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ORIGINAL ARTICLE

Annual agricultural N surplus in France over a 70-year period

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Abstract High levels of nitrogen (N) contamination of ground and surface water are still detected at European and national scales, despite the implementation of Directives, highlighting the need to improve understanding of changes in N pressure. Soil surface nitrogen balance was investigated at the county level in France over a 70-year period to identify areas with high N surpluses and trends in N pressure. Soil surface nitrogen balances were calculated for 90 NUTS3 (Nomenclature of Territorial Units for Statistics in the EU) called 'departments' (ranging from 611 to 10,145 km^2, median surface area 6032 km^2) and one NUTS2 entity. Over the whole period, the N surplus calculated for France as a whole averaged 37 kgN per ha of utilized agricultural area (UAA) and departmental N surpluses mean ranged from 10 to 86 kgN ha UAA^{-1}. Imprecision, i.e. an 80% confidence interval in N surpluses, was calculated using Monte Carlo simulation. Average imprecision for the whole period ranged from 6 to 45 kgN ha UAA^{-1} across different departments. Analysis revealed that yearly and departmental imprecision values were mainly correlated with N export ($R^2 = 0.46$). Despite this imprecision, the soil surface nitrogen balance was found to be a consistent and suitable tool to determine trends in N pressure at the department level. The model revealed an upward trend in N surplus until the 1990s for 82% of the area studied, and a downward or stable trend for more than 90% of the area since the European Nitrates Directive has been implemented.

Keywords Soil surface balance · Nitrogen · Trend analysis · Uncertainty · Surplus

Introduction

Concentrations of nitrogen compounds observed in European surface waters are higher than the reported natural values (EEA 2001). This has led to eutrophication of coastal waters and degradation of continental water in terms of quality for drinking water production. Mitigation programs and Directives have been implemented specifically to reduce nitrogen inputs, a key factor to limit eutrophication in coastal areas (Leip et al. 2011). However, the effects of these Directives regarding nitrogen concentrations remain insufficient at the European scale (Bouraoui and Grizzetti 2011). At the country scale, for example in France, river

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basins such as those of the Loire (Minaudo et al. 2015) and the Seine (Passy et al. 2013) still present signs of eutrophication. Many authors have highlighted that the response of river basins to mitigation measures is delayed for several years to decades due to long solute transfer time through soil and groundwater systems and catchment buffering (Cherry et al. 2008; Fovet et al. 2015; Ma and Yamanaka 2016). Agricultural intensification has led to an increase in nutrient inputs such as chemical and organic fertilizers. Since the 1960s, agricultural systems have constituted the main diffuse source of nitrogen in water bodies (Aquilina et al. 2012; Heathwaite et al. 1996; Öborn et al. 2003; Oenema et al. 2003) and are currently considered to be the main source of nitrogen delivered to European seas (Bouraoui and Grizzetti 2014). Long-term quantification of diffuse N pressure from agricultural systems is only available at the country scale for France (Bouraoui and Grizzetti 2011). Currently changes in N pressure at a smaller scale over the long term are not available. Furthermore, national diffuse N pressure cannot be used to identify and estimate major diffuse pollution in specific areas despite the latter was a recommendation of the European Union Water Framework Directive (WFD, 2000/60/CE).

N balances are based on the 'conservation of matter' principle and to construct them requires combining the individual N processes (Meisinger et al. 2008). As a whole, N balance is a useful tool to improve understanding of N flux at a regional scale (Galloway et al. 2004; Sutton et al. 2011). The difference between N input and output is called the N surplus and can be used to estimate N pressure (EEA 2001). This approach is common and has been used widely (Alvarez et al. 2014; Asmala et al. 2011; Salo and Turtola 2006). Depending on how the limits of the agro-system are defined, there are different types of nitrogen balance. (1) The *Farm-gate balance* considers the system as a whole farm, including cropland, grassland and livestock. The surplus defined in this type of balance does not distinguish between losses from the soil and from animal systems. (2) The *Soil system budget* considers only the total N pool of the soil itself and requires a quantification of every single N input and output flux, such as leaching, runoff, denitrification, export of N with harvested crops and variations in soil nitrogen stocks, resulting in a high degree of uncertainty due to the lack and poor quality of data available (Öborn et al. 2003). (3) The *Soil surface balance* considers that the inputs consist of the nitrogen entering the soil through fertilizer and manure application, symbiotic N_2 fixation and atmospheric deposition, and that the output is the export of N through harvested crops (Oenema et al. 2003), thus the N surplus corresponds to the nitrogen entering the soil but unused by crops. This nitrogen surplus can be stored in the pool of soil organic matter or can be lost from the soil through runoff, volatilisation, denitrification or leaching. The amount of N prone to leaching can contribute notably to N contamination of aquifers and rivers. Soil surface balances can be applied at various levels from plot to national scales (Cherry et al. 2008). At the smallest administrative scale, models using farm records can lead to spatially accurate results. These models use agricultural census data (Alvarez et al. 2014; EEA 2001) which provide details in terms of both space (Table 1) and time. For example, the French reference model NOPOLU (SoeS 2013) estimates diffuse N source emission with a statistical model based on the soil surface balance principle that allows a spatialized surplus to be calculated at the Nomenclature of Territorial Units for Statistics (NUTS4) level, which is a fine scale (Schoumans and Silgram 2003). However, this model mainly uses datasets that are not available every year: agricultural census data, land cover information and results of national surveys. At larger scales, other models can be used with countrywide or regional data (Table 1), which do not enable spatial nitrogen pressure to be clearly understood over a whole drainage basin.

