Multivariate and long-term time series analysis to assess the effect of nitrogen management policy on groundwater quality in Wallonia, BE

Elise Verstraeten[1], Alice Alonso[1], Louise Collier[2], Marnik Vanclooster[1]

[1]Earth and Life Institute - Environnemental Sciences, Université catholique de Louvain, Louvain-la-Neuve, 1348, Belgium
[2]Société Wallonne des Eaux, Verviers, 4800, Belgium

Correspondence to: Alice Alonso (alice.alonso@uclouvain.be)

Abstract. Groundwater nitrate contamination remains a significant environmental and health concern, as the effectiveness of nitrogen management programs has shown mixed results. This study leverages nearly two decades of groundwater monitoring data to assess and explain nitrate contamination trends in vulnerable zones in Wallonia, Belgium, following the implementation of the regional sustainable nitrogen management program.

Using nitrate concentration time series (2002–2020) from 36 monitoring points across four groundwater bodies, we calculated six nitrate pollution indicators that capture temporal dynamics. Spatially explicit variables describing potential contamination drivers were compiled, and their relationships with the nitrate pollution indicators were assessed using correlations and multivariate regressions.

Although mean groundwater nitrate concentrations remained stable due to nitrogen legacy effects, there was an overall improvement as nitrate concentrations decreased over the study period. Decreases were observed in the Brusselian sands, where concentrations were initially higher, while increases were found in the Geer basin chalks, typically less contaminated. These diverging trends can be explained by differences in aquifer characteristics and nitrate transfer time lags. Agricultural land cover continues to have a negative impact on nitrate contamination, even after 20 years of PGDA implementation. The limited predictive power of the regression models highlights the multifaceted nature of groundwater nitrate contamination and the challenges in representing controlling factors, mainly due to lack of data.

Overall, this study emphasizes the need for sustained and adaptive nitrogen management policies, especially in vulnerable aquifers and cropland-dominated regions, alongside long-term monitoring to address time lags and nitrogen legacy effects. It also underscores the importance of detailed datasets to support advanced non-linear machine learning approaches that can capture the complex interactions involved.

1 Introduction

For several decades, concerns have been raised about elevated nitrate concentrations in groundwaters. These high nitrate levels are harmful to the environment as they contribute to the biodiversity loss and eutrophication of the aquatic ecosystems (de Vries et al., 2024; Grizzetti et al., 2011; Hornung, 1999), and can pose serious health risks when the groundwater is used for drinking water (Bouchard et al., 1992; Comly, 1945; de Vries et al., 2024; Schullehner et al., 2018; M. Ward et al., 2018). Despite extensive research, understanding the factors underlying nitrate concentrations and trends is complicated by the diffuse, long-term and multi-causal nature of the contamination (Shukla & Saxena, 2018).

The primary anthropogenic sources of nitrates in groundwater are agricultural activities (Spalding & Exner, 1993; Wick et al., 2012). However, non-agricultural sources such as inadequate treatment and disposal of human waste and wastewater, landfill, and industrial waste can also be significant contributor to high concentrations in aquifers (Mattern et al., 2009; Vanclooster et al., 2020; Wakida & Lerner, 2005). While human activities drive increased nitrogen input to the soils, the leaching of nitrates to groundwater is governed by key natural processes, referred to as the nitrogen cycle (B. B. Ward, 2013). Specifically, nitrate formation is driven by nitrification, a process in which ammonium, introduced through fertilizer and mineralization of organic matter, is oxidized to nitrate. In contrast, nitrate concentration levels decrease through plants and microbial uptake as well as through denitrification, which transforms nitrate into nitrogen gas under anaerobic conditions. These processes are influenced by environmental factors such as climate, soil composition, geological formation, and the depth of groundwater tables.

To address the environmental and health risks associated with nitrate pollution, the European Union's Nitrate Directive (91/676/EEC) has played a central role in promoting best management practices (BMPs) that mitigate agricultural nitrate contamination. Established in 1991, the directive aims to protect water bodies by requiring member states to identify Nitrate Vulnerable Zones (NVZs) and implement targeted measures to reduce nitrate leaching, aiming to maintain surface and groundwater nitrate concentrations below 50 mg/L.

Studies evaluating the effectiveness of such policies have shown mixed results. While many report encouraging signs of reduced nitrate pollution due to improved nitrogen management practices, others highlight persistent challenges, with continued increases or limited improvement in concentrations in many locations (Ferguson, 2015; Hansen et al., 2012, 2017; Van Grinsven et al., 2012, 2016). In Wallonia (Belgium), the European Directive was transposed into a program for sustainable nitrogen management ("Programme de Gestion Durable de l'Azote en Agriculture", PGDA) in 2002. This program includes region-wide measures and additional requirements in the NVZs, including restrictions on manure spreading, mandatory soil cover, and groundwater monitoring (Picron et al., 2017). In compliance with the Water Framework Directive (WFD, 2000/60/EC) and the Groundwater Directive (GWD, 2006/118/EC), the Walloon regional authority monitors groundwater nitrate concentrations, to identify vulnerable areas and to evaluate the effectiveness of the PGDA (SPW - DEE - Direction des Eaux souterraines, 2024). The observed effectiveness so far is debated without clear overall signs of improvement (Batlle Aguilar et al., 2007; SPW - DEE - Direction des Eaux souterraines, 2024). Many control sites keep exceeding the European guide level of 25 mg/l and most of the groundwater bodies in the Nitrate Vulnerable Zones (NVZ), partially or locally, have high levels and several exceedances of the standard of 50 mg/l. Despite this, average nitrate levels in some aquifers have been shown to stabilize or even decrease in recent years.

To better understand the impacts of the PGDA on nitrogen dynamics, Sohier & Degré (2010) modeled soil nitrogen surpluses using a modified version of the process-oriented EPIC model (Williams et al., 1984). Their work demonstrated the positive effects of certain agricultural practices while also highlighting significant variability driven by weather conditions, particularly precipitation. The regional authority uses this model to assess the agricultural soil nitrogen balance (SPW, 2022). However, the results are dependent on the hypotheses and assumptions about the physical processes underlying the model. When these processes are not well defined, which is challenging due to their complexity, the accuracy of the predictions is limited. Efforts have been made to measure soil nitrate concentrations at the end of the agricultural crop season, known as potentially leachable nitrogen (APL). The in-situ APL measurements are correlated with agricultural practices such as fertilization and crop management, as well as with nitrate concentrations in soil water (Vandenberghe, 2016). However these measurements remain limited in both space and time, which constrains comprehensive assessments of nitrate contamination trends. Moreover, soil and soil water nitrate concentrations can differ from groundwater nitrate concentrations, as the latter are affected by nitrate transfer lags through soil matrices (Hansen et al., 2012; Mattern & Vanclooster, 2010; Visser et al., 2007) and the slow release of accumulated nitrogen (Ascott et al., 2017; Kyte et al., 2023; Liu et al., 2024) — a phenomenon referred to as the "nitrogen legacy effect" (Basu et al., 2022; Van Meter et al., 2016).

