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PAPER

Review: the environmental status and implications of the nitrate time lag in Europe and North America

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Abstract The efficacy of water quality policies aiming to reduce or prevent nitrate contamination of waterbodies may be constrained by the inherent delay or "time lag" of water and solute transport through unsaturated (soil) and saturated (groundwater) pathways. These delays must be quantified in order to establish realistic deadlines, thresholds and policy expectations, and to design effective best management practices. The objective of this review is to synthesise the current state of research on nitrate-related time lags in both the European and North American environmental and legislative contexts. The durations of time lags have been found to differ according to climatic, pedological, landscape and management scenarios. Elucidation of these driving factors at a watershed scale is essential where water quality is impaired or at risk. Finally, the existence of time lags is increasingly being acknowledged at a policy level and incorporated into the development of environmental legislation. However, the full impact of these time lags is not yet fully understood or appreciated, and continued outreach and education in scientific, public and policy venues is still required.

Keywords Water framework directive · Lag time · Groundwater monitoring · Unsaturated zone · Nitrate

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Introduction

The efficacy of water quality policies is influenced by both anthropogenic and non-anthropogenic controls (Bechmann et al. 2008). The latter category includes hydrologic (Fenton et al. 2011) and biogeochemical (Jahangir et al. 2013a; Fenton et al. 2017; Jahangir et al. 2017) time lags arising during the migration of excess reactive nitrogen (Nr) through subsurface pathways. These delays make any correlation between programme of measure (POM) efficacy and water quality improvements difficult (Osenbrück et al. 2006; Meals et al. 2010; Fenton et al. 2011; Hrachowitz et al. 2016; and others). Time lags have significant consequences for policy design and legislation. Bilotta et al. (2014) recommended that, following the example of the healthcare industry, environmental policies must be informed by thorough and comprehensive review of the scientific research in that field. The purpose of this review paper is to examine the current research on time lags, to explore the policy implications arising from their existence, and to compare European and North American scenarios within the context of their unique hydrologic, environmental and legislative backgrounds. Some critical studies are highlighted in Fig. 1, and are discussed within this review.

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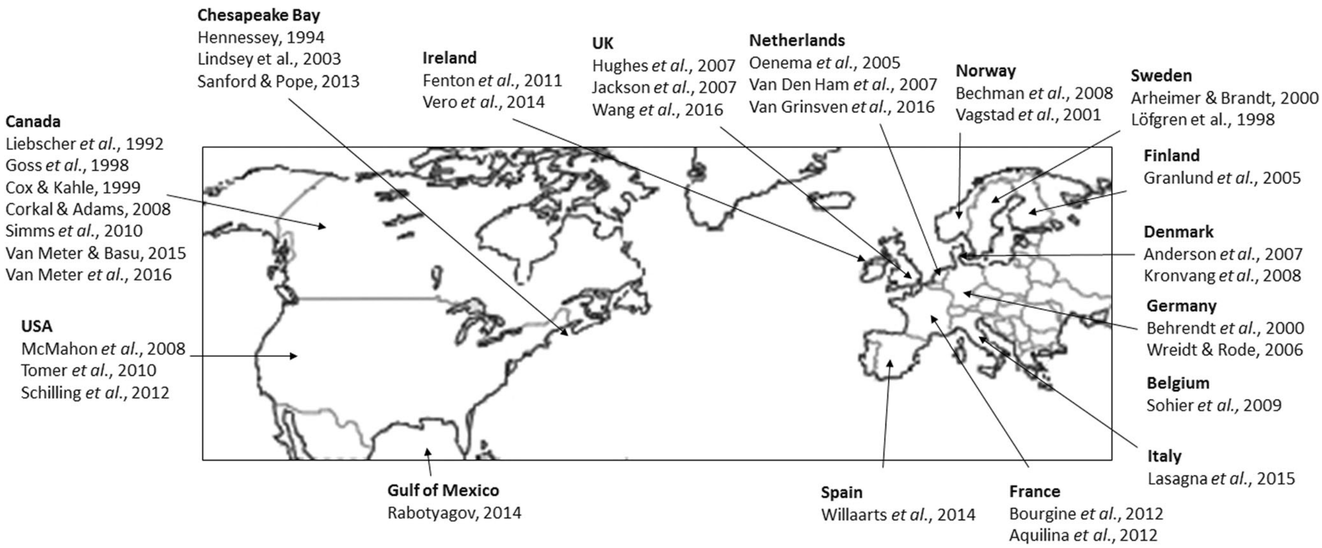

Fig. 1 A selection of key international publications evidencing time lag and water quality

What is time lag?

The transport of water and nutrients, supplied to surface waterbodies and groundwater abstraction points from agricultural, industrial or other sources, may occur via a range of hydrologic pathways. These include overland flow, interflow, shallow and deep groundwater flow, and conduit flow (Archbold et al. 2010), and may also include cycling through a variety of biogeochemical pathways (Hrachowitz et al. 2016), including uptake into plants and soil organic matter (Shen et al. 1989; Sebilo et al. 2013). Transport along these pathways imposes a delay between the implementation of POM designed to improve the quality of groundwater or surface waters (Schirmer et al. 2014), and measurable improvements in water quality. This delay is referred to by several different terms, including "memory effect", "delayed response", "residence effect", "legacy effect" or "time lag" (which is the preferred term in the current paper) (Worrall and Burt 1999; Bechmann et al. 2008; Iital et al. 2008; Wahlin and Grimvall 2008; Vero et al. 2014; Hamilton 2011; Van Meter et al. 2016a, b). Although time lags associated with delays in policy implementation may also occur (Bechmann et al. 2008; Meals et al. 2010), these lags are ignored in the present review. Determining the duration of time lag is of critical importance from a policy and monitoring perspective (Bain et al. 2012), as correlation of the success of a legislative instrument (even assuming 100% implementation) and the current water quality status is not always possible (Fenton et al. 2011), and observations may also be confounded by inter-annual meteorological variability (Bechmann et al. 2008). In addition, prediction of timelines with respect to water quality improvements or deterioration is challenging (Hering et al. 2010). The subsurface pathway, which includes both the unsaturated and saturated zones, presents particular challenges due to the spatial and temporal heterogeneity of physical properties (such as depth of the unsaturated zone, groundwater recharge rate, baseflow indices, hydrogeological properties of the soil/subsoil and geology, and dominance of either matrix and/or fracture pathways), the scope for chemical processes and transformations such as attenuation capacity (Jahangir et al. 2013a, b), difficulties in determining the length of retention times throughout the subsurface and the influence of meteorological conditions (Hocking and Kelly 2016). This pathway bears particular importance with respect to nitrate (NO_3^-) and its transformational components, e.g. organic nitrogen (Van Meter et al. 2016a) and ammonium (Vadas et al. 2007), due to the high mobility of this important agrochemical, and the hazards which it may present to water quality and ecology (Azevedo et al. 2015).

