Investigating nitrate transport in a thick sandstone unsaturated zone based on integrated modelling – the Eden Valley, UK

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ABSTRACT

Pollution of water by nitrate is not only an environmental issue but also an economic one and a threat to human health. In the Eden Valley (EV), groundwater nitrate concentrations are very variable distributed and have a rising trend, with some groundwater exceeding the limit of 50mg NO_3 l^{-1} demanded by the EU Nitrate and Water Framework Directive. This study was undertaken to investigate the lag time between nitrate loading and entering groundwater, and explore the linkage between the nitrate in groundwater and its historic surface application. This involved the application of integrated nitrate transport modelling in the unsaturated zone (USZ) of Permo-Triassic sandstones (PT-SS). Three process-based models were integrated in the study. A map of the USZ thickness of the PT-SS was derived from a groundwater flow model using GIS-Groundwater – a GIS groundwater flow model; the historic and future arrival time of nitrate at the water table were modelled using a GIS nitrate transport model in the USZ; and a GIS model for nitrate dilution in the saturated zone was developed to validate the results.

The modelled thickness of the PT-SS USZ in the Eden Valley varies from 0 – 183 m and hence this imposes a large range of nitrate transport time (lag time) in the USZ (up to c. 60 years). The variable USZ thickness is one of the major reasons for the observed variable distribution of nitrate groundwater concentrations in the EV. The peak nitrate loading at around 1983 has affected most of the study area, and will arrive at the water table in the areas with thicker USZ within the next 35 years. The results of this study, especially the lag time between nitrate surface loading and entering the groundwater, are significant in supporting decision making. The integrated model developed in this study is readily-transferable to other regions or countries for any diffuse conservative pollutant, and can be used for studying the impact of climate and landuse changes on groundwater nitrate pollution.

KEYWORDS

nitrate transport, nitrate storage, sandstone, unsaturated zone, GIS-Groundwater, integrated modelling

INTRODUCTION

Groundwater, which makes up nearly 70% of world's freshwater, plays an important role in our life, economies, and environment. In the UK, it provides one third of public water supply in England and Wales, increasing to up to 80% in Southern England; it also maintains the flow in many rivers. Nitrate groundwater contamination, which arises mainly from diffuse agricultural sources (Foster, 2000; Defra, 2006), has been identified as an environmental, economic, and human health problem (Comley, 1945; Matson et al., 1997; Hill, 1999; Pretty et al., 2000; Bryan, 2006) faced by both developing and developed countries (Campbell et al., 2004).

Motivated by the need to protect water resources, the European Union established an upper limit of 50 mg NO_3 l^{-1} for all groundwater (Nitrates Directive 91/676/EEC); and the Water Framework Directive (EU Directive 2000/60/EC) requires that this level is to be achieved by 2015 if groundwater bodies to reach at least 'good' status. However, according to Stuart et al. (2007), the average groundwater nitrate concentrations in UK have been rising with rates between 0.35 and 0.51 mg

NO_3 $l^{-1}year^{-1}$, and one third of the sites assessed in their study exceeded 50 mg NO_3 l^{-1}.

Parts of the Eden Valley catchment, located in north Cumbria, UK, have groundwater nitrate pollution problems. The Environment Agency's groundwater monitoring data show that abstracted groundwater in this area has a range of nitrate concentrations; some groundwater exceeds the limit of 50 mg NO_3 l^{-1} and exhibit a rising trend with time (Butcher et al., 2003; Butcher et al, 2005). In order to make sound decisions for groundwater quality management in the area, it is necessary to answer the scientific questions of what caused the distribution of groundwater nitrate concentration; when the nitrate in the groundwater was loaded into the unsaturated zone (USZ) and eventually reached the water table; and whether the historic peak nitrate loading has reached the water table in the EV. In order to answer these questions, it is necessary to model the nitrate travel process in the thick Permo-Triassic sandstones (PT-SS) USZ in the area. USZs impose a significant delay (years, decades and even centuries) on the arrival of recharge and hence nitrate leached from soils with different landuses at the water table. The nitrate lag time in the USZ, which is rarely considered in policy making in groundwater resource management, would be significant in supporting decision making for achieving environmental objectives in short timescales.

The transport and storage of nitrate in USZ porewater has been widely studied from the late 1970s onwards (Foster and Crease, 1974; Young et al., 1976; Spears, 1979; Oakes et al., 1981; Lawrence and Foster, 1986; Geake and Foster, 1989; Hoffmann et al., 2000; Brouyère et al., 2004; Ledoux et al., 2007; Rivett et al., 2007), and different type of numerical nitrate-pollution related modelling have been carried out (e.g. Palmer, 1987; Foster, 1993; Lake et al., 2003; Wang and Yang, 2008; Yang and Wang, 2010). Most recently, a simple process-based GIS model was developed to predict the loading of nitrate at the water table for the UK (Wang et al., 2011). This model was used in this study in conjunction with a GIS groundwater flow model and a GIS model for nitrate transport in saturated zone, to investigate the nitrate transport in the thick PT-SS USZ to provide scientific evidence for sustainable groundwater nitrate pollution management in the EV.

STUDY AREA

The Eden Valley, located in Cumbria in the north-west of England, lies between two upland areas, the Pennines to the east and the English Lake District to the west. It receives an average annual rainfall of about 1000 mm $year^{-1}$. The study area is aligned northwest-southeast and is 56 km long and 4.5 – 14 km wide (Figure 1). Pastoral farming and tourism are major economic activities in the region. Population density is relatively low at approximately 0.2 $hectare^{-1}$. The area is mainly covered by managed grassland, arable land and semi-natural vegetation with small proportions of woodland, and urban land use (Daily et al., 2006). In recent years the application of slurry to the grazed grasslands has been increased due to more intensive farming activities (Butcher et al, 2003). The River Eden, the main river in the study area, runs from its headwaters in the Pennines to the Solway Firth in the north-west, having three main tributaries, the River Eamont, the River Irthing and the River Calder.

Many geological and hydrogeological studies in the area have been carried out (e.g. Arthurton, et al., 1978; Patrick, 1978; Arthurton and Wadge, 1981; Allen et al., 1997; Millward and McCormac, 2003). In the study area, the Permian Penrith Sandstone (up to 900 m thick), dips gently eastwards and is principally red-brown to brick red in colour with well-rounded, well-sorted and medium to coarse grains. It is overlain by the Eden Shale Formation (up to 180 m thick), which is generally red in colour with brown, green and grey beds in places, and consists of mudstone, siltstone, sandstone, breccia and conglomerate. This is overlain by the St Bees Sandstone (up to 350 m thick), which consists of red-brown and grey, fine-grained, cross-bedded sandstone (Allen et al, 1997). Figure 1 shows the bedrock geology of the study area.