The aim of this paper was threefold: (1) to calculate soil surface N balances at a subnational scale focusing on estimation uncertainties, (2) to identify trends in diffuse N pressure and (3) to investigate the robustness of these trends despite uncertainties.

To determine diffuse N pressure originating from agricultural systems over more than half a century, we calculated surpluses using a soil surface balance using statistical data available at the NUTS3 level, a seldom used spatial resolution, corresponding to the French administrative departments, and Corsica (NUTS2). We estimated the associated uncertainty range based on model reliability using relevant information about current knowledge of N fluxes and available data (Uusitalo et al. 2015). We applied this method and quantified surpluses with their uncertainty for 90 departments and one NUTS2 entity in France from 1940 to 2010.

Table 1 Soil surface balance models used in France, including their spatial resolution, data sources and year of application

Model/Name	Sources	Spatial resolution (ha)	Scale of database used		Mineral fertilizer	Year of application
			Yield	Area		
NOPOLU	(1)	$\sim 170\text{–}6.4 \times 10^{7}$	(NUTS2[a])	(LAU1)	National surveys	2002, 2004, 2007, 2010
Adapted from NOPOLU	(2)	$\sim 1.5 \times 10^{7}$	(NUTS2[a])	(LAU1)	Scenarios	2001
BASCULE	(3)	2–620	Field	Field	Farm record	1992
MITERRA	(4)	$0.03\text{–}54 \times 10^{6}$	National	(NUTS2)	FAO	2000
INTEGRATOR	(5)		National	NCU	FAO	1970–2030
IMAGE	(6)		National	Country	FAO	1970–2030
IDEAg (CAPRI + DNDC)	(7)		Regional	HSMU	FAO	2002

Scales are given following the EUROSTAT classification, NUTS3 is an equivalent of a department and LAU1 of municipality. Sources in the table refer to: (1) SoeS (2013), (2) EEA (2001), (3) Benoît (1992), (4) Velthof et al. (2009) cited by deVries et al. (2011), (5) deVries et al. (2011), (6) Bouwman et al. (2005), (7) Leip et al. (2008)

HSMU Homogeneous Spatial Mapping Units

[a] *NCU*, nitroEurope Calculation Units. Units refer to clusters of 1 km^2 grid units that are characterized by a similar environment and/or farming condition

Materials and methods

Surpluses were calculated annually between 1940 and 2010 for 91 geographic entities: 90 French metropolitan departments and one NUTS 2 entity: Corsica. Paris and the neighbouring departments were not included in this study because each represented <0.01% of the French utilized agricultural area (UAA) (SSP, *Service de la Statistique et de la prospective*, 1940 to 2010).

Balancing methods

Nitrogen surpluses were determined using a soil surface balance method (Oenema et al. 2003). For each department, the soil surface balance quantifies N input such as manure and chemical fertilizers, atmospheric deposition and symbiotic fixation, and N output represented by harvested crops, including fruit, vegetables and grazing. All units are in kgN ha UAA^{-1} $year^{-1}$.

N input was calculated using Eq. (1):

$$NI = N_{Fix} + N_{Air} + N_{Min} + N_{Man} \tag{1}$$

where NI is the total nitrogen entering the soil, N_{Fix} is the symbiotic fixation of N_2, N_{Air} is the atmospheric deposition of nitrogen, N_{Min} and N_{Man} represent the nitrogen available for plants from chemical and manure respectively.

