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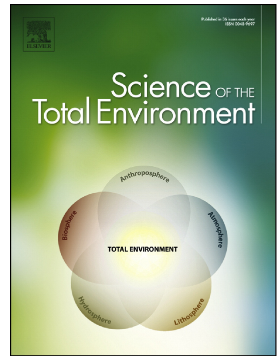

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Grey water footprint as an indicator for diffuse nitrogen pollution: The case of Navarra, Spain

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Abstract

Nitrogen is an essential element for plant growth, while its application and associated pollution is a worldwide concern. Generally, diffuse source pollution and its impacts on ecosystem health are difficult to monitor and regulate. Here we used the grey water footprint (GWF) and water pollution level (WPL) indicators, based on a soil nitrogen balance approach to differentiate between surface and groundwater, in order to better understand and quantify the pressure that nitrogen fertilisation generates on freshwater. For the first time, we compared the results of these indicators with actual nitrogen concentration data in surface and groundwater bodies, showing in both cases a positive significant correlation according to Spearman correlation coefficient. This means that the theoretical WPL results might be valuable to anticipate and identify nitrate pollution in surface and groundwater bodies. However, several factors influence the N-related processes that should be considered, such as natural attenuation. We estimated the agricultural and livestock nitrogen loads delivered to freshwater and the associated GWFs and WPLs at the municipality level in Navarra. Large GWFs are observed in southern Navarra, particularly in intensive agricultural regions such as Ribera Alta-Aragón and Ribera Baja. We estimated that 64% of the GWF related to nitrogen loads came from artificial fertilisers, 16% from manure, 11% from atmospheric deposition and the remaining 9% from biological fixation, seeds and other organic fertilisers. Among the crops, cereals had the largest contribution to the nitrogen-related GWF (54%) followed by vegetables (17%) and fodder (11%).

Keywords

Grey Water Footprint; Water Pollution Level; Nitrogen; Agriculture; Livestock; Navarra.

1. Introduction

Diffuse nitrogen (N) pollution is a major water challenge facing the twenty-first century and is likely to increase as the global population expands and with regular diet becoming more meat-intensive (Steffen et al., 2015). Inefficiencies in the management of mineral fertiliser and manure makes agriculture the largest source of N pollution worldwide (Kanter and Searchinger, 2018). More than half of the N applied to agricultural land does not contribute to plant and animal growth, instead, mixes in air and water (Kanter and Searchinger, 2018). Unintended adverse environmental impacts are caused from N leaching and runoff from agricultural soils, including groundwater contamination and eutrophication of freshwater and estuarine ecosystems (Zhang et al., 2017). Discharges of N to water bodies are projected to increase by 35-46% between 2000 and 2050 on the basis of the climate change scenarios (IFPRI and Veolia, 2015).

Point sources of pollution are still a concern in some parts of the world (Ferreira et al., 2017; Sanches Fernandes et al., 2018), but are generally easier to identify and more cost-effective to quantify, manage, and regulate. In comparison, diffuse source pollution and its impacts on ecosystems are more difficult to monitor and regulate due to its high spatial and temporal variability, the high transaction costs involved in dealing with large numbers of heterogeneous polluters and because pollution control may require co-operation and agreement within catchments and across sub-national jurisdictions and countries (OECD, 2017). Recently, various papers have addressed the impact of diffuse pollution sources on nitrate-related water quality degradation, under different land uses and anthropogenic pressures (Pacheco et al., 2015; Pacheco and Sanches Fernandes, 2016; Mockler et al., 2017). However, these papers do not make explicit the different type of crops and farming systems, using the sole category “agriculture”.

As a measure to quantify the pressure that N pollution from different sectors and crops puts on freshwater resources, we used the grey water footprint indicator (GWF). The GWF is measured as the volume of water required to assimilate the load of pollutants based on natural background concentration and existing ambient water quality standards (Hoekstra et al., 2011).

The concept of water pollution level (WPL) is used to determine if there is sufficient water flow to assimilate N pollution. WPL is defined as the GWF in a water body divided by the runoff (Hoekstra et al., 2011). The WPL quantifies the part of the pollution assimilation capacity in a water body that has been used. WPL = 1 indicates the pollution assimilation capacity has been fully utilised. WPL > 1 indicates that the environmental assimilative capacity (i.e. loading) limits have been exceeded resulting in a violation of water quality standards.

Since the evolution of the GWF concept (Chapagain et al., 2006), a number of GWF and WPL studies have been published quantifying the anthropogenic N load to fresh water from different sectors (agricultural, domestic and industrial) and scales in a theoretical way, with no comparison of the indicator results with actual N concentrations (Chukalla et al., 2018a; 2018b; Karandish, 2019; Liu et al., 2012; Mekonnen and Hoekstra, 2011; Mekonnen and Hoekstra, 2015; Pellicer-Martínez and Martínez-Paz, 2016; Miglietta et al., 2017; Zhao et al., 2019). Regarding the methodology, to the best of our knowledge, only three previous studies have used an advanced soil balance approach for estimating the diffuse N loads at the field (Chukalla et al., 2018a; 2018b) and global level (Mekonnen and Hoekstra, 2015). Among those, only the global study specifies GWFs together with WPLs by sector or crop (Mekonnen and Hoekstra, 2015). However, studies that include both GWFs and WPLs differentiating between the surface and groundwater pollution are lacking.

The purpose of this paper is to assess for the first time the robustness and accuracy of the GWF and WPL indicators applied to agriculture, comparing their results with real N concentration in water bodies, and to relate the N pollution in water bodies to specific sectors and crops. The use of detailed local data has allowed the development of a soil N balance approach and to differentiate between surface and groundwater bodies. The studied region is the Autonomous Region of Navarra, in northern Spain, essentially occupied by farmlands and forest spots, where southern water bodies are affected by nitrates of agricultural origin (Figure 1).

2. Study area

2.1. Site description and land use

The Navarra Autonomous Region is located in northern Spain on the western side of the Pyrenees. Navarra has a total population of approximately 640,000 inhabitants and an area of 10,384 km^2 (Figure 1). Agricultural land in Navarra accounts for 33% of the territory (340,127 ha), of which 68% is rainfed and 32% irrigated, mostly in southern Navarra (Government of Navarra, 2019a). Cereals represent 66% of the cultivated area, followed by fodder, 10%, vegetables, 8%, vineyards, 6%, and others, 10%. Soil erosion and nutrient export is an issue in Navarran agricultural lands (Casalí et al., 2008; Casalí et al., 2010; Mechán et al., 2018). Forests, scrubs and grasslands represent around 60% of the land cover (618,012 ha) and unproductive land the remaining 8% (80,244 ha) (Government of Navarra, 2019a).

2.2. Climate

Navarra is divided into seven agricultural regions according to different soil physical and climatic conditions (Vicente et al., 2004). The north-western part of Navarra (agricultural region I and the northern part of IV) has a temperate maritime climate, with oceanic influence. Rainfall is abundant in the north, where it varies from 1,100 to 2,500 mm, becoming progressively scarcer to the south. The Alpine Region (agricultural region II) includes the Pyrenean and Pre-Pyrenean valleys and presents more continental conditions. The central area of Navarra (agricultural regions III and V and the less mountainous and southernmost area of IV) has a Mediterranean climate. The Ebro "Ribera" (agricultural regions VI and VII) has a temperate Mediterranean climate with dry summers, few and irregular rains, with less than 500 mm per year, great temperature fluctuations and semi-arid steppe spots in its western zone.

2.3. Geology and landscape

Navarran geology is highly diverse. It is located between the Pyrenees and the Cantabrian mountain range, with its southern half almost over the Ebro massif right up to the edge of the Iberian System. Its structure is very varied, having the characteristics of each of these units. There are two differentiated landscapes in Navarra: the mountains in the northern half, as part of the Pyrenean and Cantabrian orography, and the "Ribera" in the southern half, with large plains and flat landscape located in the depression of the Ebro valley. Lands in the north are generally above 600 m altitude, whereas those in the south below 400 m (Vicente et al., 2004).