Given these complexities, leveraging long-term groundwater quality data is a promising approach (He et al., 2022; Rodriguez-Galiano et al., 2014). Such data encode all relevant processes of the nitrate contamination process, they are widely available and they enable broader insights into nitrate pollution trends and drivers. For example, Vanclooster et al. (2020) used a data-driven approach to model groundwater nitrate pollution in the Brussels-Capital Region, Belgium, focussing on spatial variability but not on the temporal evolution of nitrate concentrations.

This study aims to leverage groundwater monitoring data to interpret the effect of the PGDA on groundwater quality in Wallonia over nearly two decades. Specifically, our objectives are:

(i) To assess the long-term (2002 – 2020) evolution of nitrate concentrations following the PDGA implementation ;

(ii) To identify the factors driving the nitrate concentration changes over time and across different locations.

2. Material and Methods

2.1 Study area

Our study area focuses on key Nitrate Vulnerable Zones (NVZ) of the Region Wallonia, Belgium, where nitrate contamination levels are the highest (SPW - DEE - Direction des Eaux souterraines, 2024). Specifically, it includes all monitored sites where levels currently exceed 50 mg/l. The PGDA defines stricter measures which are applicable only in these areas, such as specific spreading periods and conditions, obligations regarding soil cover, and monitoring requirements (Picron et al., 2017). We selected four groundwater bodies where a major drinking water company supplying the monitoring data operates several extraction sites:: the Geer basin chalks, the Brusselian sands, the Haine basin chalks and the Landenian sands (Figure 1). The concept of "water body" was introduced within the Water Framework Directive to classify the various aquatic environments that characterize the European territory. A groundwater body consists of a distinct volume of groundwater within one or more aquifers. In Wallonia, the groundwater bodies were delineated by a group of experts based on hydrogeological criteria such as the extent of geological layers or the interaction with surface waters, as well as non-hydrogeological criteria such as the administrative limits. The Geer basin chalks and the Brusselian sands were part of the first nitrate vulnerable zones as defined in 1994, and have thus been subjected to the associated regulations since then. The Haine basin chalks and the Landenian sands were added in the NVZ in 2013.

The Geer basin chalks groundwater body covers an area of 440 km² and is located in the Meuse hydrographic basin (SPW, 2016). The groundwater body's aquifer, the Hesbaye aquifer, is said to have a substantial storage capacity and a high porosity. It is partly overlaid in its northeast portion by the Landenian sands groundwater body. Agricultural land covers approximately 68% of the land surface, with 14% of it being meadows and 86% crops. The region has a high population density with 340 inhabitants per square kilometre.

The Landenian sands groundwater body spans a surface of 206 km² and is located within the Scheldt hydrographic basin (SPW, 2010). The groundwater body's aquifer is the Landenian sands aquifer. Due to limited exploitation, the hydrogeological properties of this aquifer remain poorly defined. Agricultural land covers approximately 78% of the total land surface, with meadows accounting for 8% and crops making up the remaining 92%. The population density is relatively low, with 160 inhabitants per square kilometre.

The Brusselian sands groundwater body spans a surface of 964,5 km² and is situated in the Scheldt hydrographic basin (SPW, 2006b). Its aquifer is the Brusselian sands aquifer, which has a high storage capacity but a low hydraulic conductivity. Agriculture covers 71% of the land surface and another 10% is urban land.

The Haine basin chalks groundwater body covers an area of 644 km² and is situated in the Scheldt hydrographic basin (SPW, 2006a). The main aquifer of this water body is the Mons basin chalks aquifer. The aquifer's porosity has a permeability ranging from 10^{-5} to 10^{-7} m/s, while the fissures in the chalk formation entail a permeability of 2.10^{-3} to 5.10^{-5} m/s. In the northwest, the groundwater body is partially overlaid by the Haine valley sands groundwater body. The land surface area consists of 64% agricultural land and 23% urban land.

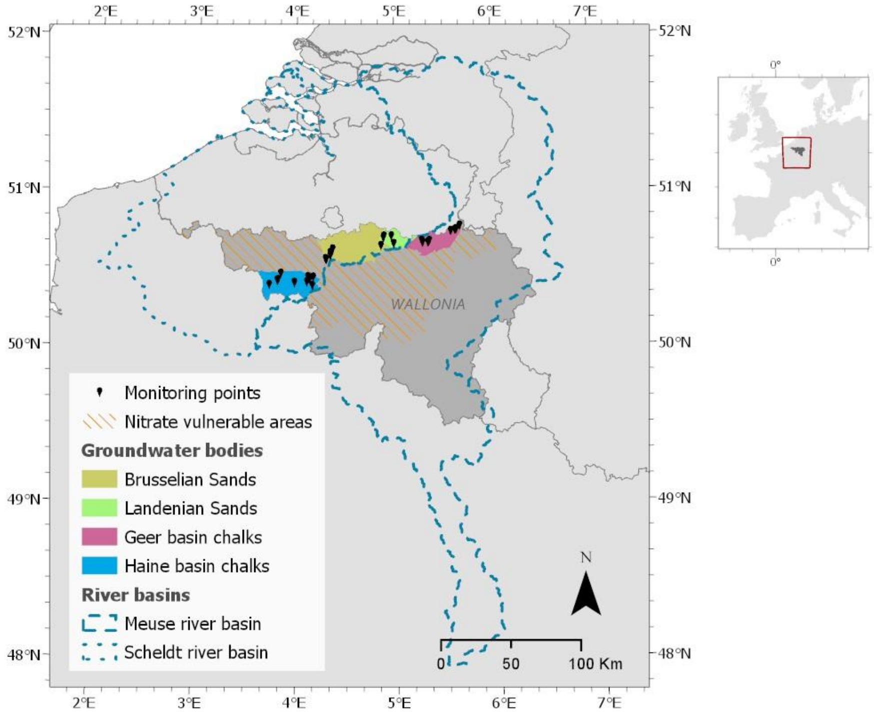

Figure 1. Location of the four groundwater bodies and the 36 monitoring points.

2.2 Groundwater nitrate concentration

2.2.1 Data collection

The monitoring points are water intake structures exploited for drinking water production by the Société Wallonne des Eaux (SWDE). They are wells, galleries, springs or drains. Only points in unconfined aquifers, with a water quality monitoring period exceeding ten years were selected. Since our focus is on vulnerable groundwater with high nitrate concentrations, we excluded one monitoring point located in anoxic groundwater, where nitrate levels are below 3 mg/l, probably resulting from high denitrification rates (Rivett et al., 2008). The final dataset included 36 points from which 13 points are in the Haine basin chalks, nine points in the Geer basin chalks, nine points in the Brusselian sands and five points in the Landenian sands (Figure 1).

Time series were available for some points from the sixties onwards, but the trend and causal analysis covers 2002-2020, starting at the onset of the sustainable nitrate management program (PGDA) and ending with the most recent year for which data were available. For nine out of the 36 points the first available data started after 2002: seven in 2003, one in 2006, and one in 2009. The temporal resolution of the nitrate measurements is variable, with the total number of measurements per point ranging from 53 to 948 over the study period. The water samples were analysed by the laboratory of the drinking water production company, the SWDE, under ISO 17025 accreditation.

