



A catchment-scale method to simulating the impact of historical nitrate loading from agricultural land on the nitrate-concentration trends in the sandstone aquifers in the Eden Valley, UK

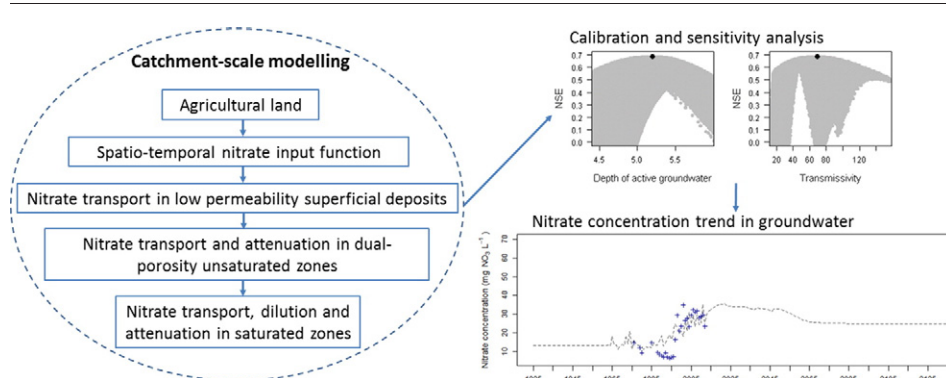
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HIGHLIGHTS

- An approach to modelling groundwater nitrate at the catchment scale is presented.
- It considers nitrate transport in glacial till and dual-porosity unsaturated zones.
- The impact of historical nitrate loading on groundwater quality is better understood.
- The modelled results are valuable for evaluating the nitrate legacy issue.
- The method is transferable and requires a modest parameterisation.

GRAPHICAL ABSTRACT



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ABSTRACT

Nitrate water pollution, which is mainly caused by agricultural activities, remains an international problem. It can cause serious long-term environmental and human health issues due to nitrate time-lag in the groundwater system. However, the nitrate subsurface legacy issue has rarely been considered in environmental water management. We have developed a simple catchment-scale approach to investigate the impact of historical nitrate loading from agricultural land on the nitrate-concentration trends in sandstones, which represent major aquifers in the Eden Valley, UK. The model developed considers the spatio-temporal nitrate loading, low permeability superficial deposits, dual-porosity unsaturated zones, and nitrate dilution in aquifers. Monte Carlo simulations were undertaken to analyse parameter sensitivity and calibrate the model using observed datasets. Time series of annual average nitrate concentrations from 1925 to 2150 were generated for four aquifer zones in the study area. The results show that the nitrate concentrations in 'St Bees Sandstones', 'silicified Penrith Sandstones', and 'non-silicified Penrith Sandstones' keep rising or stay high before declining to stable levels, whilst that in 'interbedded Brockram Penrith Sandstones' will level off after a slight decrease. This study can help policymakers better understand local nitrate-legacy issues. It also provides a framework for informing the long-term impact and timescale of different scenarios introduced to deliver water-quality compliance. This model requires relatively modest parameterisation and is readily transferable to other areas.

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1. Introduction

Excessive nitrate concentrations in water bodies can cause serious long-term environmental issues and threaten both economy and human health (Bryan, 2006; Defra, 2002, 2006a; Pretty et al., 2000; Thorburn et al., 2003; Ward, 2009). Nitrate in freshwater remains an international problem (European Environment Agency, 2000, 2007; Hinkle et al., 2007; Rivett et al., 2008; Sebilo et al., 2006; Thorburn et al., 2003; Torrecilla et al., 2005; Wang et al., 2012a; Wang and Yang, 2008; Yang and Wang, 2010). Elevated nitrate concentrations in groundwater are found across Europe. For example, the European Environment Agency (2007) reported that the proportion of groundwater bodies with mean nitrate concentration $>25 \text{ mg L}^{-1}$ (as NO_3) in 2003 were 80% in Spain, 50% in the UK, 36% in Germany, 34% in France and 32% in Italy. Despite efforts made under the EU Water Framework Directive (Directive 2000/60/EC) by 2015 to improve water quality, there is still a continuous decline in freshwater quality in the UK. For example, nitrate concentrations are exceeding the EU drinking water standard (50 mg L^{-1} (as NO_3)) and have a rising trend in many rivers (Burt et al., 2008, 2011) and aquifers (Smith, 2005; Stuart et al., 2007). It is estimated that about 60% of all groundwater bodies in England will fail to achieve good status by 2015 (Defra, 2006b).

Agricultural land is the major source of nitrate water pollution (Ferrier et al., 2004; Thorburn et al., 2003; Torrecilla et al., 2005). Point source discharges have been estimated as contributing $<1\%$ of the total nitrate flux to groundwater in the UK (Sutton et al., 2011). Agricultural yields are increased by the addition of nitrogen (N) in fertilisers, but this leads to nitrate leaching into freshwaters (groundwater and surface water). Nitrate concentrations in groundwater beneath agricultural land can be several to a hundred-fold higher than that under semi-natural vegetation (Nolan and Stoner, 2000). During the last century, the pools and fluxes of N in UK ecosystems have been transformed mainly by the fertiliser-based intensification of agriculture (Burt et al., 2011). In response to this growing European-wide problem, the European Commission implemented the Nitrates Directive (91/676/EEC) to focus on delivering measures to address agricultural sources of nitrate.

