



Provided by the author(s) and University of Galway in accordance with publisher policies. Please cite the published version when available.

Title	Review: the environmental status and implications of the nitrate time lag in Europe and North America.
Author(s)	Vero, Sara E.; Basu, Nandita B.; Van Meter, Kimberly; Richards, Karl G.; Mellander, Per-Erik; Healy, Mark G.; Fenton, Owen
Publication Date	2017-08-14
Publication Information	Vero, Sara E., Basu, Nandita B., Van Meter, Kimberly, Richards, Karl G., Mellander, Per-Erik, Healy, Mark G., & Fenton, Owen. (2018). Review: the environmental status and implications of the nitrate time lag in Europe and North America. <i>Hydrogeology Journal</i> , 26(1), 7-22. doi: 10.1007/s10040-017-1650-9
Publisher	Springer Verlag
Link to publisher's version	https://doi.org/10.1007/s10040-017-1650-9
Item record	http://hdl.handle.net/10379/7129
DOI	http://dx.doi.org/10.1007/s10040-017-1650-9

Downloaded 2024-04-26T02:44:47Z

Some rights reserved. For more information, please see the item record link above.



Published as: Vero, S.E., Basu, N.B., Van Meter, K., Richards, K.G., 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. *Hydrogeology Journal* 26: 7 – 22. DOI: 10.1007/s10040-017-1650-9

(Note: Not the final print version)

Nitrate time lag: A review of our current understanding and implications in Europe and North America

Sara E. Vero¹, Nandita B. Basu², Kimberly Van Meter², Karl G. Richards³, Per-Erik Mellander¹, Mark G. Healy⁴ and Owen Fenton³

¹ *Agricultural Catchments Programme, Teagasc, Environmental Research Centre, Johnstown Castle, Co. Wexford, Ireland*

² *Department of Earth and Environmental Sciences, University of Waterloo, 200 University Avenue West, Waterloo, ON N2L 3G1, Canada*

³ *Teagasc Environmental Research Centre, Johnstown Castle, Co. Wexford, Ireland*

⁴ *Civil Engineering, National University of Ireland Galway, Co. Galway, Ireland*

Target Journal – Water Resources Research – 25 publishing units: 1 unit = 500 words/1 table/1 figure

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 determine realistic deadlines and thresholds, and to design effective best management practices. The objective of this review is to synthesise the current state of research on time lag, in both the European and North American/Canadian environmental and legislative contexts. The durations of time lags have been found to differ according to differing climatic, pedological, landscape and management scenarios, and elucidation of these driving factors on a watershed scale is therefore essential where water quality is impaired or at risk. Finally, acknowledgement and understanding of time lag is increasingly seen at policy level, and incorporated in the development of environmental legislation. However, it is not yet ubiquitously appreciated, and continued outreach and education in scientific, public and policy venues is still required.

Keywords: Water Framework Directive; lag time; groundwater; unsaturated zone; nitrate

Introduction

The efficacy of water quality policies is influenced by both anthropogenic and non-anthropogenic controls (Bechman *et al.*, 2008). The latter category includes hydrologic (Fenton *et al.*, 2011) and biogeochemical (Jahangir *et al.*, 2013) time lags arising during the migration of excess reactive nitrogen (Nr) through subsurface pathways, which makes any correlation between programme of measure efficacy and water quality improvements difficult (Osenbrück *et al.*, 2006; Meals *et al.*, 2010; Fenton *et al.*, 2011; 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 hydrologic 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.

What is time lag?

The transport of water and nutrients, supplied to surface water bodies and groundwater abstraction points from agricultural, industrial or other sources, may occur via a range of hydrologic pathways, including overland flow, interflow, shallow and deep groundwater flow, and conduit flow (Archbold, 2010), and may also include cycling through a variety of biogeochemical pathways, including uptake into plants and soil organic matter (Sebilo *et al.* 2013). Transport along these pathways imposes a delay between the implementation of programs of measures (POM) for mitigation, which are designed to improve the quality of ground- or surface-waters (Schirmer *et al.*, 2014), and measurable improvements in water quality. This delay is referred to by a number of different terms, including a 'memory effect', 'delayed response', 'residence effect', 'legacy effect' or 'time lag' (which is the preferred term in the present manuscript) (Worrall and Burt, 1999; Bechmann *et al.*, 2008; lital *et al.*, 2008; Wahlin and Grimvall, 2008; Vero *et al.*, 2014; Hamilton 2012; Van Meter & Basu). 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 length of the 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, the scope for chemical processes and transformations (such as attenuation capacity (Jahangir *et al.*, 2013), difficulties in determining the length of retention times throughout the subsurface (Schueth *et al.*, XXXX), 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.*, 2016) 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).

Fenton *et al.* (2011) and Sousa *et al.* (2013) have divided the total time lag (t_T) via subsurface pathways into unsaturated zone (t_u) and groundwater (saturated) (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 until it reaches shallow groundwater (i.e. the water table). Nitrogen in the unsaturated zone can be either in the dissolved form, as NO_3^- , or as organic nitrogen sorbed to the soil matrix. Van Meter and Basu (2010) 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_s), porosity (n) and pore size distribution), effective rainfall or recharge (ER), and depth of the unsaturated zone. The magnitude of the biogeochemical legacy depends on the clay or mineral content of the soil as well as management factors such as the cropping pattern, levels of fertilizer application, the type of fertilizer applied, etc.

Arrival of NO_3^- at the water table takes the form of a breakthrough curve, which can be subdivided 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. 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, a more realistic approach 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).

Presumably as a result of these complexities, t_u has received less attention in the literature than the t_s portion of the time lag. 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; Jackson *et al.*, 2008; Vertes *et al.*, 2008; Van Meter and Basu 2015). Many studies have also observed t_u in the field (Lindsay *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; Rudolph *et al.*, 2015) have reported observing similar peaks in NO_3^- between 2 and 3 m below ground level in multi-layered soil profiles.

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. As such, the speed of transport (per unit area) is likely to be more rapid than in the unsaturated zone due to the greater availability of channels through which the solute may be transmitted. However, 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). Groundwater, in contrast to water in transit through the soil, 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. Furthermore, 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. Extensive research related to groundwater travel pathways has been conducted in the Chesapeake Bay, United States (Hennessey, 1994; Lindsey *et al.*, 2003; Sanford and Pope, 2013; Van Meter and Basu, In Review, and others). 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.*, 2013) means that abstracted groundwater samples may not reflect concentrations delivered through the soil to the water table, either spatially or temporally.