The nitrate concentration time series contained some problematic values that were noticeably lower or higher than their neighbours due to reported human errors. We thus filtered the time series using a moving window of two years, with an upper and lower limit being the mean of the data within the window plus and minus three times the standard deviation. Figure 2 shows the annual averages of the resulting time series.

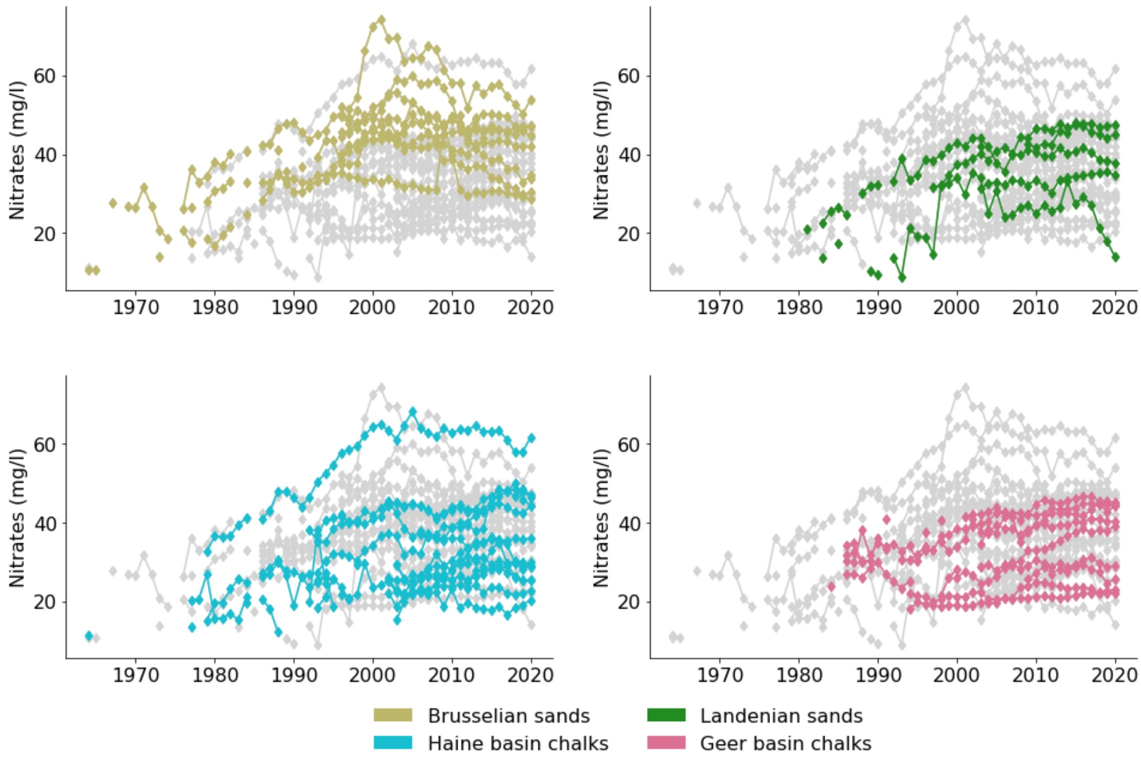

Figure 2. Time series of the yearly mean nitrate concentrations at the 36 monitoring points, sorted by groundwater body. In grey, the time series of all points. In colour, the time series of the points in each groundwater body.

2.2.2 Definition of nitrate pollution indicators

We defined a set of six indicators that capture the state and rate of change in nitrate concentration in 2002, in 2020 and the evolution in between. The indicators are used to assess the spatial and temporal evolution of nitrate contamination at each monitoring point, and, they serve as dependent variables in our data analysis to identify factors influencing nitrate concentrations. The indicators and their interpretation are defined in Table 1 and illustrated in Figure 3.

Table 1. Definition and interpretation of the nitrate pollution indicators.

Pollution indicator (I)		Unit	Usage and interpretation
I1	Average nitrate concentration in 2002	mg/l	Snapshot of nitrate concentration levels in the groundwater before the implementation of the PGDA. Serves as a baseline for comparison with future years.
I2	Average nitrate concentration in 2020	mg/l	Snapshot of nitrate concentration levels in the groundwater in a recent year. Allows for direct comparison with past data to assess changes over time.
I3	Concentration difference between 2020 and 2002 (I3 = I2-I1)	mg/l	Indicates how the concentration has changed since the implementation of the PGDA. A positive value indicates an increase in nitrate levels, while a negative value indicates a decrease.
I4	Slope in 2002	mg/l/year	Rate of change in nitrate concentration on 01-01-2002. Provides insight into how rapidly nitrate levels were changing at the beginning of the period.
I5	Slope in 2020	mg/l/year	Rate of change in nitrate concentration on 01-01-2020. Useful for understanding recent dynamics and informing current policy decisions.
I6	Difference in slope between 2020 and 2002 (I6 = I5-I4)	mg/l/year	Indicates how the rate of change in nitrate concentration has changed over the study period. A positive value indicates an accelerating increase (or decelerating decrease) in nitrate levels, while a negative value suggests a decelerating increase (or accelerating decrease).

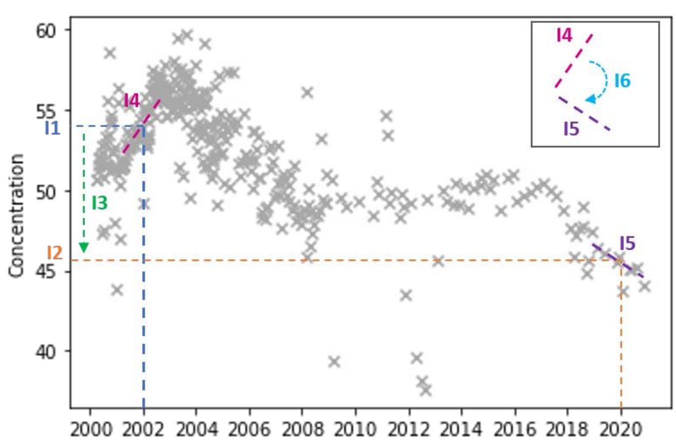

Figure 3. Illustration of the six indicators of the nitrate pollution indicators. Times series data from a monitoring station in the Brusselian sands is used for illustrative purpose.

The method used for slope computation (I4 and I5) affects the trend diagnostics. The non-parametric Mann-Kendall test has been widely applied and recommended for groundwater pollution trend assessments (Frollini et al., 2021; Grath et al., 2001; Hirsch et al., 1991; Urresti-Estala et al., 2016). This method only captures a single linear trend, making it not directly applicable to detect trend changes in long, non-linear time series. To address this limitation, Lee et al. (2010) computed trends using local regression. In our analysis, we applied two different approaches: (i) computing tangent lines to a local regression and (ii) detecting change point and applying the Mann-Kendall test. Since both methods produced similar slope indicators, we focus on the local regression approach in the main text, with details of the change-point detection approach provided in the supplementary materials.

The local regression approach consisted in smoothing the time series using locally weighted scatterplot smoothing (lowess) (Cleveland, 1979) and computing the slope of the tangent line to the smoothed time series on January 1, 2002 (=I4), and on January 1, 2020 (=I5). The lowess window length was set to eight years for time series longer than twelve years, allowing to capture the long-term fluctuations, and to 2/3 of the time series length for shorter series. Within a local window of 90 days, a linear interpolation was applied instead of a weighted regression to increase stability.