Borehole hydrographs from the Penrith Sandstone aquifer in the area show a small annual fluctuation in groundwater levels, typically less than 1 m, indicating the groundwater flow type in the aquifer is intergranular with high storage (ESI 2004). Some hydrographs from the same aquifer also show very long-term water level fluctuations (with about 10 years between the peaks and troughs) apparently as a result of long-term changes in recharge (Butcher et al, 2003). Groundwater flow in

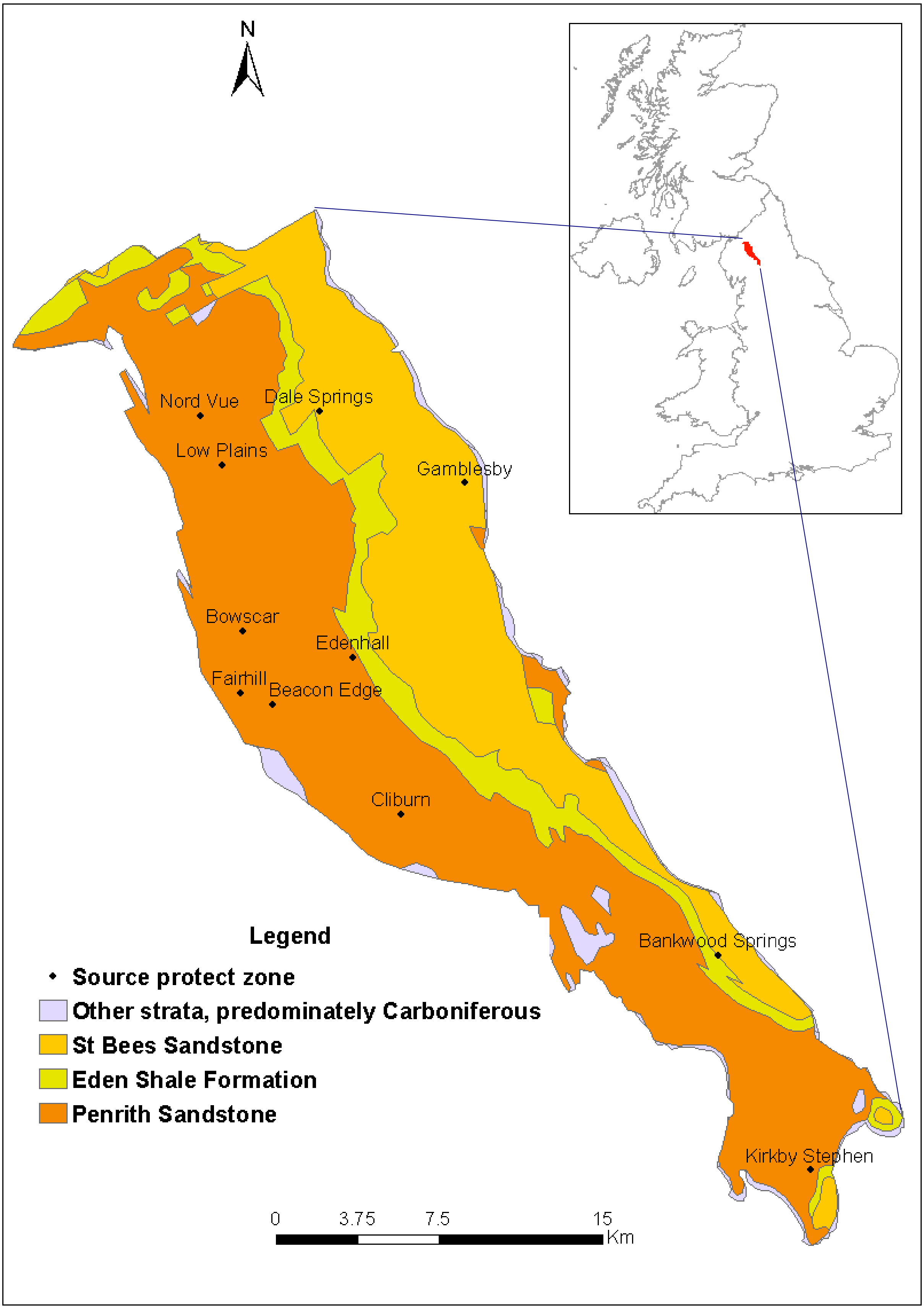

Figure 1. - The bedrock geological map for the Eden Valley.

the study area is dominated by flow to the River Eden. The hydraulic gradients in the Penrith Sandstone aquifer are generally gentle and predictable, whilst the ones in the St Bees Sandstone aquifer are generally steeper, reflecting its generally lower permeability (Butcher et al, 2003; Daily et al., 2006). The Penrith Sandstone and St Bees Sandstone form the major aquifers in the region. The hydraulic conductivity (K) values in the aquifers range from 3.5 x 10^{-5} – 26.2 m day^{-1} and 0.048 – 3.5 m day^{-1} respectively (Allen et al, 1997). Groundwater levels are close to the surface in the vicinity of the River Eden, but they are as much as 100 m below ground in the north-west part of the study area. According to Daily et al. (2006), there may be some groundwater flow between adjacent and underlying Carboniferous rocks in the area, however, the numerous springs, which arise along the faulted contact, suggest that much of the groundwater is transferred to surface flow.

METHODOLOGIES

Groundwater flow modelling

The thickness of the PT-SS USZ in the study area is needed for simulating nitrate transport in the USZ. Therefore it is necessary to simulate the long-term steady-state groundwater levels by carrying out groundwater flow modelling for the area.

GIS-Groundwater – a seamless GIS numerical finite difference groundwater flow model (Wang et al., 2010) was used in this study. GIS-Groundwater can be easily and efficiently applied to simulate groundwater flow by directly using GIS datasets in constructing groundwater flow models. The Penrith Sandstone and St Bees Sandstone were simplified as a single layer aquifer with a distribution of permeability values. The modelling extent is defined by a (100 m by 100 m) GIS layer. Users do not need to ensure that shapes of the modelling area are valid for applying groundwater flow equations, because GIS-Groundwater removes abnormal nodes if there are any, and automatically defines boundary and non-boundary modelling node types. Therefore, any spatial complex shapes for the modelling extent are supported by the model. A GIS layer containing the distributed K values was entered into the model; river nodes and river stages entered were derived from a Centre for Ecology and Hydrology (CEH) river system dataset and a CEH digital elevation model (50 m by 50 m); groundwater abstraction data were defined in the model using a GIS layer.

Nitrate input function

The nitrate input function assumes temporally varying but spatially uniform leaching of nitrate from the base of the soil. A time-varying nitrate input function derived in the work of Wang et al. (2011) was used in this study assuming a single arable landuse is covering the study area. The derived nitrate input function shows an excellent agreement with mean porewater nitrate concentrations from 300 cored boreholes across the UK in the BGS database (Figure 2). The sudden increase of porewater nitrate concentrations between 1990 and 2000 was due to an artefact of both the relatively small number of recent data points and the focus of recent studies on areas with a nitrate problem.