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. 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) (EU WFD; 2000/60/EC, OJEC, 2000). Under this legislation, member states are obliged to attain ‘good’ qualitative status of all ground 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 NO₃⁻). 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; however, 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. As an example: within the Republic of Ireland, the Nitrates Directive (European Council, 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 (DAFM), 2010) and Food Wise 2025 (DAFM, 2015)). Notably, all EU member states, apart from France, which does not operate a system which legally defines such rates, have negotiated derogations to the N application limits (van Grinsven *et al.*, 2012). 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 vary 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). As such, the EU faces a particular challenge in designing and implementing environmental policies which are both suitable and effective across its 28 diverse member states (Hering *et al.*, 2010). Bouma *et al.* (2002) have 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 (Figure 1). 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 our 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, 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, 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). In Baltic countries (Finland and Poland), cessation of state-supported agriculture in the late 1980s and early 1990s, led to reduced fertilizer use. 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 1995-1999 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 fertilizers and leaching of NO_3^- from the soil and into aquifers began in the late 1950s. These practices resulted in increased groundwater concentrations, which ultimately stabilised at c. 1-2 mm NO_3^- in 1980 (Postma *et al.*, 1991). Andersen *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). Due to time lags (t_u and t_s), however, actual decreases have not matched calculated values (Oenema *et al.*, 2005). Nitrogen surpluses on Dutch dairy farms dropped by an annual rate of c. $7 \text{ kg N ha}^{-1} \text{ year}$ between 1980 and 2005 in response to the implementation of nutrient management legislation, which led to improvements in manure N fertilizer replacement values and reduced fertilizer application (Van den Ham *et al.*, 2007). There has been some improvement in water quality in the Netherlands since this time (Anonymous, 2009); however, van Grinsven *et al.* (2016) have maintained that current deadlines are unlikely to be achieved and that regionally specific mitigation measures are required. 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-yr reporting periods.

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. Wreidt 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 and process-orientated nature of NO_3^- transport. Such a comprehensive approach was implemented by Wreidt and Rode (2006), in the German context, and indicated a time lag of c. 80-90 years in a 20 km^2 catchment. That study also concluded that 80% of the NO_3^- load was removed by denitrification before it reached a surface receptor.

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, a number of other factors may be influencing these trends, including climate change and non-agricultural sources (e.g. sewage treatment works and industry). Such sources are less significant in Ireland, which (both currently and historically) emphasises agricultural over industrial production (van Grinsven *et al.*, 2012). The British Geological Survey recently modelled aquifer response times for England and Wales (Jackson *et al.*, 2008). The results of that investigation indicated a timescale of decades (8 and 46 years to reach 50% breakthrough in alluvial and chalk aquifers, respectively) for most aquifer classes, during which time NO_3^- pollution will continue to reach discharge points, despite reductions in contemporary surface loadings (Jackson *et al.*, 2008). In this report, the modelled time for groundwater NO_3^- to decrease in limestone aquifers in the U.K. by 25 and 50 % were <10 years and between 18 and 22 years for high and low rainfall regions. Responses for alluvial aquifers in the UK were more rapid, with the time for NO_3^- to reduce by 50% being 8-11 years. These results reflect the generally small, shallow aquifers with short groundwater travel pathways observed in such regions. A study of river water quality in the eastern part of the Humber Basin, NE England, showed that reductions in fertilizer 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, compared to 9 years on the nearby river Piddle (Howden and Burt, 2009). This demonstrates 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 .

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 fertilizer, water quality may also remain unchanged due to lag time effects. Wang *et al.* (2016) have presented a long-term (125-yr) modelling approach to NO_3^- transport at regional and national scales in the UK. That approach demonstrated that 'turning-point' changes in

groundwater NO₃⁻ 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-yr period, despite increased N loading from agriculture diffuse sources (Zhou *et al.*, 2000). Other examples of such a scenario have been observed in the U.K. over a 15-yr period (Tomlinson, 1970) and in a North American site over a 35-yr period (Keeney and DeLuca, 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 (EEA), 2013) indicated poor chemical status of 25% (by area) of EU groundwater; 16% of member states exhibit poor status in >10% of groundwater bodies, while Luxembourg, Belgium, Malta and the Czech Republic are failing in excess of 50% of their groundwaters (EEA, 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 NO₃⁻ is only now reaching the watertable as a result of prolonged time lag. Analyses of these sites (EPA, 2015) indicated that these trends were significant in two locations, and likely to increase mean N concentration above the threshold by 2021.

North American Context

Similar to Europe, North America saw dramatic increases in 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 fertilizers (Novotny 2002). Initially, there was little concern for the environmental implications of these changes. In the 1960s, however, a convergence of factors led to a new environmental awareness in the North American public, including the publication of Rachel Carson's *Silent Spring* (1962), the growth of the social justice movement (Bouleau 2008), growing problems of eutrophication in Lake Erie (Neerls 2011), and a series of high-profile reports of burning rivers, fish kills and declining shellfish populations (Hines, 2013). This new awareness led to the passing of sweeping new environmental legislation, including the 1970 *Canada Water Act* (1970),

the 1972 Clean Water Act (CWA) in the United States (1972) and the 1972 International Joint Commission's Great Lakes Water Quality Agreement (GLWQA) (Bouleau 2008).

These new policies were unprecedented in their scope and focus on water quality. In the U.S., 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 U.S. 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, which came about as the culmination of multiple studies and reports suggesting that nutrient enrichment was the primary cause of eutrophication in the Great Lakes, 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 done initially to address nonpoint source nutrient pollution. Under the U.S. CWA, farmers were not required to meet waste discharge requirements (Dowd, XXXX), and under the Canada Water Act, nutrient pollution was addressed explicitly only as it related to phosphorus in detergents (Hines, 2013). Although the binational GLWQA represented a clear shift from point-source clean-up efforts to an emphasis on nonpoint source pollution (Donahue, 1999), phosphorus (P), not N, was identified as the nutrient of concern, and targets were not set for reductions in N loading. In Canada, both federal, provincial, and local agencies now play a role in managing nonpoint 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 best management practices. In 1987, the U.S. amended Section 319 of the CWA, to cover nonpoint sources, but, as in Canada, the actions under Section 319 include only indirect control measures, with state management programs providing information, grants, and technical assistance (Griffiths and Wheeler, 2005).