The total time lag (t_T) via subsurface pathways includes both unsaturated zone (t_u) and groundwater (saturated zone) (t_s) components. The former represents the movement of NO_3^- from the soil surface (where it may have been applied as fertiliser) vertically through soil and unsaturated bedrock (Hillel 2004) until it reaches shallow groundwater (i.e. the water table). Nitrogen in the unsaturated zone may be either in the dissolved form, as NO_3^-, or as organic nitrogen sorbed to the soil matrix. Van Meter and Basu (2015) refer to the former as the hydrologic legacy, while the latter is referred to as the biogeochemical. Travel of NO_3^- through the unsaturated zone is dependent upon soil hydraulic properties such as saturated hydraulic conductivity (k_{sat}), porosity (n) and pore size distribution, effective rainfall or recharge (ER), and depth of the zone. Due to the variety of pore sizes and complexities exhibited in soil and unsaturated bedrock, both rapid preferential flow and slow matrix flows are frequently observed within the same area. While at certain times either preferential or matrix flow will prevail, at others both rapid flow through cracks and macropores, and slow filtration through the interaggregate matrix will occur. The implications of this dual-porosity are that (1) t_u associated with the physical, convective transport will vary, and (2) t_u associated with sorbing of N onto soil and rock surfaces will be affected by the surface area of the pore classes through which it is in transit (Jarvis 2007). In reality, diffuse transport between the various pore regions is common, and transport is rarely confined to either preferential or matrix flow (Gerke and van Genuchten 1993). The magnitude of the biogeochemical legacy further depends on the clay or mineral content of the soil as well as management factors such as the cropping pattern, levels of fertiliser application, the type of fertiliser applied, etc. (McLauchlan 2006).

Arrival of NO_3^- at the water table takes the form of a breakthrough curve, which can be divided into the initial breakthrough (IBT), peak concentration (*peak*), centre of mass (COM) and solute exit (*exit*; Vero et al. 2014). IBT represents the first arrival of the solute at the water table, and is of particular interest from a policy perspective, as it indicates the trajectory of groundwater quality changes in response to management practices. The middle stages (peak and COM) are indicative of the bulk of nutrient transport, while *exit* represents total flushing of the solute from the profile. As such, although this latter marker may be considered to indicate the fullest extent of t_u, it is in reality, difficult to discern from environmental monitoring, as the signal may be extremely low relative to background noise.

The groundwater travel time (t_s) begins once the contaminant “breaks through” the water table and becomes available for lateral transport through the saturated zone. The total distance to the receptor may vary from very short (leading to a brief t_s, where the source is adjacent to a receptor), to decadal (where the groundwater pathway is long). Similarly to the unsaturated zone, the hydraulic characteristics of the saturated zone will determine the speed with which water and hence, solutes such as nitrate are vectored to a receptor—for example, karstic aquifers may exhibit extremely brief t_S even when distances from source to receptor are relatively long, due to high conductivity within fissures, whilst simultaneously exhibiting much slower transport within the pore matrix (Huebsch et al. 2014; Fenton et al. 2017). In such scenarios, ranges of t_S can better reflect the nature of nitrate transport than single figures. In contrast to water in transit through the soil, groundwater is considered as both a vector for nutrient transport to a receptor (such as surface waterbody) and a receptor itself, from which drinking water may be abstracted (Frind et al. 2006). Accordingly, qualitative thresholds are established for groundwater that are distinct from those specified for surface waters. These qualitative thresholds typically specify concentrations of NO_3^- (and also for other chemicals), above which the water is considered to be at poor quality or unpotable. Any specific point within an aquifer is liable to receive or transmit water and solutes in three dimensions. The measured concentration of a contaminant at an abstraction point consequently reflects the mean value for the area from which the well is recharged. A greater density of sampling wells in a given area will therefore provide a better indication of actual groundwater quality. Concentrations of groundwater NO_3^- may be relatively high (Vadas et al. 2007), which has implications for receptor quality, although denitrification in groundwater “hotspots (Jahangir et al. 2013a, b) means that abstracted groundwater samples may not reflect concentrations delivered through the soil to the water table, either spatially or temporally (Rudolph et al. 2015).

Denitrification is one of the dominant nitrate attenuation processes in the subsurface. It involves the reduction of nitrate via a chain of microbial reduction reactions to nitrogen gases (nitrous oxide - partial denitrification, and di-nitrogen - full denitrification) (Knowles 1982). The extent of natural attenuation occurring along a subsurface continuum depends on many factors such as (but not limited to) oxygen and electron donor concentration and availability, NO_3^- concentration, pH, temperature and electrical conductivity (Rivett et al. 2008; Fenton et al. 2009; Rudolph et al. 2015). Throughout subsurface transport, nitrate will encounter combinations of the aforementioned factors; hence, concentrations discharging to the receptor at a delivery point will be a composite of all of these interactions. Prolonged t_T therefore facilitates higher opportunities for attenuation by simply increasing the likelihood of NO_3^- encountering optimum conditions for denitrification. Counterintuitively, a deadline-based approach to water quality improvements may thus not take full advantage of the potential for natural remediation.