2.4. Nitrate vulnerable zones

Currently, there are three designated nitrate vulnerable zones (NVZ) in Navarra: Cidacos river basin, Robo river basin and Ebro-Aragón river basin (Government of Navarra, 2018a)(Figure 1).

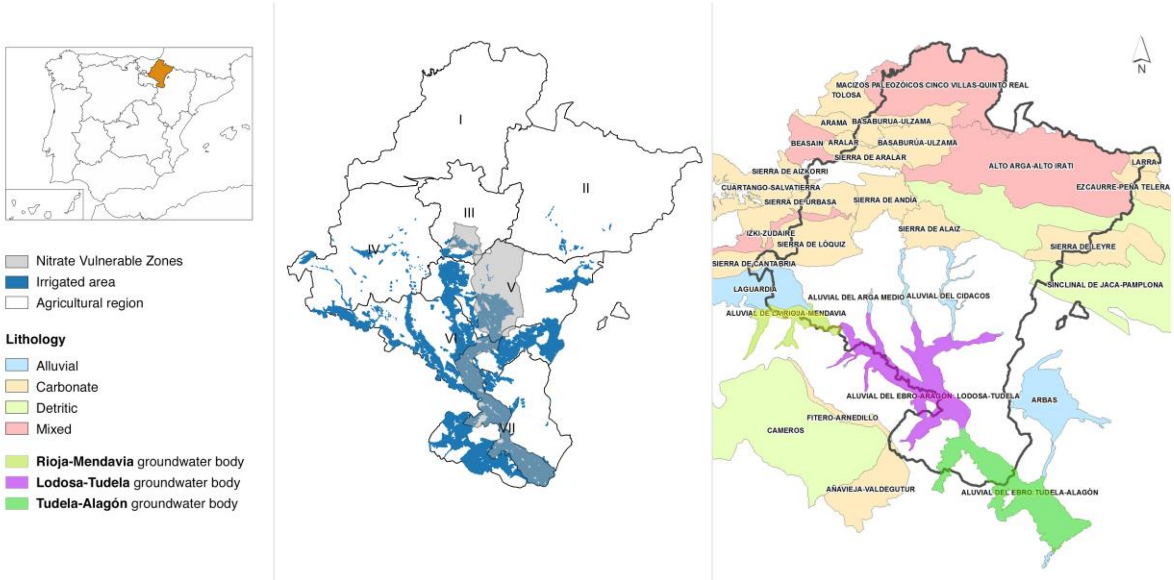


Figure 1. Location and main features of the study area: Navarra, Spain. Agricultural regions I Noroccidental, II Pirineos, III Cuenca de Pamplona, IV Tierra Estella, V Navarra Media, VI Ribera Alta-Aragón, VII Ribera Baja (left). Map of groundwater bodies in Navarra highlighting the Ebro alluvial aquifer (Rioja-Mendavia; Lodosa-Tudela; Tudela-Alagón) (right). Source: Government of Navarra (2018d).

3. Material and methods

3.1. Agricultural nitrogen load to freshwater

Diffuse N loads to fresh water from agriculture were estimated for 60 crops separately following the method described by the Spanish Ministry of Agriculture, Fisheries and Food (MAPA, 2018).

We considered six inputs: application of artificial fertiliser, animal manure, other organic fertilisers different from manure (compost from solid urban waste and sewage sludge), seeds, wet and dry atmospheric deposition, and biological N fixation. The outputs in the N balance included N withdrawal from the field through crop harvesting and N output through taking away crop residues (straw-plant), annual growth of woody crops (wood and roots), burning of crop residues, fertiliser volatilisation and gaseous losses from soil.

We took spatial crop distribution per agricultural region for 2017 from the Government of Navarra (2019a) and the distribution per municipality for the agricultural year 2017/2018 from the POCTEFA PyrenEOS project (PyrenEOS, 2019). The application rate of artificial fertiliser per crop was obtained from the Government of Navarra (2018a), which provides limits for the application of N fertilisers to crops in NVZs in Navarra. In the case of cereals, the practical recommendations for rainfed and irrigated winter cereals that were followed in Navarra were obtained from the regional Institute of Agrifood Technology and Infrastructure (ITGA, 1996). N fertiliser inputs for crops, total and per agricultural region, where also analysed for the period extending from 2000 to 2018.

The manure input was calculated at the agricultural region level by multiplying livestock density registered at the beginning of 2018 (Government of Navarra, 2019b) with the animal-specific excretion rates (MAPA, 2018). The animal-specific excretion rate per animal age, orientation (milk/meat), and destination was sourced from MAPA (2018). MAPA (2018) does not provide the fraction of the manure spread on the cropland per agricultural region and livestock type. We assumed that all non-volatilized manure was applied to cropland within each agricultural region, except for the poultry manure, which is mainly used outside Navarra − approximately 20% for composting -mainly exported to the rest of Spain, Portugal and France and 80% for mushroom production in Pradejón and Autol in La Rioja, Spain (Redondo-Izal, personal communication). The allocation of manure was performed according to the crop and managed grassland area in each agricultural region (Government of Navarra, 2019a). In this study, we considered manure inputs on croplands (including managed grasslands); however, we did not conduct further study on manure inputs on grazing lands. Data on compost from solid urban waste applied to vineyard and cereals and sewage sludge applied to vegetables for Navarra were obtained from MAPA (2018). Their allocation was performed according to the crop area mentioned in each agricultural region.

The N input from seeds was calculated for rainfed and irrigated croplands separately by multiplying the crop area (ha) (Government of Navarra, 2019a) with the seed dose (kg/ha) and N richness (% N of seeds) per crop as in MAPA (2018). Wet and dry atmospheric deposition rates were calculated by adding the pre-fertilisation volatilisation and fertilisation volatilisation divided by the district area and multiplied by the agricultural area in line with MAPA (2018). The biological N fixation rates were sourced from MAPA (2018).

The N removal through harvested crops and crop residues (straw-plant) was estimated by multiplying the crop production (Government of Navarra, 2019a) by the crop-specific N content, which was interpolated between maximum/minimum crop yields (Government of Navarra, 2019a) and minimum/maximum extraction coefficients for each crop (kilograms of N to produce a ton of crop) following MAPA (2018). The annual growth of woody crops (wood and roots) was based on the crop-specific N extraction coefficients from MAPA (2018).

The fertiliser volatilisation includes organic fertilisers (manure and other organic fertilisers) from their production (i.e. from the pre-fertilisation), mineral fertilisation and crop residue burning. The mineral fertiliser volatilisation rate for Navarra is 13.71% and the organic fertiliser volatilisation rate is 20%, which is in line with MAPAMA (2017). The crop residue burning was based on the crop-specific N extraction coefficients from MAPA (2018). The gaseous losses from soil were calculated following MAPAMA (2017).

Leaching and runoff of N -the movement of N from the soil to ground or surface waters was estimated by assuming balance of N in the soil in the long term.

Finally, to estimate the N loads to surface and groundwater, we used the historic average surface runoff per municipality (mm) and historic average aquifer recharge data per municipality (mm) for the period 1980/81-2005/06 from the SIMPA Model (MITECO, 2019).

3.2. Grey water footprint

Following Hoekstra et al. (2011) the GWF (m^3/y) is calculated by dividing the N load (Load, kg/year) by the difference between the ambient water quality standard for N (the maximum acceptable concentration C_{max}, mg/l) and the natural concentration of N in the receiving water body (C_{nat}, in mg/l) (Box 1):

$$GWF = \frac{Load}{(C_{max} - C_{nat})} \tag{1}$$

The natural concentration is the concentration in a water body if it were in pristine condition, before human disturbances in the catchment. For this study we selected a value of 10 mg NO_3/l as the very good status class change limit from the Royal Decree 817/2015 (BOE, 2015).

We have used a maximum allowable concentration value of 50 mg NO_3/l for groundwater in line with the European Nitrates Directive (EC, 1991). The maximum allowable concentration for surface water was assumed to be 25 mg NO_3/l, except for the Basque Pyrenean rivers, which was 15 mg NO_3/l, as the limit of good-moderate status of the Royal Decree 817/2015 (BOE, 2015).