In the freshwater cycle, nitrate leached from soil is subsequently transported by surface runoff to reach streams or by infiltration into the unsaturated zone (USZ – from the base of the soil layer to the water table). Nitrate entering the groundwater system is then slowly transported through the USZs downwards to groundwater in aquifers. Recent research suggests that it could take decades for leached nitrate to discharge into freshwaters due to the nitrate time-lag in the USZs and saturated zones (Ascott et al., in press; Burt et al., 2011; Howden et al., 2011; Jackson et al., 2007; Wang et al., 2012a, 2016). This may cause a time-lag between the loading of nitrate from agricultural land and the change of nitrate concentrations in groundwater and surface water. For example, Dautrebande et al. (1996) found that the anticipated decrease in nitrate concentrations in the aquifer following the reduction of nitrate loading from agricultural land was not observed. However, current environmental water management strategies rarely consider the nitrate time-lag in the groundwater system (Burt et al., 2011; Collins et al., 2009).

The Eden catchment, Cumbria, UK (Fig. 1) is a largely rural area with its main sources of income being agriculture and tourism (Butcher et al., 2003; Daily et al., 2006). The Environment Agency's groundwater monitoring data show that some groundwater exceeds the limit of 50 mg L^{-1} (as NO_3) in the Eden Valley (Butcher et al., 2005). In recent years, the increasingly intensive farming activities, such as the increased application of slurry to the grazed grassland, have added more pressures on water quality in the area (Butcher et al., 2003, 2005). Efforts have been made to tackle agricultural diffuse groundwater pollution in the area. For example, the River Eden Demonstration Test Catchment (DTC) project (McGonigle et al., 2014) was funded by the Department for Environment, Food & Rural Affairs (Defra) to assess if it is possible

to cost-effectively mitigate diffuse pollution from agriculture whilst maintaining agricultural productivity (<http://www.edendtc.org.uk/>). The Environment Agency defined Groundwater Source Protection Zones (SPZs) (<http://apps.environment-agency.gov.uk/wiyby/37833.aspx>) in the Eden Valley to set up pollution prevention measures and to monitor the activities of potential polluters nearby. However, without evidence of the impact of nitrate-legacy issues on groundwater quality, it is difficult to evaluate the effectiveness of existing measures or to decide whether additional or alternative measures are necessary. So a key question for nitrate-water-pollution management in the area is how long it will take for nitrate concentrations in groundwater to peak and then stabilise at an acceptable level ($<50 \text{ mg L}^{-1}$ (as NO_3)) in response to historical and future land-management measures. Therefore, it is necessary to investigate the impacts of historical nitrate loading from agricultural land on the changing trends in nitrate concentrations for the major aquifers in the Eden Valley.

Wang et al. (2013) studied the nitrate time-lag in the sandstone USZ of the Eden Valley taking the Bowscar SPZ as an example. Outside of the study area, efforts have been made to simulate nitrate transport in the USZ and saturated zone at the catchment scale. For example, Mathias et al. (2006) used Richards' equation (a nonlinear partial differential equation) to explicitly represents fracture–matrix transfer for both water and solute in the Chalk, which is a soft and porous limestone. Price and Andersson (2014) combined a simple USZ nitrate transport model with fully-distributed complex groundwater flow and transport models to study nitrate transport in the Chalk. These catchment specific models, which require a wide range of parameters and are computationally-demanding, are of limited value for application to catchment-scale modelling for nitrate management. There is a need to develop a simple but still conceptually feasible model suitable for simulating long-term trend of nitrate concentration in groundwater at the catchment scale. In addition, the nitrate transport in low permeability superficial deposits has rarely been considered in existing nitrate subsurface models. Low permeability superficial deposits, however, overlay about 20.7% of the major aquifers in England and Wales (BCS, 2015a, 2015b), and 54% of the Permo-Triassic sandstones in the Eden Valley (Section 2.1).

Based on a simple catchment-scale model developed in this study, the impact of historical nitrate loading from agricultural land on the nitrate-concentration trends in sandstones of the Eden Valley was investigated. By considering the major nitrate processes in the groundwater system, this model introduces nitrate transport in low permeability superficial deposits and in both the intergranular matrix and fractures in the USZs. Nitrate transport and dilution in the saturated zone were also simulated using a simplified hydrological conceptual model.

2. Methodologies

2.1. Site setting

The Eden Catchment (2308 km^2) lies between the highlands of the Pennines to the east and the English Lake District to the west. The River Eden, which is the main river in the catchment, runs from its headwaters in the Pennines to the Solway Firth in the north-west. The area is mainly covered by managed grassland, arable land and semi-natural vegetation.

Carboniferous limestones fringe much of the Eden Catchment (Fig. 1) and have very low porosity and permeability, thus making a negligible contribution to total groundwater flow. Therefore, their storage and permeability rely almost entirely on fissure size, extent and degree of interconnection (Allen et al., 2010). They only constitute an aquifer due to the presence of a secondary network of solution-enlarged fractures and joints (Jones et al., 2000). Ordovician and Silurian intrusion rocks form the uplands of the Lake District and can also be found in the south-east of the catchment (Fig. 1).

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