In this nitrate input function, a low and constant value (25 kg N ha^{-1} $year^{-1}$) between 1925 to 1940 reflects the pre-war low level of industrialisation with very limited use of non-manure-based fertilizers; from 1940 to 1955, there was a 1 kg N ha^{-1} $year^{-1}$ rise in nitrogen input to 40 kg N ha^{-1} in 1955. This was the result of the gradual intensification of agriculture during and just after World War II; a more rapid rise of 1.5 kg N ha^{-1} $year^{-1}$ from 40 kg N ha^{-1} in 1955 to 70 kg N ha^{-1} (a peak value between 1975 and 1990) in 1975 was due to increases in the use of chemical based fertilizers to meet the food needs of an expanding population; the nitrogen input declines with a rate of 1 kg N ha^{-1} $year^{-1}$ from 1991 to 2020 (from 70 kg N ha^{-1} in 1991 to 40 kg N ha^{-1}) as a result of restrictions on fertilizer application in water resource management; finally, there is a constant 40 kg N ha^{-1} nitrogen application from 2020 to 2050, assuming a return to nitrogen input levels similar to those associated with early intensified farming in the mid-1950s.

GIS nitrate transport model in the USZ

A process-based GIS nitrate transport model in the USZ (Wang et al., 2011), which has been applied in predicting the arriving time for peak nitrate loading at the water table of the UK, was employed in this study. This model links a nitrate input function, USZ thickness, and lithologically dependent rate of nitrate USZ transport to estimate the arrival time of nitrate at the water table. The assumptions of this model include: nitrate loading is from the base of the soil; nitrate moves vertically from the land surface to the water table; nitrate movement is through the matrix only in dual-porosity strata; the mass of nitrate in the USZ is preserved; nitrate moves at a constant velocity through the USZ; and there is no hydrodynamic dispersion of nitrate in the USZ. Denitrification is the dominant nitrate attenuation process in the subsurface (Rivett et al., 2007), but Kinniburgh et al., (1994) regarded this as insignificant beneath the soil zone in the USZ of UK aquifers. Butcher et al (2005) found no evidence of denitrification in sampled groundwater in the Eden Valley.

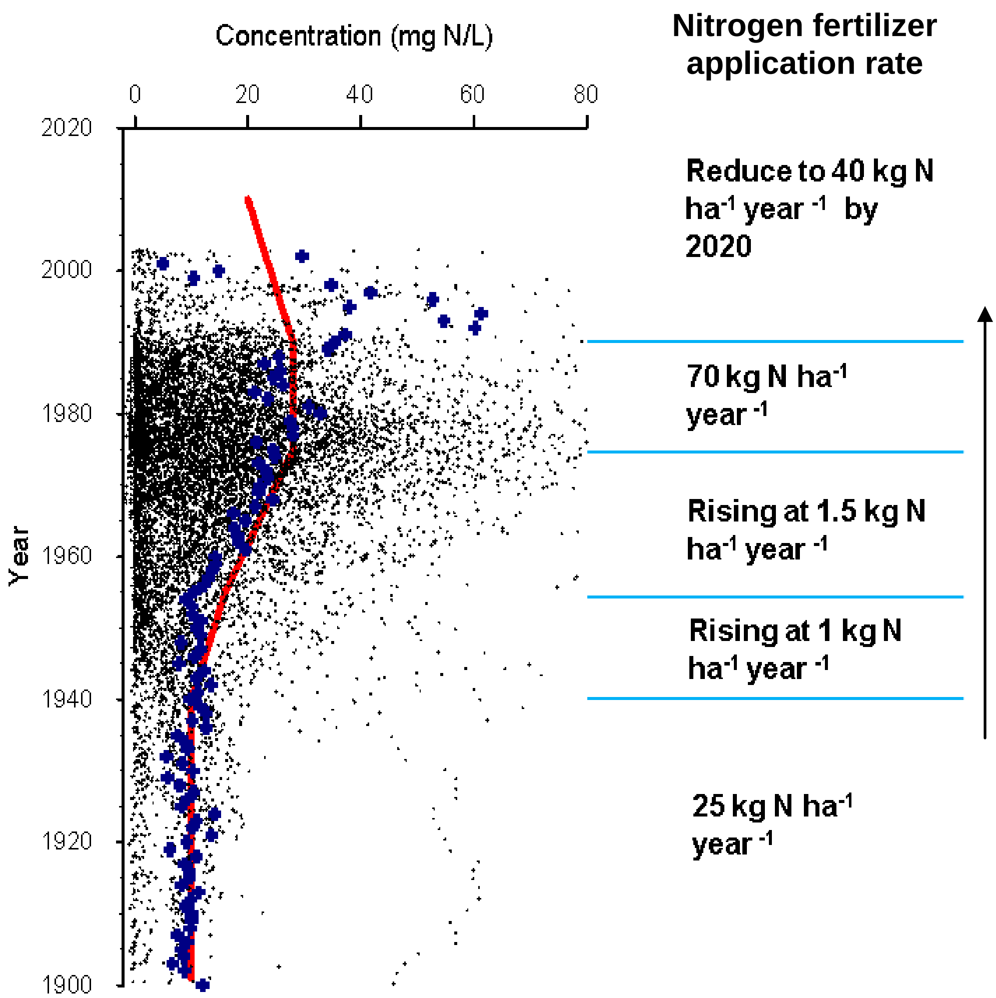

Figure 2. - Nitrate input function. Red solid line shows nitrate input spans derived from literature data. Black dots show individual porewater nitrate concentrations from 300 cored boreholes in the BGS database that have been back-plotted to give nitrate concentrations at the base of the soil zone at their year of recharge calculated using depth in the profile and estimated USZ travel time. Blue crosses show average nitrate concentration for a given year calculated from the porewater data. The nitrogen application rate on the right reflects the historic different levels of industrialisation and the introduction of measures to reduce the fertilizer application rate (Adapted from Wang et al. (2011))

Modelling nitrate dilution in the saturated zone

A GIS model of nitrate transport in the saturated zone was developed to simulate yearly nitrate concentration at a borehole by considering the process of nitrate leaching from the bottom of soil zone, the nitrate movement in the USZ and dilution in the saturated zone. The simulated pumped nitrate concentration in boreholes were compared with observed ones to validate the numerical modelling parameters such as the nitrate transport velocity in the USZ, the thickness of the USZ, and the aquifer hydraulic conductivity values used for deriving the thickness of the USZ.

Consideration of the dilution process was simplified by assuming that nitrate arriving at a borehole dilutes in water pumped out of the borehole, and the groundwater flow within a groundwater Source Protection Zone (SPZ) (a groundwater catchment for a pumping borehole), reaches a steady-state, i.e., the long-term recharge volume within a SPZ equals to water pumped out of the borehole in the SPZ. In this study, only the abstraction sources of the SPZ are indicated in the figures. Not all leached nitrate reaches the abstraction borehole due to attenuation processes in the USZ and the saturated zone. Nitrate concentration can be reduced due to denitrification and absorption in USZ; nitrate in the saturated zone will be absorbed by small porous or transports outside of SPZ due to diffusion or dispersion. Therefore a nitrate attenuation coefficient (*NAC*) was introduced into this model. With this conceptual model, the depth of the saturated zone, the thickness of active groundwater zone can be ignored, and the nitrate dispersion and diffusion processes can be simplified in simulating yearly nitrate concentration at a borehole in the SPZ.