3.3. Water pollution level

The WPL, which measures the degree of pollution within a catchment, is estimated as the ratio of the total of GWF in a catchment to the runoff from that catchment (R_{act}, m^3/year) (Hoekstra et al., 2011) (Box 1):

$$WPL = \frac{GWF}{R_{act}} \tag{2}$$

The historic average total runoff data (i.e. the data from the gauging station that accounts for both surface and groundwater contribution to the river flow) (mm) and average groundwater flow (mm) at municipality level for the period 1980/81-2005/06 were obtained from the SIMPA Model (MITECO, 2019).

Box 1. Grey water footprint and water pollution level: concepts and usefulness

The grey water footprint (GWF) associated with a human activity is an indicator of the severity of water pollution. It refers to the volume of freshwater that is required to assimilate a load of pollutants based on the ambient water quality standards (Hoekstra et al., 2011).

Building on the GWF, the water pollution level (WPL) in a catchment area is the degree of pollution of the run-off flow, measured as the fraction of the pollution assimilation capacity of runoff actually consumed. A WPL of 1 or higher means that the waste assimilation capacity of the runoff flow has been fully consumed (Hoekstra et al., 2011).

Both concepts look at water quality from a pollution load perspective from the ecosystem standpoint, moving beyond the sectorial emission or discharge viewpoint. In comparison and complementary to other sophisticated and robust models and tools that calculate the emissions of nitrogen to water, such as MONERIS (Venohr, 2011), SWAT (Arnold et al., 1998) or GREEN (Grizzetti et al., 2008), the GWF and WPL indicators are able to quantify the dimension and severity of the pressure from different sectors and crops to water bodies in a disaggregated manner, and improve the understanding of the relation between these pressures and the impact on water bodies.

The GWF and WPL indicators are also complementary to other models and methods such as the “eutrophication potential”, which considers the interaction between nitrogen and phosphorus in eutrophication.

3.4. Correlation between water pollution level and actual N content

The non-parametric Spearman’s rank correlation coefficient is computed using XLSTAT to assess the association between the water pollution level (see section 3.3) and the actual N concentration in water bodies (Government of Navarra, 2018b; 2018c, 2018d) as data do not follow a normal distribution.

4. Results

4.1. Grey water footprint

The N load to fresh water systems from croplands is estimated at 11,609 tonnes in the year 2017. The grey surface water footprint related to agricultural N loads was 1,961 hm^3/year and the grey groundwater footprint 282 hm^3/year in the year 2017, that is, 87% and 13%, respectively (Figures 2 and 3).

The spatial variation of GWF correlates to the spatial variation of the nutrient loads, which are highest in areas of intensive agriculture in southern Navarra, particularly in agricultural regions Ribera Alta-Aragón and Ribera Baja.

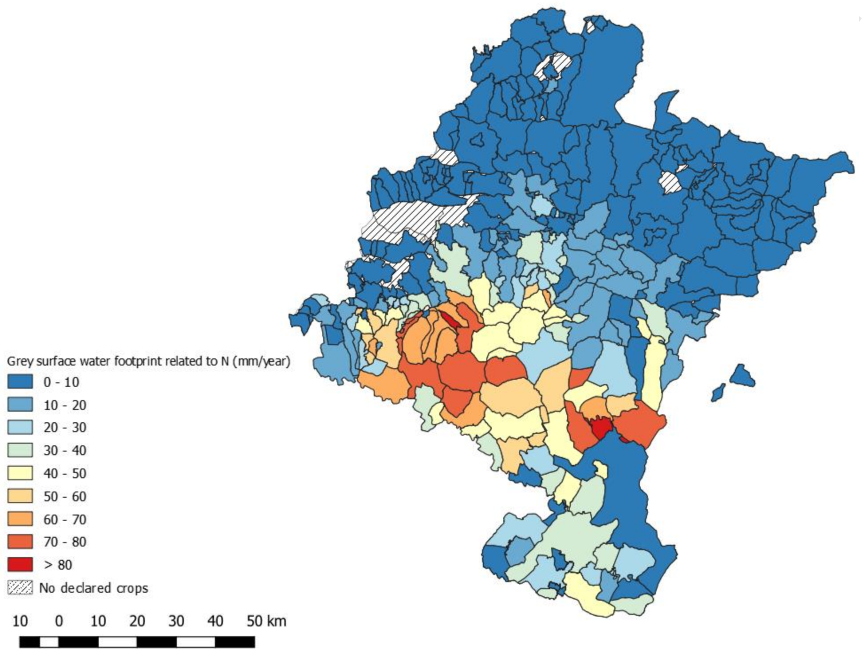


Figure 2. Grey surface water footprint related to nitrogen (N) loads from diffuse sources. Year: 2017/2018. The data are shown in mm/year at the municipality level.

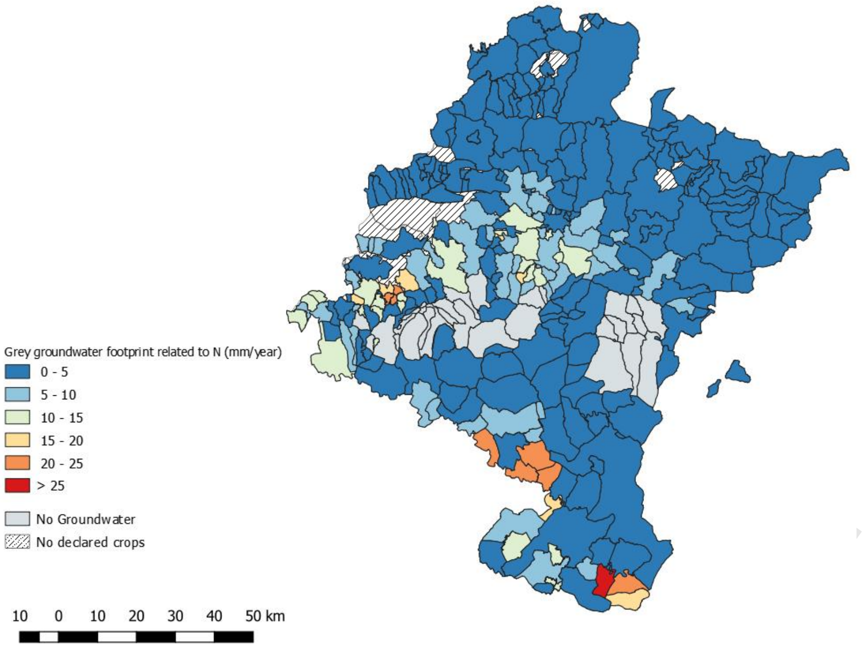


Figure 3. Grey groundwater footprint related to nitrogen (N) loads from diffuse sources. Year: 2017/2018. The data are shown in mm/year at the municipality level.

The contributions of different product categories and agricultural regions to the global GWF related to N loads to fresh water are presented in Figure 4. The largest share (54%) comes from cereal production (wheat 21%, barley 19% and maize 8%), 17% from the production of vegetables (broccoli 6%), and 11% from fodder (polyphyte meadows 4% and fodder maize 3%).

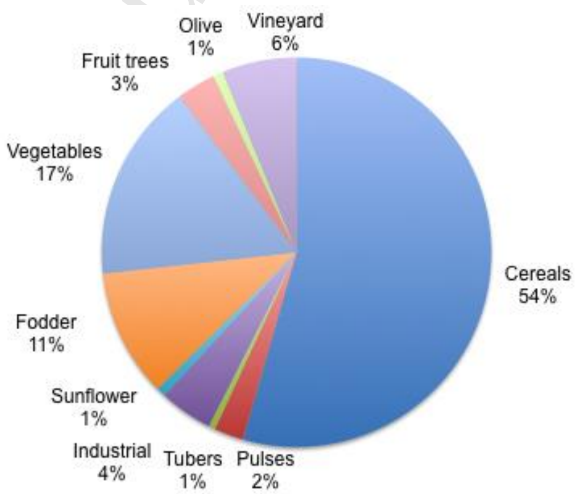


Figure 4. Relative contribution of different product categories to total grey water footprint (GWF) related to nitrogen loads. Year 2017.