For the nine monitoring points whose first measurements were taken after 2002, the slope of the trend in 2002 (I4) was taken as the slope at the first data point, and the mean absolute nitrate concentration in 2002 (I1) was obtained by hindcasting that trend. All time series cover a period longer than eight years, the minimum length recommended by Grath et al. (2001) for groundwater pollution trend assessments.

2.3 Candidate explanatory variables

We computed a set of potential explanatory variables standing for environmental and anthropogenic factors which may impact the nitrate concentration dynamics. We used these variables as independent variables in our data analysis to identify factors influencing nitrate concentrations. The variables encompass both the inherent vulnerability to pollution and the anthropogenic influence, which include human activities that could cause or affect punctual and/or diffuse pollution. The definitions of these indicators, computation means and data sources are given in Table 2. The descriptive statistics of the values of these variables for the monitored locations are given in

Table 3.

2.3.1 Delineation of the influence zones

The risk of groundwater contamination at a specific location is influenced by the traits of the land surface area that can potentially transport pollutants to it. Therefore, delineating this land surface area, here referred to as the 'influence zone', is crucial in the analysis of groundwater nitrate concentrations (Mattern et al., 2009). We defined the influence zones as the topographic surface watersheds of the legal protect zones of each water intake structure (SPW, 2020). These zones correspond either to the groundwater table area with a maximum transfer time of 50 days to the water intake structure as estimated through geological modelling, or they are defined as circular areas centred around the structure location, whose radius depends on the aquifer substrate: 100 meters for sandy aquifers, 500 meters for gravel aquifers and 1000 meters for karstic aquifers. The boundaries of these zones are available on the regional institution's geographical data portal (SPW, n.d.). We delineated the watersheds with the ArcGIS watershed toolbox, using a 2-meter resolution raster of flow direction and flow accumulation generated by the LIDAX project (SPW, 2019).

2.3.2 Inherent vulnerability

To quantify the natural vulnerability, we considered the seven factors of groundwater natural vulnerability to pollution as defined in the DRASTIC model of the U.S. Environmental Protection Agency (Aller et al., 1985): aquifer depth, recharge, aquifer media, soil type, the topography, impact of the vadose zone, and hydraulic conductivity. We used the depth of the water intake structures as a proxy of the depth to the groundwater table since piezometric measurements were not available for all structures. We used the mean annual rainfall as a proxy for the net recharge. We used a single categorical variable, namely the groundwater body of the water intake structure, as a proxy for the three DRASTIC vulnerability factors aquifer media, impact of the vadose zone and hydraulic conductivity. The variable standing for the topography was the mean slope in the influence zone calculated using a 2 m resolution digital slope product derived from a 1 m digital elevation model. We did not include the soil in the set of explanatory variables since the main soil type of all influence zones was identical, namely loam. We considered all the vulnerability variables to be time-invariant over the studied period.
To be able to include the categorical variable aquifer media (GWbody) in our analysis, we replaced it by four binary variables using one-hot encoding. We called the new variables GWbodyLS, GWbodyBS, GWbodyHBC and GWbodyGBC, they indicate respectively the monitoring points in the Landenian sands, the Brusselian sands, the Haine basin chalks and the Geer basin chalks. We do not expect any change of these variables over the study period.

2.3.3 Land use characteristics

Agricultural land is a significant source of nitrate leaching to groundwater (Cameron et al., 2013; Strebel et al., 1989), with nitrates originating from nitrogen fertilizers applied on cropland, and from grazing livestock on meadows. Consequently, both crop land and meadow area were considered as potential drivers in this study. We included specifically potato crop cover, as it is known to leave a high concentration of potentially leachable nitrates in the upper soil layer after the growing season (Bah et al., 2015). In contrast, forested and green areas are generally less prone to nitrate leaching (Cameron et al., 2013; Zhang et al., 2013), hence we also considered it as driver, but expecting them to have a mitigating impact. Urban wastewater losses represent an additional nitrate source (Torres-Martínez et al., 2020). We used built infrastructure area as a proxy to estimate wastewater production. We visually examined the trends in these variables over the study period, finding that only the meadow area exhibited a trend. Therefore, we included a sixth variable that accounts for the change in meadow cover over the study period.

2.3.4 Point pollution sources

We considered three potential point pollution sources. Graveyards can contribute to nitrate pollution through the decomposition of organic materials (Mattern et al., 2009). Farm buildings are included because they are often associated with manure storage and handling, which can be a direct source of nitrates if not managed properly. Finally, buildings not connected to the collective sewage system indicate potential nitrate sources from septic tanks or dry wells, which can leach untreated wastewater. The datasets used capture the situation in 2020 (Table 2). We expect little change for these variables over the studied period.

Table 2. Definition of the potential explanatory variables used in the statistical models. IZ: influence zone. SPW: Service Public de Wallonie. SPF: Service Public Fédéral. SPGE : Société Publique de Gestion de l'Eau. SWDE : Société Wallonne de Gestion de l'Eau. IRM : Institut Royal Météorologique.

Variable	ID	Definition and unit	Source dataset	Provider
Water extraction depth	Depth	Depth of the bottom part of the water intake structures (m)	Documentation of water intake structures	SWDE
Groundwater body	GWBody	Groundwater body (Brusselian sands, Landenian sands, Haine basin chalks and Geer basin chalks)	Documentation of water intake structures	SWDE
Mean annual rainfall	Rainfall	Interannual (1961-2019) average of annual rainfall (mm/year)	1961-2019, 5000 m resolution climate dataset for Belgium	IRM
Topography	TerrainSlope	Average of the terrain slope in the IZ (%)	1 m resolution digital slope model 2013-2014	SPW
Crop cover	CropLU	Interannual (1998-2019) mean percentage of IZ area with crop cover (%)	Anonymous agricultural land registry (annual data from 1998-2019)	SPW
Potato crop cover	PCropLU			
Meadow cover	MeadowLU			
Change in meadow cover	MeadowReg	Trend in meadow area calculated as the slope of the linear regression of the yearly meadow area percentage between 2002 and 2020 (%/year)		
Built area	BuiltLU	Interannual (1998-2019) mean percentage of IZ area with built infrastructures (%)	Walloon land registry (annual data from 1998-2019)	SPF

Forested and green areas	GreenLU	Percentage of IZ area with forests and green spaces in 2003 (%)	Land cover map 2003	SPF
Presence of farm(s)	Farms	Number of farms in the IZ in 2020 divided by the surface of the IZ (nb/km^2)	Continuous cartographic mapping project	SPW
Presence of graveyard(s)	Graveyards	Number of graveyards in the IZ in 2020 divided by the surface of the IZ (nb/km^2)	Continuous cartographic mapping project	SPW
Buildings with automonous sewage regime	NoSewage	Number of buildings not connected to the sewage system in 2020 divided by the surface of the IZ (nb/km^2)	Walloon land registry (2020) Wastewater management plan	SPF SPGE

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Table 3. Descriptive statistics of the independent variables for the 36 monitoring points. NA: not applicable.

