become more evident and transparent only if modelling studies routinely report and quantify N build-up in the inactive ON pool (Sarrazin et al., 2022). Hence, modelling studies should include, report and critically analyse model-internal N fluxes and stores in addition to concentrations in the receiving waters.

The scarcity of field data certainly poses limitations to a thorough evaluation of simulated legacy stores in water quality models (Sarrazin et al., 2022). Long-term data on the build-up of ON in soils are scarce and often limited to a few locations and the top soil (e.g. Jenkinson, 1991; Sebilo et al., 2013; Van Meter et al., 2016). Similarly, although specific groundwater monitoring networks have been established (e.g. in Europe in the context of the Water Framework Directive), measurements of groundwater nitrate concentration remain patchy in space (point measurements) and sporadic in time (typically annual frequency). Hence, we need field data that are more tailored to characterization of N legacies such as (i) soil ON measurements over longer periods to quantify biogeochemical legacy; (ii) nitrate concentrations in groundwater to evaluate simulated IN in the subsurface; (iii) tracer data to constrain simulated TTDs (Benettin et al., 2017; Rodriguez et al., 2021; Visser et al., 2019); (iv) field assessments of denitrification in groundwater (Eschenbach et al., 2018); (v) detailed spatial and temporal data on N input from point and diffuse sources to facilitate quantification of N legacy from N budgets (Ehrhardt et al., 2021; Van Meter & Basu, 2017) or trajectories of N seasonality (Ebeling et al., 2021); and (vi) "soft data" such as geochemical information on denitrification potential in the soil and deeper subsurface. Regardless of the quantity and quality of these observational data, uncertainties in simulated legacies associated with model input data, structure and parameterisation need to be clearly addressed and communicated (Sarrazin et al., 2022).

In summary, we want to instigate a broader discussion in the water quality modelling community about the need for explicit simulation and assessment of legacy components in their models, and call for concerted efforts between and among modellers and experimental scientists to gather field data that can inform the modelling of N legacies (Basu et al., 2022; Fu et al., 2020). Such efforts can only thrive once we overcome disciplinary boundaries between water quantity and quality modelling as well as between modelling and experimental research on N in soils and groundwater. We believe that the explicit simulation and reporting of biogeochemical and hydrological legacy will provide essential insights into model functioning and realism, and eventually result in more vigorous efforts to collect field data on N legacy. Interdisciplinary and transdisciplinary efforts to quantify legacy components will greatly benefit not only the scientific community but also society as a whole, as they will lead to more credible estimates of the hidden legacy components and make their impact quantifiable for water managers, lawmakers, farmers and other stakeholders. This is one of the overarching goals of water quality modelling and paves the way for a fundamental contribution to mitigating human perturbations of the global N cycle.

AUTHOR CONTRIBUTIONS

This paper emerged from a team effort by Andreas Musolff, Fanny J. Sarrazin, Pia Ebeling, Rohini Kumar, Stefanie R. Lutz and Tam V. Nguyen within a water quality modelling initiative led by Sabine Attinger and Andreas Musolff. Fanny J. Sarrazin, Stefanie R. Lutz and Tam V. Nguyen performed a model review serving as basis of the manuscript. Rohini Kumar and Tam V. Nguyen designed the figure. Rohini Kumar and Stefanie R. Lutz directed the writing process. Andreas Musolff, Fanny J. Sarrazin, Kimberley J. Van Meter, Nandita B. Basu, Jan H. Fleckenstein, Pia Ebeling, Rohini Kumar, Stefanie R. Lutz and Tam V. Nguyen contributed to the final manuscript.

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