The nitrate travel time form the loading point to a borehole is calculated using equations:

$$TTT_{i,j} = UTT_{i,j} + STT_{i,j} \tag{1}$$

$$STT_{i,j} = \frac{Dist_{i,j}}{VS_{i,j}} \tag{2}$$

$$VS_{i,j} = \frac{K \times G_{i,j}}{\Phi} \tag{3}$$

where $TTT_{i,j}$ (years) is the total nitrate travel time from the ground surface at the modelling cell (i, j) to a borehole; $UTT_{i,j}$ (years) is the nitrate travel time from the loading point at the bottom of soil zone to the water table at the modelling cell (i, j) in the USZ; $STT_{i,j}$ (years) is the nitrate travel time from the water table at the modelling cell (i, j) to a borehole within the saturated zone; Dist (m) is a 3D distance between the water table at the modelling cell (i, j) and the screen level of a borehole; $VS_{i,j}$ is the velocity of nitrate transport in saturated zone; K (m day^{-1}) is the hydraulic conductivity for the saturated media in a SPZ; Gi,j is the average hydraulic gradient between the water table at the modelling cell (i, j) and the screen level of the borehole; and Φ is the porosity of aquifer media in the SPZ.

The amount of nitrate reaching at a borehole in a year *N* from a modelling cell (i, j) is the nitrate loading in the year (*N* minus $TTT_{i,j}$) in the cell (determined by nitrate input function); and the total amount of nitrate reaching at the borehole in the year *N* (TTN_N: mg NO_3) is the sum amount of nitrate for all the modelling cells from different loading years within the SPZ. The actual total nitrate arriving at the borehole ($ATTN_N$: mg NO_3) can be calculated using Equation 4.

$$ATTN_N = TTN_N \times (1 - NAC) \tag{4}$$

Hence, an average nitrate concentration in year *N* can be calculated from:

$$Ncon_N = \frac{ATTN_N}{Vol} \tag{5}$$

$$Vol = PumpRate \times 365 \tag{6}$$

where $Ncon_N$ (mg NO_3 l^{-1}) is the average nitrate concentration in the water pumped out of a borehole in the year *N*; *Vol* (litre) is the volume of water pumped out from the borehole in a year; and *PumpRate* (l day^{-1}) is the groundwater pumping rate of the borehole.

RESULTS

A constant value for groundwater recharge of 1 mm day^{-1} was used in the groundwater flow modelling in the area. The groundwater flow model was calibrated by comparing the simulated long-term average groundwater heads with observed ones in 39 boreholes. Figure 3 shows that the model was well calibrated. In this study, the K values for modelling the groundwater flow in Penrith Sandstone and St Bees Sandstone are 3.5m day^{-1} and 0.6m day^{-1} respectively. The distributed USZ thickness map for the area (Figure 4) was then derived by subtracting the modelled long-term average groundwater heads from the DEM.

The calculated USZ thickness, groundwater heads, and the K values from the calibrated groundwater flow model were used in modelling nitrate dilution in saturated zone in the Bowscar SPZ to the northwest of the study area (Figure 1). It is understood that nitrate is travelling with a velocity of around 3 m year^{-1} in the PT-SS USZ from previous study in the area (Butcher et al 2008); a 400-day zone in Bowscar SPZ was used to simulate the yearly nitrate concentration in its borehole (with a pumping rate of 1.5 Ml day^{-1} and a screen level of about 117 m AOD); and the nitrate input function described in previous section was also used in the simulation. The model was calibrated by comparing the simulated with observed yearly nitrate concentrations in the Bowscar borehole. The modelled results can well represent the trend of nitrate concentration in the borehole (Figure 5). This implies that the understanding of the nitrate travel velocity in the P-T SS USZ is correct; the method for deriving the thickness of the USZ is reliable; and the nitrate input function can be used for this study. All these data were then fed into the GIS nitrate transport model to model the nitrate transport in the USZ.

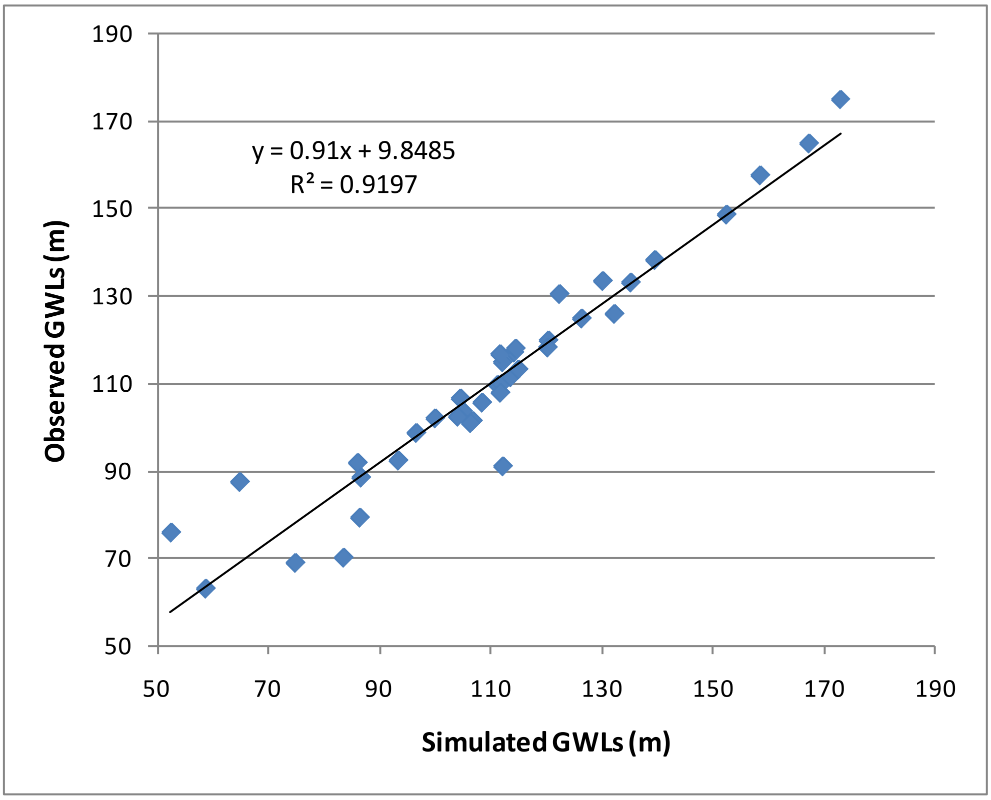

Figure 3. - Correlations between observed and modelled long-term steady-state groundwater heads.

In the study area, the modelled thickness of the USZ is greatest, 183 m in the northwest of the Eden Valley, and reduces to 0 m (i.e. groundwater heads are the same elevation as the river stages) along the River Eden and its tributaries. SPZs generally have a thicker USZ than other parts of the study area (Figure 4). This is partly because public supply abstraction boreholes were sited in elevated positions in order to contribute to a gravity fed supply system.