At the regional and local scales, significant efforts have been made to reduce nonpoint source NO_3^- pollution. For example, in 1987, the Chesapeake Bay Program, a partnership of multiple states and the U.S. EPA, committed to reducing "controllable" loading of N to the Chesapeake Bay by 40% by 2000 (Van Meter & Basu, in review). Although this commitment led to extensive implementation

of nutrient-based best management practices in agricultural areas (Sharpley, 1999, Hennessey, 1994), a 2011 report evaluating the success of these efforts notes that progress has been limited and that 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 calling for a reduction of the Gulf of Mexico's summer hypoxic zone to less than 5,000 km² by 2015 (Rabotyagov, 2014; USEPA, 2008). In 2014 and 2015, however, the dead zones were 3-4 times the stated goal (13,085 km² and 16,768 km², respectively), despite the millions of dollars spent on watershed BMP. The goal has now been postponed to 2035. Across, the U.S., the EPA has reported impairment of more than 33,000 waterbodies due to nonpoint source pollution (USEPA, 2011), and successful remediation of only 354 (or 1%) these waterbodies, despite spending on the order of \$200 million annually to improve water quality under CWA Section 319.

In the following two sections, we will focus on 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 & Basu, submitted). Decreases in N concentrations and loads, however, have been slow to follow, and elevated NO₃⁻ levels in both surface and groundwater are increasingly a threat to drinking water quality, particularly in rural areas (Goss *et al.* 1998; Sousa *et al.*, 2013; Corkal & Adkins, 2008; Wassenaar, 1995). A cross correlation analysis between N inputs and stream N concentrations revealed lag times across the GRW to range between 15-30 years, with the longest lags being associated with summer concentrations (**REF**). In a study carried out in upland areas of the GRW, Pearce and Yates (2015) 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.

A detailed study of NO₃⁻ lag times was conducted in Ontario for the Thornton well field, a site providing approximately 6000 m³/day of water to the town of Woodstock (Sousa *et al.*, 2013; County of Oxford, 2011). Application of N fertilizer in agricultural areas around the well field had led, in the 1990s, to NO₃⁻ concentrations above the maximum allowable concentration (MAC) of 10 mg-N/L. 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 fertilizer application rates. Concurrently, detailed site characterization was conducted to evaluate

BMP performance by monitoring NO_3^- t_u . Average soil pore water NO_3^- concentrations dropped from approximately 20 mg/L to 10 mg/L $\text{NO}_3\text{-N}$ within 2 years of a 50% reduction in fertilizer application, while NO_3^- in groundwater wells reduced approximately linearly from ~ 10 mg/L in 2002 to 7 mg/L in 2013 (Rudolph et al. XX). 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 ranging from 7-40 years, with anywhere from 26-82% of that time being within the unsaturated zone (Sousa et al. 2013).

Lag times associated with NO_3^- contamination of drinking water wells have 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 $\text{NO}_3\text{-N/L}$ 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 $\text{NO}_3\text{-N/L}$ (Liebscher et al., 1992). In 1992, the government of British Columbia signed into law the Code of Agricultural Practice for Waste Management (B.C. Ministry of Water, Land and Air Protection, 1992; Wassenaar et al., 2006), which required implementation of a variety of new BMPs, including improvements in livestock waste management and optimization of fertilizer 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 (Schreir *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 $\text{NO}_3\text{-N/L}$. When both domestic and monitoring wells are considered, 59% exceeded NO_3^- drinking water standards (Wassenaar, 2006).

United States

One of the largest U.S. programs to improve water quality has been the U.S. Conservation Reserve Program (CRP), which subsidises farmers for the voluntary removal of environmentally sensitive land from agricultural production (USDA, 2016). The CRP was enacted in 1985, and, as of 2006, operates at an annual cost of approximately \$1.7 billion (Ribaud, 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 lag times have been found to be a primary cause of such short-term lack of success (Meals et al., 2010). Field studies indicate that NO_3^- lag times 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 lag times > 30 years in two 30 ha Iowa watersheds, such that groundwater NO_3^- concentrations in 2003 was being influenced by fertilization in the 1970s. Owens et al (2008) reported that the response in groundwater quality to changes in fertilization rates can range from 4-10 years, even in very small (<2ha) watersheds, and that briefer lag times (c. 3 yrs) 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 fertilizer applications did not result in stream NO_3^- reductions, which was corroborated by groundwater age dating that indicated travel times of 15-39 years (Galeone, 2005).

In the Walnut Creek Restoration NNSMP project in Iowa, 1224 ha of row-crop land was converted to native prairie over a period of 14 years (1991-2005) in the 5218 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 approach (GIS), groundwater travel times were estimated to range between 2 – 308 yrs, with mean travel times between 10-14 years (Schilling and Wolter, 2007; Basu et al 2012). It was also determined that increased density of tile drainage would decrease t_u , and hence t_r , due to faster routing of flows through the subsurface pathways (Schilling 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 N legacy (N in a dissolved form, present in both soil and groundwater) and a biogeochemical N legacy (N that has undergone biogeochemical transformation to organic N and is retained within the soil matrix). Those results suggest that while it may take 5 yrs 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_r) would be strongly dependent on the spatial distribution of converted sites within the watershed and could range from 8 yrs to multiple decades.

In the Chesapeake Bay (CB) watershed, efforts to reduce NO_3^- loading have been ongoing for the last two decades, but have yielded limited success. Groundwater travel times have been documented to range from years to decades, and a modelling study in the Delmarva peninsula to the East of CB revealed t_T of the order of several decades before concentration reductions will be achieved (Sanford and Pope, 2013). Van Meter et al (2016) used the ELeMENT travel time distribution model coupled with 200 yrs 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 CB) were older than 10 years. In another study, Kauffman et al. (2008) developed a three-dimensional groundwater flow model to examine the effects of land use and groundwater travel times on the distribution of NO_3^- in surface- and groundwater across an approximately 1000 km² area of the southern New Jersey Coastal Plain in the eastern U.S. 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 fertilizer 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 yrs 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 utilized 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 United States e.g. North Dakota (Vecchia, 2003)). By characterising catchments exhibiting poor or declining water quality with respect to their associated time lags, we may be better able 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.

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.

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 reliance on popular publications was rather than any of the national EPA water quality reports (EPA, 2005a; EPA, 2005b), 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. 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 fertiliser 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 Department of Environment, Food and Rural Affairs (DEFRA) recognised in their 2008 report that prolonged t_T within catchments may obscure improvements in water quality, and the Parliamentary Office of Science and Technology (POST) (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-yr 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 (Sustainable Europe Research Institute, 2011). Accordingly, a trend-based approach may provide a more effective measure of interventions designed to decrease NO_3^- levels in receiving water bodies. Burt *et al.*, (2010) highlighted the need for long-term water quality monitoring in order to quantify these lags, as monitoring programs <10 yrs in duration were found to not only fail to capture the full extent of prolonged t_T , but also, are strongly influenced by inter-annual meteorologic variability. 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 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 specific targets and deadlines prescribed by current policies may need review in light of current research.

