Spatial distribution and uncertainties of nitrogen budgets for agriculture in the Tagus river basin in Portugal – Implications for effectiveness of mitigation measures

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ABSTRACT

The present study describes a methodology to quantify the gross soil nitrogen balance (SNB) for agricultural land use in the Tagus Nitrate Vulnerable Zone (TVZ) between 1989 and 2016, including effects of implementation of the EC Nitrates Directive (ND, 91/676/EEC) since 2004. The study uses decadal information from National Agricultural Census at parish level and is supported by a Geographical Information System (GIS). The average SNB of the TVZ decreased significantly ($p < 0.001$) from $125\,\mathrm{kg\,ha^{-1}}$ in 1989 to $63\,\mathrm{kg\,ha^{-1}}$ in 2016 (on average $1.8\,\mathrm{kg\,ha^{-1}\,yr^{-1}}$). The N input via irrigation water was particularly high in seven municipalities where it represents up to 35% of the total inputs. Using a Monte Carlo approach, the average uncertainty of the SNB was calculated at 12%. The most important sources of uncertainty were mineral fertilization, nitrate concentration in groundwater used for irrigation and the percentage of farmers complying with ND measures. GIS analysis shows that, at the scale of municipalities, the SNB has decreased since the ND implementation in regions dominated by irrigated intensive crop production, while the SNB has stabilized in municipalities in the south of TVZ with intensive livestock production (75% of area). We conclude that the implementation of ND and national measures to reduce the SNB was not equally effective across the TVZ. Our results indicate that the most effective measures to reduce the SNB in the future are increasing compliance of farmers with the ND related measures and measures increasing the nitrogen use efficiency of livestock manures.

1. Introduction

Agricultural soils receive considerable amounts of mineral fertilizers and animal manures each year. An imbalance between these nitrogen (N) inputs and crop uptake will result in N surpluses and increased losses of N to different environmental compartments. Excess N in the environment affects soil, air and water quality, with negative impacts on climate, human health, and biodiversity (Sutton et al., 2011; Erisman et al., 2013).

Mitigation programmes and Directives have been implemented to reduce nitrogen loads to environmental compartments, namely to water bodies. A key strategic approach is the EU Water Framework Directive (WFD) (2000/60/EC) which aims to tackle pollution at source. With the objective of reducing water contamination through nitrates from agricultural sources, the European Community Nitrates Directive (ND)

(Council Directive, 1991/676/EEC) is one of the earliest pieces of EU legislation aimed at controlling pollution and improving water quality, thus supporting the aims of the WFD. The ND defines Vulnerable Zones as areas of land draining into ground and surface waters vulnerable to pollution from nitrogen compounds. These water bodies are identified making use of the following criteria: a) for surface freshwaters, in particular those used or intended for the abstraction of drinking water, if they contain or could contain, more than the concentration of nitrates laid down in accordance with Directive 75 /440/EEC (directive concerning the quality required of surface water intended for the abstraction of drinking water); b) for groundwater, if they contain or could contain more than $50\,\mathrm{mg\,L^{-1}}$ nitrates. The ND required that within two years following the notification of the directive, the Member States (MS) establish Codes of Good Agricultural Practices (CGAP) to be implemented by the farmers on a voluntary basis, and Action Programs

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(AP) with mandatory measures for their vulnerable zones. Within four years of the establishment of AP, measures initially prescribed in the CGAP became mandatory. The ND was transposed into Portuguese legislation by decree law 235/97. The most recent AP, which covers the nine existing Nitrate Vulnerable Zones, was established in Ordinance 259/2012, where a set of mitigation measures defined by official authorities must be implemented by farmers (Annex I). However, reports by the MS regarding the status of their water bodies show that the effects of the AP measures remain insufficient at the European scale (Bouraoui et al., 2011). Therefore, it is crucial to select more efficient nutrient management measures, by identifying the drivers of N imbalance in each case.

Nitrogen budgets (NB) facilitate the understanding of nutrient cycling in agro-ecosystems (Oenema et al., 2003) and are used to assess the efficacy of measures and policies regarding nutrient management and to identify surpluses. By accounting for the major N input and output fluxes (Cherry et al., 2012), a NB provides insight into links between agricultural N use and losses to the environment. Furthermore, as the NB can take account of current agricultural practice and be used to investigate the potential effectiveness of proposed mitigation measures, future trends can be predicted and the impact of management practices upon the ecosystems assessed (Alvarez et al., 2014), enabling improvement of N use efficiency (NUE) (Spiertz, 2009).

Different approaches can be used to calculate N budgets and related N surpluses, depending on available data and the purpose of the study, with farm gate, soil-surface and soil-system approaches reported in previous studies (e.g. Oenema et al., 2003; van Beek et al., 2003; Hoang and Alauddin, 2010). These approaches use different spatial boundaries of the agricultural system investigated. Whereas a farm-gate balance assesses the difference between N flows entering and leaving the farm, a soil surface balance quantifies the N flows entering the soil via the surface and leaving it via crop uptake (Oenema et al., 2003). The soil system approach also includes nutrients gains and losses within the soil (Hoang and Alauddin, 2010). These approaches can be applied at different scales, for example a farm gate or a soil balance, can also be calculated at national scale (rather than an individual farm or catchment), without having access to detailed individual farm and field data.

The data required for estimating a NB using the soil surface approach are simpler and less uncertain than those required for the soil system approach. The soil surface balance can produce a gross or a net N surplus, depending on whether gaseous N emissions to the atmosphere during fertilizer application are included in the balance. The gross N surplus is the amount of N not being used by the production system, representing the risk of N losses to the environment (Hansen et al., 2012). It is the sum of the N losses to the water bodies, to the air and includes N accumulation in soil. According to the European Environment Agency (EEA), N surpluses above 25 Kg N ha^{-1} carry a potential risk for aquifer contamination (EEA, 2018). However, this is a very generic and indicative value which does not take in account the specificity of different catchment properties or situations. Published studies using the soil surface balance include, among others, Alvarez et al. (2014) for a region of Argentina, Bechmann et al. (2014) for a small catchment in Nordic-Baltic countries, Poisvert et al. (2017) at the county (département) level in France, Jan et al. (2017) for 210 farms in Switzerland. De Girolamo et al. (2017) and Lassaletta et al. (2012) apply the soil system NB for Italian and Spanish watersheds, respectively. The farm gate NB is considered by Lord et al. (2002) and Macgregor and Warren (2016) in the UK.

N balances at country scale, where both extensive and intensive agriculture systems are present, may dilute the N surplus results (Reinhard and Velthof, 2015) by averaging between intensive and extensive regions. This can be an issue for meaningful results, depending of the purpose of the calculation. For example, if the objective is to determine potential loads to large rivers, a country scale evaluation may be adequate. On the other hand, if the objective is to finding hotspot of groundwater pollution, a cumulative value over a large and potentially heterogeneous area will not be adequate.

For N budgets in Mediterranean regions, intensive irrigated agriculture can play an important role for N surpluses (Arauzo et al., 2011), where the main source of irrigation is groundwater with high levels of nitrates, such as in a NVZ. Studies of N budgets in Mediterranean regions with strong influence of irrigation are scarce. Lassaletta et al. (2012) and De Girolamo et al. (2017) present two detailed studies at the watershed level. Nevertheless, the N inputs associated with irrigation were not quantified. Also, it was not an objective of these studies to relate N surpluses with changes in agricultural management practices imposed by the ND. According to van Grinsven et al. (2012), there are considerable differences in N surpluses and losses between countries and regions. There is also a lack of knowledge concerning the effects of spatial variation in N surplus at the watershed (Bartoli et al., 2012) and landscape levels (Drouet et al., 2012) and also at the individual NVZ level. Worrall et al. (2009) showed that 69% of the NVZs implemented in the EU during the 1990s showed no significant improvement in surface water nutrient concentrations, even after 15 years. Thus, they conclude that the control strategies need to be rethought, especially if the NVZs are small. On the other hand, Kristensen et al. (2016) emphasised the importance of any policy framework to be adopted at local socio-economic and physical conditions since the opportunity and ability for farmers to respond to mitigation initiatives and other policy instruments varies considerably. Thus, a localized and detailed analysis of NBs provides a useful basis for further studying the N dynamics and transfer between the different environmental compartments, for designing effective spatially targeted mitigation measures (Hansen et al., 2017; Richard et al., 2018).

For Portugal, N surplus statistics are available only at country scale, based on Eurostat estimates (Eurostat, 2016). Recently, Portugal has been involved in the International Nitrogen Management System (INMS, Sutton et al., 2016), in the UNECE Task Force on Reactive Nitrogen (http://www.clrtap-tfrn.org/), and in the H2020 Twinning project NitroPortugal coordinated by the University of Lisbon (grant number 692331). As a result of these collaborative opportunities, this study presents a methodology to quantify the N surpluses associated with agricultural practices as well as their evolution after the implementation of the EC Nitrates Directive. A case study is presented for the Tagus NVZ, located to the east and northeast of Lisbon, Portugal. The specific objectives were to (i) identify the main inputs of N in the NVZ, in particular to investigate the importance of N additions through groundwater used for irrigation; (ii) calculate time series of soil surface gross soil N surpluses (SNB) at a high spatial resolution (parish level) and quantify their uncertainties; (iii) identify statistically significant temporal trends of the N surpluses; (iv) analyse the spatial distribution of the N surpluses and identify critical areas and specific causes for any imbalances, and in particular to analyse intensively irrigated crop production *vs* livestock production areas. The results are discussed to assess the effectiveness of specific Action Program measures and the potential for improvement. In addition, some adjustments to current policies are proposed.

2. Materials and methods

2.1. Study area characterization

The Tagus Nitrate Vulnerable Zone (TVZ) is located in the catchment of the river Tagus in Portugal (Fig. 1a). A simplified N assessment of the Tagus catchment presented in Cordovil et al. (2018) concludes that the sustainability of the Tagus River Basin can only be guaranteed through reductions in input loads complemented by an effective transnational management process of water flows.