Variable	**Unit**	**Mean**	**Min**	**Median**	**Max**
CropLU	%	57	6	53	91
PCropLU	%	5	0,1	5	14
MeadowLU	%	8	0,4	7	19
MeadowReg	%/year	-0,02	-0,66	-0,09	0,92
BuiltLU	%	3	0,2	2	8
GreenLU	%	6	0,1	3	63
Farms	nb/km^2	0,3	0	0,4	1,2
Graveyards	nb/km^2	0,06	0	0	0,3
NoSewage	nb/km^2	0,2	0,03	0,1	0,5
Depth	m	29	0,6	19	120
Rainfall	mm/year	814	760	807	867
TerrainSlope	%	4,6	2,6	4,4	9,0
Groundwater body	-	NA	NA	NA	NA

2.4 Data analysis

2.4.1 Evolution of the nitrate concentrations

We computed descriptive statistics and visuals to depict the past and present status and temporal changes of the nitrate concentrations in the studied groundwater bodies, using the nitrate pollution indicators (Table 1).

2.4.2 Bivariate analysis to identify the controlling factors

We tested the strength and direction of the association between the nitrate pollution indicators (Table 1) and each independent variable (Table 2) separately by computing the Kendall rank correlation (also known as Kendall's tau coefficient, Kendall, 1938). The value of Kendall's tau ranges from -1 to 1 and the closer the coefficient is to either -1 or 1, the stronger the association. A higher positive value indicates a strong positive association, while a higher negative value indicates a strong negative association.

2.4.3 Multivariate linear regressions to identify the controlling factors

We used multiple linear regression to assess the individual contribution of each independent variable (Table 2) to account for potential confounding effects.

Before applying the regression models, we addressed multicollinearity by removing highly collinear variables, hence ensuring that the remaining variables provide reliable contributions to the analysis. First, we replaced the four binary variables representing the groundwater bodies, which were highly collinear, with one single binary variable ‘Aquifer’, distinguishing the aquifer media of the groundwater bodies: a value of 1 for the Brusselian and Landenian sands and a value of 0 for the Geer and Haine basin chalks. We then removed one variable at a time until the variance inflation factor (VIF) values (Mansfield & Helms, 1982) of all remaining variables were below a threshold of 5 (James et al., 2013). The variable to remove at each iteration was selected based on its VIF value, correlation with other variables and importance as explanatory variable, assessed by the authors’ expert judgment.

We built nine multiple linear regression models with the remaining independent variables, one for each indicator and slope calculation method. We used the ordinary least square (OLS) function of the Python *statsmodels* library (Seabold et al., 2010). We standardized the independent variables to a mean of zero and a standard deviation of one, facilitating the comparison of their respective impacts on the nitrate concentration indicators. We applied a stepwise multiple linear regression to identify the most important predictor variables, eliminating at each iteration the independent variable with the highest p-value until the p-values of all remaining variables were below 0.05.

3. Results

3.1 Evolution of the nitrate concentrations

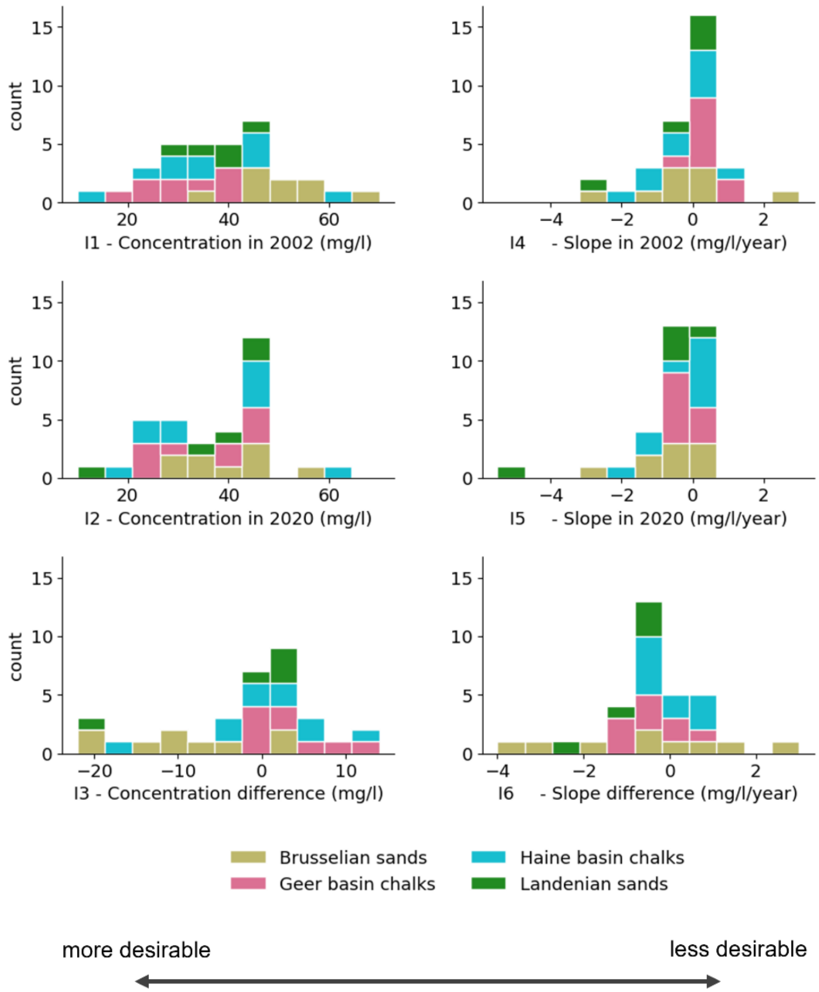
Table 4 shows statistical summaries for the six pollution indicators. It shows that the average concentration of nitrates in 2002 was 37.7 mg/l with a standard deviation of 12.2 mg/l and hardly decreased in 2020 with an average of 36.5 mg/l and a standard deviation of 10.8 mg/l. The mean change in concentration levels between 2002 and 2020 (I3) exhibits a slight decrease of 1.2 mg/l on average, but with a wide variation (standard deviation of 8.8 mg/l), ranging from a decrease of 21.3 mg/l to an increase of 12.5 mg/l. Forty seven percent of the monitored locations have witnessed a decrease in concentration, while the other 53% have seen an increase (Figure 4). The average rate of change in nitrate concentrations (I4 and I5) are slightly negative whichever the method, but with variations ranging from a negative to a positive rate. They are slightly more negative in 2020 than in 2002. The maximum rate of change has decreased from +2.7 mg/l/year in 2002 to 0.6 mg/l/year in 2020, which indicates an overall deceleration of the rate of change over the study period. This is confirmed by the negative values of the averaged I6.

While these statistics indicate a slightly mild decrease in nitrate concentrations since 2002, the distributions of the indicators color-coded by the type of aquifer in Figure 4 suggest that the decrease has mainly been significant in the Brusselian sands, while the pink stacks of the histogram for I3 indicate an increase in the Geer basin chalks.

Table 4. Descriptive statistics of the six nitrate indicators.