Monitoring and modelling approaches

Attempts to quantify t_u have, to date, been challenging, leading to simplifications, such as an assumption of saturated soil conditions (Fenton et al. 2011); however, more realistic approaches must acknowledge the unsaturated nature of this component; numerical models incorporating the Richards equation (such as Hydrus 1D, VLEACH, or Hydrogeosphere) and site-specific meteorological and soil parameters have been proposed as a viable tool for this approach (Izbicki et al. 2015; Vero et al. 2017). Other complications include (but are not limited to) denitrification, dispersion and dilution (Nishikawa et al. 2003), equations for which may be incorporated into model supplements (e.g. PHREEQC; Parkhurst and Appelo 1999). Hrachowitz et al. (2016) discussed the challenges associated with modelling the dynamics of time lags and weighed the performance of physically or conceptually based models, of which there are many and varied approaches. A move towards a holistic approach, incorporating unsaturated, saturated and surface components, biochemical factors, and appropriate scales would likely help (Hrachowitz et al. 2016) but may be too intrinsically "data-hungry" to be readily implemented in many scenarios. Recent research has been encouraging, however—for example, Wang and Burke (2017) presented the "nitrate time bomb" (NTB) model, which, taking the Eden Valley basin (UK) as an example, incorporated both unsaturated and saturated zone components, and both matrix and fracture flow to produce aquifer NO_3^- concentration time series at catchment scale between 1925 and 2150. After demonstrating the national applicability of the NTB model, Wang et al. (2016) concluded that annual estimates produced in this fashion are useful for quantifying the effects of historical practices at relatively large scales; however, the introduction of more site-specific data and incorporation of complexities such as areas exhibiting multiple porosities, denitrification processes, climate projections etc., would improve the applicability of the model at a more localised scale. It is likely that such extensions to sound hydrologic modelling frameworks will be forthcoming in the near future. Another strong example of an integrated modelling framework is that presented by Wriedt and Rode (2006). In that study, N fertiliser inputs were distributed at various rates (ranging from 0 to 225 kg N/ha^{-1}) across a simulated catchment, exhibiting heterogeneous management, soil and aquifer characteristics. The distribution of the N source influenced t_T as a result of heterogeneous loading at the soil surface, which correlated with loads entering subsurface pathways. Assumption of uniform distribution of N sources overestimated t_T compared to the site-specific distribution approach. Furthermore, shifting from a conservative to a reactive modelling approach revealed opportunities for N transformations such that 80% of the NO_3^- load was denitrified before it reached a surface receptor. While that study demonstrates a high level of model integration and site-specific data, further work is required regarding transport and transformation processes within the hyporheic zone and in the receiving surface waterbody. It was also acknowledged that given the limitations in availability and resolution of data (soil, management, geology etc.) increases in model complexity may not trump uncertainty arising from input parameters. For the Walloon region (16.9 km^2) of Belgium a model integrating soil type and depth, slope, land use and meteorological data was used to evaluate t_u with a view to commenting on the efficacy of POM (Sohier et al. 2009). This approach has the benefit of integrating data (such as land use and soil descriptions) which are increasingly readily available.

Simulations of solute transport in baseflow-dominated systems suggest that groundwater NO_3^- concentrations are unlikely to decline for several decades after input has been reduced or stopped, and that increases resulting from historical nutrient loading are inevitable within the short term (Jackson et al. 2007, 2008; Vertes et al. 2008; Van Meter and Basu 2015). Many studies have also observed t_u in the field (Lindsey et al. 2003; Osenbrück et al. 2006)—one such example is provided by Vadas et al. (2007), who found that although soil NO_3^- increased with depth, there were no temporal fluctuations in groundwater NO_3^- over a 2.5-year period subsequent to the implementation of prescribed management practices. These results indicate that t_u was still ongoing, and that the effects of changed management practices in 1997 were yet to be observed at groundwater level, although decreases in NO_3^- could be observed in the upper layers of the soil. Other studies (Bekeris 2007; Sousa et al. 2013) have reported observing similar peaks in NO_3^- between 2 and 3 m below ground level in multi-layered soil profiles. These concentration gradients which increase with depth indicate scenarios in which a load of NO_3^- is moving vertically towards the water table, but which may have a delayed effect on water quality. In the absence of leaching and the associated time lag, it would be expected that the gradient would be reversed, with higher NO_3^- concentrations decreasing with depth. The gradual movement of such NO_3^- loads has led to time lag being alternately referred to as the "nitrate time bomb", both in peer-reviewed (Wang et al. 2012, 2013; Ward et al. 2013) and popular publications (Addiscott 2005).

European context

Subsequent to the Second World War, increased food production became a major European policy objective, and corresponding increases in fertiliser application and land use intensity were observed (Howden et al. 2010; Aquilina et al. 2012). However, beginning in the early 1970s, the attention of policymakers shifted towards environmental concerns, with the implementation of the first Environmental Action Program in 1973 (EC 1974). The primary legislation currently governing water quality in the European Union (EU) is the Water Framework Directive (WFD; 2000/60/EC, European Commission 2000). Under this legislation, member states are obliged to attain "good" qualitative status of all groundwater and surface waters, relative to fixed chemical thresholds (for example, a maximum allowable concentration of 11.3 mg L^{-1} in drinking water and of 37.5 mg L^{-1} in groundwater is specified for NO_3^-). The WFD specified an initial deadline (2015) by which this objective was desired; however, observed evidence of time lag as well as general scientific consensus indicated that attainment of this objective across all waterbodies was unachievable within the specified timeframe (Craig and Daly 2010; Fenton et al. 2011; Schulte et al. 2006). Extended deadlines (2021 and 2027) have been implemented in such instances; nevertheless, without considering time lags, it is not possible to anticipate the likely efficacy of POM with respect to these later reporting periods (Chyzheuskaya 2015). National policies have been implemented in response to the WFD, with a view to meeting water quality objectives; these policies specify management practices to control or offset nutrient delivery to receptors (Collins and McGonigle 2008)—as an example, within the Republic of Ireland, the Nitrates Directive (European Commission 1991) is the main policy mechanism in place intended to avert point and diffuse nutrient losses from agricultural land to water and minimise the risk of eutrophication. The suite of POM specified by this policy includes implementation of buffer strips around surface waters, prescribed livestock stocking rates, closed periods for fertiliser application corresponding to seasonally high rainfall rates, regulations pertaining to storage capacity for agricultural slurry and manures, and limits to fertiliser application rates. Derogation to this latter stipulation (which allows nitrogen (N) application of up to 230 kg ha^{-1} in Denmark and 250 kg ha^{-1} in all other countries), however, have become critical to attaining the production goals outlined by the current agri-environmental plans—Food Harvest 2020 (Dept. of Agriculture, Food and the Marine 2010) and Food Wise 2025 (Dept. of Agriculture, Food and the Marine 2015). Notably, all EU member states, apart from France, which does not operate a system legally defining such rates, have negotiated derogations to the N application limits (van Grinsven et al. 2013). Such negotiations suggest a discrepancy between legislative stipulations and the requirements of the burgeoning European agricultural sector. The percentage of agricultural land subject to these exemptions varies between member states, from only 1.5% of agricultural land in the United Kingdom to 50% in the Netherlands (Grant 2009). Appraisal of the efficacy of Nitrates Directive POM, at catchment scale, is conducted in Ireland by the Agricultural Catchments Program (ACP; Wall et al. 2012; Shore et al. 2013; Mellander et al. 2016); similar programs exist in other member states such as the National Agricultural Environmental Monitoring Program (JOVA) in Norway and the Demonstration Test Catchments in the UK. As such, the EU faces a particular challenge in designing and implementing environmental policies which are both suitable and effective across its 28 politically, environmentally and economically diverse member states (Hering et al. 2010). Bouma et al. (2002) reported heterogeneity in water-quality-policy efficacy resulting from soil and land use differences in the Netherlands, and have concluded that "fine-tuning" of regulatory policy and incorporation of geographical information systems are required due to these differences. Such a necessity observed in a relatively small member state (4.15 million ha) is correspondingly greater when similar water quality policies and targets are enacted across the entire EU (432.5 million ha).