Looking at the regional distribution, the Tierra Estella agricultural region is the largest GWF contributor, with approximately 37% to the total grey surface water footprint and 39% to the total grey ground water footprint (Figure 5).

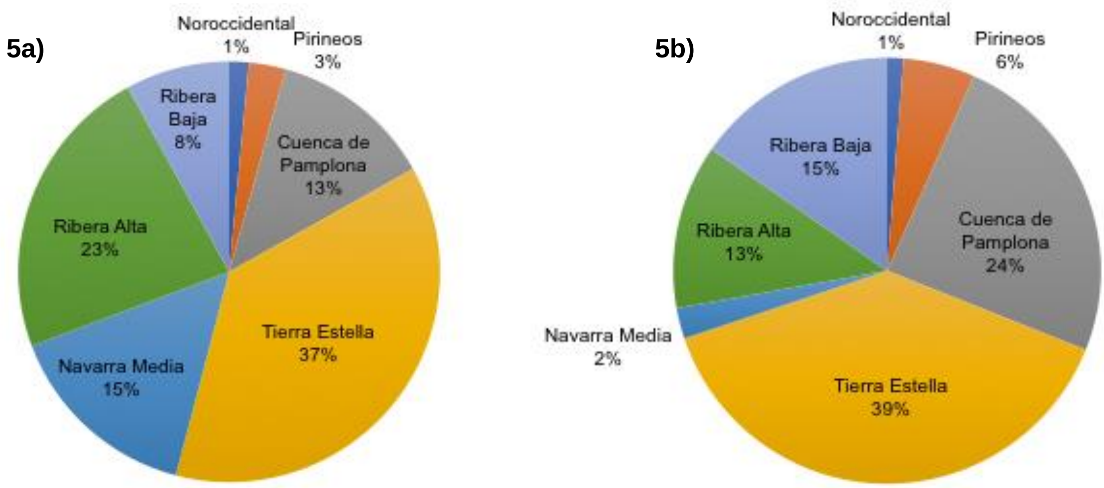


Figure 5. Relative contribution of different regions to total grey surface water footprint (5a) and groundwater footprint (5b) related to nitrogen loads. Year 2017/2018.

The largest share of the N input in croplands comes from artificial fertilisers, which accounts for approximately 64% of the total input. N input from manure accounts for 16% of the total input, atmospheric deposition, 11%, biological fixation, 7%, seeds, 2% and other organic fertilisers account for 1%.

Cereal crops account for approximately 73% of the N input from artificial fertiliser, 59% from manure, 67% from other organic fertilisers, 81% from seeds and 65% from atmospheric deposition. These large numbers are due to their large cultivated area and relatively high fertiliser use rates. Approximately 67% of N input from bio-fixation came from fodder legumes, 18% from vegetable legumes and 15% from pulses.

Cereal crops account for the largest N removal with harvested crops and crop residues (69%) and are responsible for 69% of the N fertiliser volatilisation and 66% of the gaseous losses from soil. Fruit trees, olive groves, and vineyards are responsible for 100% growth of woody crops and burning of crop residues.

4.2. Water pollution level

The WPL related to anthropogenic N loads are shown in Figures 6 and 7. Both surface and groundwater bodies in the rainy region north of Navarra meet water quality standards for N (< 50 mg NO_3/l for groundwater bodies; < 25 mg NO_3/l for surface water bodies, except for Basque Pyrenean rivers, which is < 15 mg NO_3/l).

The municipalities with surface WPL > 1 cover approximately 40% of the total land area and are mainly located in southern Navarra, 13% in Ribera Baja, and 12% in Ribera Alta-Aragón agricultural regions. In these areas pollution assimilative capacity is insufficient to assimilate the actual N pollution resulting in a theoretical violation of water quality standards.

The municipalities with ground WPL > 1 cover approximately 25% of the total land area of Navarra, which is a little less than half of the total area covered by groundwater in Navarra (62%). These areas are placed over the Ebro alluvial aquifer in southern Navarra (Figure 1), where most irrigated agricultural land is located, 9% in Ribera Baja and 12% in Ribera Alta-Aragón agricultural regions. The alluvial aquifer is significantly recharged by the infiltration of irrigation water and discharges into the river with several months of deferral (LIFE Nitrates, 2015).

On the whole, the actual water quality data in terms of annual average NO_3 in surface and groundwater bodies and NVZs are consistent with the theoretical N-related water pollution levels (Figures 6 and 7). Currently, groundwater has good quality in Navarra, except for the Ebro alluvial aquifer, which shows a moderate nitrate status (25-50 mg/l), achieving higher concentrations (50-100 mg/l) in the Tudela-Cortes area due to the development of intensive farming (Government of Navarra, 2018b). In general, nitrate concentrations in surface water bodies in Navarra are lower than those measured in the Ebro alluvial aquifer. Only low-flow rivers such as Cidacos, Iranzu (tributary of the Ega river), Odrón or Robo presents average annual N concentrations above 25 mg/l (Government of Navarra, 2018b).

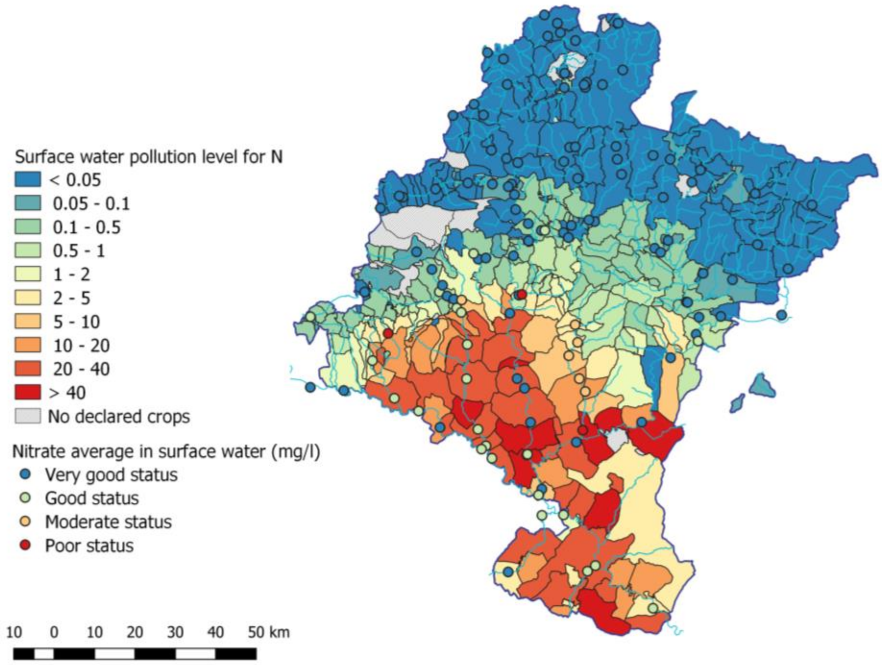


Figure 6. Surface water pollution level per municipality related to nitrogen (N) loads from diffuse sources (year 2017/2018) and annual average NO_3 concentration (mg/l) in surface water bodies (year 2018). Source: Government of Navarra (2018b and c). The indicators are: very good status (<10 mg NO_3/l), good status (10-25 mg NO_3/l), moderate status (25-50 mg NO_3/l), poor status (> 50 mg NO_3/l). For the Basque Pyrenean rivers: Very good status (<8 mg NO_3/l), good status (8-15 mg NO_3/l), moderate status (15-50 mg NO_3/l), and poor status (> 50 mg NO_3/l).