Pollution indicator (I)	Unit	Mean ± standard deviation	Minimum	Percentile 25 (Q1)	Median Q2	Percentile 75 (Q3)	Maximum	IQR (Q3-Q1)
I1 – Concentration in 2002	mg/l	37,7 ± 12,2	15,4	28,3	38,1	44,3	69,2	16,0
I2 - Concentration in 2020	mg/l	36,5 ± 10,8	14,1	28,6	37,0	44,7	61,6	16,1
I3 - Concentration difference	mg/l	-1,2 ± 8,8	-21,3	-4,9	0,1	3,6	12,5	8,5
I4 – Slope in 2002	mg/l/year	-0,1 ± 1,1	-3,1	-0,5	0,1	0,5	2,7	1,1

I5 – Slope in 2020	mg/l/year	-0,5 ± 1,1	-5,2	-0,7	-0,2	0,1	0,6	0,8
I6 – Slope difference	mg/l/year	-0,4 ± 1,2	-3,6	-0,8	-0,4	-0,0	2,7	0,7



275 **Figure 4. Histograms of the six pollution indicators. The black arrow at the bottom indicates that, for each indicator, lower values are more desirable than higher values, as this entail lower nitrate concentrations, decreasing trends and a decrease in concentration from 2002 to 2020.**

3.2 Identification of the controlling factors

3.2.1 Bivariate analysis

The analysis reveals a consistent positive correlation between the crop and potato crop area and all the pollution indicators, and hence an undesirable effect of these variables (Figure 5). This effect is only significative on the nitrate concentration in 2020 (I2) and rate of change in 2002 (I4). On the other hand, the analysis reveals a consistent negative correlation between the forest and green space area and all the pollution indicators, and hence a desirable effect of this variable. This effect is only significative on the nitrate concentration in 2020 (I2). Results also indicate a significant positive relationship, or undesirable effect, of the meadow area on the rate of change in 2020 (I5). Temporal trend in the meadow area (MeadowReg) shows a significant negative correlation with concentrations in 2020 (I2), suggesting a desirable effect of an increase in meadow area on I2, but shows a positive correlation with the change in concentrations (I3), suggesting an undesirable effect of an increase in meadow area on I3.

The results show a positive relationship and hence undesirable effect of the number of farms on all indicators, and of the number of graveyards on the change in concentrations (I3) and the rate of changes in 2002 and 2020 (I4 and I5). Conversely, graveyards displayed a significant negative relationship with concentrations in 2002 (I1). Finally, there is no detected influence of the presence of building area, a proxy for population density, and buildings not connected to wastewater treatment plants.

The depth of the water intake structure, serving as a surrogate for groundwater table depth, shows a significant correlation with the indicator of change in concentration (I3). However, it does not exhibit any correlation with the other indicators. There is a significant negative relationship between the annual rainfall, a proxy for recharge, and the indicator of change in concentrations (I3), while there is a significant positive relationship with the concentration in 2002 (I1). A weak negative relationship (positive effects) was found between the terrain slope and all the indicators.

Results also confirm a clear influence of the aquifer media. While concentrations in 2002 and 2020 were higher in Brusselian sands and lower in the chalks aquifers, the decrease and rate of decrease has been more prominent in the sands.

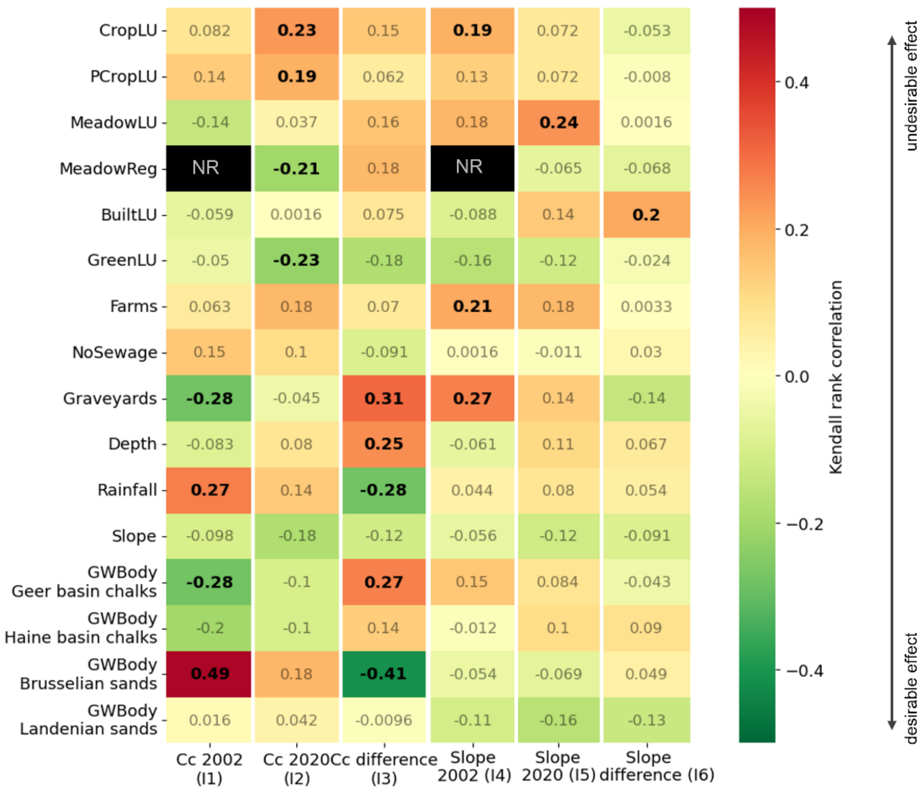


Figure 5 Heatmap of the Kendall rank correlation coefficients between the explanatory variables and the six pollution indicators. Coefficients in bold indicate a significant relationship (p-value<0.1) NR: not relevant. The black arrow on the right indicates that low correlation is more desirable than high correlation, as low correlation entails that high values of the explanatory variables are correlated to low values for the six pollution indicators and vice versa.

3.2.2 Multivariate linear regression analyses

In the process of refining our multivariate regression models, we made several adjustments to address issues of multicollinearity among the variables. We removed the variable ‘Rainfall’ for its high VIF of 242. The variables ‘CropLU’ and ‘PCropLU’ had a VIF of respectively 20 and 12, and were highly correlated (R^2=0.77). Considering the importance of 'CropLU' as explanatory variable, we chose to retain it and remove 'PCropLU.' The next variables we removed were ‘Slope’ and ‘NoSewage’ as they each had the highest VIF among the remaining variables (9 and 7 respectively). Finally, we removed the variables ‘Farms’ and ‘Graveyards’, allowing to satisfy the condition of all remaining variables having a VIF < 5. The decision to exclude 'Farms' was based on its redundancy with ‘CropLU’ and ‘MeadowLU’, which already represent agricultural activity. As for ‘Graveyards’, their very sparse distribution in the considered areas led us to expect a limited effect.

The results of the multivariate regression models with the selected independent variables are presented in Table **5**. The table shows the coefficients of the independent variables for each model, after stepwise removal of all non-significant variables (p-value of coefficient < 0.05). Note that the interpretation of the coefficient value is difficult because the independent variables have been normalized. In this normalized context, the coefficient indicates the expected change in the dependent variable per standard deviation change in the independent variable. However, it allows to interpret the relative importance of the variables, as a higher coefficient indicates a higher change of the pollution indicator per standard deviation change.