Evidence for nitrogen-related time lags in both the unsaturated and saturated zones across Europe is extensive, with peer-reviewed publications originating from many EU member states—Fig. 1; Table S1 of the electronic supplementary material (ESM). The implications of time lag have been recognised by the scientific community for decades; a 1974 paper depicted increased groundwater N concentrations in an arable, chalk aquifer in which groundwater age was found to exceed 10 years (Foster and Crease 1974). Despite this, time lag remains poorly understood by the general public, and specifically, by some of the groups advocating more extensive legislation and POM. In 2010, for example, the European Environmental Bureau purported that time lag was a "generic excuse" to escape more stringent policy measures (Scheure and Naus 2010). This misconception must be overcome in order to facilitate informed and realistic environmental policies. The following sections will summarise knowledge of time lags, as observed across the EU.

Eastern Europe

Long-term fertilisation experiments have shown that important processes related to N turnover operate on a time scale of decades up to a century, and in several major Eastern European rivers there is a remarkable lack of response to the dramatic decrease in the use of commercial fertilisers that began in the late 1980s (Grimvall et al. 2000): for example, data from 1987 to 1998 in four rivers in this region—the Emajogi and Ohnejogi (Estonia), the Daugava (Latvia), and the Tisza (Hungary)—showed that the nutrient response to management changes was slow and limited in many rivers. Time lags were evident in medium-sized and large catchment areas, suggesting that factors other than reduced fertiliser application influenced the inertia of the water quality response (Stålnacke et al. 2003, 2004). Further studies in this geographical region, where nutrient inputs have substantially decreased, have found either no downward trends in NO_3^- concentrations (Procházková et al. 1996; Berankova and Ungerman 1996), or only limited downward trends (Tumas 2000) in large river catchments after years of observation.

Northern Europe

In Northern Europe, the water quality response to relatively recent decreases in N input to a catchment may be muted by the greater response to post-world war increases in input (Grimvall et al. 2000) and the time lag required for flushing of accumulated nutrients. In 1988, HELCOM in Sweden reached an agreement to reduce pollution transport from land via all pathways (including subsurface and overland), to 50% of the 1984 level by 1995. Success in reducing agricultural leaching, however, has been limited, with only a 15% reduction being achieved during this period (Arheimer and Brandt 2000). Cessation of state-supported agriculture in Baltic countries (Finland and Poland) during the late 1980s and early 1990s, led to reductions in fertiliser use (Granlund et al. 2005); nevertheless, river N still increased in some areas, which may indicate the ongoing arrival of N that had been migrating through the unsaturated and saturated zones. A review of water quality data from 1981 to 2000 shows that little or no reduction of riverine nutrient loads was achieved from

1995 to 1999 in response to POM implemented through the Finnish Agri-Environmental Programme in 1995, which intended to reduce N and phosphorus (P) loads by 50% (Granlund et al. 2005). Bechmann et al. (2008) have also observed strong inertia in environmental response in Norwegian catchments to mitigation strategies, including limits regarding stocking and fertiliser application rates. Greater annual N losses have been observed in Norwegian catchments than in other Nordic countries (Vagstad et al. 2001), which may correspond to nationally high application rates and measured soil N surpluses (Bechmann et al. 2008); in such scenarios, it is difficult to disentangle the direct effects of current practices from legacy effects. In Denmark, intensive application of fertilisers and leaching of NO_3^- from the soil and into aquifers began in the late 1950s, and these practices resulted in increased groundwater concentrations, which ultimately stabilised at ca. 1–2 mg L^{-1} NO_3^- in 1980 (Postma et al. 1991). Andersen Søgarrd et al. (2007) demonstrated that long travel times of groundwater make it difficult to relate current land use and NO_3^- leaching from soils to the discharge of NO_3^- to the marine environment. Interestingly, stringent Danish measures, which go beyond the requirements of the Irish Nitrates Directive (Botta and Kozluk 2014), have resulted in a decline in NO_3^- leaching from the root zone. Reductions in Denmark's surface-water NO_3^- concentrations (differentiating point from diffuse sources) were identified between 1992 and 2002 (Kronvang et al. 2008), but it took more than 20 years for these reductions to be observed. It therefore remains unclear as to whether such increases in stringency could facilitate achievement of WFD goals within the currently specified timeframes, suggesting that current deadline-based approaches may be less appropriate than a focus on trends and trajectories in water quality.

Mainland Europe

In the Netherlands, NO_3^- leaching to groundwater and N discharges to surface waters have been found to be related to N surpluses, hydrological condition, land use, and soil type (Oenema et al. 2005). Indeed, it has been calculated that decreasing the national N surplus by 1 kg ha^{-1} could on average decrease NO_3^- leaching to groundwater by 0.08 kg ha^{-1} and transport to surface waters on average by 0.12 kg ha^{-1} (Oenema et al. 2005). However, that study found that large reductions in N loading from agricultural sources (60% of surplus) produced relatively limited reductions in N concentrations of small rivers and groundwaters (20%). This is as a result of the joint confounding factors of time lag and alternative N point sources (industrial and domestic; Oenema et al. 2005). Oenema et al. (2005) proposed that nutrient surpluses are appropriately considered as indicators of "potential", rather than "actual" loss. Nitrogen surpluses on Dutch dairy farms dropped by an annual rate of ca. 7 kg N ha^{-1} year between 1980 and 2005 in response to the implementation of nutrient management legislation, which led to improvements in manure N fertiliser replacement values and reduced fertiliser application (Van den Ham et al. 2007). This reduction can be in part attributed to the implementation of nutrient management plans incorporating farm-gate balance accounting—the Mineral Accounting System (MINAS; Oenema et al. 2005). There has been some improvement in water quality in the Netherlands since this time however, van Grinsven et al. (2016) have maintained that current deadlines are unlikely to be achieved and that regionally specific mitigation measures are required. Results of modelling conducted by Sohier et al. (2009) for the Walloon region of Belgium indicated that 8% of the area would require in excess of 15 years for the effects of best management practices (BMPs) to be observed in groundwater. Such results demonstrate the decadal scale of N-related time lags, which is unlikely to be wholly reflected or observed in the current WFD 6-year reporting periods. Despite this, correcting imbalances between fertiliser application and crop nutrient offtake will reduce soil N surpluses, and bears relevance for the long-term attainment of ecological goals (Oenema et al. 2005).