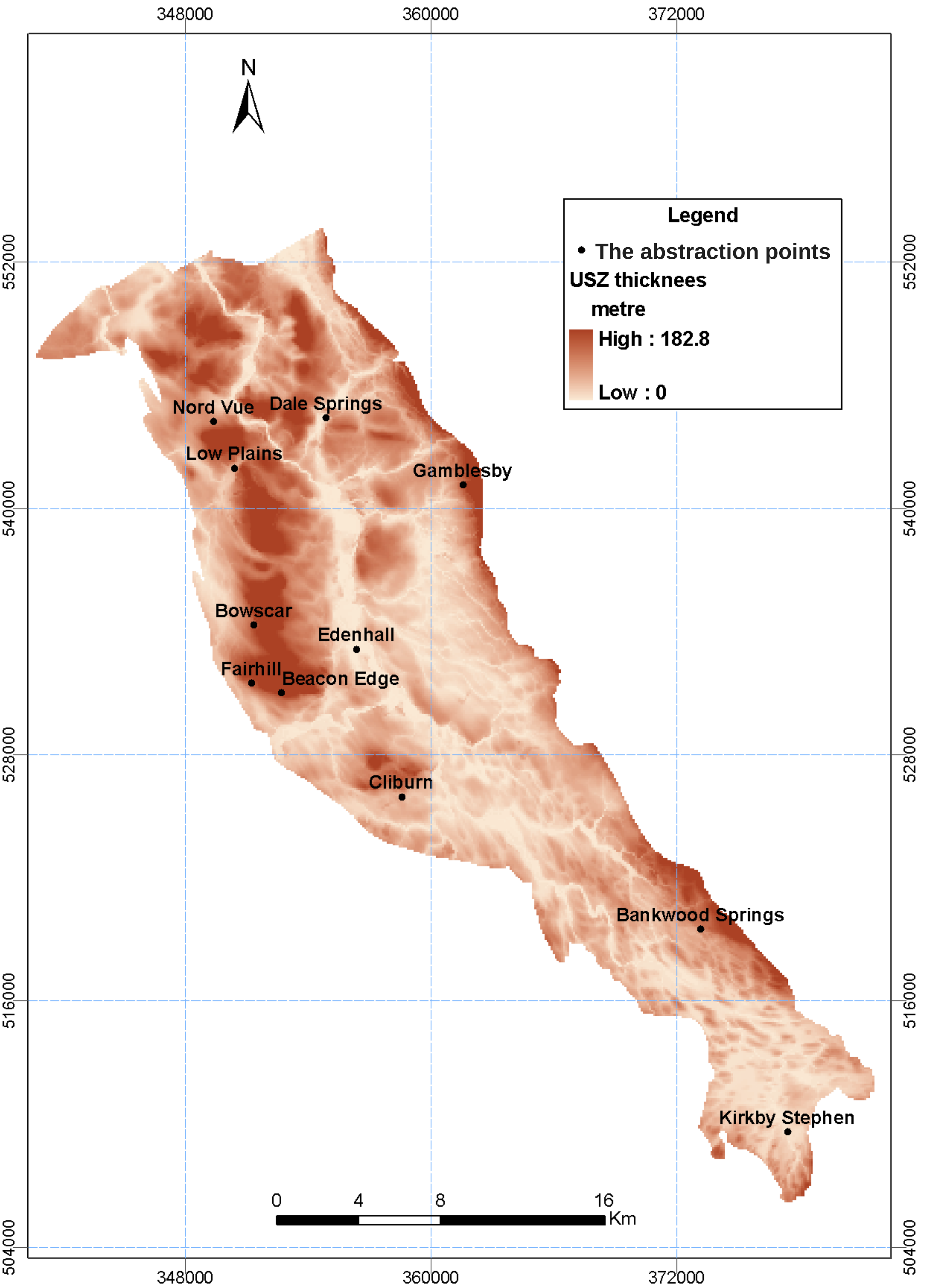

Figure 4. - Calculated thickness of USZ for the Eden Valley

The nitrate travel time in the USZ (which correlates with the USZ thickness), ranges from 0 to 61 years with a mean value of 12 years. Figure 6 suggests that strip areas along streams have short travel times (0-1 year) due to thin USZs, whilst mountainous areas in the east and west of the Eden Valley have longer nitrate travel times.

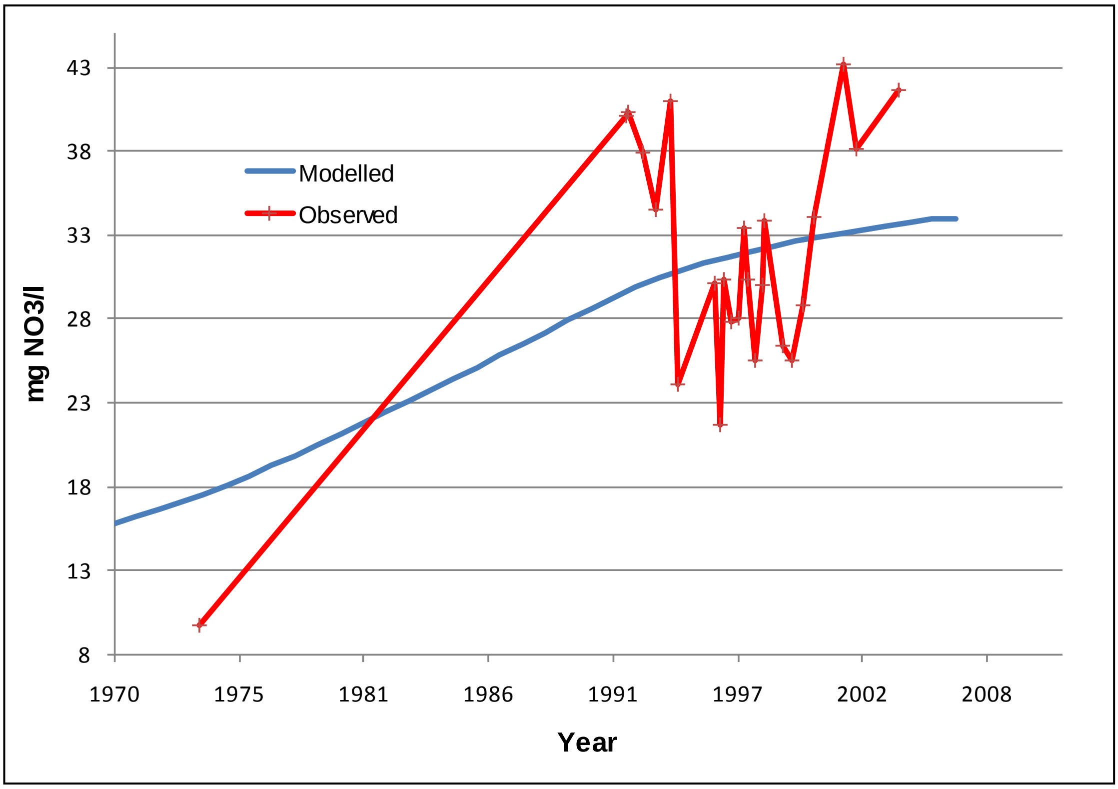

Figure 5. The modelled and observed yearly nitrate concentration in Bowscar borehole.