The regression models highlight the significant role of the aquifer media in explaining the variability of multiple indicators. Sandy aquifers tend to have higher nitrate concentrations but have also shown more desirable concentration changes and rates of change over the study period. The land use variables (CropLU, MeadowLU, GreenLU) exhibit varying influences across the response indicators. Larger crop areas correlate with higher concentrations and less desirable rate changes in 2020 (I2 and I5), while more forested and green areas are associated with more favorable concentration changes (I3). The models explain only 18 to 46% of the variance in the indicator values, as indicated by the R^2 coefficients.

Table 5. Coefficients of the variables used in the multiple linear regressions. Only statistically significant coefficients (p-value < 0.05) are shown. NR : not relevant.

	Cc 2002 (I1)	Cc 2020 (I2)	Difference cc (I3)	Rate of change 2002 (I4)	Rate of change 2020 (I5)	Difference rate of change (I6)
Constant	+ 37,7	+ 36,5	-	-	- 0,49	-
CropLU	-	+ 5,3	-	-	+ 0,66	-
MeadowLU	-	+ 4,0	-	-	-	-
MeadowReg	NR	-	-	-	-	-
BuiltLU	-	-	-	-	+ 0,62	-
GreenLU	-	-	- 3,9	-	-	-
Aquifer	+ 6,2	+ 5,4	- 4,8	-	- 0,38	-
Depth	-	+ 5,6	-	-	-	-
R^2	0,26	0,40	0,46	-	0,31	-

4. Discussion

4.1 Evolution of the nitrate concentrations

Since the implementation of the PDGA, the average nitrate concentration across monitoring sites has shown relative stability (Table 4). However, this overall trend conceals variations: 53% of the sites recorded an increase in nitrate levels, while 47%

experienced a decrease. The most significant reduction was observed in the Brusselian sands, while concentrations increased at most sites in the Geer basin chalks (Figure 4). These findings are consistent with those reported by (SPW - DEE - Direction des Eaux souterraines, 2024). The observed disparity in nitrate trends between these regions reflects differences in the hydrogeological characteristics of the aquifers. The Brusselian sands, characterized by higher permeability and lower nitrogen storage capacity, may exhibit shorter response times to changes in surface nitrogen loading, resulting in more immediate declines in nitrate concentrations. Conversely, the Geer basin chalks, with greater capacity for nitrogen storage and slower groundwater flow, may exhibit a delayed response to reduced nitrogen inputs. This lag effect suggests that, despite reductions in nitrogen loading (SPW, 2022), improvements in nitrate levels in the Geer basin chalks may only become evident in the coming years, provided that nitrogen management measures continue to be implemented. As Liu et al. (2024) emphasize, addressing the delayed response caused by lag effects and nitrogen legacy requires the implementation of sustained, long-term strategies.

Encouragingly, the nitrate concentration trends show improvement, with lower rates of change in 2020 (I5) compared to 2002 (I4), and a drop in the maximum rate of increase from 2,7 mg/l/year in 2002 to only 0,6 mg/l/year in 2020 (Table 4). This suggests that, although nitrate levels continue to rise at some sites, the pace has slowed, indicating a positive outcome of the measures implemented under the PDGA.

4.2 Identification of the controlling factors

Our analysis confirms the anticipated relationship between agricultural land use and nitrate contamination, with nitrate concentrations were positively correlated with cropland area (Figure 5 and Table 5, I1 and I2) (Gurdak & Qi, 2012; Wick et al., 2012). However, while this correlation was expected for 2002 (I1, prior to the full implementation of the PGDA), its persistence in 2020 (I2) is concerning. Nearly two decades after the introduction of the PGDA, which was designed to reduce nitrogen inputs and nitrate leaching, the correlation between cropland and high nitrate concentrations suggests limited effectiveness of the measures in altering the relationship between land use and groundwater quality. This lack of decoupling highlights the challenge of mitigating "legacy nitrogen" effects, whereby nitrate accumulated in soils and aquifers from past agricultural practices continues to leach into groundwater long after inputs have been reduced (Basu et al., 2022; Van Meter et al., 2016).

Additionally, one might have expected the croplands to be negatively related to the rate of nitrate concentration change (I5 in Table 5) due to the implementation of nitrogen management measures under the PGDA. Contrary to this expectation, the observed positive relationship suggests that nitrate accumulation in groundwater is ongoing. This may reflect the combined effects of legacy nitrogen and potentially insufficient compliance or enforcement of PGDA measures in some regions (Ascott et al., 2017; Hansen et al., 2012).

These findings emphasize the importance of considering both historical nitrogen loads and ongoing agricultural practices. Efforts to enhance the effectiveness of nitrate reduction policies should consider the incorporation of measures to accelerate the recovery of aquifers, such as the promotion of deep-rooted crops that reduce leaching. Enhanced monitoring and stricter enforcement of fertilizer application limits may also help to mitigate further contamination. Furthermore, given the evident lag effects, long-term policy evaluations should account for the temporal dynamics of nitrate transfer and accumulation within aquifers.

Forested and green areas exhibit a negative association with nitrate pollution, showing a lesser contribution to nitrate leaching. This aligns with findings by Zhang et al. (2013) and (Cameron et al., 2013), who noted that forests act as natural buffers by promoting nitrogen uptake and reducing runoff. Moreover, the data suggests that nitrate contamination evolution (I3) was slightly better in more forested zones (Table 5). This implies that PGDA measures might currently be more effective in areas with lower inherent vulnerability. Interestingly, while reductions in meadow area over time correlate with lower nitrate concentrations, meadow area itself does not show a significant impact on nitrate levels. This may result from the failure to differentiate between pasture—typically associated with high nitrate leaching—and other types of meadows (Sacchi et al., 2013). Addressing this distinction in future assessments could enhance the specificity of land-use-related policies.

No significant negative effect was observed from built infrastructures, which contrasts with the findings of Mattern et al. (2009) who identified residential land as having negative influence on nitrate concentrations in the Brusselian sands. This discrepancy may be due to our study's focus on semi-rural areas, where built-up areas are limited (under 8% of total land use, Table 3) and the associated sewage pressure is likely low. Expanding monitoring to include more urbanized areas could clarify the influence of residential land use on nitrate trends.

Aquifer type emerges as a critical predictor of nitrate vulnerability, with sandy aquifers showing higher contamination levels than chalk aquifers. This suggests that policies should prioritize sandy aquifer regions, particularly those under cropland, for targeted measures like stricter nitrogen application limits and buffer zones. However, aquifer type also predicts changes over time (I3), with a general decrease in nitrate concentrations in the Brusselian sands contrasting with increases in the Geer basin chalks (Figure 5). This finding underscores the importance of tailoring strategies to the hydrogeological characteristics of aquifers. For example, in chalk aquifers, where response times to management measures are longer due to nitrogen storage and slower groundwater flow, additional interventions may be needed to address legacy nitrogen.

Depth also plays a significant role, with shallow groundwater points showing more improvement in nitrate concentrations compared to deeper ones. This delayed response in deeper groundwater bodies to the PGDA measures reflects a time lag in nitrate transfer through the vadose zone. Supporting evidence from (Masetti et al., 2008) in Northern Italy highlights how higher annual precipitation and recharge rates can shorten lag times and improve nitrate trends.