References

ADAS, 2007. Nitrates consultation supporting paper D1 on implementation of the Nitrates Directive in England. Nitrates in water – the current status in England (2006).

Almasri, M.N. and Kaluarachchi, J.J. 2007. Modelling nitrate contamination of groundwater in agricultural catchments. *Journal of Hydrology*. 343: 211-229

Andersen Søgarrd, M., Baron, L., Gudbjerg, J., Gregersen, J., Chapellier, D., Jakobsen, R. and Postma, D. 2007. Discharge of nitrate-containing groundwater into a coastal marine environment. *Journal of Hydrology*. 336: 98-114

Anonymous, 2009. Planbureau voor de Leefomgeving, Bilthoven, The Netherlands, 248 pp.

Aquilina, L., Vergnaud-Ayraud, V., Labasque, T., Bour, O., Molenat, J., Ruiz, L., de Montety, V., De Ridder, J., Roques, C. and Longuevergne, L. 2012. Nitrate dynamics in agricultural catchments deduced from groundwater dating and long-term nitrate monitoring in surface- and groundwaters. *Science of the Total Environment*. 435-436: 167-178

Archbold, M., Bruen, M., Deakin, J., Doody, D., Flynn, R., Kelly-Quinn, M., Misstear, B. and Offerdinger, U. 2010. Contaminant movement and attenuation along pathways from the land surface to aquatic receptors. A review. STRIVE Report Series No. 56. https://www.epa.ie/pubs/reports/research/water/STRIVE_56_Archbold_Contaminant_sPathwaysReview_web.pdf Last downloaded 1/11/15

Arheimer, B. and Brandt, M. 2000. Watershed modelling of non-point nitrogen pollution from arable land in the Swedish coast in 1985 and 1994. *Ecological Engineering*. 14: 389-404.

Azevedo, L.B., van Zelm, R., Leuven, R.S.E.W., Jan Hendricks, A. and Huijbregts, M.A.J. 2015. Combined ecological risks of nitrogen and phosphorus in European freshwaters. *Environmental Pollution*. 200: 85-92

Bain, D.J., Green, M.B., Campbell, J.I., Chamblee, J.F., Chaoka, S., Jennifer M. Fraterrigo, J.M., Kaushal, S.S., Martin, S.I., Jordan, T.E., Parolari, A.J., Sobczak, W.V., Weller, D.E., Wollheim, W.M., Boose, E.R., Duncan, J.M., Gettel, G.M., Hall, B.R., Kumar, P., Thompson, J.R., Vose, J.M., Elliott, E.M. and Leig, D.S. 2012. Legacy effects in material flux: Structural catchment changes in predate long-term studies. *Bioscience*. 62(6)

Basu, N.B., Jindal, P., Schilling, K.E., Wolter, C.F. and Takle, E.S. 2012. Evaluation of analytical and numerical approaches for the estimation of groundwater travel time distribution. *Journal of Hydrology*. 475: 65-73

Baumgartner, F.R. and Jonas, B.D. 1991. Agenda dynamics and policy subsystems. *The Journal of Politics*. 53(4): 1044-1074

Bechmann, M., Deelstra, J., Stålnacke, P., Eggestad, H.O., Øygarden, L. and Pengerud, A, 2008. Monitoring catchment scale agricultural pollution in Norway: policy instruments, implementation of mitigation methods and trends in nutrient and sediment losses. *Environmental Science & Policy*. 11: 102-114.

Behrendt, H., Huber, P., Kornmilch, M., Opitz, D., Schmoll, O., Scholz, G. and Uebe, R. 2000. Nutrient Emissions into river basins of Germany. UBA-Texte, 23/00, p 266.

Bekeris, L. 2007. Field-scale evaluation of enhanced agricultural management practices using a novel unsaturated zone nitrate mass load approach. Master's thesis, Department of Earth Sciences, University of Waterloo, Waterloo, Ontario.

Berankova, D. and Ungerman, J. 1996. Nonpoint sources of pollution in the Morava river basin. *Water Science and Technology*. 33: 127-135

Bilotta, G.S., Milner, A.M. and Boyd, I. 2014. On the use of systematic reviews to inform environmental policies. *Environmental Science and Policy*. 42: 67-77

Botta, E. and T. Kozluk, T. 2014. Measuring environmental policy stringency in OECD countries: A composite index approach. OECD Economics Department Working Papers No. 1177, OECD Publishing

Bouma, J., Van Alphen, B.J. and Stoorvogel, J.J. 2002. Fine tuning water quality regulations in agriculture to soil differences. *Environmental Science and Policy*. 5: 113-120

Bourgine, B., Lopez, B. and Baran, N. 2012. Characterisation of groundwater nitrate long term trends at regional scale using statistical and geostatistical tools. Ninth International Geostatistics Congress, Oslo, Norway June 11 – 15, 2012

Browne, W.P. 1990. Organised interests and their issue niches: A search for pluralism in a policy domain. *Journal of Politics*. 52(2): 477-509

Burt, T.P., Howden, N.J.K., Worrall, F. and Whelan, M.J. 2008. Importance of long-term monitoring for detecting environmental change: lessons from a lowland river in south east England. *Biogeosciences* 5: 1529-1535

Burt T.P., Howden, N.J.K., Worrall, F. and Whelan, M.J. 2010. Long-term monitoring of river water nitrate: How much data do we need? *Journal of Environmental Monitoring* 12: 71–79

Canada Water Act 1970

Carson, R. 1962. *Silent Spring*. Houghton Mifflin Company. United States

Cherry, K.A., Shepard, M., Withers, P.J.A. and Mooney, S.J., 2008. Assessing the effectiveness of actions to mitigate nutrient loss from agriculture: A review of methods. *Science of the Total Environment*. 406: 1-23.

Chyzheuskaya, A. 2015. Modelling the economics of water pollution improvement in the agricultural context. Ph.D thesis. National University of Ireland, Galway.

Clean Water Act 1972. Clean Water Act.

Collins, A.L., McGonigle, D.F., 2008. Monitoring and modelling diffuse pollution for agriculture policy support: UK and European experience. *Environmental Science and Policy*. 11: 97-101.

Corkal, D.R. and Adkins, P.E. 2008. Canadian agriculture and water. IWRA World Water Congress 1st-4th September. Montpellier, France

County of Oxford 2011

Cox, S.E. and Kahle, S.C. 1999. Hydrogeology, ground-water quality, and sources of nitrate in lowland glacial aquifers of Whatcom County, Washington, and British Columbia, Canada. Water-Resources Investigations Report 98-4195. United States Geological Survey

Craig, M. and Daly, D. 2010. Methodology for establishing threshold values and the assessment of chemical and quantitative status of groundwater, including an assessment of pollution trends and trend reversal. Hydrometric and Groundwater Section, Office of Environmental Assessment, EPA, Dublin.