The initial designated TVZ area (Ordinance 1100/2004) comprised 19 000 ha but was subsequently extended (Ordinance 1433/2006) to 100 000 ha. Another extension (Ordinance 1366/2010) increased the TVZ to 241 686 ha (Fig. 1b). Currently the TVZ contains 60% of the
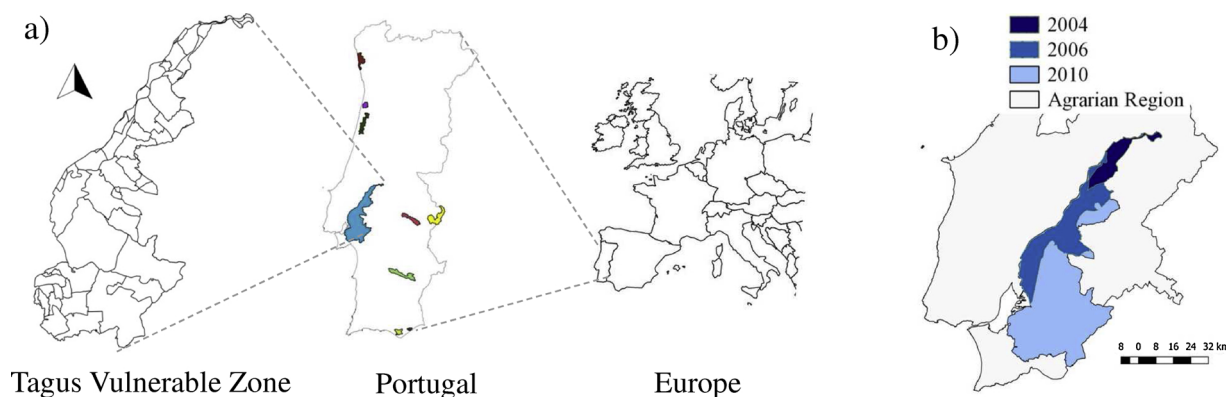
Fig. 1. a) Location of the Tagus Nitrate Vulnerable Zone in Portugal and Europe; b) Evolution of the Tagus Nitrate Vulnerable Zone area.

Table 1
Agriculture selected features in the TVZ compared with the national (Portuguese) and the EU-28 average, minimum and maximum values.

	TVZ	Portugal[*]	EU-28[*]	Minimum[*]	Maximum[*]
Average Farm area (ha)	37	12.	14	1 (Malta)	152 (Czech R.)
UAA (%)	52	47	46	9 (Finland)	76 (Denmark)
Irrigated area (% UAA)	60	13	6	0 (Est, IRL, Lux)	34 (Malta)

[*]Eurostat (2018).

total NVZ area in Portugal. It comprises 72 parishes organized into 20 municipalities. Agriculture accounts for the majority of land use and is of great economic importance in the area. The Utilized Agricultural Area (UAA) is on average 52%, slightly above the national and EU-28 averages (Table 1). Nevertheless, it varies between 6% and 83% among the municipalities in the TVZ. Irrigated land in the municipalities varies between 30 and 90% of the UAA, well above the national and the EU-28 average (Table 1). These figures highlight the importance of irrigated intensive agriculture in the study region. About 14% of the UAA is occupied by small farms with areas between 5 and 20 ha, 12% by farms with areas between 20 and 50 ha and 62% by farms larger than 50 ha. Table 1 shows that the average farm area is considerably larger in the TVZ, compared to the whole country, which allowed the implementation of mechanized agricultural practices and the intensive crop production in the region. The TVZ is part of the Agrarian Region of Ribatejo e Oeste (Fig. 1b). It was at the level of the Agrarian Region that data were available and obtained to complement the decennial data at the parish level, since the former are available at a higher frequency (once every 1–5 years, depending on the variable).

The agricultural systems of the region have become more intensive during the past decades and currently the most common crops are irrigated grain maize and horticulture for industrial processing (mostly tomatoes), with the rest of the area under vineyards, olive groves and permanent pastures. About 48% of the soils in the TVZ are of high and moderate permeability (sandy texture - Regosols and sandy loam texture - Haplic Podzols, FAO (2006)). The climate is Mediterranean with hot dry summers and mild wet winters, classified as Csa according to the Köppen system (Peel et al., 2007). Annual precipitation (30-year average) ranges from 697 mm in the north to 559 mm in the south of the TVZ. Long term daily mean and maximum air temperatures are 13 and 29 °C, respectively. About 90% of the landscape is flat and only in 6% of the territory there is a mild slope. The TVZ extends over a hydrogeologically complex area, comprising two layered aquifers separated by an aquitard, i.e., a geological layer with reduced permeability (Almeida et al., 2000). In the deeper semi-confined aquifer (5 to 50 m below soil surface) 17% of the wells from the national monitoring network (SNIR, 2017) record nitrate concentrations higher than

50 mg L^{-1}, but concentrations have also been slowly increasing in wells with low concentrations. Above the aquitard, in the upper aquifer (2 to 14 m below soil surface), 55% of the monitoring wells record nitrate concentrations higher than 50 mg L^{-1}. This is considered to be a greater threat than the lower aquifer due to its proximity to the soil surface which makes it react rapidly to agricultural management.

2.2. Data sources

The study covers a 27 years period, from 1989 to 2016. It was carried out using decadal information from the National Agricultural Census (INE, 2017) with detailed parish level data available for the years 1989, 1999 and 2009 (crop areas, irrigated areas and livestock populations). For the intermediate years 1995, 2005, 2013 and 2016, data were collected at the geographic level of the Agrarian Region and scaled down to the parish level. Crop yield data were collected at the level of the Agrarian Region (INE, 2017) for all the studied years.

Fertilizer sales data were considered the best available proxy for fertilizer use (Tomich et al., 2016; Poisvert et al., 2017). However, only overall country values (per unit of agricultural area) were available and collected (INE, 2017). Nitrate concentrations in irrigation water from groundwater and surface waters were taken from the National Water Resources Information System database (SNIR, 2017) and Nuncio (2013), respectively. Average irrigation volumes based on long time meteorological and crop data for the region were taken from Sousa and Morais (2011).

Geographical base data included the geographical boundaries of the parishes and municipalities as determined by the Portuguese Official Administrative Maps, CAOP (DGT (2017). The NVZ map and the water quality monitoring network data were made available by the National Environmental Information System (SNIAmb, 2017).

Several coefficients used in the calculations were extracted from local and European reports. Biological fixation of N by leguminous crops was estimated using coefficients found in LQARS (2006) and Baddeley et al. (2014). N input from crop residues left in and on the soil was estimated using information form the Rural Development National Agency DGADR (2017) and Agneessens et al. (2014).

Livestock excretion rates from the Intergovernmental Panel on Climate Change (IPCC, Dong et al., 2006) were used to estimate the N excreted by different livestock types. Country specific animal weights and emission factors from manure management obtained from the Portuguese Environment Agency report submitted under the UNECE convention (APA, 2016) were used to estimate ammonia emissions.

Data for the estimation of atmospheric N deposition were provided by the Co-operative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP, 2017) at a 50 × 50 km resolution. Several studies rely on this source, e.g. Lassaletta et al. (2012); Dalgaard et al. (2012); Andersen et al. (2016); Poisvert et al. (2017).

Table 2

Information regarding irrigation and N management from farms located in the Tagus Vulnerable Zone.

Farm #	Area (ha)	Soil texture	Crop	Irrigation		Nitrogen (kg ha^{-1})				Yield ton ha^{-1}	Reference
				Type	Amount (mm)	Fertilization	Irrigation water	Previous crops	Crop export		
1	1.0	sandy	Forage maize	Solid set	430	Mineral: 342 Organic: 140	0	58.2	283	18.5	Cameira et al. (2007)
2	1.0	silty loam	Grain maize	Level Basin	350	217	30.7	–	321	12.7	Cameira et al. (2007)
3	8.0	sandy loam	Grain maize	Pivot	605	231	20	104	268	10.0	Sousa et al. (2007)
4	51.0	loam	Forage maize	Pivot	438	187	45	35	245	na	Sousa et al. (2007)
5	3.6	sandy loam	Potato	Solid set	765	186	137	37	198	48	Sousa et al. (2007)
6		loam	Paddy rice	Basin	Na	120	(9.5 mg NO$_3$ L^{-1})	na	115	5.6	Figueiredo (2011)
7	8.0	loam	Tomato for processing	Drip	800	265	na	na	na	106	Damásio (2017)
8	35.0	silty loam	Tomato for processing	Drip	≈ 637	241	na	na	198	na	Data from 2018 not published

Na-not available.

Table 3

Maximum N application rates (total N) allowed in the Tagus Vulnerable Zone (Portuguese Action Program- Ordinance 83/2010).

Crop	N fertilizer application rate (kg N ha^{-1})	Crop	N fertilizer application rate (kg N ha^{-1})
Grain maize	300	Fresh fruit orchards	107
Winter cereals	162	Citrus trees	200
Paddy rice	180	Nut trees	127
Fodder crops	128	Olive orchards	120
Vegetables	147	Vineyards	110

In addition, data from in situ experiments developed at several farms located in the TVZ were used to validate some of the estimates (Table 2).

2.3. Gross soil nitrogen balance

The gross soil nitrogen balance (SNB) was calculated annually for representative groups of crops, at the highest spatial resolution available (parish level), using the soil surface balance method according with the OECD methodology (Oenema et al., 2003; Eurostat, 2016). The crop groups selected were grain maize, winter cereals, paddy rice, legume crops, vegetables for fresh consumption and processing, fruit trees, citrus trees, nut trees, olive trees and vineyards. The N surplus was calculated for the years 1989, 1995, 1999, 2005, 2009, 2013 and 2016. For the years 1995, 2005, 2013 and 2016, Eq. 1 was used to downscale from agrarian region to parish level (as explained in Section 2.2).

$$\text{Data P}_{(i)} = \frac{\text{Data AR}_{(i)}}{2} \cdot \left[\left(\frac{\text{Data P}}{\text{Data AR}} \right)_{(j)} + \left(\frac{\text{Data P}}{\text{Data AR}} \right)_{(j+1)} \right] \tag{1}$$

where Data P is the data for the parish, Data AR is the data for the agrarian region, (i) indicates periods for which there are no detailed data and (j) indicates the periods with detailed data (j < i < j + 1).

The balance equation (Eq. 2) was applied separately for each crop group in each parish to calculate the N surplus at the highest spatial resolution possible (all terms in kg ha^{-1} yr^{-1}).

$$N_{surplus} = MF + MN + IW + AD + BF + CR - EXP, \tag{2}$$

where MF is the N in mineral fertilizers, MN is the livestock manure N, IW is the N in the irrigation water, AD is the atmospheric N deposition, BF is the biological N fixation, CR is the crop residues N and EXP is the N uptake by the crops.

The N surplus (in kg (ha UAA)$^{-1}$) for each parish (N$_{SP}$) was calculated as shown in Eq. 3:

$$N_{SP} = \frac{\sum_{i=1}^{10} N_{surplus\,(i)}}{UAA_P} \tag{3}$$

where UAA$_P$ is the utilized agricultural area of the parish (ha) and (i) is the crop type.