Residence times of injected and environmental tracers in unconfined aquifers across the EU have been found to be as high as 10 years for a variety of catchment sizes and climatic regions—Switzerland, Greece, Germany and France) (Worthington 2007). These aquifers all exhibit triple porosity but with contrasting flow and storage in their matrices, fractures and channels. The matrices of such aquifers have high storage, leading to increased time lags, whereas briefer time lags occur in the channels and fractures. Wriedt and Rode (2006) have reported that NO_3^- concentrations reaching surface waters in lowland areas of northern Germany failed to reflect the initial levels of N losses from the surface due to denitrification and long residence times within the groundwater. Intensification of land-use in that member state, subsequent to the 1950s, allowed prolonged loading. The mixing of groundwater of different ages may therefore influence the present delivery of N to surface water. Attempts have been made to model time lags from soil to groundwater (Sousa et al. 2013; Vero et al. 2014) and discharge to surface water (Almasri and Kaluarachchi 2007) accounting for the three-dimensional (3D) and process-orientated nature of NO_3^- transport. Such a comprehensive modelling approach was implemented by Wriedt and Rode (2006), in the German context, and indicated an average t_T of 80 years in a 20 km^2

groundwater dominated catchment; however, given the high heterogeneity of soil and aquifer properties and their influence on both physical transport and denitrification capacity, the actual distribution of t_T may vary greatly.

Western Europe

Due to a history of glaciation and its position relative to the Atlantic Ocean, Western Europe exhibits a complex unsaturated zone, including highly heterogeneous soil and unsaturated bedrock, and, frequently, sedimentary aquifers. Consequently, catchments in this region (including Ireland and the United Kingdom) exhibit unique timescales and patterns of NO_3^- transport from sources to receptors. In the UK, the most rapid responses occur in alluvial sands and gravels, and limestone aquifers (e.g. Lincolnshire Limestone), while responses in deeper sandstone (e.g. Sherwood sandstone) and chalk aquifers (Hughes et al. 2007) can be on the order of decades. In parts of England, groundwater NO_3^- concentrations are increasing, not due to current practices, but due to the legacy of agricultural intensification over the past 50 years (ADAS 2007). In addition to agricultural intensification, other factors may be influencing these trends, including climate change and non-agricultural sources (e.g. sewage treatment works and industry). Such sources may be less significant in Ireland, which (both currently and historically) emphasises agricultural over industrial production (van Grinsven et al. 2013). The results of uniquely long-term monitoring of the Thames river basin over 140 years (Howden et al. 2010) revealed increasing NO_3^- concentrations in discharge over the study period, with sharpest increases as a result of land use and management changes in the 1940s and 1960s. Concentrations of NO_3^- in the Thames peaked in the 1980s, prior to the implementation of controls on N fertiliser application. Major trend reversals have not, however, been observed despite stabilisation of N loading and remediation efforts over the subsequent decades (Howden et al. 2010), which indicates the obstinate effects of time lag, particularly in large river basins, and further highlights the limitations of hydrologically short-term monitoring when used without appropriate historical context. Howden et al. (2010) noted that both the continued release of N bound within the soil, and the delay caused by slow movement of water and nutrients through the hydrologic system contribute to this effect. A study of river water quality in the eastern part of the Humber Basin, NE England, showed that reductions in fertiliser application did not correspond to a proportionate reduction in NO_3^- concentrations in the rivers due to soil processes within the catchment (Neal et al. 2008). Analysis of NO_3^- concentrations from long-term monitoring (1965–2007) of the River Frome in southern England, coupled with modelling exercises, indicated that it would take 12 years to increase mean NO_3^- concentrations by 1 mg L^{-1}, compared to 9 years on the nearby River Piddle (Howden and Burt 2009), demonstrating that within a relatively small area, subject to the same meteorological drivers, other factors such as soil type, land use and biochemical interactions will influence t_T. For a small number of sites both biogeochemical and hydrologic time lags operate concurrently. In some instances, the biogeochemical component may further prolong achievement of deadlines or conversely, may offer scope for the remediation of some contaminant transport (Fenton et al. 2017). In that study, biogeochemical time lags were on the order of decades, whereas hydrologic time lag at the same site ranged from months to years.

As nutrient management policies are designed to bring fertiliser application rates in line with plant requirements, it is likely that the continued elevation of river N concentrations will be driven, at least in part, by baseflow contributions (t_T; Spahr et al. 2010). Concomitantly, when increases occur in application rates of fertiliser, water quality may also remain unchanged due to lag time effects. Wang et al. (2016) presented a long-term (125-year) modelling approach to NO_3^- transport at regional and national scales in the UK. That study incorporated the nitrate time bomb model (Wang et al. 2013), with a national scale conceptual model for N loading and 28 zones defining the range of aquifer characteristics across England and Wales. Results demonstrated that "turning-point" changes in groundwater NO_3^- concentrations are to be anticipated in many catchments, on a multi-decadal scale, with IBT alone exceeding 11 months in 21 of the 28 study sites; for 13 of those study sites, the turning-point was predicted subsequent to 2020. In Lough Erne, located in the north-west Ireland, no temporal trend was found over a 25-year period, despite increased N loading from agriculture diffuse sources (Zhou et al. 2000). Other examples of limited groundwater NO_3^- responses to changed nutrient management policies have been observed in the UK over a 15-year period (Tomlinson 1970) and in a North American site over a 35-year period (Keeney and De Luca 1993). It should therefore be recognised that future water quality hazards may be as yet undetected due to the time lag currently affecting potential contaminants. Fenton et al. (2011) examined time lags through both the unsaturated and saturated zones in Ireland, and concluded that t_u alone could preclude attainment of WFD targets within the first reporting period (2015), thus highlighting the need for environmental trend assessment rather than fixed deadlines, in the design of water quality policies.

The European Indicator Assessment (European Environment Agency 2013) reported poor chemical status of 25% (by area) of EU groundwater. Poor status in >10% of groundwater bodies was exhibited by 16% of member states, while in excess of 50% of the groundwaters of Luxembourg, Belgium, Malta and the Czech Republic are failing to meet chemical targets (European Environment Agency 2013). Within this context, Ireland's performance as regards groundwater quality should be regarded as highly successful; however, 15% of Irish groundwaters exhibit a trend of increasing concentrations, which is likely to be the legacy of past management practices, in which leached NO_3^- is only now reaching the water table as a result of prolonged time lag. Analyses of 11 sites demonstrating increasing concentrations (Environmental Protection Agency, EPA 2015) indicated that these trends were significant in two locations (Fethard, Co. Tipperary, and Redcross, Co. Wicklow), and likely to increase mean N concentration above the 37.5 mg L^{-1} threshold for "good" quality by 2021.