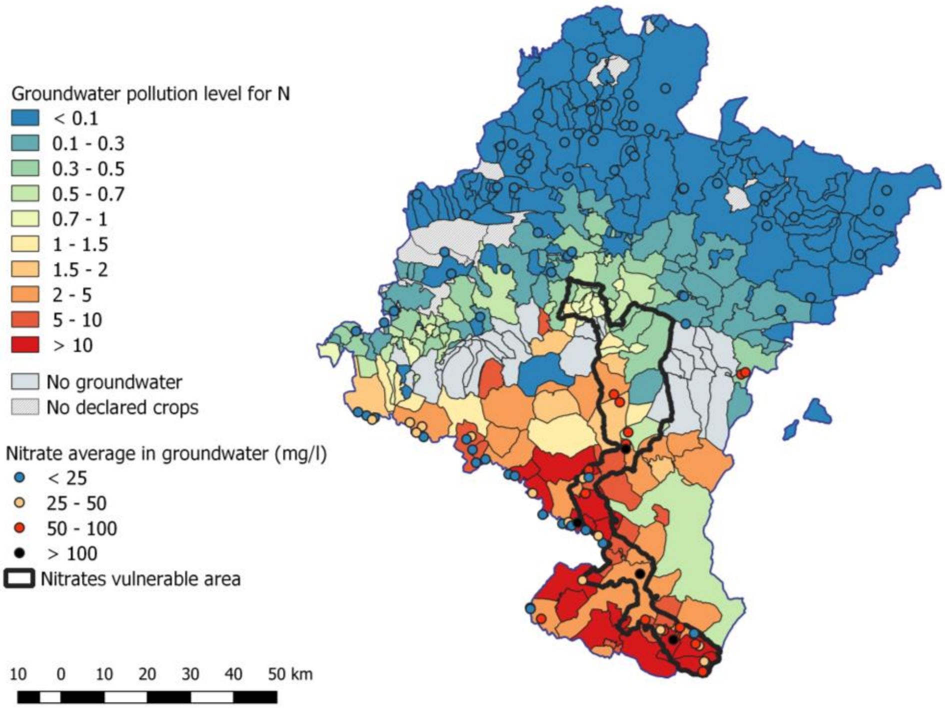


Figure 7. Groundwater pollution level per municipality related to nitrogen (N) loads from diffuse sources (year 2017/2018) and annual average NO_3 concentration (mg/l) in groundwater bodies (year 2018). Source: Government of Navarra (2018d).

4.3. Correlation between water pollution level and actual N content

There is a significant positive correlation ($r_s = 0.742$, $n = 103$, $P < 0.001$) between the ground WPL and the actual N concentration in groundwater bodies. In the case of surface water, there is also a significant positive correlation ($r_s = 0.706$, $n = 116$, $P < 0.001$) between the surface WPL and the actual N concentration in surface water bodies. This means that both variables, WPL and actually measured N concentration, move in the same direction.

4.4. Crop fertilisation: analysis and trends

In general terms, total fertiliser use in Navarra has remained relatively stable since 2000 in a range of 35 and 40 thousand tonnes (Figure 8). More than 40% of total fertiliser use happens in the Ribera Alta-Aragón (VI) and Ribera Baja (VII) agricultural regions, 23% and 17% respectively (Figure 9). These two regions, VI and VII, are located over the Ebro alluvial aquifer (Figure 1), where nitrate groundwater pollution is mainly related to excessive fertilisation and surplus irrigation water (LIFE Nitrates, 2015).

Most N fertiliser input comes from artificial fertilisers; however, manure can be a relevant source in several agricultural regions: Noroccidental (70%), Ribera Alta-Aragón (30%) and Ribera Baja (10%).

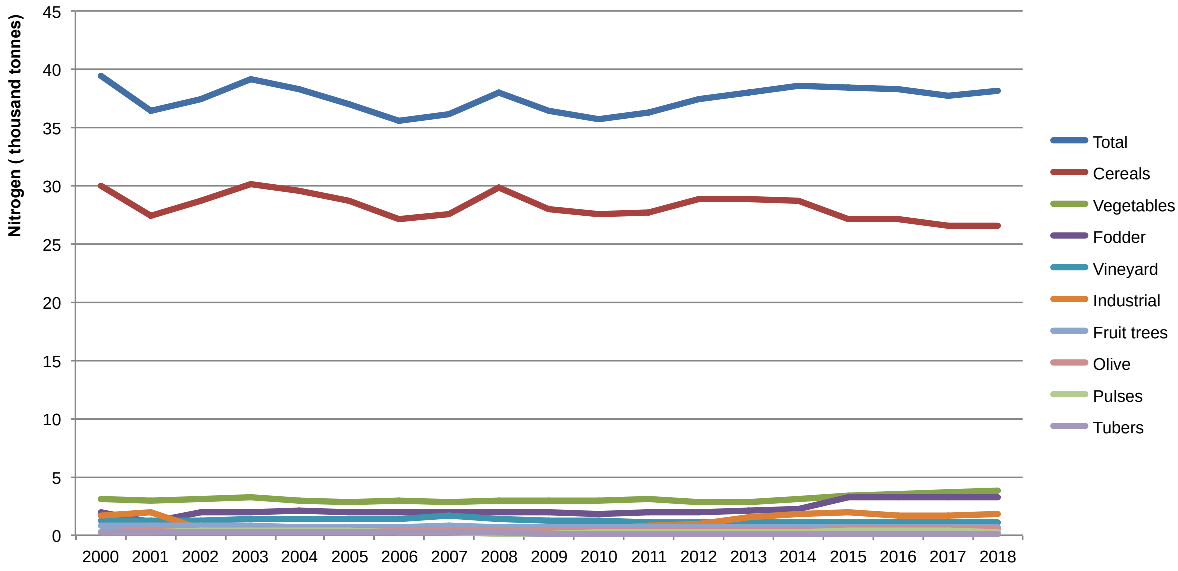


Figure 8. Nitrogen fertiliser input (thousand tonnes) for crops, irrigated and rainfed, in Navarra. Period 2000–2018.

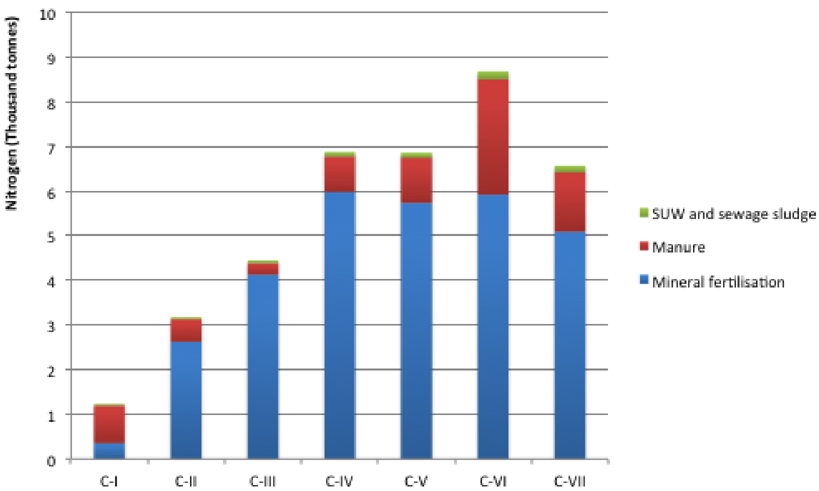


Figure 9. Nitrogen fertiliser input (thousand tonnes) for crops per agricultural region in Navarra. Year 2017. C-I Noroccidental, C-II Pirineos, C-III Cuenca de Pamplona, C-IV Tierra Estella, C-V Navarra Media, C-VI Ribera Alta-Aragón, C-VII Ribera Baja.

Except for the Noroccidental agricultural region, cereals are the main N fertiliser users in Navarra, followed by vegetables (Figure 10). Rainfed wheat and barley are the largest N inputs in northern agricultural regions, while rainfed barley and maize prevail in southern areas.