The nitrate arriving at the water table and entering the saturated zone in the area in 2010 was loaded/leached during 1940s – 2000s (Figure 7). The groundwater nitrate in the area was mainly from 1980s – 2000s, whilst the groundwater nitrate in the most of SPZs leached into the system during 1940s-1970s. The peak nitrate loading around 1983 has affected most of the study area. For areas around the SPZs of Bowscar, Beacon Edge, Low Plains, Nord Vue, Dale Springs, Gamblesby, Bankwood Springs, and Cliburn, the peak nitrate loading will arrive at the water table in the next 34 years (Figure 8). Statistical analysis shows that 8.7% of the Penrith Sandstone and 7.3% of the St Bees Sandstone have not been affected by peak nitrate.

The distributed maps for nitrate concentration at the water table for each year between 1925 and 2050 were produced. The results show that the average nitrate concentration at the water table across the EV has reached its peak and will decrease over the next 40 years. It is worth noting that some of unaffected areas with thicker USZs around Beacon Edge, Fairhills, Bowscar, Nord Vue, Low Plains, Gamblesby, and Bankwood Springs, will be affected by peak nitrate loadings between 2020 and 2030, and then retain a high nitrate concentration level (170 mg NO_3 l^{-1}) (before any groundwater dilution) from 2030 to 2050.

Two time series of the average nitrate concentration at the water table of the two major aquifers of the Eden Valley have been produced (Figure 9). The results suggest that the Penrith Sandstone and St Bees Sandstone have almost the same modelled trend of average nitrate concentration (before any groundwater dilution) at the water table. The nitrate concentrations at the water table of both aquifers reached the peak around 1995, and have declined since then. It is also worth noting that the unrealistic high nitrate concentration in Figure 9 is not the bulk groundwater nitrate concentration but the one at the water table (For modelling purposes it was assumed that nitrate stays at a very thin layer at the water table before the dilution process). However, the nitrate concentration at the water table is a good indicator of the trend of nitrate present in the groundwater regime.

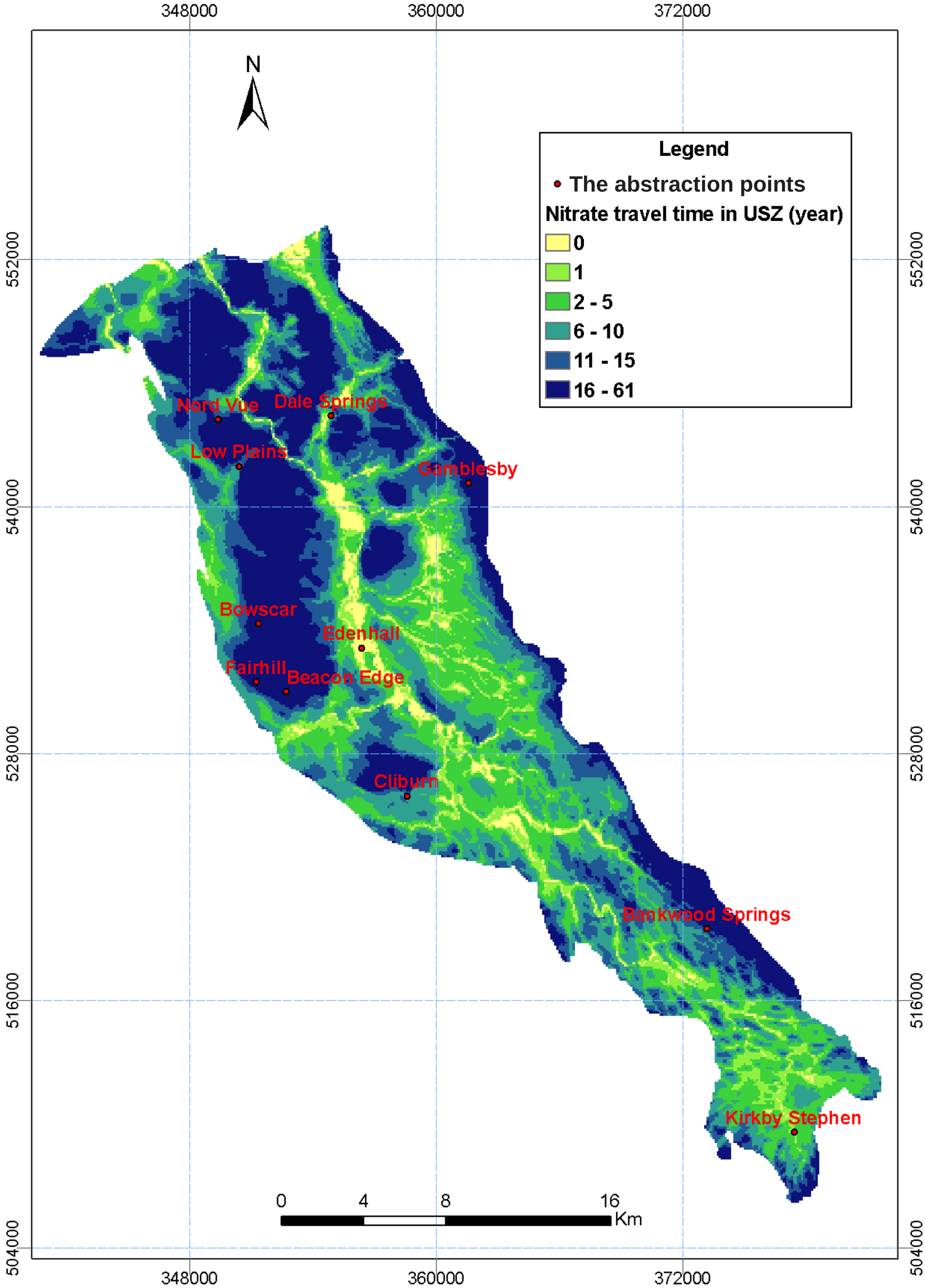

Figure 6. Modelled nitrate travel time in the USZ of the Eden Valley.