Mineral fertilizer use at the parish level (MF, kg N yr^{-1}) was estimated based on the national trend, which was the best information available. Two periods were defined for this estimation, one before the designation of TVZ (pre-2004) when there were no limitations to fertilizer applications, nor other N-related rules the farmers were obliged to comply with, and the other period after the implementation of the NVZ Action Program. The estimation method started by considering that in the designation year (2004), the amounts of fertilizer applied were the maximum N application rates allowed in the TVZ for each crop group (Table 3), given that this is a highly productive area with intensive farming and irrigation. These values were extrapolated backward and forward in time, taking account of the national trend which reflects the fluctuation in fertilizer sales. For the period after 2004, the following additional considerations were taken into account as imposed by the AP (Annex I, measures 4, 5 and 11): (1) The amount of mineral N applied to crops and estimated as describe above, was corrected by deducting the N entering via irrigation water and the N provided by the crop residues left in the field after the harvest of previous crops; (2) The maximum amount of manure applied was limited to 170 kg N ha^{-1} and it was assumed that all the farmers complied with this rule. The Portuguese Action program contains the maximum amounts of manure from different animal sources that can be applied to comply with this limit; (3) crop N requirements were satisfied by taking account of livestock manures first, and then, if needed, topped up with mineral N fertilizers; (4) It was assumed that in 2004 only 25% of the farmers in the TVZ improved the crop N fertilization requirements by following the Action Program measures, although these were mandatory, and that this percentage gradually increased to 75% for the most recent year (2016). From informal discussions with farmers in the area, it is clear that compliance is not at 100%, but it is not possible to quantify this with data.

The manure derived N (MN, kg N yr^{-1}) (Velthof et al., 2015) was estimated by subtracting N losses due to NH$_3$ volatilization and NO$_3$-leaching in stables and manure storage facilities (LE$_{loss}$) from the annual N excretion (LE), i.e. pre-application to the soil surface. The calculations were carried out using the Tier 1 method from the IPCC guidelines for national greenhouse gas inventories (Dong et al., 2006). Calculations (Eqs. 4, 5) were based on the assumption that 85% of the livestock population was housed year-round. It was assumed that the N produced in each parish was equally spread over the UAA of the parish, as farmers share their manure locally.

$$LE = \sum_i \left(N_{(i)} \cdot N_{rate(i)} \cdot \frac{TAM_{(i)}}{1000} \cdot 365 \right) \quad (4)$$

where LE is the N in the livestock excretion, N$_{(i)}$ is the number of animals per livestock type, N$_{rate\ (i)}$ is the IPCC Tier 1 rate of N excreted by each livestock type (kg N (1000 kg animal.day)$^{-1}$, TAM$_{(i)}$ is the country specific average animal mass, and (i) is the livestock type (cattle, pigs, sheep, goats and poultry).

The amount of N lost during manure storage was estimated as:

$$LE_{loss} = \sum_j \sum_i [LE_{(i,j)}(EF_{NH3(i,j)} + EF_{Lx(i,j)})] \quad (5)$$

where (j) is the manure management system (liquid or solid), (i) is the livestock type, EF$_{NH3}$ is the volatilisation factor (dim), and EF$_{Lx}$ is the leaching factor (dim);

The excretion coefficients and emission factors which depend on the livestock type and on the manure management system used are shown in Table 4 and are default values from Dong et al. (2006). For the leaching factor, an average value of 0.1 was used. Only for TAM, country specific data was used (APA, 2016).

The N supplied via irrigation (IW, kg N yr^{-1}) may be an important balance input in a NVZ where most of the irrigation relies on groundwater with high concentrations of NO$_3$-. In most of the TVZ about 80% of the irrigation water comes from groundwater, except for the southern part where this percentage decreases to 60%. For the estimation of this input (Eq. 6), long term annual irrigation volumes per crop were used together with the N-NO$_3$ concentrations in water from the wells located in each parish or, if that was the case, from surface waters (reservoirs/dams).

$$N_{Irrig(i)} = Irrig_{(i)} \cdot A_{C(i)} \cdot [N - NO_3] \cdot 10^{-6} \quad (6)$$

where I$_{rrig(i)}$ is the average irrigation volume (L m^{-2}) applied per year to the crop i, A$_{C(i)}$ is the area (m^2) occupied by crop i in the parish, and [N-NO$_3$] is the nitrate N concentration (mg L^{-1}) in the wells or surface waters.

The calculations of atmospheric N deposition (AD, kg N yr^{-1}) were carried out by applying the N deposition rates provided by EMEP for the 50 x 50 km grid cells covering the TVZ, for the areas occupied by the different crops.

Biological N fixation by leguminous crops (BF, kg N yr^{-1}) was

Table 4
Coefficients related to livestock N excretion.

Livestock	TAM* kg	N$_{rate}$ # kg N (1000 kg d)$^{-1}$	EF$_{NH3}$ (dim)## Liquid	Solid	Frac$_{MS}$ (dim)+ Liquid	Solid
Dairy cattle	550	0.48	0.40	0.30	0.35	0.65
Other cattle	400	0.33	0.40	0.35	0.35	0.65
Pigs	120	0.465	0.48	0.45	0.91	0.09
Poultry	3.5	0.835	###	0.45	0	1
Sheep	50	0.85	###	0.19	0	1
Goats	25	1.28	###	0.19	0	1
Horses	307	0.26	###	0.19	0	1

* average animal mass, # N excretion rate, ##NH$_3$ volatilization rate, + fraction corresponding to each manure management system, ###not applicable.

Table 5
Nitrogen amounts left in the soil by the previous crop: DGADR (2017); Agneesen et al., 2014.

Current Crop	Previous crop	Nitrogen in previous crop residues
Vegetables	Vegetables	30 kg ha^{-1}
Grain maize	Winter cereal	1.5 kg of N/t green mass incorporated
Grain maize	Legumes	2.5 kg of N/t green mass incorporated
Winter cereal	Grain Maize	1.5 kg of N/t green mass incorporated
Winter cereal	Legumes	2.5 kg of N/t green mass incorporated

estimated using typical average values of 10.1, 165 and 133 kg N ha^{-1} for temporary pastures, permanent pastures and grain legume crops, respectively (LQARS, 2006; Baddeley et al., 2014).

After the harvest of crops such as cereals and vegetables, considerable amounts of stalks are left on the field, in some cases containing high concentrations of nitrogen in their tissues (Agneessens et al., 2014). To estimate the amount of N left on fields in the form of crop residues (CR, kg N yr^{-1}), the most common crop sequences in the TVZ were identified, and the data presented in Table 5 for previous crops were considered as inputs for the current crops.

The export of N (EXP, kg N yr^{-1}), i.e. the N uptake by crops, was estimated from the statistics of crop productivity (INE, 2017) and the N concentration in the plants at the time of harvest provided in the literature. This method was possible for maize, paddy rice, and tomatoes for industrial processing and potatoes (Sousa et al., 2007; Cameira et al., 2007 and others presented in Table 2). For the remaining crops, no data were available regarding N content at the time of harvest. Thus, N export was calculated by Eq. 7:

$$N_{exp} = P_{act} \cdot \left(\frac{N_{upt}}{P_{ref}} \right) \quad (7)$$

where P$_{act}$ is the actual crop productivity (INE, 2017), and N$_{upt}$ is the N removal by crops for the reference productivity (P$_{ref}$) (LQARS, 2006).

The assumptions and simplifications necessary to apply the described methodology can be sources of uncertainty for the N surplus and these are summarized below:

- Spatial data for fertilizer practices for each crop type do not exist, so national trends were used and downscaled for the TVZ;
- The N in the manure produced in one parish was assumed to be used in that parish in the first instance. When the maximum allowed application rates were reached, the manure was distributed first to other parishes of the same municipality and then to the neighboring municipalities;
- The adherence of farmers to the measures proscribed in the Action Program is unknown. Therefore, expert judgement was used to estimate this adherence;
- For some crops no coefficients are available to convert the production into N content. The method used was to combine scientific research, where available, with expert judgement. Few experimental data exist to validate these coefficients;
- N deposition estimates taken form the EMEP represent average values for the 50 x 50 km grid and may underestimate atmospheric N input in municipalities with a high livestock density or intensive crop fertilization (Dalgaard et al., 2012), and overestimate for more extensive pastures. This is largely due to ammonia deposition decreasing rapidly away from source areas, i.e. within a few km (e.g. Dragosits et al., 2002, 2006; Vogt et al., 2013);
- It is difficult to estimate the N export by grassland/pastures. It was assumed that the N content in unfertilised grassland is roughly equal to the N content of the excreta of the grazing animals. A similar approach was used by Lord et al. (2002);
- Most of the coefficients used in the estimation of the manure N applied to the crops are dependent upon the livestock management (namely feeding practices) which was not considered in this study

due to lack of information.

The N surplus data (in kg N yr^{-1}) calculated by parish were then aggregated to the municipality (N_{SM}) level for further analysis, and then calculated per unit area for the UAA (Eq. 8):

$$N_{SM} = \frac{\sum_{j=1}^{72} N_{SP(j)} \cdot UAA_{P(j)}}{UAA_M} \tag{8}$$

where UAA_P and UAA_M are the utilized agricultural areas of the parishes and the municipalities, respectively, obtained from the official statistics (INE,2017), and (j) is the parish number.

To assess the spatial distribution of the N balance inputs, outputs and N surplus across the TVZ, the base data and the calculation procedures of the gross soil N balance were integrated in a Geographical Information System (QGIS). The different balance terms of gross soil N balance, obtained for the different years considered, were spatially distributed at the municipality level.

2.4. Statistical methods

2.4.1. Uncertainty and sensitivity analysis

For the deterministic model used to quantify the N soil surface balance (Eqs. 2–6), the key uncertainties affecting N surplus outputs result from the uncertainly in the input data and parameters. To evaluate the uncertainty in each component of the balance and in the average N surplus of the TVZ, a simple Monte Carlo simulation analysis was carried out, using a Latin Hypercube sampling strategy (Marino et al., 2008). For each studied year and municipality, 1000 simulation runs of the model were carried out, assuming independent variables and parameters. A Latin Hypercube sample of standard uniform distributions was initially generated and then transformed to the statistical distributions assigned to each of the input variables/parameters (Table 6). Data collected from national surveys and other sources (as described in Section 2.2) provided the average values, while their relative uncertainty (given by the coefficients of variation, CV) was estimated based on published literature (e.g., Dong et al., 2006; Gu et al., 2017; Poisvert et al., 2017) and authors expert judgment. Variables where data were derived from national census statistics have the lowest CVs, while variables and parameters obtained from international publications were ascribed the highest CVs.

To identify the most critical input factors (variables and parameters) of the model, a global sensitivity analysis (SA) was also performed. The extended version of the Fourier Amplitude Sensitivity Test (eFAST) proposed by Saltelli et al. (1999) was used. This method, based on the decomposition of the variance of the output, allows the computation of two sensitivity indices for each factor: the first order or main effect index (S_{xi}) and the total effect index (S_{Txi}). Both indices are normalized by the variance of the output, providing therefore scaled sensitivity measures in [0,1]. The maxima and minima of the 92 uncertain input factors used in the SA were established based on their statistical distributions and respective parameters (Table 6).