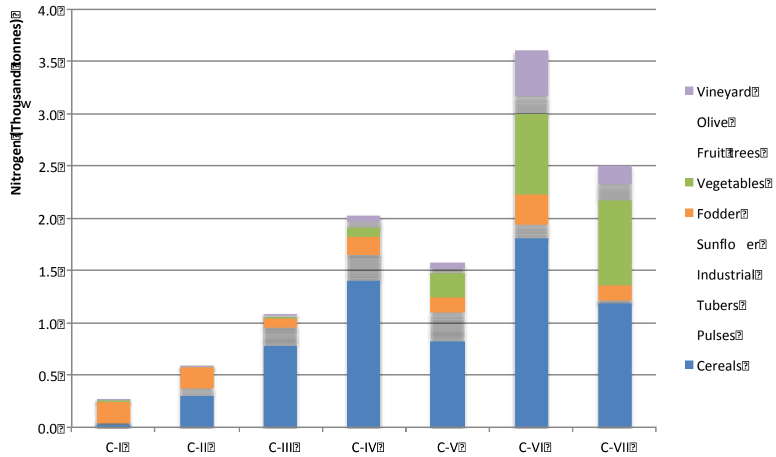


Figure 10. Relative nitrogen fertiliser input (thousand tonnes) by different product categories and agricultural regions. Year 2017. C-I Noroccidental, C-II Pirineos, C-III Cuenca de Pamplona, C-IV Tierra Estella, C-V Navarra Media, C-VI Ribera Alta-Aragón, C-VII Ribera Baja.

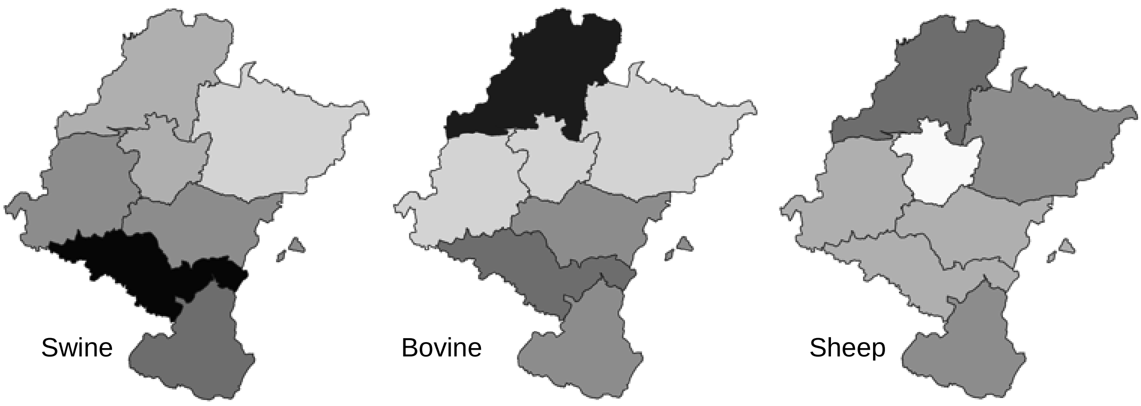
As regards the crops in agricultural regions VI and VII, with the highest WPL and actual NO_3 concentrations, cereal and vegetables experience the main N inputs. In the case of agricultural region VI, maize, rainfed barley and, with increasing relevance, vegetables are the main contributors. While in the case of agricultural region VII, vegetables, with increasing relevance, and maize are the N-related main crops.

4.5. Organic fertiliser: analysis and trends

Sixteen percent of the total N input in croplands comes from manure (Figure 9). In the context of the European Nitrates Directive (91/676/EEC) (EC, 1991), N from manure is treated more cautiously because its agronomic behaviour is different from that of N from mineral fertilisers. Nitrogen from manure is not readily available for crops as the one from mineral fertilisers. It needs to be mineralised before being absorbed by the crops. The time gap between mineralisation and crop utilisation increases the risk of N losses to water. The Directive only sets out a maximum N limit for manure to be applied on land that is 170 kg/ha/year. This limit only applies to areas identified as NVZs (i.e. polluted or at risk of pollution) (Figure 1).

Large number of local animal concentration poses high risks to the environment when manure production is out of balance with land availability and crop needs. This imbalance creates a surplus of nutrients, a large amount of which is sooner or later lost to water and air, if not exported out of the region, sometimes leading to additional pressures in the receiving areas (EC, 2018).

Livestock farming pollution (from stabled livestock farms) can be significant at local level in the Ebro alluvial in Navarra (LIFE Nitrates, 2015). The largest livestock manure N inputs (kg/year) are located in the Ribera Alta-Aragón agricultural region, mainly due to pork production (Figure 11). Pork is the main contributor to N loads in southern agricultural regions VI and VII, 52% and 35% respectively, without accounting for poultry manure, which is mainly used outside Navarra (Redondo-Izal, personal communication).



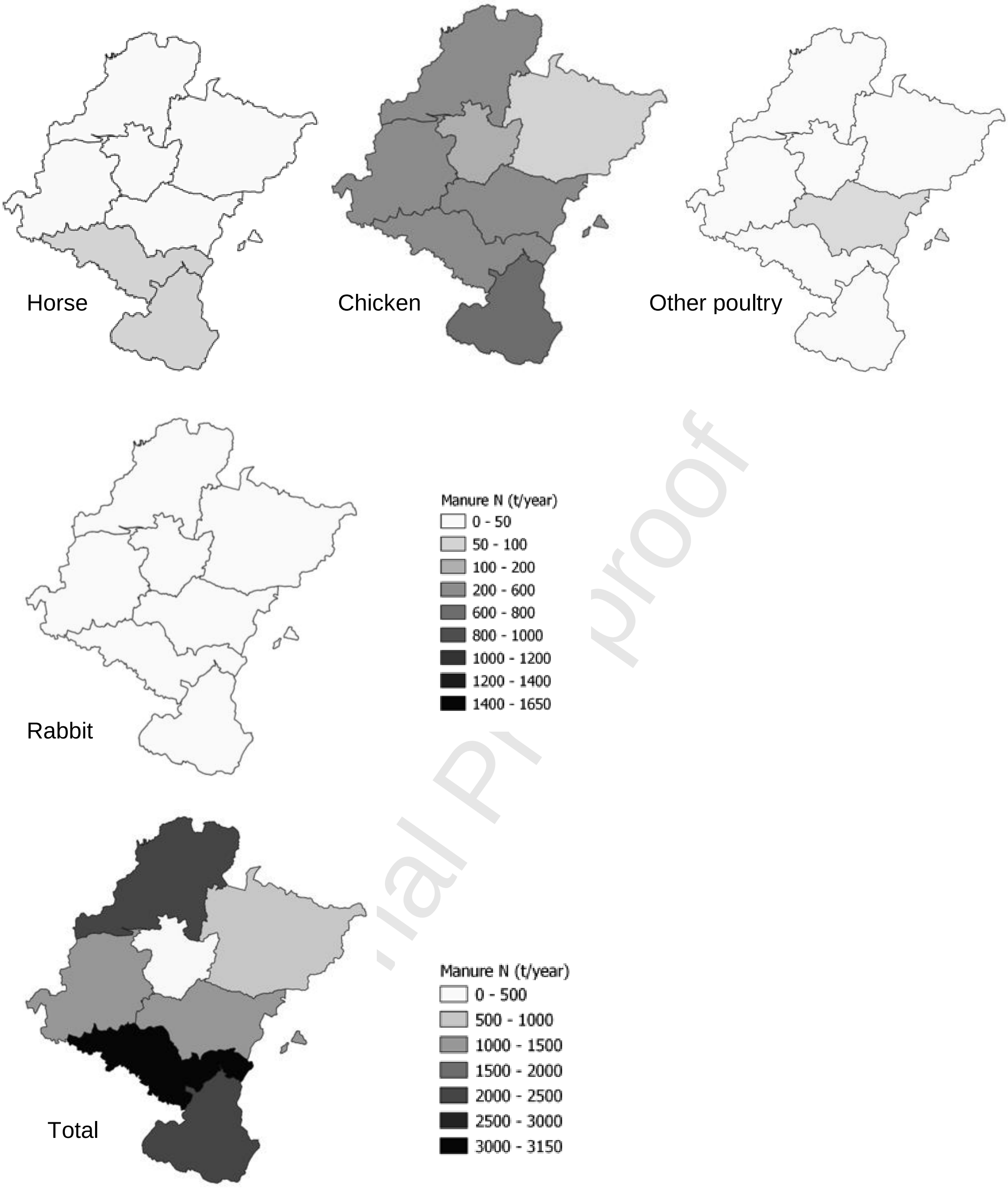


Figure 11. Pork, bovine, ovine, equine, chicken, other poultry, rabbit and total indoor livestock manure nitrogen content (tonnes/year) per agricultural region in Navarra (2018).