N input from manure was calculated according to Eq. (2). It was determined from the estimated excretion rates of livestock, taking into account N loss through processes such as denitrification and volatilisation to calculate the amount of nitrogen that actually entered the soil.

$$N_{Man} = \sum_{i=1}^{n} \sum_{j=1}^{m} Nb_j * E_j * C_i \Big/ A_{UAA} \tag{2}$$

In Eq. (2), *i* is the livestock type (cattle, sheep,…), *j* the livestock class (dairy cow, bull, lamb, duck…), Nb_j the annual population of animals in each livestock class, E_j excretion per individual animal of that class (kgN $head^{-1}$ $year^{-1}$) (Table 2), and A_{UAA} the utilized agricultural area (ha) per department. N excretion was adjusted for losses through volatilization of ammonia, denitrification and N_2 loss for each animal class, multiplying the total nitrogen in livestock excretion by a coefficient C_i. C_i refers to N in livestock excretion that was not lost to the atmosphere i.e. $C_i = 1$—value in Table 3.

N volatilisation for each type of N chemical fertilizer was taken into account in accordance with EMEP-Corinair cited in CORPEN (*Comité d'ORientation pour des Pratiques agricoles respectueuses de l'ENvironnement*) (2006) based on SoeS report (2013) resulting in a calculation of N input from chemical fertilizers using Eq. (3):

$$N_{Min} = \sum_{j=1}^{m} F_j * K_j \Big/ A_{UAA} \tag{3}$$

Table 2 N excretion per livestock category (kg N $head^{-1}$ $year^{-1}$)

Livestock category	Mean	Min.	Max.	Source	Factors taken into account
Bovine animal					
Animal over 2 years old					
Dairy cow	111.6	72.4	161.3	(1)	Milk yield (4000–10000 kg/an); diet (harvested herbage—hay and grass silage-, grass, corn silage)
Suckler cow + calf	79.5	47.3	125.0	(2)	Animal size (600–740 kg); diet (harvested herbage—hay and grass silage-, grass, corn silage)
Plough oxen	101.6	62.6	147.4	(2)	Animal size 900 kg ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Dairy heifer	50.8	31.3	73.7	(2)	Animal size 450 kg ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Replacement heifer for suckler cow	62.1	38.3	90.1	(2)	Animal size 550 kg ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Slaughter heifer	56.5	31.3	90.1	(2)	Calculated following results for dairy heifer and replacement heifer
Cull cow	24.2	13.8	37.6	(2)	Diet (grass silage or corn silage); fattening duration (2–4 month)
Fattening steer	80.0	54.6	106.5	(2)	Animal size 650 kg ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Bull	101.6	62.6	147.4	(2)	Animal size 900 kg ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Animal between 1 and 2 years old					
Dairy heifer	45.2	27.8	65.5	(2)	Animal size 400 ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage) (harvested herbage - hay and grass silage-, grass, corn silage)
Replacement heifer for suckler cow	50.8	31.3	73.7	(2)	Animal size 450 ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Slaughter heifer	48.0	27.8	73.7	(2)	Calculated following results for dairy heifer and replacement heifer
Fattening steer (Male)	67.7	46.2	90.1	(2)	Animal size 550 ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Bull	56.5	34.8	81.9	(2)	Animal size 500 ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Animal under 1 year of age					
Veal calf	7.2	5.8	8.6	(2)	Fed with milk powder
Other animals	23.2	7.0	39.0	(2)	Animal size 250 kg ± 20% and 300 ± 20%; diet (harvested herbage—hay and grass silage-, grass, corn silage)
Sheep					
Ewe-ram	14.3	11.4	17.1	(3)	
Lamb	5.7	4.3	7.1	(3)	
Goat					
Goat (more than 1 year)	14.3	11.4	17.1	(4)	
Kid (under 1 year of age)	5.7	4.3	7.1	(4)	
Horse					
Horse, Donkey, Mule	56.0	26.0	73.0	(5)	
Poultry					
Cock and hen	0.1430	0.0220	0.0680	(6)	
Duck	0.1853	0.1110	0.2960	(6)	

Table 2 continued

Livestock category	Mean	Min.	Max.	Source	Factors taken into account
Turkey	0.3107	0.1430	0.5730	(6)	
Goose	0.4187	0.1770	0.6710	(6)	
Guinea fowl	0.1460	0.0870	0.2590	(6)	
Quail	0.0255	0.0250	0.0260	(6)	
Pigeon	0.8270	0.6616	0.9924	(6)	
Pig					
Young pig (20–50 kg)	0.59	0.56	0.62	(7)	Simple or bi-phase feeding
Sow (more than 50 kg)	22.5	20.4	24.6	(7)	Simple or bi-phase feeding
Boar (more than 50 kg)	4.2	3.8	4.6	(7)	Simple or bi-phase feeding
Fattening Pigs	4.2	3.8	4.6	(7)	Simple or bi-phase feeding
Rabbit					
Adult	3.3	1.9	4.6	(5)	
Young	0.06	0.05	0.08	(5)	

Sources in the table refer to: (1) Corpen (1999), (2) Corpen (2001), (3) Corpen (1988), (4) Circular DERF/SDAGER/C2002-3013, (5) JOFR, 2011, (6) Corpen (2006), (7) Corpen (2003)

Hypotheses for factors which affect N excretion such as animal size or diet are mainly based on Soes (2013)

where F_j is the amount of each type of chemical fertilizer delivered per department and K_j the fraction of N provided through chemical fertilizer that was not lost to the atmosphere.