2.4.2. Trend and correlation analyses

Nonparametric techniques were used to detect monotonic trends in the longitudinal N surplus data and to estimate the intensity of association between N surplus and other variables. Bartels' rank test of randomness (Bartels, 1982) was applied to analyse the significance of a trend in a sequence of observations. Bartels' test is the rank version of the von Neumann's ratio test (von Neumann, 1941) where the null hypothesis of randomness (independence) can be tested against the existence of a trend (Mateus and Caeiro, 2014). Due to the small size of the samples, the exact distribution of the statistics and the exact probability values (p-value) of the test could be calculated. Whenever a significant trend was detected, the slope of the trend line was estimated by the Theil–Sen's method (Theil, 1950; Sen, 1968) and the intercept by the estimator proposed by Hettmansperger et al. (1997) (Hollander et al., 2014).

A similar approach was followed to analyse the association between N surplus and the amount of mineral or organic N applied in each year. After sorting one of the variables, Bartels' test was applied to the other variable. Whenever randomness was rejected, Spearman's rho (ρ, rank correlation coefficient) was calculated to determine the signal and strength of correlation between the two variables. This methodology avoids the problems of non-normal data, heteroscedasticity and serial correlation that could affect the reliability of the results of other statistical tests. All the analyses were carried out in R (R Core and Team, 2018) with RStudio software (RStudio Team, 2018) and using the packages 'lhs' (Carnell, 2018), 'sensitivity' (Pujol et al., 2018) and 'randtests' (Mateus and Caeiro, 2014).
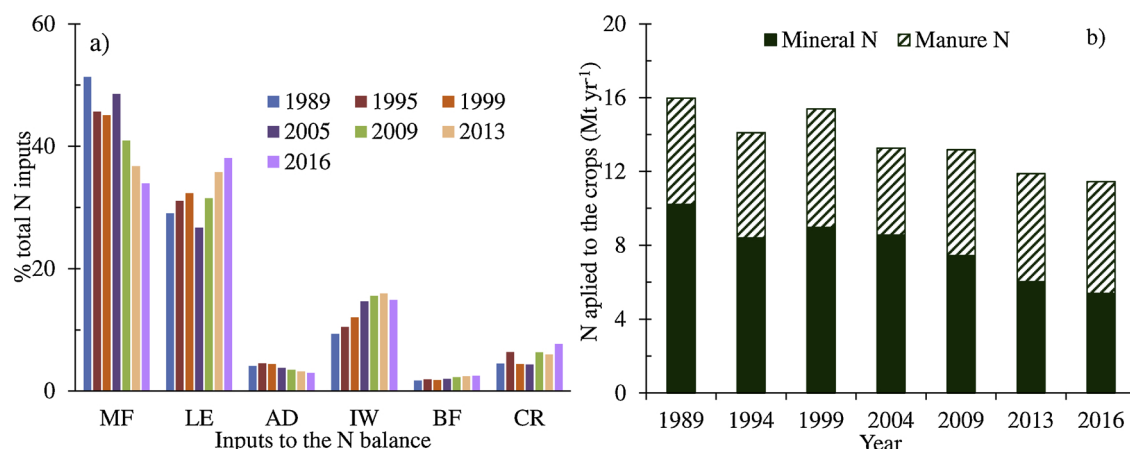
Table 6
Variables and parameters used in the gross soil N surplus balance equation and statistical distribution and coefficient of variation (CV) used in the uncertainty analysis.

Variable/Parameter description	Units	Distribution	CV (%)
Crops areas	ha	Truncated Normal	5
Crops productivity	kg ha^{-1}	Normal	5
Trend in fertilizer sales	–	Uniform	30
% farmers complying with ND	–	Uniform	15
Animal population	–	Truncated Normal	5
Average animal mass	kg	Truncated Normal	15
N excretion rate for animals	kg N (1000 animal day)$^{-1}$	Normal	30
Manure solid fraction	–	Normal	15
Manure volatilization rate (solid fraction)	–	Normal	30
Manure volatilization rate (liquid)	–	Normal	30
Manure leaching coefficient	–	Normal	30
Atmospheric N deposition rate	kg N ha^{-1} yr^{-1}	Truncated Normal	30
Average irrigation depth	mm yr^{-1}	Normal	30
Fraction of irrigation water from GW	–	Uniform	10
Groundwater nitrate concentration	mg L^{-1}	Truncated Normal	25
Surface water nitrate concentration	mg L^{-1}	Normal	25
Symbiotic N fixation rate	kg N ha^{-1} yr^{-1}	Truncated Normal	30
N left in the soil by previous crops	kg N ha^{-1} yr^{-1}	Normal	25
Fate of straw from previous crops	–	Beta	50,1 [a]
Crops reference productivity	kg N ha^{-1} yr^{-1}	Normal	5
Crops N export rate	kg N ha^{-1} yr^{-1}	Normal	5

[a] Used shape parameters of the Beta distribution.

Fig. 2. a) Contribution of the different sources to the N balance; b) Evolution of mineral and organic livestock fertilizers use in the Tagus Vulnerable Zone (MF-mineral fertilizers, MN – manure N production, AD - atmospheric deposition, IW - irrigation water, BF - biological fixation and CR - crop residues).

3. Results and discussion

3.1. Gross soil nitrogen balance for the Tagus nitrate vulnerable zone

3.1.1. Nitrogen inputs

Fig. 2a shows the contribution of the N inputs to the SNB. Fertilizers (mineral and organic) represent the major N flow for the study period, averaging at 75% of total N inflows. Until 2009, the inputs through mineral fertilization were larger than those from livestock manure (Fig. 2b), although showing a declining trend. The reduction in the mineral fertilizer inputs between 1989 and 2005 reflects the decrease in fertilizer sales registered at country level. In 2005, mineral fertilizer inputs show an increase, again following the country level trend. After 2005, with the designated NVZ area becoming progressively larger (as described in Section 2.1), mineral fertilization started to decrease where the livestock production was high. This was probably due to the limits imposed by the Action Program to the maximum total N that can be applied to each crop, and the preference of farmers for applying livestock excreta. Overall, the amount of mineral fertilizers as N input decreased by 34% from 1989 to 2016. N inputs through mineral fertilizers have also decreased during this period in some European countries, e.g. Croatia (50%), Italy (30%) and Greece (25%), while for other countries they have increased. This was the case of Bulgaria (133%), Latvia (4%) and Austria (58%). At the EU-28 level there was an increase from 67.4 kg ha^{-1} in 2006 to 74.4 kg ha^{-1} in 2016 (Eurostat, 2018).

The changes in mineral fertilizer use were influenced by the utilized agricultural area (UAA), its occupation by the different crops, and economic and legal constraints such as the ones introduced by the Common Agricultural Policy (CAP) or imposed by the Nitrates Directive and the National Action Plan from 2004. Fig. 3a) shows UAA occupation during the years under analysis. The area of arable crops decreased between 1999 and 2004, mainly due to the CAP set aside policy and quotas that limited production. As a result, the area occupied by permanent pastures increased. Following the country trend, arable land had increased again by in 2009, in the TVZ, due to the rise in the market values of the agricultural products, leading to land use change, from set aside to cereal production. While annual crops overall showed just a small increase for the study period, the percentage of winter cereals decreased considerably while grain maize and vegetable production, mainly for industry, increased (Fig. 3b). The latter two are annual crops with high N input. Permanent crop areas, which are low N input crops, slightly decreased between 1989 and 1999 and then remained fairly steady. However, after 2004, mineral fertilizer use decreased due to the NVZ designation and the implementation of the mitigation measures from the Action Program which the farmers increasingly adhered to.

Table 7 compares the agricultural land use in the TVZ, Portugal, the EU-28 and the EU countries with maximum and minimums values. In the TVZ, the percentage of UAA occupied by arable land is higher than the country average, which is particularly due to grain maize and vegetable crops. On the other hand, the high shares of arable land presented by some European countries, which are reflected in the EU-28 average, are occupied essentially by rainfed cereals.

The overall tendency in the TVZ during the study period was for an increase of the irrigated area (by 16%) which is a good indicator of N consumption, since irrigated crops are the most demanding in terms of
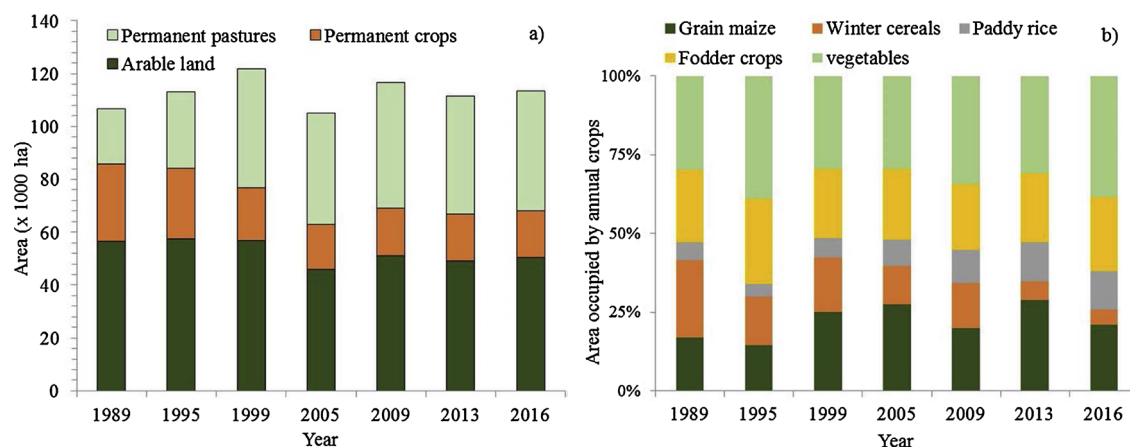

Fig. 3. a) Utilized agricultural area (UAA) by main crop types; b) Evolution of the area occupied by main annual crops in the Tagus Vulnerable Zone.

Table 7
Forms of agricultural land use in the TVZ, Portugal and the EU-28 (data refers to 2016).

Land use (% UAA)	TVZ	Portugal	EU-28[*]	Minimum[*]	Maximum[*]
Arable land	44	30	60	21 (Ireland)	98 (Finland)
Permanent Pastures	40	49	32	0 (Malta)	80 (Ireland)
Permanent crops	16	20	6	0.2 (UK, FIN, S, IRL)	25 (Cyprus)

[*] Eurostat (2018).

nutrients.

Manure N almost matched mineral fertilizer application in 2013 and even exceeded it in 2016, (Fig. 2a), with total values of 6.1 and 5.9 Mt yr^{-1} respectively (Fig. 2b). This corresponded to shares of 36 and 39% of the total N inputs, respectively. This increase was associated with the increase in livestock production between 2005 and 2016 (Fig. 4a), in particular of cattle (46%), pigs (38%) and poultry (70%). At the EU level, there was a decrease in cattle and pig populations between 2005 and 2016 (Eurostat, 2018). As shown in Fig. 5, only 10 countries presented an increase in the cattle population and only 6 countries had an increase in their pig population. Nationally, for the same period, Portugal had increases of 9 and 11% for the total number of cattle and pigs, respectively.