Dept. of Agriculture, Food and the Marine. 2010. Food Harvest 2020. A Vision for Irish Agri-Food and Fisheries. <https://www.agriculture.gov.ie/media/migration/agri-foodindustry/foodharvest2020/2020FoodHarvestEng240810.pdf> Last downloaded 1/11/15

Dept. of Agriculture, Food and the Marine. 2015. Food Wise 2025. A 10-year vision for the Irish agri-food industry. <http://www.agriculture.gov.ie/media/migration/agri-foodindustry/foodwise2025/report/FoodWise2025.pdf>. Last downloaded 15/10/15

Dept. of the Environment, Food and Rural Affairs 2008

Donahue 1999

Dowd

Environmental Protection Agency. 2005a. The quality of drinking water in Ireland: A report for the year 2004. EPA, Johnstown Castle, Wexford, Ireland

Environmental Protection Agency. 2005b. Water Quality in Ireland 2001-2003. EPA, Johnstown Castle, Wexford, Ireland

European Community, 1974. Declaration on the environmental action program. *International Legal Materials*. 13(1): 164-216

European Commission, 1991. Council Directive (1991/676/EC). EC Directive of the European Parliament and of the Council of 12 December 1991 on the protection of waters against pollution caused by nitrates from agricultural sources. Brussels. pp 1-8

European Commission, 2000. Council Directive (2000/60/EC). EC Directive of the European Parliament and of the Council 200/60/EC establishing a framework for community action in the field of water policy. Brussels. pp72

European Environment Agency. 2013. Chemical status indicators for water across Europe. <http://www.eea.europa.eu/data-and-maps/indicators/wfd-indicator-chemical-status/assessment>
Last accessed 26/10/15

Fenton, O., Richards, K.G., Kirwan, L., Khalil, M.I., Healy, M.G. 2009. Factors affecting nitrate distribution in shallow groundwater under a beef farm in South Eastern Ireland. *Journal of Environmental Management*. 90: 3135–3146.

Fenton, O., Schulte, R.P.O., Jordan, P., Lalor, S.T.J. and Richards, K.G. 2011. Time lag: A methodology for estimation of vertical and horizontal travel and flushing timescales to nitrate threshold concentrations in Irish aquifers. *Environmental Science and Policy*. 14(4): 419-431

Frind, E.O., Molson, J.W. and Rudolph, D.L. 2006. Well vulnerability: A quantitative approach for source water protection. *Groundwater*. 44(5): 732-742

Goss, M.J., Barry, D.A.J., Rudolph, D.L. 1998. Contamination in Ontario farmstead domestic wells and its association with agriculture: 1. Results from drinking water wells. *Journal of Contaminant Hydrology* 32: 267–293.

Granlund, K., Räike, A., Ekholm, P., Rankinen, K. and Rekolainen, S., 2005. Assessment of water protection targets for agricultural nutrient loading in Finland. *Journal of Hydrology*. 304: 251-260.

Grant, R. 2009. Manure management in a European context. Efficient and reliable utilisation of manure nutrients. Manure application management conference, Teagasc, Johnstown Castle, November 2009.

Great Lakes Water Quality Agreement 1972

Griffiths, C. and Wheeler, W. 2005. Benefit-cost analysis of US regulations affecting surface water quality. In R. Brouwer and D. Pierce (Eds.) Cost-benefit analysis and water resources management. Cheltenham, United Kingdom.

Grimvall, A., Stålnacke, P. and Tonderski, A., 2000. Timescales of nutrient losses from land to sea - a European perspective. *J. Ecological Engineering*. 14: 363-371.

Hamilton, S.K. 2012. Biogeochemical lags may delay responses of streams to ecological restoration. *Freshwater Biology*. 57(1): 43-57

Hansen, L. 2007. Conservation reserve program: Environmental benefits update. *Agricultural and Resource Economics Review*. 36(2): 1-14

Hennessey, T.M. 1994. Governance and adaptive management for estuarine ecosystems: The case of Chesapeake Bay. *Coastal Management*. 22: 119-145

Hering, D., Borja, A., Carstensen, J., Carvalho, L., Elliott, M., Feld, C. K., Heiskanen, A.-S., Johnson, R. K., Moe, J., Pont, D., Solheim, A. L., De Bund, W. V. 2010. The European Water Framework Directive at the age of 10: A critical review of the achievements with recommendations for the future. *Science of the Total Environment*. 408: 4007-4019.

Hines, 2013

Hocking, M. and Kelly, B.F.J. 2016. Groundwater recharge and time lag measurement through vertisols using impulse response functions. *Journal of Hydrology*. 535: 22-35

Howden, N.J.K. and Burt, T.P. 2009. Statistical analysis of nitrate concentrations from the Rivers Frome and Piddle (Dorset, UK) for the period 1965–2007. *Ecohydrology* 2(1): 55-65

Hughes, A., Chilton, J. and Williams, A. 2007. Review and categorisation of nitrate transport in groundwater systems: Final report on the work to end of March 2007, Report to DEFRA contract NIT18, British Geological Survey, London, 20 pages.

lital, A., Pachel, K. and Deelstra, J., 2008. Monitoring of diffuse pollution from agriculture to support implementation of the WFD and the Nitrate Directive in Estonia. *Environmental Science & Policy*. 11: 185-193.