North American context

Similar to Europe, North America dramatically increased food production after the Second World War, as the farming economy shifted to a new reliance on intensive production practices and a widespread use of pesticides and chemical fertilisers (Novotny 2002). Initially, there was little concern for the environmental implications of these changes until a convergence of factors in the 1960s led to a new environmental awareness in the North American public. These factors included the publication of Rachel Carson's *Silent Spring* (Carson 1962), the growth of the social justice movement (Bouleau 2008), burgeoning problems of eutrophication in Lake Erie (Makarewicz and Bertram 1991), and a series of high-profile reports of burning rivers, fish kills and declining shellfish populations (Hines 2013). This new awareness led to the introduction of sweeping new environmental legislation, including the 1970 Canada Water Act, the 1972 Clean Water Act (CWA) in the United States (also referred to as the "Federal Water Pollution Control Act Amendments"; United States Congress 1972) and the 1972 International Joint Commission's Great Lakes Water Quality Agreement (GLWQA; Bouleau 2008). The latter agreement came about as the culmination of multiple studies and reports suggesting that nutrient enrichment was the primary cause of eutrophication in the Great Lakes (Donahue 1999).

These new policies were unprecedented in their scope and focus on water quality. In the US, the dumping of pollutants into common waterways prior to the 1972 CWA was regulated only under "nuisance" law, meaning that it was not treated as problematic unless it was proved to cause unreasonable harm to another's property right or to the public interest (Hines 2013). Under the CWA, however, the US established a long-term goal of eliminating *all* discharge of pollutants to all navigable waterways and, to this end, adopted a variety of effluent limitations for pollutants of concern (Hines 2013). The Canada Water Act extended the federal reach of Canadian water law to include issues of water quality, specifically giving it the authority to regulate concentrations of nutrients in cleaning agents and water conditioners (1995). The GLWQA secured a binational commitment to water quality in the Great Lakes, establishing water quality objectives and providing for the development of water quality monitoring programs (Donahue 1999).

Despite this recent attention to water quality in North America, little was initially done to address non-point source nutrient pollution. Under the US CWA, farmers were not required to meet waste discharge requirements (Dowd et al. 2008), and under the Canada Water Act, nutrient pollution was addressed explicitly only as it related to phosphorus (P) in detergents (Hines 2013). Although the GLWQA represented a clear shift from point-source clean-up efforts to an emphasis on non-point source pollution (Donahue 1999), P, not N, was identified as the nutrient of concern, and targets were not set for reductions in N loading. In Canada, federal, provincial, and local agencies now play a role in managing non-point source pollution, in some cases under source-water-protection plans (Simms et al. 2010). Most strategies within such plans involve voluntary participation by farmers, with local agencies promoting and providing support for implementation of BMPs. In 1987, the US amended Section 319 of the CWA to cover non-point sources, but, similar to Canada, the actions under Section 319 include only voluntary, indirect control measures with state management programs providing information grants, and technical assistance (Griffiths and Wheeler 2005). Accordingly, the amendment has been criticised as relying too heavily on voluntary measures and not adequately addressing the impact of non-point sources on groundwater quality (Fentress 1988).

At regional and local scales, significant efforts have been made to reduce non-point source NO_3^- pollution—for example, in 1987, the Chesapeake Bay Program, a partnership of multiple states and the US EPA, committed to reducing "controllable" loading of N to the Chesapeake Bay by 40% by 2000 (Van Meter et al. 2016b). Although this commitment led to extensive implementation of nutrient-based BMPs in agricultural areas (Sharpley 1999; Hennessey 1994) progress has been limited and the nutrient reduction goals have still not been attained (Reckhow et al. 2011). Similarly, in 2008, the Mississippi River/Gulf of Mexico Watershed Nutrient Task Force created an action plan with the intent of reducing the Gulf of Mexico's summer hypoxic zone to less than 5,000 km^2 by 2015 (Rabotyagov 2014; USEPA 2008). In 2014 and 2015, however, the dead zones were 3–4 times the stated goal (13,085 km^2 and 16,768 km^2, respectively), despite the millions of dollars spent on establishing watershed BMPs. The goal has been postponed to 2035. Across the US, the EPA reported impairment of more than 33,000 waterbodies due to nonpoint source pollution (US Environmental Protection Agency 2011). Successful remediation of only 354 (or 1%) of these waterbodies was achieved, despite spending on the order of \$200 million annually to improve water quality under CWA Section 319. The following two sections examine current research and recent case studies regarding N-related time lags in Canada and the United States.

Canada

In Southern Ontario's Grand River Watershed (GRW), N surplus values have decreased on the order of 10–40% since reaching a peak in the mid- to late 1970s, due to a combination of improved nutrient management, increasing crop yields, and decreases in atmospheric N deposition (Van Meter et al. 2016a). Decreases in N concentrations and loads, however, have been slow to follow, and elevated NO_3^- levels in both surface water and groundwater are increasingly a threat to drinking water quality, particularly in rural areas (Goss et al. 1998; Sousa et al. 2013; Corkal and Adkins 2008; Wassenaar 1995). Research in upland areas of the GRW found no association between the abundance and location of agricultural BMPs and improvements in water quality during the summer season over a period of 3–15 years after their implementation (Pearce and Yates 2015).

A detailed study of NO_3^- lag times was conducted in Ontario for the Thornton well field, a site providing approximately 6,000 m^3/day of water to the town of Woodstock (Sousa et al. 2013). Application of N fertiliser in agricultural areas around the well field had led, in the 1990s, to NO_3^- concentrations above the maximum allowable concentration (MAC) of 10 mg L^{-1}. In response, the County of Oxford purchased approximately 100 ha of land in the capture zone of the well field in 2002 and then rented this land back to local farmers, with a strict cap being placed on fertiliser application rates. Concurrently, detailed site characterisation was conducted to evaluate BMP performance by monitoring the t_u of NO_3^-. Average soil pore water NO_3^- concentrations dropped from approximately 20 to 10 mg L^{-1} within 2 years of a 50% reduction in fertiliser application, while NO_3^- in groundwater wells reduced approximately linearly from ~10 in 2002 to 7 mg L^{-1} in 2013 (Rudolph 2015). The slower reduction in the wells compared to the pore water concentrations is reflective of t_u. Numerical modelling conducted at the site (Sousa et al. 2013) indicated total time lags in the range 7–40 years, with anywhere in the range 26–82% of that time being within the unsaturated zone (Sousa et al. 2013).