Pork industry is undergoing a tremendous development in recent years (Figure 12). Currently, in Navarra there are as many pigs as inhabitants. This development has been so rapid that its positive or negative consequences or externalities have not been examined. The effects of animal manure generally are a combination of positive and negative effects. Manure can have negative effects on water and air pollution, but can also bring gains. For instance, in Navarra, manure and slurries, together with other agro-food wastes, are used in three biomethanisation or biogas plants and seven composting plants. In 2016, 38% of the compost was used in Navarra, mainly in agriculture and, to a lesser extent, in gardening (Government of Navarra, 2018e).

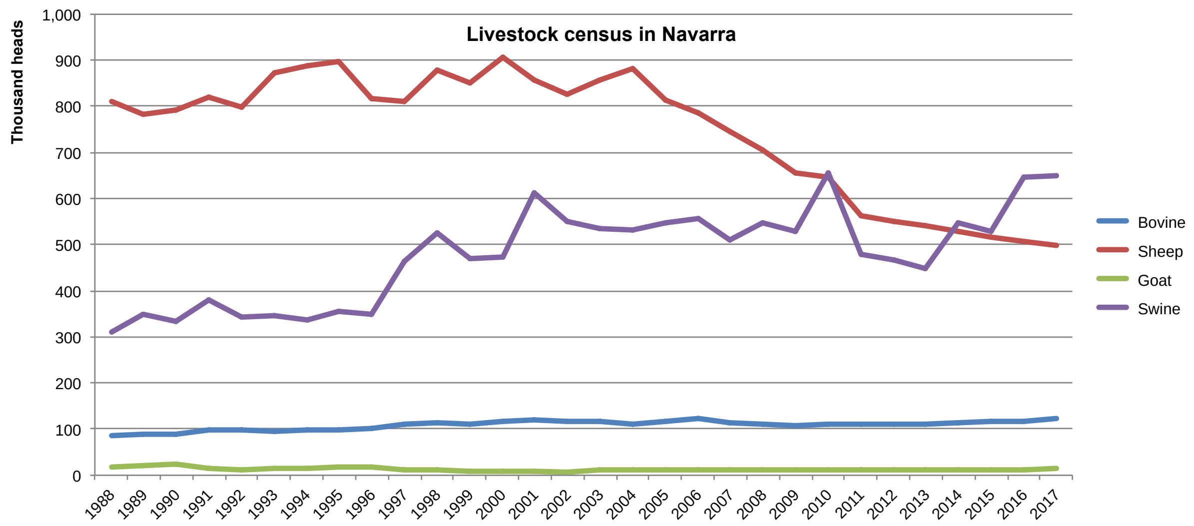


Figure 12. Livestock census in Navarra. Source: Statistical Yearbook (MAPA, 2018) and Government of Navarra (2019c).

The bovine manure production, in terms of N per year, is also significant in Noroccidental agricultural region I, where 70% of the total N input in croplands comes from manure (Figures 9 and 11).

This study accounts for the livestock manure N input to croplands but not for the input to grazing lands. The indoor and outdoor livestock manure N input to grazing lands accounts for 71% of the total N input in the Noroccidental agricultural region I and for 24% in the Pirineos agricultural region II - without accounting for seeds, biofixation and atmospheric deposition.

In the remaining agricultural regions, III, IV, V, VI and VII, manure N input to grazing lands is lower, representing 5%, 7%, 3%, 7% and 7%, respectively.

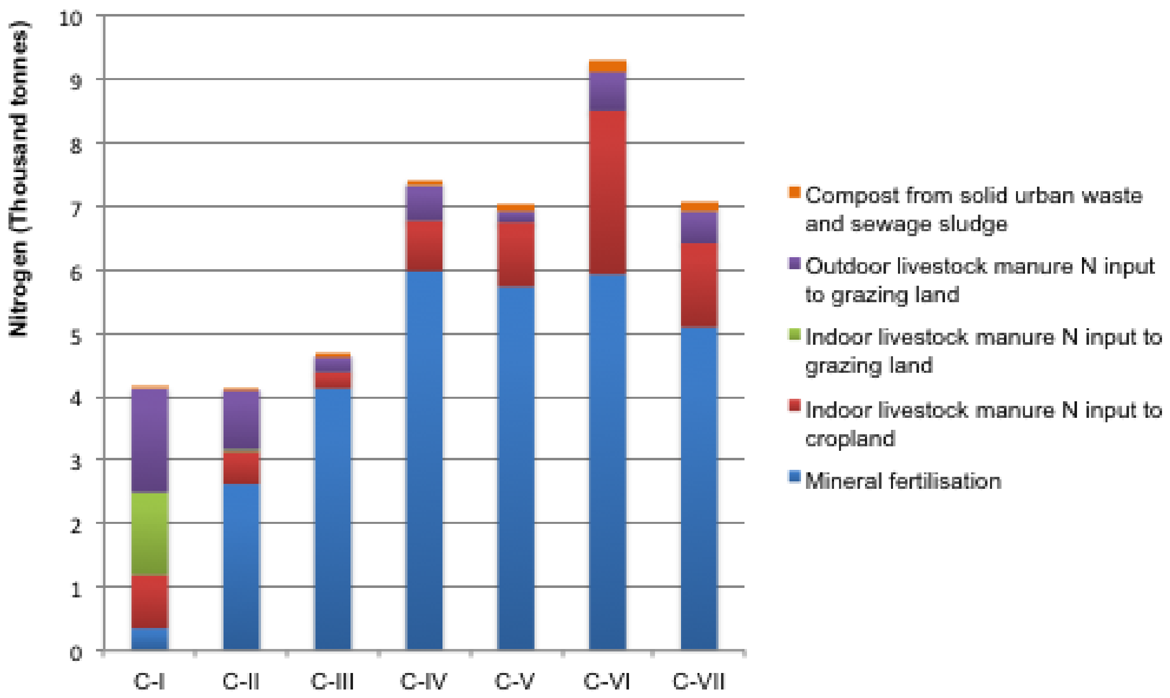


Figure 13. Nitrogen fertiliser inputs (thousand tonnes) to cropland and grazing land per agricultural region in Navarra – without accounting for seeds, biofixation and atmospheric deposition. Year 2017/2018. C-I Noroccidental, C-II Pirineos, C-III Cuenca de Pamplona, C-IV Tierra Estella, C-V Navarra Media, C-VI Ribera Alta-Aragón, C-VII Ribera Baja.

5. Discussion

5.1. Consistency between water pollution level and actual N content

The study demonstrates a significant positive correlation between the theoretical WPL results and the actual N concentration in water bodies, suggesting a predictive relationship between them. However, several are the factors that might influence the N-related processes. According to the results of this study, the correlation is stronger in groundwater bodies, probably because in the case of surface water bodies other factors come into play, such as natural attenuation of pollution by the riparian vegetation or denitrification in river banks (Burt et al., 2007; Rivett et al., 2008; Antiguedad et al., 2017). When these processes occur, river basins are able to assimilate more N and need less water to dilute. This might be the case of some of the 29 surface water spots in very good and good status for nitrates (NO_3 < 10 mg/l and 10-25 mg/l, respectively) that fall within areas polluted beyond the natural assimilative capacity according to the theoretical surface WPL values (WPL > 1) (Figure 6). This might also explain some of the differences between the WPL results and real NO_3 values in groundwater bodies. There are 20 groundwater spots in good status for nitrates (NO_3 < 25 mg/l) that fall within areas polluted beyond the natural assimilative capacity according to the estimated ground WPL values (WPL > 1) (Figure 7). There seems to be an overestimation of the WPL as compared to measured concentrations, partly explained by natural attenuation processes. The GWF and WPL results can be considered a worst-case scenario.