- Izbicki, J.A., Flint, A.L., O'Leary, D.R., Nishikawa, T., Martin, P., Johnson, R.D. and Clark, D.A. 2015. Storage and mobilisation of natural and septic nitrate in thick unsaturated zones, California. *Journal of Hydrology*. 524: 147-165
- Jackson, B.M., Wheeler, H.S., Wade, A.J., Butterfield, D., Mathias, S.A., Ireson, A.M., Butler, A.P., McIntyre, N.R. and Whitehead, P.G. 2007. Catchment-scale modelling of flow and nutrient transport in Chalk unsaturated zone. *Ecological Modelling*. 41-51.
- Jackson, B.M., Browne, C.A., Butler, A.P., Peach, D., Wade, A.J. and Wheeler, H.S. 2008. Nitrate transport in Chalk catchments: monitoring, modelling and policy implications. *Environmental Science and Policy*. 125-135.
- Jahangir, M.M.R., Johnston, P., Addy, K., Khalil, M.I., Groffman, P.M. and Richards, K.G. 2013. Quantification of in situ denitrification rates in groundwater below an arable and a grassland system. *Water Air and Soil Pollution*. 224:1693
- Kauffman et al 2008
- Keeney, D.R. and De Luca, T.H. 1993. Des Moines River nitrate in relation to watershed agricultural practices.: 1945 versus 1980s. *Journal of Environmental Quality* 22(2): 267-272
- Kronvang, B., Andersen, H.E., Børgesen, C., Dalgaard, T., Larsen, S.E., Bøgestrand, J. and Blicher-Mathiasen, G., 2008. Effects of policy measures implemented in Denmark on nitrogen pollution of the aquatic environment. *Environmental Science and Policy*. 11: 144-152.
- Lasagna, M., Franchino, E. and De Luca, D. 2015. Areal and vertical distribution of nitrate concentration in Piedmont Plain aquifers (North-Western Italy). In: *Engineering Geology for Society and Territory: Vol. 3. Chapter: 81*. Springer International Publishing. Eds: Lollino, G., Arattano, M., Rinaldi, M., Giustolisi, O., Marechal, J-C. Gordon E. Grant, G.E. pp.389-392
- Liebscher, H., Hii, B. and McNaughton, D. 1992. Nitrogen and pesticides in the Abbotsford aquifer, southwestern British Columbia. Inland Waters Directorate, Environment. North Vancouver, British Columbia, Canada
- Lindinger, H. and Scheidleder, A. 2004. Indicator fact sheet. (WEU1) Nitrate in groundwater. European Environmental Assessment.
- Lindsey, B.D., Phillips, S.W., Donnelly, C.A., Speiran, G.K., Plummer, L.N., Böhlke, J-K., Focazio, M.J., Burton, W.C. and Busenberg, E. 2003. Residence times and nitrate transport in ground water discharging to streams in the Chesapeake Bay watershed. United States Geological Survey. Water-Resources Investigation Report 03-4035

Löfgren, S., Steineck, S. and Jacobsson, C., 1998. Model analysis of environmental impact from two hypothetical agricultural production systems in Sweden, Denmark and Lithuania in the years 2010 and 2030. Baltic Sea Agenda 21. JTI-report, nr 243, Institute of Agricultural Engineering, Uppsala, Sweden.

McMahon, P.B., Böhlke, J.K., Kauffman, L.J., Kipp, K.L., Landon, M.K., Crandall, C.A., Burrow, K.R. and Brown, C.J. 2008. Source and transport controls on the movement of nitrate to public supply wells in selected principal aquifers of the United States. *Water Resources Research*. 44: W04401, doi:10.1029/2007WR006252

Meals, D.W., Dressing, S.A. and Davenport, T.A. 2010. Time lag in response to best management practices: A review. *Journal of Environmental Quality*. 39: 85-96

Mellander, P-E., Jordan, P., Shore, M., McDonald, N.T., Wall, D.P., Shortle, G. and Daly, K. 2016. Identifying contrasting influences and surface water signals for specific groundwater phosphorus vulnerability. *Science of the Total Environment*.

Ministry of Water, Land and Air Protection, British Columbia 1992

Mitchell, M.J. 2001. Linkages of nitrate losses in watersheds to hydrological processes. *Hydrological Processes*. 15(17): 3305-3307

Neal, C., Davies, H. and Neal, M. 2008. Water quality, nutrients and the water framework directive in an agricultural region: The lower Humber Rivers, northern England. *Journal of Hydrology*. 350: 232-245.

Nishikawa, T., Densmore, J.N., Martin, P. and Matti, J. 2003. Evaluation of the source and transport of high nitrate concentrations in groundwater, Warren Sub-basin, California. United States Geological Survey. Water Resources Investigations Report 03-4009. Sacramento, California.

Novotny, V. 2002. Water quality, diffuse pollution and watershed management 2nd Edition. Wiley

Oenema, O., van Liere, L. and Schoumans, O., 2005. Effects of lowering nitrogen and phosphorus surpluses in agriculture on the quality of groundwater and surface water in the Netherlands. *Journal of Hydrology*. 304: 289-301.

Osenbrück, K., Fiedler, S. Knöller, K., Weise, S.M., Sültenfuß, J., Oster, H. and Strauch, G. 2006. Timescales and development of groundwater pollution by nitrate in drinking water wells of the Jahna-Aue, Saxonia, Germany. *Water Resources Research*. 42(12)

Owens, L., Shipitalo, M.J. and Bonta, J.V. 2008. Water quality response times to pasture management changes in small and large watersheds. *Journal of Soil and Water Conservation*. 63(5): 292-299

Parkhurst, D.L. and Appelo, C.A.J. 1999. User's guide to PHREEQC (Version 2) – A computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations. U.S.G.S. Water-Resources Investigations Report. 99-4259

Parliament 2004

Pearce, N.J.T. and Yates, A.G. 2015. Agricultural best management practice abundance and location does not influence stream ecosystem function or water quality in the summer season. *Water*. 7: 6861-6876

Postma, D., Boesen, C., Kristiansen, H. and Larsen, F. 1991. Nitrate reduction in an unconfined aquifer: water chemistry, reduction processes, and geochemical modeling. *Water Resources Research*. 27: 2027-2045

Procházková, L., Blažka, P. and Kopáček, J. 1996. Impact of diffuse pollution on water quality of the Vltava river (Slapy reservoir), Czech Republic. *Water Science and Technology*. 33: 145-152

Rabotyagov, S.S. 2014. Cost-effective targeting of conservation investments to reduce the northern Gulf of Mexico hypoxic zone. *Proceedings of the National Academy of Sciences of the United States of America*. 111(52): 18530-18535

Reckhow et al 2011

Ribaudo, M.O. 1989. Targeting the conservation reserve program to maximise water quality benefits. *Land Economics*. 65(4): 320-332

Rudolph et al ??

Rudolph, D.L., Devlin, J.F. and Bekeris, L. 2015. Challenges and a strategy for agricultural BMP monitoring and remediation of nitrate contamination from unconsolidated aquifers. *Groundwater Monitoring and Remediation*. 35(1): 97-109

Sanford, W.E., and Pope, J.P. 2013. Quantifying groundwater's role in delaying improvements to Chesapeake Bay water quality. *Environmental Science & Technology*. 47(23): 13330–13338

Scheure, S. and Naus, J. 2010. 10 Years of the Water Framework Directive: A toothless tiger? A snapshot assessment of EU environmental ambitions. Ed. Hontelez, J. European Environmental Bureau.

Scheuth et al ???