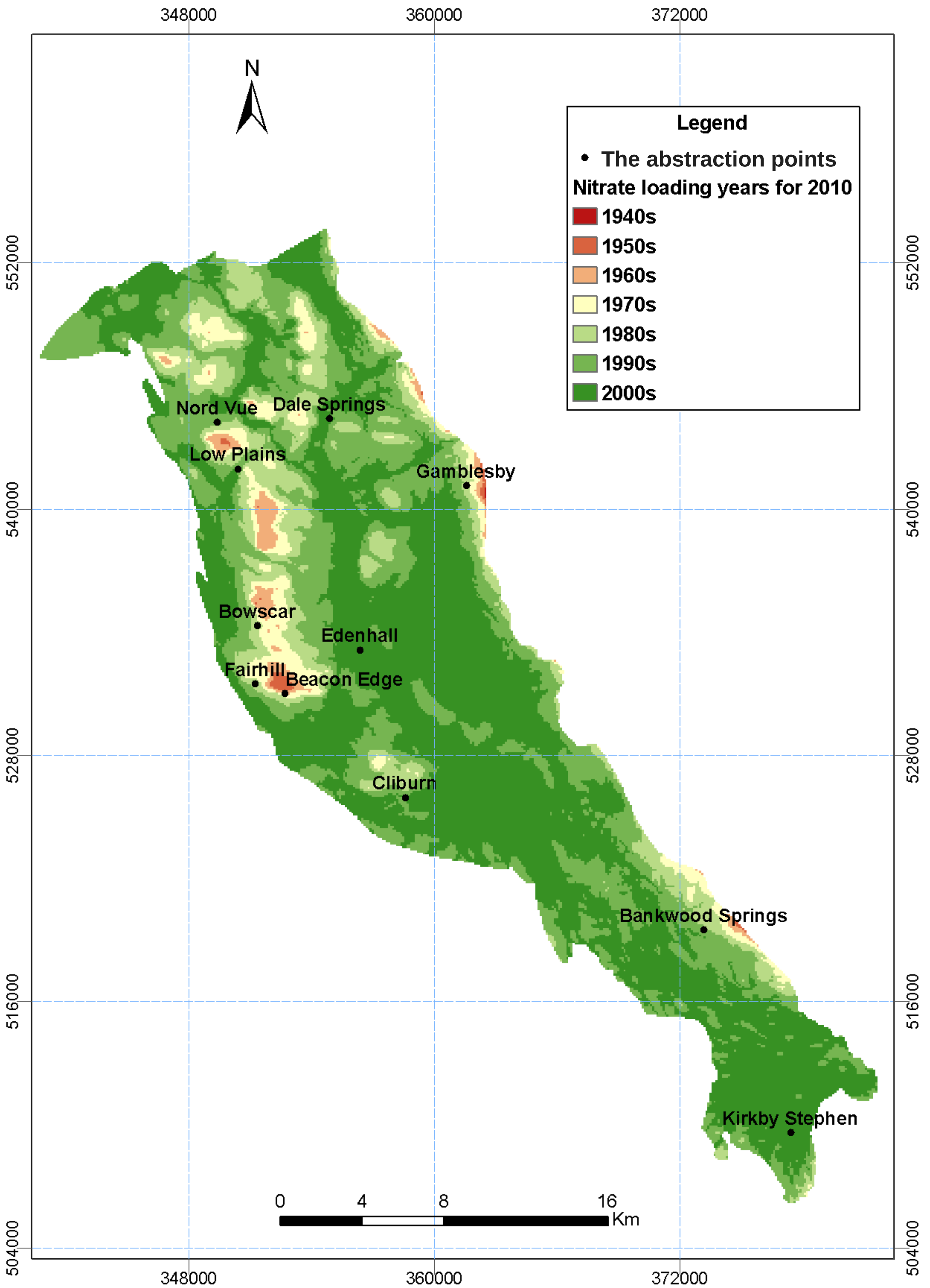

Figure 7. The loading time for nitrate arriving at the water table of the Eden Valley in 2010.

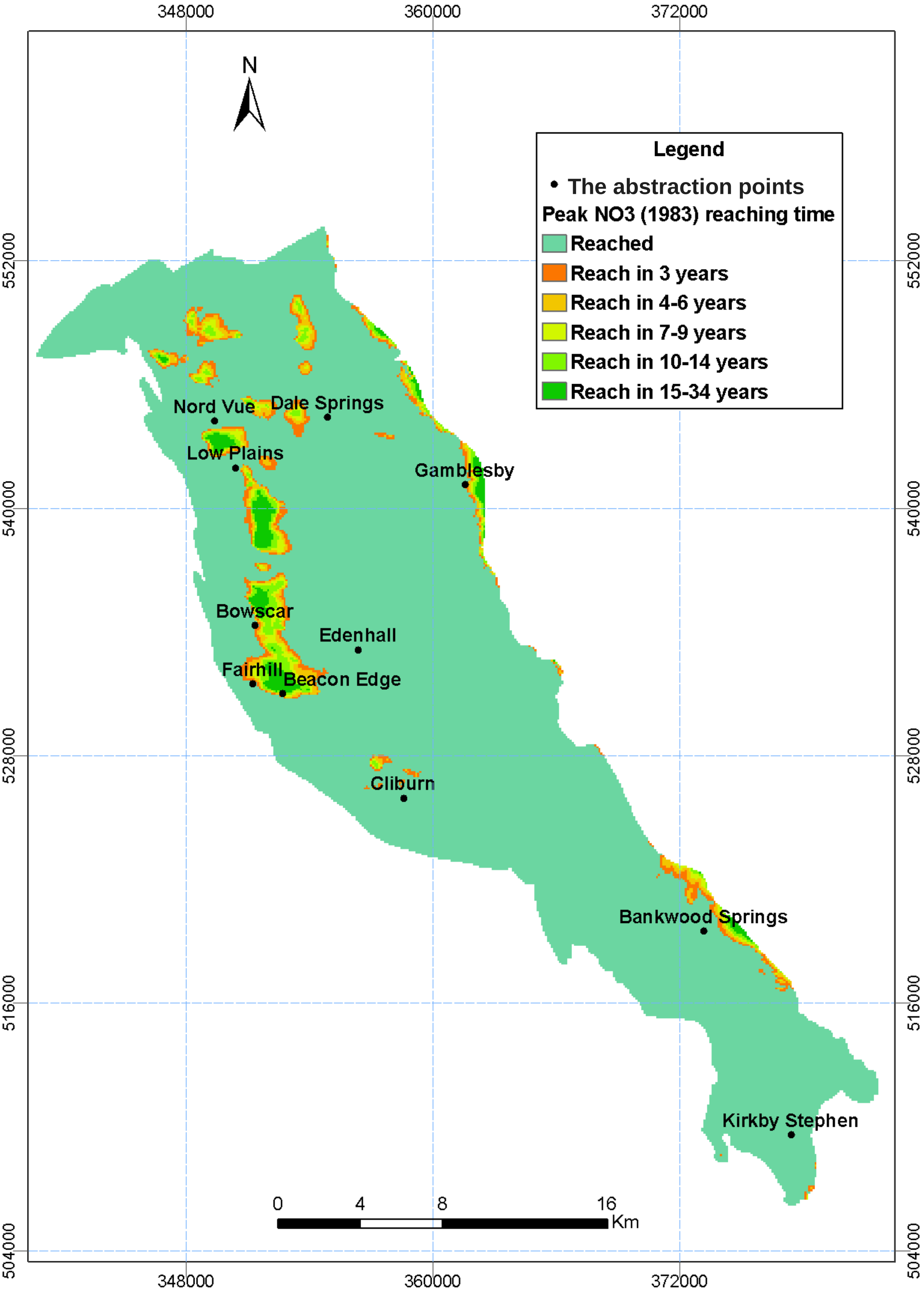

Figure 8. The future arrival time for the peak nitrate loading (around 1983) from 2010