The correlation between graveyards and less favorable nitrate trends warrants careful consideration, as sparse distribution of the graveyards may obscure localized effects. Targeted monitoring near such potential point sources could help identify specific mitigation needs.

The low predictive power of the multivariate models (R^2 = 18–46%) reflects the complexity of groundwater systems and the factors influencing nitrate concentrations Groundwater systems are influenced by numerous natural and anthropogenic elements, some of which have not been fully captured due to data limitations or to unaccounted sources of nitrate leachate (Masetti et al., 2008). This complexity is compounded by non-linear relationships and time lags between surface changes and groundwater response, which our linear regression models may not fully capture (Wick et al., 2012). Furthermore, the limited number of groundwater data points (36) restricts the model's ability to account for the full variability of the system (Ishwaran, 2007).

To mitigate multicollinearity, variables with high Variance Inflation Factor (VIF) values were removed from the multivariate regression, which likely improved the stability of the coefficient estimates but could also have led to the exclusion of significant predictors. Rainfall was removed as it exhibited highest collinearity, as expected, given its regional variability, which also applies to other variables such as aquifer type, crop land use, and depth. Potato cropland was quite obviously highly collinear with cropland and therefore excluded. Interestingly, land use variables were not as collinear as anticipated, which can be explained by the fact that built infrastructure only accounts for buildings rather than the total urban area, and that the land use variables (crop + forest + built) thus not encompass the entire area. The methodological trade-off of removing potential controlling factors to avoid collinearity further contributes to the relatively low model performance (Ishwaran, 2007).

4.3 Challenges in defining the pollution indicators and the independent variables

We provide six nitrate pollution indicators that collectively capture nitrate pollution state and trends over time. However, the use of these indicators, comes with certain limitations. The indicator on the difference in nitrate concentration (I3) may miss short-term fluctuations, while indicators I4 and I5, representing the slopes at the beginning and end of the study period, may not be representative of the longer-term trend. The indicator I6, the difference in slopes, is quite abstract and harder to interpret. The reliability of these indicators depends on the quality and completeness of the underlying data, as well as on the methods used to compute them. The indicators of rate of change (I4, I5 and I6) demonstrate robustness, as they are not very sensitive to the chosen method for defining local slope, as evidenced by the similar statistics (see supplementary materials - Table S1), which increases confidence in their values.

The precise delineation of the influence zones is essential for effectively characterizing and quantifying the independent variables representing the factors affecting nitrate pollution, as highlighted by Nobre et al. (2007) and Mattern et al. (2009). However, defining these zones is fraught with challenges, due to complex subsurface geology, the ever-changing dynamics of groundwater flow, and constraints related to data availability. In our study, we used methodological simplifications to delineate these zones, acknowledging the resulting approximation. Another pitfall is the spatial overlap of the influence zones of some monitoring points. This overlap implies that the monitoring points are not entirely independent, leading to similar values of potential explanatory variables for these points. This dependency in our dataset could have influenced the data analysis.

A key strength of our study lies in the comprehensive computation of a broad array of independent variables representing potential controlling factors. However, some choices made to characterize these variables, driven by data-availability limitations, may have introduced additional noise or uncertainty to the data analysis. For instance, we used the variable 'Depth Structure,' rather than the more precise variable ‘Groundwater table depth’, unavailable at the necessary spatial scale, to account for the travel time in the saturated zone. Similarly, ‘Rainfall’ was used as a proxy for ‘Precipitation surplus’, also unavailable at the necessary spatial scale, to approximate net recharge. These substitutions, while necessary, can affect that accuracy of our analysis, and mainly the lack of more significant relationships between the independent variable and the nitrate pollution indicators. Furthermore, some potentially important controlling factors, such as agricultural practices, mitigation measures, livestock density, and manure and fertilizer storage practices, could not be included in our analysis due to the lack of regionally available proxies for these variables.

4.5 Perspectives

The findings of our current study open several pathways for future research. A primary direction, contingent to the data-availability, is the enhancement of independent variables representing controlling factors. Future work should refine existing variables by distinguishing between more specific crop types and rotations, as well as differentiating grazed from non-grazed meadows. Additionally, incorporating data on fertilizer application rates, livestock densities, nitrogen surplus estimations, in-situ leachable nitrogen measurements, the state of sewage systems, actual groundwater table depth and precipitation surplus, and concentrations of other pollutants, representative of nitrate-generating activities, would help increase the representation of all factors potentially influencing nitrate contamination. However, these advancements depend heavily on the availability of comprehensive and open datasets.

Our data-driven approach could be combined with complementary methodologies. Groundwater dating and chemical and isotopic analyses (Böhlke & Denver, 1995; Christiaens et al., 2023; Mattern et al., 2011; Vanclooster et al., 2020) could offer critical temporal perspectives on the source and evolution of groundwater contamination. Numerical process-oriented models could allow to better take into account the mechanisms known to affect nitrate leaching to groundwater.

Expanding the groundwater nitrate concentration dataset by incorporating additional monitoring points would strengthen the representativeness and predictive power of models.

With a larger dataset and better quality independent variables, more advanced, non-linear machine learning techniques could be employed to uncover new insights and capture the complexities of nitrate contamination. These approaches could provide a deeper understanding of the underlying processes and hence help guide future best management practices. However, expanding the dataset and collecting better-quality independent variables is a challenging task due to the limited availability of long-term and spatially explicit data, and the complexities associated with data collection across multiple stakeholders. Addressing these challenges will be crucial for the development of more robust and advanced analytical approaches in future studies.

5. Conclusions

This study contributes to global scientific knowledge by investigating the long-term spatial and temporal dynamics of nitrate contamination in groundwater, linking these trends to land use, hydrogeological conditions, and nitrogen management policies. By leveraging two decades of data, it provides insights into persistent contamination sources, such as cropland, and highlights the critical role of aquifer characteristics and depth in mediating nitrate responses. To enhance policy effectiveness, our results suggest the relevance of targeted interventions by prioritizing vulnerable aquifers and cropland-dominated zones. They also highlight the need to address time lags and nitrogen legacy effects by sustaining and intensifying measures over extended periods.

The limited predictive power of the regression models reflects the inherent complexity of groundwater nitrate contamination and the difficulty of adequately representing its controlling factors. Constraints such as data availability and required simplifications in defining independent variables limit model reliability. This underlines the need for more detailed and accessible datasets that better capture the controlling factors, and the need to sustain and perhaps expand the monitoring systems to better capture spatial variability and localized impacts of land use, and long-term changes. This will facilitate the development of advanced machine-learning modelling approaches capable of better addressing the multifaceted and nonlinear processes governing nitrate pollution.

Data availability

The data used in this study are not publicly available. For data access, please contact the corresponding author.

Author contribution

EV and AA conceptualized the study. EV performed the data curation and formal analysis. LC provided the data. MV did the funding acquisition and project administration. AA and MV carried out the supervision. AA and EV wrote the original manuscript.

Competing interests

One of the coauthors is a member of the editorial board of Hydrology and Earth System Sciences.

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