Fig. 4b shows the trends of manure N applied to the TVZ cropland. The amounts of N lost during livestock housing and storage (by type of livestock) were estimated to be 3.7, 5.1, 5.1, and 5.4 Mt for the four studied years between 2005 and 2016. It was assumed that all N remaining in the manure after losses by volatilisation and/or leaching during livestock housing and manure storage was applied to croplands.

The third most important N input flow (12%) corresponds to N contained in irrigation water (Fig. 2a). It is important to note that this inflow was highly variable within the TVZ (Fig. 6), with seven municipalities showing values above the average for the TVZ since 2005 (20.3 kg ha^{-1}). This input is particularly high for three municipalities where it represents between 30 and 35% of the total N inputs. As it can be seen in Fig. 6, these municipalities are located in the northern part of the TVZ, and are spatially associated with the distribution of irrigated crops, in particular grain maize and vegetables. Atmospheric deposition (AD) represented, on average, 4% of total inputs, while biological fixation (BF) and crop residues accounted for 2 and 5%, respectively. Lassaletta et al. (2012) showed that for another Mediterranean watershed (Ebro River Basin, NE Spain), AD represented 7% of total inputs. De Girolamo et al. (2017) present contributions of 9 and 6% for BF and AD in another Mediterranean catchment (Celone River Basin, South Italy). However, neither of these studies account for N inputs through irrigation water.

3.1.2. Nitrogen output

Fig. 7a shows that the N export by crop production increased on average during the study period, except from 1989 to1995, and 1999 to 2005. These variations are likely due to changes in the area occupied by the crops, but also, particularly in 2005, due to adverse weather conditions (severe drought) that probably influenced crop productivity and thus N uptake. Fig. 7a also shows that the increase in crop productivity and the more rational application of N fertilizers contributed to an overall improvement in N use efficiency (NUE). Irrigation with groundwater containing considerable amounts of nitrates recycles part of the leached NO_3^- and can be contributing for the overall TVZ NUE increase. The use of improved crop varieties, the increase in irrigation efficiency (which decreased leaching losses), and new technologies associated with fertilizers (e.g. slow release N fertilizers), may also have contributed to the NUE improvement. Similar increases in NUE were also reported for Denmark, by Dalgaard et al. (2014), associated with NVZ designations.

N uptake varied among crops, with the dominant share of total N export relating to grain maize and vegetables for processing (Fig. 7b). Since 1999 these crops represent more than 50% of the total removal, reaching values close to 70% for the last two periods of the study. Permanent crops (fruit trees, olives groves and vineyards) present a much lower share, remaining fairly constant. By contrast, the removal of nitrogen by winter cereals decreased considerably due to the decrease in the occupied area.

3.1.3. Nitrogen surplus

Fig. 8 shows the evolution of the gross soil N surplus between 1989 and 2016 calculated for the different territory units (municipalities) within the TVZ. The average N surplus represents the area weighted average of the different municipalities' values calculated in this work, while the country values were estimated by Eurostat (Eurostat, 2016; not available for all study years).

Bartels' statistical test showed a highly significant decreasing trend (p < 0.001) of the N surplus at the level of the TVZ for the study period (encompassing 27 years). The slope of the trend line indicates a decrease of 1.8 kg N per hectare of UAA and per year. These results may be affected by the fact that a downscaling procedure was used to obtain data for some years of the study period. However, when these years are removed from the trend analysis the average decrease in the N surplus is even higher (slope = - 2.3). It is interesting to note that, similar to several other EU countries (e.g. Belgium, Denmark, Germany, Greece, Italy, Netherlands) there is a considerable decrease of N surplus prior to the implementation of the Nitrates Directive (Baraui et al., 2011). This can be associated with the previously mentioned increase in the price of
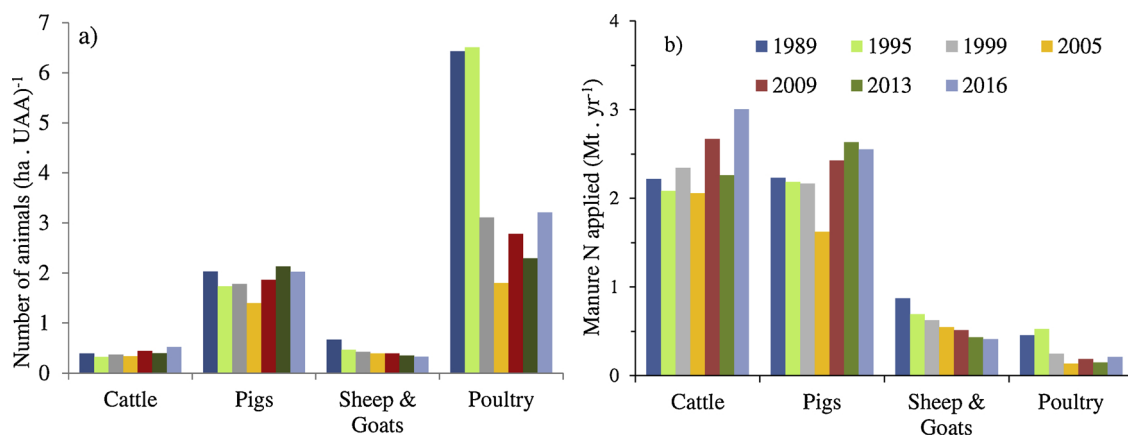

Fig. 4. a) Livestock population per unit of utilized agricultural area (UAA) in the Tagus Vulnerable Zone; b) manure N applied to cropland (excluding losses during storage and management).
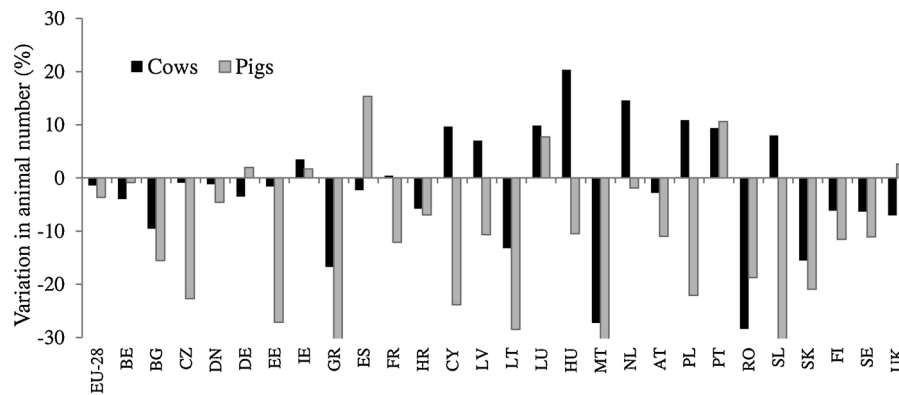
Fig. 5. Variation in the number of cows and pigs in the EU-28 countries, between 2005 and 2016 (Eurostat, 2018).

mineral fertilizers and the set aside policies. The increase of NUE due to use of improved crop varieties can also be a cause for this, although the increase of NUE can also be an effect of the decrease in N inputs. The most important decrease (26%) in N surplus occurred between 1989 and 1999 and relates to the reduction in fertilizer sales at country level. After 2005, N surplus continued to decrease but at a lower rate. During this period, livestock populations (in particular cattle, pigs and poultry) started to increase (Fig. 4a), which is reflected in the N surplus. However, there are considerable variations in the N surplus among municipalities due to local differences.

Erisman et al. (2011) and Eurostat (2016) report surpluses for Portugal per unit of UAA and per year of 42 kg, which is substantially lower than the 64 kg calculated in the present study for the TVZ in 2016. The overall value presented by Eurostat encompasses a great diversity of agricultural systems, with a high representativeness of extensive systems with reduced fertilizer inputs. Therefore, the higher N surplus in intensive systems that occupy smaller areas (such as the TVZ) becomes diluted. However, for countries such as the Netherlands and Denmark (Dalgaard et al., 2014), where NVZ rules have been implemented over the entire country area, show values of N surplus with magnitudes similar to those presented in this work for the TVZ. Hansen et al. (2012) report N surplus reductions of 30–55% in Denmark, with a national average value of 117 kg N ha^{-1} in 2007. Lord et al. (2002) present an overall N surplus of 115 kg ha^{-1} for the 12,000,000 ha of UK agricultural land, using the farm gate method. De Girolamo et al. (2017) present a value of 60 kg ha^{-1} year^{-1} for a Mediterranean catchment in Italy (for the 2010–2011 period), for productive land where the mineral fertilization is the main source of N (72%).

These results may indicate that ND measures can have a positive impact on the reduction of N surplus in very different countries, when transposed to those countries through their specific Action Programs, suitable for their policies and practices.

Table 8 summarizes the descriptive statistics for the N surplus results for the TVZ, where the averages and the standard deviations were weighted by the areas of the municipalities.

The reported average values are associated with a considerable variation expressed by maximum and minimum values, standard deviation and coefficient of variation. Lassaletta et al. (2012) also reported such variability for their Spanish watershed. Lord et al. (2002) reported N surpluses for the UK varying between 0 and 200 kg ha^{-1}.

3.2. Spatial analysis of the N surplus within the TVZ

From absolute values and the trend analysis of N surplus, it can be concluded that agriculture pressure on diffuse N losses decreased over the studied period but remains high in the TVZ relative to the 25 kg ha^{-1} EEA reference, which means that there may still be an aquifer pollution potential. Thus, the spatial distribution of the N surplus was analysed to determine local issues and try to identify causes for N budget imbalances.