Time lag associated with NO_3^- contamination of drinking water wells has also been a significant problem in western Canada—for example, in the permeable sands and gravel of southwestern British Columbia's Abbotsford aquifer, an unconfined transboundary aquifer that has supported intensive agricultural activities for decades (Wassenaar 1995; Mitchell 2001; Cox and Kahle 1999; Zebarth et al. 2015). Elevated NO_3^- concentrations as high as 12.0 mg L^{-1} were first reported for the aquifer in the 1950s, and in 1989 63% of sampled wells were found to have concentrations exceeding the Canadian MAC of 10 mg L^{-1} (Liebscher et al. 1992). In 1992, the government of British Columbia signed into law the Code of Agricultural Practice for Waste Management (Ministry of Water, Land and Air Protection, British Colombia 1992; Wassenaar et al. 2006), which required implementation of a variety of BMPs, including improvements in livestock waste management and optimization of fertiliser application rates. The development of alternate markets for manure has led to the export of manure from the area and resulted in an estimated 50% reduction in the agricultural N surplus between 1991 and 2001 (Schreier et al. 2003). Despite such interventions, intensive monitoring has shown little improvement in mean groundwater NO_3^- concentrations (Zebarth et al. 2015). A comparison of NO_3^- concentrations between 1993 and 2004 in 31 monitoring wells showed increases in 64% of all wells, with a mean increase of approximately 6 mg L^{-1}. When both domestic and monitoring wells are considered, 59% exceeded NO_3^-drinking water standards (Wassenaar et al. 2006).

United States

One of the largest US programs to improve water quality has been the US Conservation Reserve Program (CRP), which subsidises farmers for the voluntary removal of environmentally sensitive land from agricultural production. The CRP was enacted in 1985, and, as of 2006, operates at an annual cost of approximately \$1.7 billion (Ribaudo 1989; Hansen 2007). Assessments of program benefits, however, have been mixed. Water quality improvements have been valued at approximately \$389 million per year (Hansen 2007), but how these modelled economic benefits translate into actual reductions in NO_3^- loads, particularly at the watershed scale, remain largely unclear.

Although there has been widespread implementation of agricultural BMPs in recent decades, most studies within the US have found only minimal improvements in water quality, and time lag has been found to be a primary cause of such short-term lack of success (Meals et al. 2010). Field studies indicate that NO_3^- lags in small and large watersheds across US can range from 4 years to more than 50 years (Meals et al. 2010). Tomer and Burkart (2003) report time lags >30 years in two 30 ha Iowa watersheds. As a result, the groundwater NO_3^- concentrations observed in those watersheds in 2003 were influenced by the application of agricultural fertilisers during the 1970s. Owens et al. (2008) reported that the response in groundwater quality to changes in fertilisation rates can range from 4 to 10 years, even in very small (<2 ha) watersheds, and that briefer lag times (ca. 3 years) occur when clay confining layers forced groundwater discharge pathways to be shallow. In another field study in the Pequea and Mill Creek Clean Water Act Section 319 National Nonpoint Source Monitoring Program (NNPSMP) Project (1994–2003) Pennsylvania, changes in fertiliser applications did not result in stream NO_3^- reductions, which was corroborated by groundwater age dating that indicated t_S of 15–39 years (Galeone 2005).

In the Walnut Creek Restoration NNSMP project in Iowa, 1,224 ha of row-crop land was converted to native prairie over a period of 14 years (1991–2005) in the 5,218 ha Walnut Creek watershed. Studies of stream and groundwater quality (Schilling and Spooner 2006; Tomer et al. 2010) showed statistically significant decreases in NO_3^- concentrations in both waterbodies, although the decreases lagged the establishment of reconstructed prairie by 3 years. Notably, this lag was clearly observed at a catchment scale, but became obscured across the entire river basin as a result of heterogeneous land uses. Using a coupled groundwater modelling (MODFLOW) and geographical information system (GIS) approach, t_S was estimated to range between 2 and 308 years, with mean travel times of 10–14 years (Schilling and Wolter 2007; Basu et al. 2012). Van Meter and Basu (2015) developed a reactive transport model for the same site, and found the trajectories for NO_3^- recovery at the catchment outlet could best be described by considering both the hydrologic and biogeochemical N legacy. Results of that study suggest that while it may take 5 years to achieve a 50% concentration reduction at the water table beneath converted sites, the time frame for achieving a similar reduction at the catchment outlet (reflecting t_T) would be strongly dependent on the spatial distribution of converted sites within the watershed and could range from 8 years to multiple decades.

Extensive research related to groundwater travel pathways has been conducted in the Chesapeake Bay watershed, United States (Hennessey 1994; Lindsey et al. 2003; Sanford and Pope 2013; Van Meter et al. 2016b; and others). Efforts to reduce NO_3^- loading in Chesapeake Bay have been ongoing for the last two decades, but have yielded limited success. The duration of t_S has been documented to range from years to decades, and a modelling study in the Delmarva peninsula to the east of Chesapeake Bay revealed t_T of the order of several decades before concentration reductions will be achieved (Sanford and Pope 2013). Van Meter et al. (2016a) used the ELeMENT travel time distribution model coupled with 200 years of N input data to estimate that 55 and 18% of the current annual N loads in the Mississippi River and the Susquehanna River Basins (the latter of which drains into Chesapeake Bay) were older than 10 years. In another study, Kauffman et al. (2008) developed a 3D groundwater flow model to examine the effects of land use and t_S durations on the distribution of NO_3^- in surface water and groundwater across an approximately 1,000 km^2 area of the southern New Jersey Coastal Plain in the eastern US. Model results showed a continual elevation of NO_3^- over background levels for multiple years even in scenarios where there was an immediate ban in fertiliser application. In another multi-site modelling study, McMahon et al. (2008) used groundwater tracer data (tritium, helium, and sulphur hexafluoride) along with a MODFLOW model to assess potential changes in water quality in public supply wells in California, Nebraska, Connecticut, and Florida. Time lag between contaminant inputs and the arrival of peak concentrations at the supply wells ranged between 6 and 30 years, with briefer t_T in wells with larger fractions of young water. The times required to flush 99% of the total NO_3^- mass after a complete cessation of inputs ranged from a mean of 17.5 years in the fastest contributing areas to more than 200 years in the slowest contributing areas.