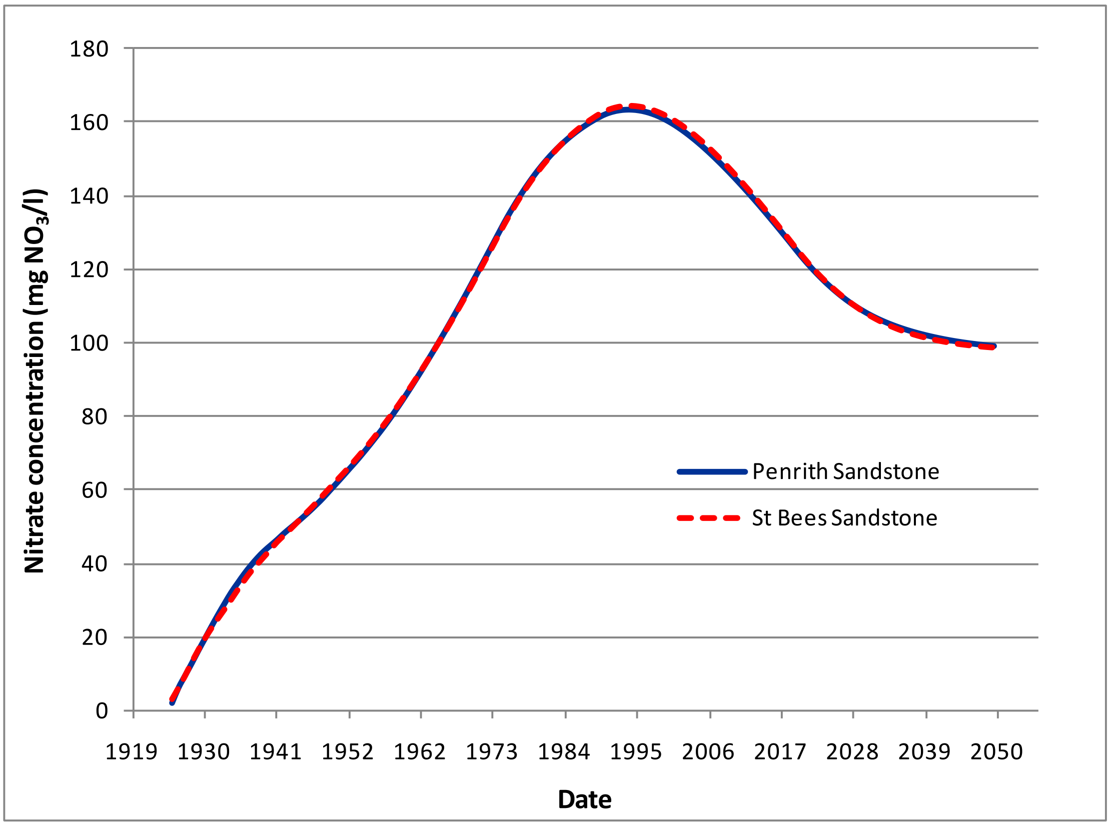

Figure 9. - The time series of the average nitrate concentration at the water table (before groundwater dilution) of two major aquifers in the Eden Valley

DISCUSSION

A significant and spatially variable thickness of the USZ, which determines the nitrate transport time in the USZ, is one of the major controls on nitrate groundwater concentrations in the area. According to groundwater quality observations, whilst most have low nitrate concentrations, there are a significant number of boreholes where nitrate concentrations are above 50 mg NO_3 l^{-1} but there does not appear to be a systematic distribution of these higher nitrate groundwaters (Butcher et al., 2003). Most parts of the study area have been affected by the peak nitrate loading (around 1983), and the nitrate entering the groundwater system is now declining. This explains the low nitrate concentration in the most of the study area; but for those SPZs with variable thicker USZs, some of them are being affected or will be affected by the peak nitrate loading showing locally high or increasing nitrate groundwater concentrations. This explains why some boreholes have high and (or) increasing nitrate concentrations.

Butcher et al., (2003) argued that localised nitrate point sources near to small volume abstractions might be another reason for the unevenly distributed groundwater nitrate concentrations. It was assumed in this study that the source of nitrate is from agricultural diffuse source and the landuses in the area were simplified as one single average type hence a single nitrate input function. It would be useful to consider detailed land use types as well the nitrate point sources. In addition, a constant groundwater recharge value was used in this study, and detailed distributed groundwater recharge will be introduced in a future study.

The lag time between surface nitrate loading and entry to groundwater is rarely taken into account in current environmental management strategies, but it is critical to effective management and control of nutrient pollution. The linkages between the nitrate historic loading and the nitrate in groundwater have been established; and the trend of nitrate in the groundwater of the Eden Valley was modelled in this study. These results are significant in supporting decision making for achieving environmental objectives in much shorter timescales. For example, the decreasing trend of the average groundwater nitrate concentration is good news, however, special attention should be paid to the areas where the historic peak nitrate loading has not yet arrived; the better appreciation of the nitrate lag time in the USZ in this study could mean that inappropriate controls are avoided as a result of removing conflicts between decision makers, environmentalists and industry. The results of studies like this should also help decision makers to define a reasonable timescale to witness the effect of an action.

The methods used in this study are readily-transferable to other regions for any diffuse conservative pollutant, and can be used for studying the impact of climate and landuse changes on groundwater nitrate pollution.

CONCLUSIONS

The nitrate transport process and its lag time in the thick PT-SS USZ of the Eden Valley were successfully investigated through an integrated modelling method.

The study area has a variable thickness of the USZ (0 – 183 m) hence a large range of nitrate transport time (lag time) in the USZ (up to c. 60 years). Groundwater nitrate in most of the area was from the 1980s – 2000s, whilst the groundwater nitrate in some of SPZs was loaded into the system during the 1940s – 1970s; the peak nitrate loading around 1983 has affected most of the study area, and will arrive at the water table in some of SPZs within the next 35 years.

Large and spatially variable thickness of the USZ is one of the major reasons for unevenly distributed groundwater nitrate concentrations in the study area. The average nitrate concentration in the whole area, which reached the peak value around 1995, has a declining trend, but the areas with thicker USZs, which have not been affected by the peak nitrate loading, will be subject to a localised high or increasing groundwater nitrate concentrations in the next few decades.

The results of this study, especially the lag time between nitrate loading and it entering the groundwater, are significant in supporting decision making for achieving environmental objectives in shorter timescales and in defining a reasonable timescale before seeing groundwater quality improvements due to management actions.

The integrated modelling method used in this study, which involves three simple process-based models, is readily-transferable to other regions or countries for any diffuse conservative pollutant, and can be a base for studying the impacts of climate and landuse changes on groundwater nitrate pollution.

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