Schilling, K.E., and Spooner, J. 2006. Effects of watershed scale land use change on stream nitrate concentrations. *Journal of Environmental Quality*. 35: 2132–2145. doi:10.2134/jeq2006.0157

Schilling, K.E. and Wolter, C.F. 2007. A GIS-based groundwater travel time model to evaluate stream nitrate concentration reductions from land use change. *Environmental Geology*. 53(2): 433-443

Schilling, K.E., Jones, C.S., Seeman, A., Bader, E. and Filipiak, J. 2012. Nitrate-nitrogen patterns in engineered catchments in the Upper Mississippi River basin. *Ecological Engineering*. 42:1–9. doi:10.1016/j.ecoleng.2012.01.026

Schirmer, M., Luster, J., Linde, N., Perona, P., Mitchell, E.A.D., Barry, D.A., Hollender, J., Cirpka, O.A., Schneider, P., Vogt, T., Radny, D. and Durisch-Kaiser, E. 2014. Morphological, hydrological, biogeochemical and ecological challenges in river restoration – the Thur river case study. *Hydrological Earth Systems Sciences*. 18: 2449-2462

Schulte, R.P.O., Richards, K., Daly, K., Kurz, I., McDonald, E.J. and Holden, N.M. 2006. Agriculture, meteorology and water quality in Ireland: A regional evaluation of pressures and pathways of nutrient loss to water. *Biology and Environment: Proceedings of the Royal Irish Academy*. 106 (2): 117-133

Seanad Éireann, 2006. *Seanad Éireann Debate*. 182(46).

Sharpley, A. 1999. Modern agriculture and the environment. *Journal of Environmental Quality*. 28(1): 356

Shen, S.M., Hart, P.B.S., Powlson, D.S. and Jenkinson, D.S. 1989. The nitrogen cycle in the Broadbalk Wheat Experiment: 15N labeled fertilizer residues in the soil and in the soil microbial biomass. *Soil. Biol. Biochem*. 21: 529–533.

Shore, M., Murphy, P.N.C., Jordan, P., Mellander, P-E., Kelly-Quinn, M., Cushen, M., Mechan, S., Shine, O. and Melland, A.R. 2013. Evaluation of a surface hydrological connectivity index in agricultural catchments. *Journal of Environmental Modelling Software*. 47: 7-15.

Simms, G., Lightman, D. and de Loë. R. 2010. Tools and approaches for source water protection in Canada, Governance Source Water Prot. Can. Rep., 1, Water Policy and Governance Group, Waterloo, Ont., Canada.

Spahr, N.E., Dubrovsky, N.M., Gronberg, J.M., Franke, O.L. and Wolock, D.M. 2010. Nitrate Loads and Concentrations in Surface-Water Base Flow and Shallow Groundwater for Selected Basins in the United States, Water Years 1990–2006. United States Geological Survey. National Water Quality Assessment Program. Scientific Investigations Report 2010-5098

Sohier, C., Dautrebande, S. and Degre, A. 2009. Hydrological modelling of the EU Nitrates Directive action programme: new developments in the Walloon region (Belgium). International workshop: Towards new methods to manage nitrate pollution within the Water Framework Directive 10 & 11 December, 2009, UNESCO – Paris

Sousa, M.R., Jones, J.P., Frind, E.O. and Rudolph, D.L. 2013. A simple method to assess unsaturated zone time lag in the travel time from ground surface to receptor. *Journal of Contaminant Hydrology*. 144: 138-151

Stålnacke, P., Grimvall, A., Libiseller, C., Laznik, M., Kokorite, I. 2003. Trends in nutrient concentrations in Latvian rivers and the response to the dramatic change in agriculture. *Journal of Hydrology*. 283: 184-205.

Stålnacke, P., Vandsemb, S.M., Vassiljev, A., Grimvall, A. and Jolankai, G. 2004. Changes in nutrient levels in some Eastern European rivers in response to large-scale changes in agriculture. *Water Science Technology*. 49: 29-36.

Stets, E.G., Kelly, V.J. and Crawford, C.G. 2015. Regional and temporal differences in nitrate trends discerned from long-term water quality monitoring data. *Journal of the American Water Resources Association* 51(5): 1394-1407

Sustainable Europe Research Institute, 2011. Establishing Environmental Sustainability Thresholds and Indicators. Fact sheet on non-renewable resource use.

Tomer, M.D. and Burkart, M.R. 2003. Long-term effects of nitrogen fertilizer use on ground water nitrate in two small watersheds. *Journal of Environmental Quality*. 32: 2158–2171

Tomer, M.D., Schilling, K.E., Cambardella, C.A., Jacobson, P. and Drobney, P. 2010. Groundwater nutrient concentrations during prairie reconstruction on an Iowa landscape. *Agriculture, Ecosystems and Environment*. 139: 206-213

Tomlinson, T, E. 1970. Trends in nitrate concentrations in English rivers in relation to fertilizer use. *Water Treatment and Examination*. 19: 277-293

Tumas, R. 2000. Evaluation and prediction of non-point pollution in Lithuania. *Ecological Engineering*. 14: 443-451.

United States Department of Agriculture 2016

USEPA 2008

USEPA 2011

- Vadas, P.A., Srinivasan, M.S., Kleinman, P.J.A., Schmidt, J.P. and Allen, A.L. 2007. Hydrology and groundwater nutrient concentrations in a ditch-drained agroecosystem. *Journal of Soil and Water Conservation*. 62(4): 178-188
- Vagstad, N., Stalnacke, P., Andersen, H.E., Deelstra, J., Gustavson, A., Iital, A., Jansons, V., Kyllmar, K., Loigu, E., Rekolainen, S., Tumas, R. and Vuorenmaa, J. 2001. Nutrient losses from agriculture in the Nordic and Baltic countries. Measurements in small agricultural catchments and national agro-environmental statistics. TemaNord Report 2001, 591
- Van Beek, C.L., van der Salm, C., Plette, A.C.C., van de Weerd, H., 2009. Nutrient loss pathways from grazed grasslands and the effects of decreasing inputs: experimental results from three soil types. *Nutrient Cycling in Agroecosystems*. 83 (2): 99-110.
- Van den Ham, A., Daatselaar, C.H.G, Doornewaard, G.L. and de Hoop, D.W. 2007. Bodemoverschotten op landbouwbedrijven. Deelrapportage in het kader van de Evaluatie Meststoffenwet 2007. Rapport 30705, LEI, The Hague, The Netherlands, 90 pp.
- van Grinsven, H.J.M., Holland, M., Jacobsen, B.H., Klimont, Z., Sutton, M.A. and Willems, J. 2013. Costs and benefits of nitrogen for Europe and implications for mitigation. *Environmental Science and Technology*. 47(8): 3571-3579
- van Grinsven, H.J.M., Tiktak, A. and Rougoor, C.W. 2016. Evaluation of the Dutch implementation of the Nitrates Directive, the Water Framework Directive and the national emission ceilings directive. *NJAS-Wageningen Journal of Life Sciences*.
- Van Meter, K. and Basu, N. 2015. Catchment legacies and time lags: A parsimonious watershed model to predict the effects of legacy storage on nitrogen export. *PlosOne* 10(5): <http://dx.doi.org/10.1371/journal.pone.0125971>
- Van Meter, K., Basu, N., Veenstra, J.J. and Burras, C.L. 2016. The nitrogen legacy: Emerging evidence of nitrogen accumulation in anthropogenic landscapes. *Environmental Research Letters* 11(3): <http://dx.doi.org/10.1088/1748-9326/11/3/035014>
- Van Meter and Basu, In Review
- Vecchia, A.V. 2003. Water-quality trend analysis and sampling design for streams in North Dakota, 1971-2000. United State Geological Survey. Water Resources Investigation Report 03-4094
- Vero, S.E., Ibrahim, T.G., Creamer, R.E., Grant, J., Healy, M.G., Henry, T., Kramers, G., Richards, K.G. and Fenton, O. 2014. Consequences of varied soil hydraulic and meteorological complexity on unsaturated zone time lag estimates. *Journal of Contaminant Hydrology*. 170: 53-67