Implications

Efficacy of intervention strategies

As evidenced by examples from the international literature, time lag cannot be dismissed as a "generic excuse" (Scheure and Naus 2010) considering the diverse soil, groundwater and landscape scenarios which influence its duration. Hence, where a waterbody fails to achieve water quality targets within a designated timeframe, it is critical to ascertain the influence of time lag in that catchment, in order to determine whether there has been an implementation or an efficacy issue. In other words, assuming full and timely implementation, it is impossible to observe the effects of intervention measures before the minimum lag time has elapsed, and before sufficient amounts of legacy N accumulated within the soil and groundwater have been flushed (Fenton et al. 2011). Where desired improvements in water quality are not met, it may be tempting to impose changes to the utilised intervention measures, particularly in response to political or cultural pressure. Indeed, a lack of understanding of hydrological principles may understandably lead the public to believe that current measures are not strenuous enough. Such a scenario may be particularly likely when qualitative thresholds and fixed deadlines are the primary evaluation tool (as in the EU), rather than trend analysis—as in parts of the US, e.g. North Dakota (Vecchia 2003). By characterising catchments exhibiting poor or declining water quality with respect to their associated time lags, it is possible that now there is better ability to evaluate the appropriateness of a given suite of intervention measures, and to determine whether either changes or extended deadlines are required. As demonstrated by Wang et al. (2016) and Vero et al. (2017), a modelling approach coupled with appropriate soil, geological and meteorological data may provide an optimal means to address this need. It should also be recognised that prolonged t_T may offer opportunities where biogeochemical processes are favourable (Rivett et al. 2008; Schilling et al. 2012; Jahangir et al. 2013b) for natural remediation, and so, could contribute to the effectiveness of intervention measures under appropriate circumstances.

Policy

In their commentary on agenda and policy dynamics, Baumgartner and Jonas (1991) noted that government policy typically exhibits prolonged durations of stability interspersed with briefer periods of extreme instability and change. Such changes may be reactive and strongly influenced by changes in prevailing influences on public opinion. Baumgartner and Jonas (1991) further noted that changes in public understanding of a scenario may be influenced not only by scientific research, but also by emotive or dramatic events. Water quality is obviously a politically sensitive issue, with almost insurmountable importance to sustainable growth and support of the growing global population. It is therefore important that the design of stable and effective environmental policies not be disproportionately influenced by any one group or opinion (Browne 1990), but rather, should incorporate both public participation and scientifically reliable information. It is also vital that policies which may be strongly market-driven (e.g. land conversion) should be considered in light of multi-decadal hydrologic implications (Howden et al. 2010).

Taking an example from the Republic of Ireland, in a 2006 debate on water quality in the upper house of government, a senator claimed to obtain his scientific information on the issue of nitrates as follows; “One does not have to be a scientist to appreciate the reality of the situation…I rely on information in the newspapers for this debate, some of which is very well-informed” (Seanad Éireann 2006). This demonstrated a reliance on or preference for popular publications, as opposed to appropriate scientific publications including the national EPA water quality reports (EPA 2005a, b), or one of the many peer-reviewed articles on water quality published, both nationally and internationally, that year. While popular publications no doubt have a place in environmental discourse, they cannot be assumed to be free of bias and opinion, nor to provide a comprehensive synthesis of the scientific literature. Hence, the former source should not supersede the latter. In this example, it is clear that despite extensive monitoring and research surrounding the issue by several different agencies, there remains a gap in effective knowledge transfer to key stakeholders, which must be addressed. The consequences of time lag in relation to NO_3^- contamination were similarly dismissed by the European Environmental Bureau (Scheure and Naus 2010); however, it is encouraging to note that such a stance is not ubiquitous in the realm of policy arena groups. The European Environmental Assessment acknowledged that “It is very difficult to prove a direct context between the application of nitrogen fertilizer in agriculture and the NO_3^- content in groundwaters as there is often a significant time lag between changes in agricultural practices and changes in NO_3^- concentrations in groundwater of up to 40 years, depending on the hydrogeological conditions” (Lindinger and Scheidleder 2004). Likewise, the United Kingdom Environment, Food and Rural Affairs Committee recognised in their 2008 report that prolonged t_T within catchments may obscure improvements in water quality (Environment, Food and Rural Affairs Committee 2008), and the Parliamentary Office of Science and Technology (2004) stated that “the complexity and geographical variability of catchments across the country [UK] means that no single set of DPW [diffuse pollution of water] reduction measures will be universally applicable.”

It is well established within scientific literature that time lags may present a significant impediment to achieving NO_3^- reduction goals within the designated reporting periods (Cherry et al. 2008). Nevertheless, there remains a discrepancy between the current legislative timeframes of the WFD (6-year periods) and the decadal to multi-decadal timescales associated with the physical movement of NO_3^- through the subsurface. The multiple difficulties posed by a threshold-based approach to environmental assessment have also been noted in relation to air emissions from EU member states (Ecologic Institute and Sustainable Europe Research Institute 2010). Accordingly, a trend-based approach may provide a more effective measure of interventions designed to decrease NO_3^- levels in receiving waterbodies. Furthermore, water quality monitoring regimes typically make evaluations based upon concentrations measured (1) at relatively low temporal resolution and (2) at few locations within a catchment, usually a surface waterbody or abstraction point. While increasing temporal resolution can be costly, it allows a more realistic assessment of the nitrate status of a catchment, as demonstrated by Shore et al. (2016) and Mellander et al. (2012). Similarly, implementing monitoring earlier within the pathway using shallow groundwater wells, or soil pore-water sampling can allow trends to be anticipated in advance of the receptor and can facilitate more detailed analysis of patterns and processes in hydrologically or biologically important areas (e.g. critical source areas and hotspots). Burt et al. (2010) highlighted the need for long-term water quality monitoring in order to quantify time lag, as monitoring programs <10 years in duration not only fail to capture the full extent of prolonged t_T, but also, are strongly influenced by inter-annual meteorologic variability. Similarly, Howden et al. (2010) demonstrated that monitoring programs of <15 years in duration may be insufficient to adequately inform long-term policy. This is a sentiment echoed throughout the literature and the efficacy of such extensive datasets with respect to t_T and trend analysis has been demonstrated both in the European (Burt et al. 2008) and North American (Stets et al. 2015) contexts. A commitment to integrated, long-term monitoring should therefore foster improved assessment of t_T, and hence, facilitate the design of implementable, effective, realistic and timely water quality policies.

Conclusions

The present review has demonstrated the ubiquity of N-related time lags in both the saturated and unsaturated zones as a hydrological occurrence, and the consequences of these lags across national and international scales. There seems ample evidence to suggest that a consideration of time lags must now become standard in the design of water quality policies, and that certain targets and deadlines prescribed by current policies may need review in light of current research. In light of the varied response to the issue of time lag in the policy domain, new strategies for the effective communication of both the theory and realities of time lag must be investigated.

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