On the other hand, there is one surface water spot in poor status for nitrates (NO_3 > 50 mg/l) that falls within a river stretch where a certain amount of assimilative capacity is available according to the theoretical WPL values (WPL < 1). This is the case of the Robo river downstream in Puente La Reina. The Robo catchment is a NVZ that comprises a set of agricultural plots located in a slope that drains the water towards the Robo tributaries (Government of Navarra, 2018b). There are also three groundwater spots in poor status for nitrates (NO_3 > 50 mg/l) within areas where a certain amount of assimilative capacity is available according to the theoretical WPL values (WPL < 1): one in the Cidacos alluvial aquifer, which is a NVZ, and the other two in springs near the city of Sangüesa (Manantial de Balrrollada and Fuente de la Alpargata). These discrepancies seem to be due to several factors not considered in the methodology. For instance, the Robo and Cidacos were both recently transformed to irrigation, and their boundaries do not match with the municipal ones.

5.2. Uncertainties and limitations of the study

The evaluations in this study are assumed to be inside the expected uncertainty range and taken as acceptable for the current purposes. The reality is complex, and there are inevitable uncertainties to be dealt with in water resources management and planning.

First of all, the GWF and WPL results strongly depend on the ambient water quality standards, which might vary depending on the jurisdiction. Ambient water quality standards are not available for all substances and for all places (Hoekstra et al., 2011). Therefore, the results of the present study might not be comparable with other GWF assessments under different jurisdictions.

Second, this study does not account for the manure input to grazing lands. This might have caused a potential underestimation of the N flow to surface and groundwater bodies, particularly in the Noroccidental agricultural region I, where livestock manure input to grazing land is significant. In this region, for the moment, the real measured NO_3 concentrations in surface or groundwater bodies do not exceed the prevailing legal limits. In southern agricultural regions, VI and VII, where these limits are exceeded, manure inputs to grazing lands, even if less than a tenth of the total inputs (see section 4.5), could represent an additional pressure on water bodies.

Third, temporal dynamics is a factor that affects the nutrient balance results. To estimate N leaching and runoff, this study assumes a steady state condition in the long-term in the soil regarding N content, which might not hold true in all places. Unlike phosphorus, which remains in the soil for a long period, N leaching is mainly in nitrate form, which is soluble and thus highly mobile in soil. This introduces an element of uncertainty into the overall nutrient balance.

Fourth, this study does not take into account N legacy, that is, the N stored in the vadose zone, which is an important source of groundwater pollution (Ascott et al., 2017; Sebilo et al., 2013; Wang et al., 2019). According to Ascott et al. (2017) N storage per unit area in the vadose zone is relatively large in Europe, including Spain, where there are relatively thick vadose zones and extensive historical agriculture. This means that the current trends of nitrate concentration increases observed in water bodies associated with agricultural areas might be the result of both current and past activities throughout the last decades. In the case of Navarra, there might be a link between the high N concentrations present in areas with low WPL levels and the presence of those vadose zones. Extreme precipitation events under climate change may accelerate the transport of nitrate through the vadose zone in the future, moving nitrate into the deep soil and aquifers (Wang et al., 2019).

Fifth, the surface and ground grey water footprint assessment here uses the grid-based hydrologic Precipitation-Contribution Simulation model (SIMPA) (Alvarez et al., 2004; MITECO, 2019). This model, based on the monthly soil-moisture balance (Temez model), simplifies soil processes, underestimating groundwater recharge, and partly distorting total run-off and tending to undervalue it (UPC, in press). As this study uses the SIMPA model for the estimation of N loads to surface and groundwater bodies using a percentage of the surface runoff and aquifer recharge data, the ground GWF and WPL may be underestimated, while surface GWF and WPL overestimated.

There might also be some additional distortions on the ground WPL results as the SIMPA model assumes that the system is in a natural state without extractions from the aquifers, which is not the case in Navarra, where groundwater is moderately used for urban use and irrigation purposes. Thus, the groundwater flow might have been assumed to be slightly larger than actual flow. Because the groundwater flow is used as denominator in the ground WPL calculation, the ground WPL might have been additionally underestimated.

This study assumes that the EU Nitrates Directive fertilisation limits are met. This is not entirely true in the non-vulnerable or vulnerable areas, partly due to the lack of control by the administration.

Finally, this study does not consider the natural attenuation of pollution, such as assimilation by the riparian vegetation or denitrification in the river banks (Burt et al., 2007; Rivett et al., 2008; Antiguedad et al., 2017). As the way forward, field verifications in those areas with high WPL values are needed. There might be a link between the low N concentrations found in high WPL value spots and areas with environmentally-friendly farming techniques such as natural barriers, corridors, vegetated buffer strips and vegetative cover conservation. And vice versa, a linkage might be found between the high N concentrations present in areas with high WPL levels and the “lack” of those agri-environmental schemes. In this way, concrete measures might be identified to improve N attenuation performance in farms, and a clear methodology to determine and prioritise which places should be assessed – those with the highest WPL.

5.3. Policy implications

In the context of the European Nitrates Directive (91/676/CEE), which aims at preventing and reducing the pollution of water caused by nitrates from agricultural sources, an analysis of the GWF and WPL evolution and trends might be a useful tool to support the process of anticipating and designating NVZs and identifying key sectors, taking into account the limitations mentioned in Section 5.2. The use of the GWF and WPL might me particularly interesting beyond Europe, in those countries with limited or no data on the actual N concentration in water bodies.

In water bodies identified as polluted or at risk of pollution, the establishment of Codes of Good Agricultural Practice and action programs have proved to be effective tools, such as in the case of the Rioja-Mendavia alluvial aquifer, which was formerly designed as NVZ (LIFE Nitrates, 2015).

Reducing nitrate levels below the levels required by Community legislation might be challenging in highly agricultural watersheds. Some systems respond quickly to the Codes of Good Agricultural Practice, whereas others need more time due to the inertia (LIFE Nitrates, 2015). Different situations call for different approaches. In line with the present results, and in agreement with the LIFE Nitrates (2015), in addition to the actions to improve fertilization and irrigation, there seem to be other ways to reduce the nitrogen loading from irrigation return flows in intensive agricultural areas. First, the establishment of riparian vegetation using species with high nutrient binding capacity can reduce nitrogen and phosphorus transfers from land to water (Burt et al., 2007; Rivett et al., 2008; Antiguedad et al., 2017). Second, the placement of artificial wetlands or the use of the existing natural wetlands can reduce nitrate levels in aquatic systems.

The analysis of the different sectors over time invites to make a reflection on the evolution of the swine sector. If the prices and foreign demand remain high and other limitations do not arise, there might be a clear rise in the number of pigs in the future, which might cause further pressure in the already polluted agricultural area VI. The livestock loads should be monitored closely and the externalities of the sector further analysed.

Finally, the GWF looks at the water quality issue from a pollution load perspective using the ecosystem assimilation capacity approach rather than from the emission standard viewpoint. The emission standard approach alone might not be sufficient for protecting water quality (Hoekstra et al., 2011; Zhang et al., 2014).

6. Conclusions

This study, for the first time, analyses the usefulness of the grey water footprint as an indicator of nitrogen diffuse-pollution to water and compares its results with actual nitrogen concentration in water bodies in Navarra.

There is no “one-size-fits-all” solution for dealing with N concerns in agriculture, since agri-environmental conditions and regulations differ across countries. The GWF and WPL indicators could help improve the understanding on the association between anthropogenic pressures and policies and nitrogen impacts on water. Where no actual data are available, the GWF and WPL, could potentially be useful theoretical indicators for roughly predicting and anticipating N-pollution “hotspots”, where water quality is degraded due to nitrogen pollution from diffuse sources. There approaches, combined with field information on agri-environmental measures, might possibly be helpful to identify policies to improve the environmental performance of the agricultural sector. When tailored to the livestock sector, they could be valuable to articulate mechanisms for a more balanced distribution of livestock farms.

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Graphical abstract
Highlights

- Diffuse pollution and its impacts on ecosystems are underreported and underregulated
- Assimilative capacity of water bodies is a quantitatively useful approach
- Understanding the link between N pressures and impacts provides insight on solutions
- In Navarra, 64% of the N-related grey water footprint came from artificial fertilizers
- The grey water footprint assessment might help anticipate N-pollution hotspots