Figs. 9 and 10 show livestock N excretion produced per municipality and the percentage of the UAA which is irrigated, respectively. These are two important drivers for N surplus, since the first constitutes an inexpensive N source and the latter reflects the production of high N demand crops. While irrigation is more prevalent in the northern and central regions of the TVZ, as expressed by a higher percentage of UAA under irrigation, livestock production is concentrated in the southern part.
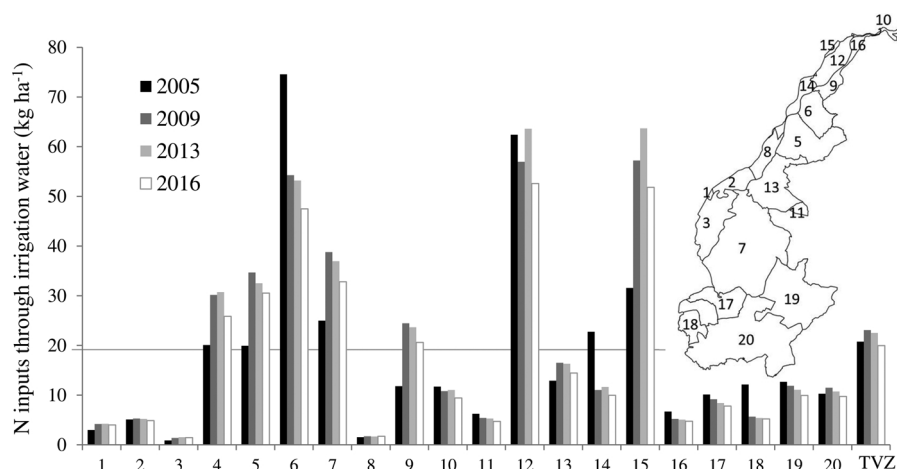

Fig. 6. N inputs through irrigation water for each municipality in the Tagus Vulnerable Zone (TVZ). The horizontal line represents the average value for the TVZ since 2005. The x axis shows the code of the municipalities presented in the map.
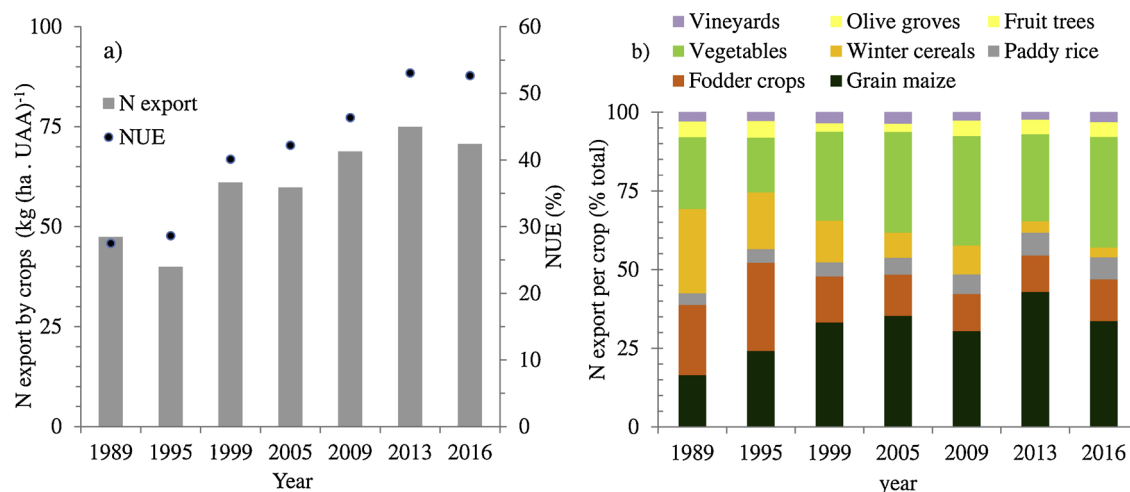
Fig. 7. a) N export rate by the crops grown in the Tagus Vulnerable Zone (uptake) and N use efficiency (NUE); b) contribution of each crop group to the overall N export.

Fig. 11 shows the spatial distribution of N surplus and its temporal dynamics. N surplus was initially high across the TVZ, and showed a wide spatial variation. In the northern part, characterized by a high proportion of irrigated agriculture (grain corn and vegetables mainly supported by mineral fertilization) and medium livestock densities, the intensification of the crop production seemed to be efficiently controlled in terms of N inputs. Indeed, the regions more dependent on mineral fertilises showed a higher decrease in N surplus during the first period, due to constraints in fertilizer purchase, and after 2005, due to the restrictions imposed by the Nitrates Directive and the Action Program regarding the maximum amount of total N that can be applied to each crop. In 2016, average N surplus for the TVZ as a whole was around 50 kg N ha^{-1}, with higher values in the southern part, where livestock agriculture dominates. Although there was a general decrease over time, more recently a stabilization between 100 and 150 kg N ha^{-1} was reached in this part of the TVZ. Four hotspot municipalities, with an N surplus above the average for the TVZ remain (Benavente – code 7, Moita -code 18, Montijo – code 19 and Palmela- code 20), located in the central and southern part of the TVZ, and making up 57% of the TVZ area.

3.2.1. Trend and correlation analysis

Despite the highly significant decreasing trend ($p < 0.001$) of the average N surplus for the TVZ as a whole, in 40% of the territory (9 municipalities) the trend was not significant ($p > 0.05$), instead indicating stabilization for the study period. On the other hand, Fig. 12

Table 8
Descriptive statistics for the N surplus calculated for the Tagus Vulnerable Zone.

Statistics	1989	1994	1999	2004	2009	2013	2016
Av$_W$ (kg N (ha UAA)$^{-1}$)	126	100	92	83	80	67	64
Max (kg N (ha UAA)$^{-1}$)	206	144	175	169	149	95	94
Min (kg N (ha UAA)$^{-1}$)	78	39	45	47	24	21	23
Med (kg N (ha UAA)$^{-1}$)	105	94	81	83	76	62	59
StD$_W$ (kg N (ha UAA)$^{-1}$)	35	19	36	18	34	24	22
CV (dim.)	0.28	0.18	0.38	0.22	0.43	0.36	0.34

Av$_W$ is the weighted average, StD$_W$ is the weighted standard deviation, Med is the median value, Max and Min are the maximum and minimum values, respectively, and CV is the coefficient of variation.

shows that after TVZ designation, more than 75% of the territory did not present a significant (decreasing) trend. Almost 90% of this area is located in the southern part of TVZ:

Bartels' test showed that, although the average N surplus for the entire period presented a significant ($p = 0.026$) positive correlation with mineral N fertilizer use, in 87% of the area the decrease in the N surplus is not correlated with the amount of mineral N applied. On the other hand, for the entire study period, the average TVZ N surplus was not significantly ($p = 0.165$) correlated with the N applied through livestock excreta, although for 27% of the TVZ area this correlation was significant ($p < 0.05$) and positive ($p > 0.85$).

The previous analysis shows that the results for the TVZ as a whole
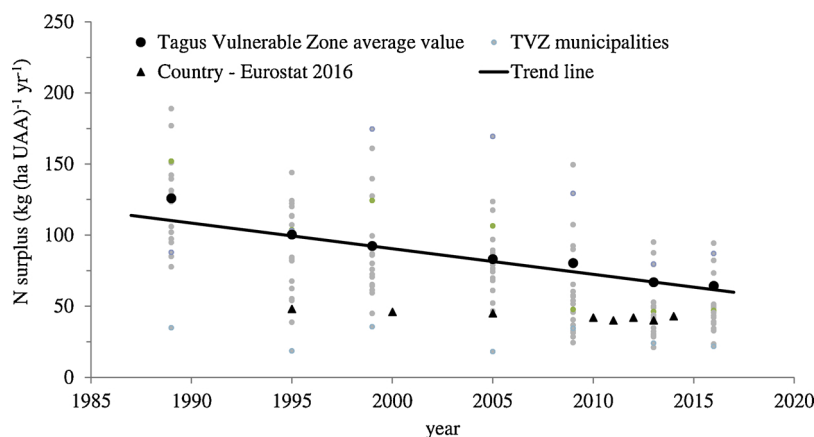

Fig. 8. Evolution of the annual agricultural N surplus in the Tagus Vulnerable Zone and of the overall values for Portugal from Eurostat (2016). Trend line equation: y = 3686.6-1.8 x.
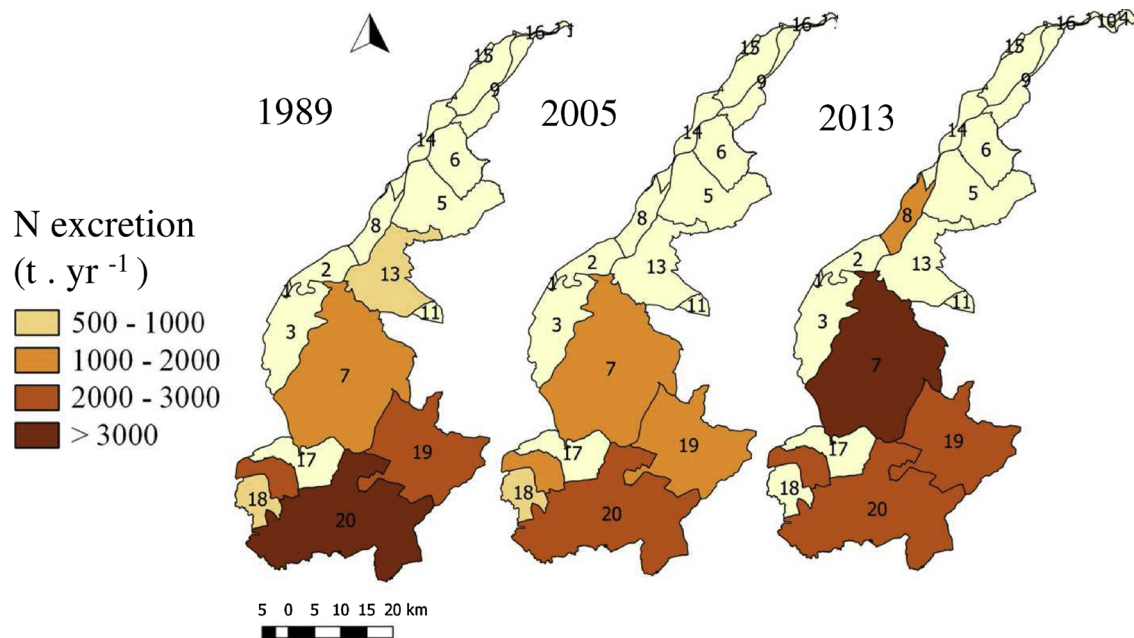
Fig. 9. N in the livestock excretion produced in each municipality of the Tagus Vulnerable Zone.

are misleading and can induce inadequate conclusions regarding fertilizer related drivers for the N surplus. In fact, different spatial patterns were detected within the TVZ associated with crop, livestock and irrigation intensity. Seven municipalities stand out, where irrigation water contributes 30% of the inputs for the N surplus. In these municipalities the irrigation water contribution has been decreasing due to the improvement in irrigation system efficiency (on average from 69 to 86%). On the other hand, there are municipalities where the different N input types contributed more evenly to the high N surplus.