Vero, S.E., Healy, M.G., Henry, T., Creamer, R.E., Ibrahim, T.G., Richards, K.G., Mellander, P-E., McDonald, N.T. and Fenton, O. 2017. A flexible modelling framework to indicate water quality trends and unsaturated zone time lag ranges at catchment scale. *Agriculture, Ecosystems and Environment* 236: 234-242

Vertes, F., Trevisan, D., and Gascuel, C. and Dorioz JM. 2008. Capacities and limits of two French grassland systems (intensive and extensive) to comply with WFD - Developing tools between science and policy to improve grassland management. In Proceedings of: Water Framework Directive Conference: "Sustainable grassland systems in Europe and the EU Water Framework Directive". Teagasc, Johnstown Castle, Wexford. 12th -14th November 2008.

Wahlin, K. and Grimvall, A. 2008. Uncertainty in water quality data and its implications for trend detection: lessons from Swedish environmental data. *Environmental Science and Policy*. 11: 115-124.

Wall, D., Jordan, P., Melland, A.R., Mellander, P-E., Buckley, C., Reaney, S.M. and Shortle, G. 2012. Using the nutrient transfer continuum concept to evaluate the European Union Nitrates Directive National Action Programme. *Environmental Science and Policy*. 14(6): 664-674

Wang, L., Stuart, M.E., Lewis, M.A., Ward, Skirvin, D., Naden, P.S., Collins, A.L. and 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. *Science of the Total Environment*. 542: 694-705

Wassenaar, L.I. 1995. Evaluation of the origin and fate of nitrate in the Abbotsford Aquifer using the isotopes of ^{15}N and ^{18}O in NO_3^- . *Applied Geochemistry* 10: 391-405

Wassenaar, L.I., Hendry, M.J. and Harrington, N. 2006. Decadal geochemical and isotopic trends for nitrate in a transboundary aquifer and implications for agricultural beneficial management practices. *Environmental Science and Technology* 40(15): 4626-4632

Worrall, F. and Burt, T.P. 1999. A univariate model of river water nitrate time series. *Journal of Hydrology*. 214:74-90.

Worthington, S.R.H. 2007. Groundwater residence times in unconfined carbonate aquifers. *Journal of Cave and Karst Studies* 69 (1): 94-102.

Wriedt, G. and Rode, M., 2006. Modelling nitrate transport and turnover in a lowland catchment system. *Journal of Hydrology*. 328: 157-176.

Zebarth, B.J., Ryan, M.C., Graham, G., Forge, T.A. and Neilsen, D. 2015. Groundwater Monitoring to Support development of BMPs for groundwater protection: The Abbotsford-Sumas Aquifer Case Study. *Groundwater Monitoring and Remediation* 35(1): 82-96

Zhou, Q., Gibson, C.E. and Foy, R.H. 2000. Long term changes of nitrogen and phosphorus loadings to a large lake in North West Ireland. *Water Research*. 34: 922-92

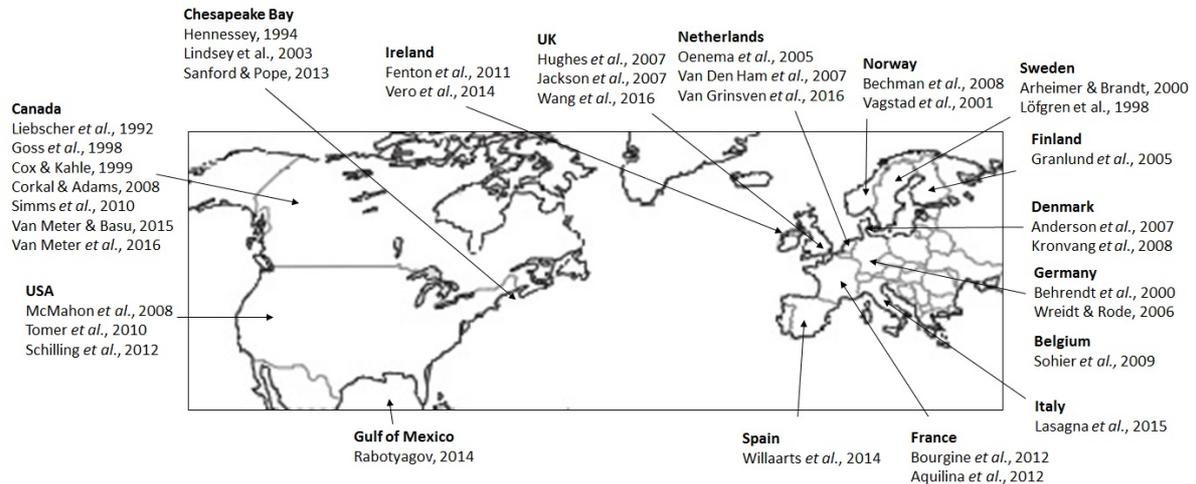


Fig. 1: A selection of international publications evidencing time lag and water quality.

Location	Scale of Study	Treatment	Lag Time	Reference
				Vadas et al. 2007
				Zebarth et al., 2015
				Wassenaar et al., 2006
				Wang et al., 2016
				Stålnacke et al., 2003 & 2004
				Grimvall et al., 2000
				Tomer and Burkhart, 2003
				Schirmer et al., 2014
				Schilling et al., 2012
				Hocking and Kelly, 2016
				Aquilina et al., 2012