3.3. Uncertainties in the calculation of the gross soil N balance

The TZV overall uncertainty associated with the N surplus

estimates, for all the years after ND implementations determined by the Monte Carlo approach is 12%. When estimated for each municipality, the uncertainty varied from 12 to 62% between municipalities. Fig. 13 shows the boxplots for the key N balance components and for the 1000 simulations performed using the Monte Carlo approach described in Section 2.4. For N inputs via irrigation water (IW), the uncertainty can override the possible differences between years. Nevertheless, for mineral fertilization (MF) and N export by crops (EXP), uncertainty analyses confirm the differences discussed in Sections 3.1.1. and 3.1.2. Also, for MN (manure N) and IW, the outliers (open circles in Fig. 13) are predominantly high values. The (average) values used to calculate MN and IW may lead to their underestimation, and consequently to the underestimation of the average N surplus for the TVZ.
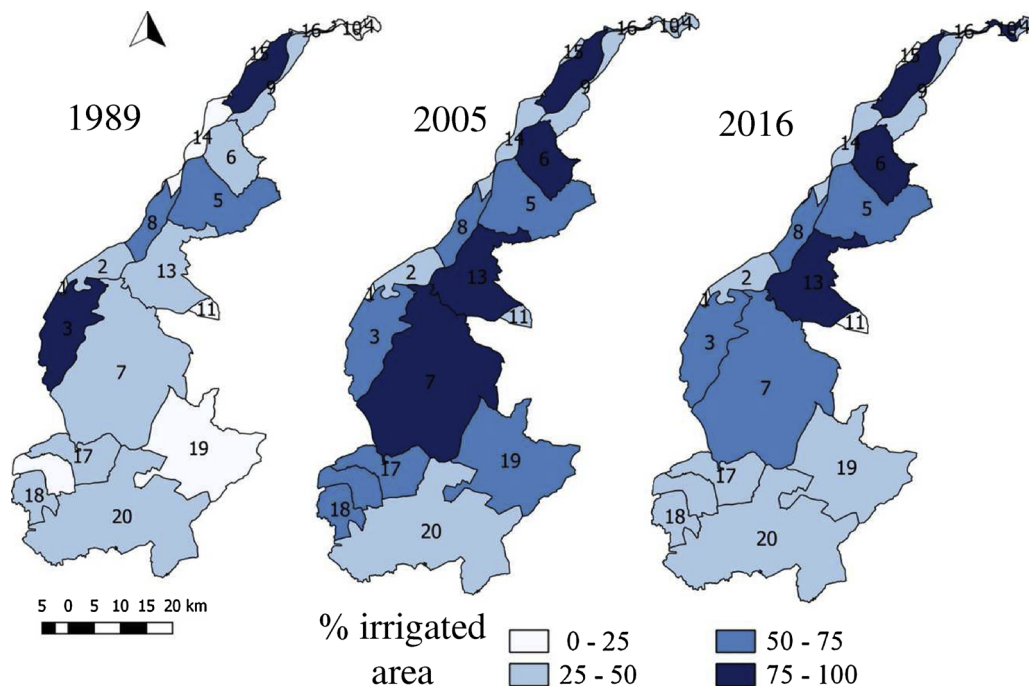

Fig. 10. Proportion of irrigated areas per municipality of the Tagus Vulnerable Zone (% of utilized agricultural area, UAA).
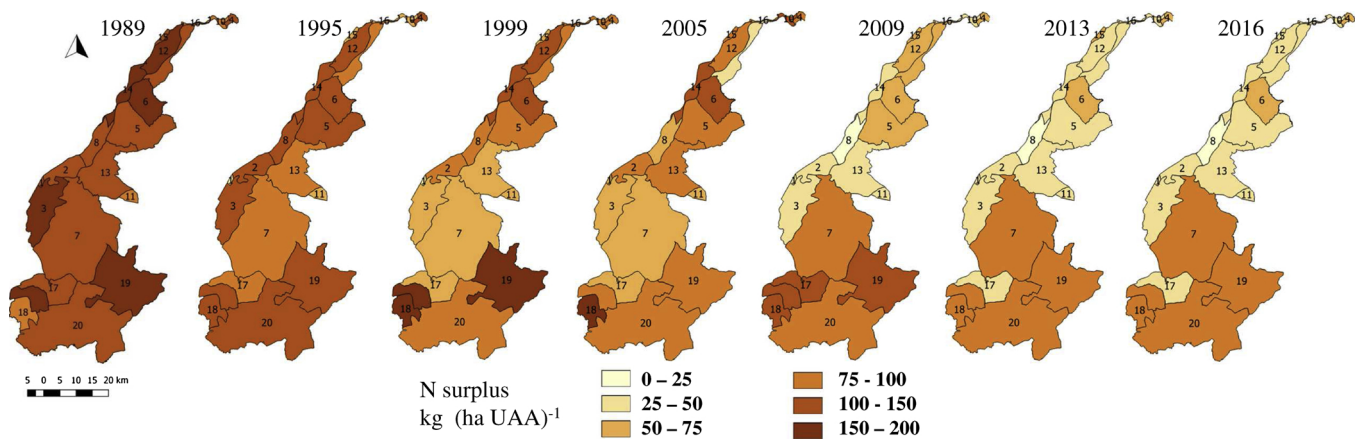
Fig. 11. Spatial distribution of the N surplus within the Tagus Vulnerable Zone for the study period.1989–2016.

Fig. 14 shows, for the model used in this study, the expected reduction in the variance of the calculated N surplus if each individual input factor (Fi) could be exactly known (main effect), and if all input factors but Fi could be known with precision (interactions) (Saltelli et al., 2004). Results show that the two more relevant factors influencing model results are the mineral fertilizer sales (F1) and the nitrate concentrations in groundwater used for irrigation (F2). About 39% of the variability in N surplus can be reduced if mineral fertilizer sales are accurately quantified. A similar percentage (36%) was found for nitrate concentrations in groundwater used for irrigation. The latter value relates to the fact that this factor influences two major inputs of the N budget, IW and MF. For the particular case of the TVZ, where 80% of the irrigation relies on groundwater with high nitrate concentrations (and in some municipalities 90% of the UAA is irrigated), the importance of this factor is very high regarding model sensitivity. This reinforces the specificity of N management in intensive, irrigated crop production systems. The other factors influencing the calculated N surplus variability, and requiring careful quantification, are the percentage of farmers that are complying with measures related to the ND (8%) (Section 2.3), the N excretion rate for animals (5 and 3% for pigs and cattle, respectively), and crop productivity, particularly for maize (2%). The importance of this last factor relates to the N export (crop uptake) estimations. The other factors listed in Table 6 which do not appear in Fig. 14 were found to contribute very little to the N surplus variability.

3.4. Environmental management and policies

Previous studies demonstrated that ND measures (Annex I) are only partially successful in terms of impact on groundwater quality (e.g. Oenema, 2009). As shown in the present work, different regions of the TVZ produce different dynamics within a soil surface N budget. In areas
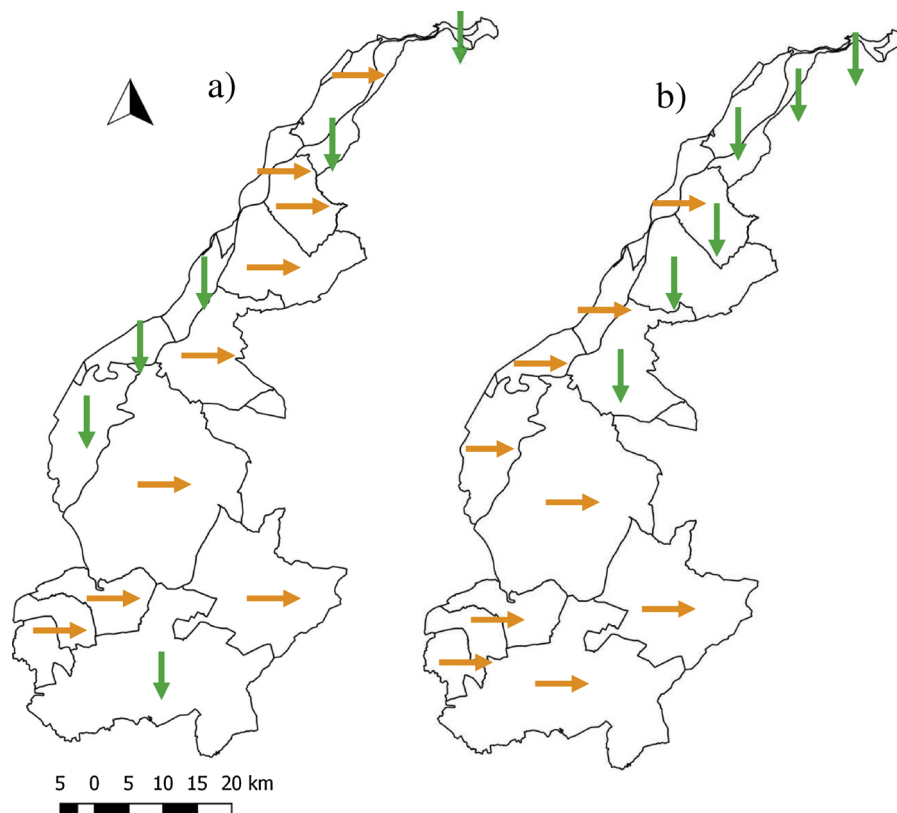

Fig. 12. Statistical trends in the N surplus for the periods: a) 1989–2005 and b) 2005–2016 (→stabilization, ↓decrease).
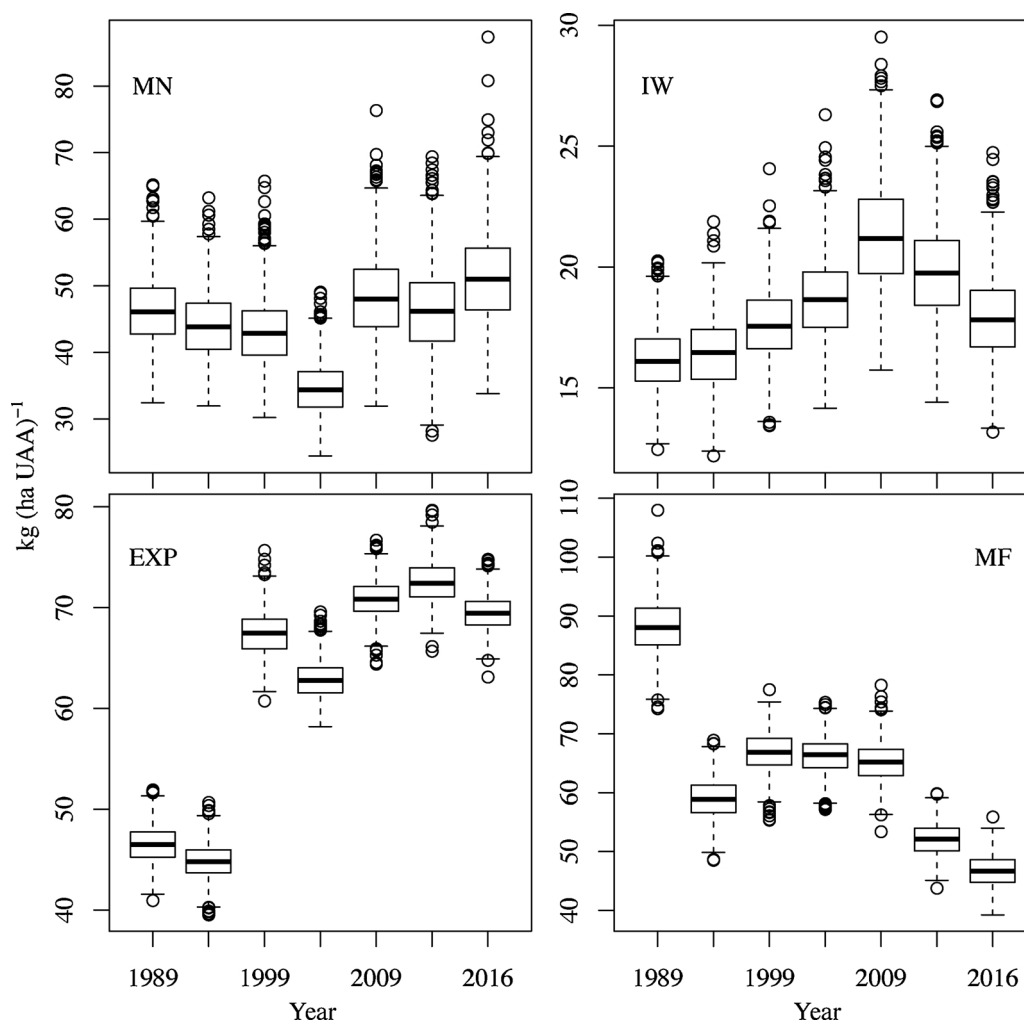
Fig. 13. Uncertainties in the calculation of the main N balance components according to the Monte Carlo analysis: Manure N (MN), irrigation water (IW), N export by crops (EXP), mineral fertilizer (MF). The circles represent the outlier values.
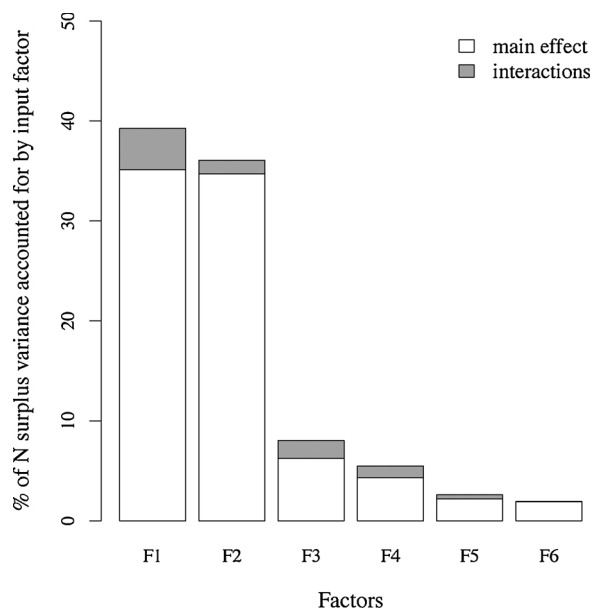

Fig. 14. Main factors influencing N surplus variance (F1 – Fertilization sales, F2 – Nitrates in groundwater used for irrigation, F3 – Percentage of farmers that comply with the ND/AP measures, F4 – N excretion coefficient for pigs, F5 – N excretion coefficient for cattle, F6 – Maize crop productivity).

where intensive irrigated crop production dominates, a balanced fertilization accounting for inputs from other sources in addition to fertilizers (e.g. N from previous crop residues, N in irrigation water) seems to be the most appropriate mitigation measure. Furthermore, using irrigation water from an aquifer contaminated with considerable amounts of nitrates will contribute to clean the aquifer if the fertilization plan takes account of this input. Therefore, the group of farmers whose main activity is cash crop production (maize, vegetables including processing tomatoes, potatoes etc.), for whom the ND/AP measures are suitable for their farm management activities, can optimally implement these measures to reduce the pressure on groundwater quality.

In some municipalities, manure N production is the most important N input and, in some cases, there is no room (or need) for mineral fertilization, with enough N available from manures. N surplus is invariably higher where the livestock production is more intensive, even if it is assumed that all the farmers comply with the mandatory maximum application rate of 170 kg N ha^{-1} for organic N. For the group of farmers whose main activity is livestock production, the implementation of mandatory good manure management practices may be necessary. The Action Program should take into account the specificity of their activity, namely the production of high amounts of manure and slurry that need to be managed. In the future, increasing mineral N fertilizer prices may provide an additional economic incentive to reduce their use and thus to increase the NUE of organic N from livestock excreta (van Grinsven et al., 2012). Focusing in reducing N inputs (tackling at source), specific approaches that may work in this context

would be the creation of sufficient manure storage capacity to store the excess N excreted using low-emission techniques to minimise volatilization and leaching losses. Composting facilities to produce composts with higher available C/N and anaerobic digesters for treatment of livestock excreta are also possibilities. Nevertheless, spatial modelling (at field and catchment scales) is fundamental to support the above, to assess the environmental and economic consequences of the different options.

In both livestock and crop farming systems there are farmers that do not (yet) understand the rationale behind mitigation measures and the reasons for their adoption (which often include cost savings if nutrients are properly accounted for and applied at optimal times for crop growth). For those farmers an action learning program would be very important. Others remain unconvinced about the balance between farm management and environmental benefits (Barnes et al., 2011; Buckley, 2012). For this group, economic supporting measures would be an incentive for taking action. In fact, regulation and mitigation options to reduce the N surplus are not always compatible with practices of farm management (Richard et al., 2018). From the policy and societal point of view, farm advice needs to be coordinated and consistent with the Action Program mitigation measures. Individual compliance with the Action Program will foster both farm and public benefits. As pointed out by Smith et al. (2017), a major challenge is that this is needed from the highest levels of governance and across the regional and local government. According to the same authors, the strategic use of payment schemes may be an efficient way to incentivise the adoption of suitable measures.

However, agriculture is not the only source of nitrogen in the TVZ. For situations were no relationship was found among the studied factors, other sources may be of importance, such as urban and the industrial pressures should also be considered and investigated (which was not possible as part of this study).

4. Conclusions

A soil surface gross nitrogen balance (SNB) was calculated for the Tagus Nitrate Vulnerable Zone for the period between 1989 and 2016. The annual N surpluses were obtained for 72 parishes and then aggregated and analysed at the municipality level for 7 "snapshot" years. N inputs and outputs were modelled using a spatial/GIS database and their uncertainty was assessed using a Monte Carlo approach. The following conclusions can be drawn from the results obtained:

(i) Fertilizers (mineral and organic) are a major N flow in the TVZ, representing on average 75% of total N inputs. Mineral fertilizers have a higher share than manure N production for the early years, however, the situation was reversed in the later years. A more moderate N input flow is provided by N contained in irrigation water (taken mainly from nitrate-rich groundwater aquifers). However, N from irrigation water represented on average 75 kg ha^{-1} year^{-1} in one municipality with intensively irrigated crop

production. The remaining N balance inputs (atmospheric deposition, biological fixation and residues from the previous crop) together are estimated to add 11% to N inputs.

(ii) The average N surplus was calculated for the study period with an uncertainty of 12%. The main factors of uncertainty in the SNB requiring a more accurate quantification are mineral fertilization, nitrate content in groundwater used for irrigation and the percentage of farmers compliance to ND measures;

(iii) A statistically significant decrease of the average N surplus in the TVZ was found over the entire study period (1989–2016), and 75% of the TVZ area seems to have reached a stabilization level over the last 10 years, leading to the conclusion that diffuse agricultural N pressure decreased but remained high.

(iv) Results indicate that the implementation of the Action Program measures is not equally effective throughout the TVZ. This confirms that national scale pressures cannot be used to identify and estimate the N loads in each specific NVZ with the objective of identifying mitigation measures. The spatial variability of the N surplus seems to be closely related with the specialization of the agricultural production in terms of crops/livestock production within the NVZ. Intensive pig, cattle and poultry farming in the southern part of the TVZ can result in very large local N surplus, reflecting the difficulty in using manure-derived N efficiently. Thus, for these areas, results identify opportunities for improved manure management, namely the improvement of manure N availability.

The spatially explicit method presented in this study was effective for evaluating mitigation measures and their potentials, and to identify N emission hotspots. The results support the need for ensuring that farmers are complying with ND/AP related measures, and the need for improved measures with respect to efficient use of livestock N excreta, with minimal losses to the environment.

The results obtained can also help water management stakeholders to implement the ND/AP measures and to evaluate the suitability of those measures across the diverse range of agricultural management practices. Nevertheless, the uncertainties associated with the method should be reduced by complementing with other assessment methods, including more detailed N source assessment, *in situ* measurements and information and process modelling. Following on from this initial study, further mechanistic and comprehensive analysis of N flows, nutrient balances and losses are required to identify further areas of improvement and to increase nitrogen use efficiency.

Acknowledgments

Annex I – Compulsory measures defined in the Nitrates Directive and its transposition to the Portuguese case (Action Program-ordinance 83/2010)

Nitrates Directive	Portuguese Action Program*
	Following the ND the following measures are mandatory
Definition of periods when the land application of fertilizer is inappropriate;	(1) Application of chemical and organic fertilizers in forbidden between Nov 1st and Feb 15th.
Guarantee of sufficient storage capacity for livestock effluents;	(2) The storage capacity for livestock effluents must be calculated for a period of 120 days, if no alternative system is proven to exist.
Definition of land limitations for application of fertilizers;	(3) Application of fertilizers is forbidden on steeply sloping ground; flooded ground; near water courses, groundwater abstraction wells and water reservoirs;
Calculation of balanced fertilizations considering inputs other than fertilizers;	(4) Calculation of fertilizer application rates must include crop requirements and N in soil at planting, carried by irrigation water, from crop residues mineralization, in livestock effluent and chemical fertilizers;
Amount of manure N application is limited to 170 kg N ha^{-1} year^{-1};	(5) Amount of manure N that can be applied is limited to 170 kg N ha^{-1} year^{-1};

Measures during storage of manure effluents	(6) Prevention of water pollution by run-off and seepage into groundwater and surface water by waterproofing of pavements and installing drainage systems separate from rainwater;
Application of chemical fertilizers and livestock effluents	(7) Chemical fertilizers must be applied at an appropriate rate and uniformity of spreading.
	(8) Injection into soil of liquid effluents (slurry), soil incorporation immediately after application of the solid effluents (manure);
Land use management-related practices	(9) Use of crop rotation systems, maintenance of a minimum quantity of vegetation cover during (rainy) periods;
Prevention of water pollution from run-off and the downward water movement beyond the reach of crop roots in irrigation systems.	(10) Correct water management must be ensured, mainly through the selection of adequate irrigation methods/systems for given soil and climate; appropriate timing of irrigation;
	Additional mandatory measures in the AP, as foreseen in ND
	(11) Maximum amount of total N that can be applied to each crop (Table 3 in the text).

*More details in (ordinance 259/2012)

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