N input through plant symbiotic fixation was calculated in accordance with Anglade et al. (2015)(Eq. 4).

$$N_{Fix} = \sum_{crop\_fix} \left(\left[\alpha_{crop\_fix} * \frac{Y_{crop\_fix}}{NHI} + \beta_{crop\_fix} \right] * BGN * A_{crop\_fix} \right) / A_{UAA} \quad (4)$$

where α_{crop_fix} and β_{crop_fix} coefficients depend on culture type, Y_{crop_fix} and A_{crop_fix} the harvested yield (kgN ha^{-1} year^{-1}), and area (ha) covered by each crop capable of fixing N_2 NHI is the N harvest index and BGN a multiplicative factor to take into account belowground contributions. Leguminous plants can be grown in mixed cultures. According to SoeS (2013), the proportion of leguminous plants was set at 0.15 for permanent pasture and 0.3 for temporary grassland.

N export (N_{Exp}) is the sum of N export for each crop (Eq. 5).

$$N_{Exp} = \sum_{crops} \left(Pdt_{crop} * N_{crop} \right) \Big/ A_{UAA} \quad (5)$$

where Pdt_{crop} is the crop yield (ton),and N_{crop} the N content (kgN ton^{-1}) for each type of crop (Suppl. 1).

Data collection

Data required to calculate soil surface N balance were collected from 12 institute publications, 7 reference papers and official French documents (Table 4).

Agronomic information originated from two sources: the agronomical annual statistics of the SSP

Table 3 N losses to the atmosphere according to organic fertilizer type

Type	Reference	N loss (% N spread)		
		Lower limit	Reference	Upper limit
Bovine	(1)	37.1	19.8	9.1
Pig	(1)	88.3	31.2	14.7
Sheep, Goat	(2)	50	30	10
Poultry	(1)	51.1	27.9	11.6
Horse	(2)	50	30	10

Sources in the table refer to (1) Gac et al. (2006), (2) personal communication UMR Pegase

(*Service de la Statistique et de la Prospective*, 1940 to 2010) and UNIFA (*Union des Industries de la Fertilisation*) (Table 4). The SSP database provides yearly data for livestock numbers, crop yields and agricultural areas (e.g. UAA) for each department. The SSP data were gathered from databases of more than 1.35×10^6 registers mostly including information for cash crop area and production (~23 and 21% respectively) and livestock (~18%). Vegetable and fruit production represented about 12 and 6% of the data respectively, vegetable and fruit area were almost the same, each representing nearly 6%. Data for chemical fertilizers were obtained from the amounts delivered in each department, which were assumed to be equivalent to the quantity used in the same department.

Crop production and livestock classes differed over the period studied. Therefore, they were reorganized into more homogenous classes when necessary. When no data was available, data series were completed using the following simple rules: the missing value prior to the first given value was assumed to be equal to this first given value. The missing value following the last given value was assumed to be equal to this last known value. If there were missing values within series, the values were calculated using linear interpolation. Some data given at a regional scale (NUTS2) were downscaled to the department scale. In this case, departmental data (d_{NUTS3_year}) were computed using regional data (d_{NUTS2_year}) multiplied by the mean ratio between departmental and regional figures calculated for other years ($d_{NUTS3\_year} = d_{NUTS2\_year} * d_{NUTS3\_other\_years}/d_{NUTS2\_other\_year}$). In the end, approximately 37% of the production and livestock database was reconstructed following the above-mentioned rules, leaving 63% of raw data that originated from SSP statistics. Grass production required adjustments because in the SSP database, it was considered that all the natural grassland production was harvested and removed while part of permanent grassland was grazed. This led to an overestimation of N export since grazing does not export as much grass and thus nitrogen as cutting. Therefore, values for grass dry matter produced in natural meadows were corrected according to livestock needs per department. The corrected value was calculated as the difference between livestock fodder needs (5,2tMS/LSU, http://ec.europa.eu/eurostat/) and the sum of temporary grassland, artificial grassland, annual fodder, and dry matter content of root and tuber fodder production (Table 5). In 2012, a review published by Peyraud et al. found that about a quarter of livestock farms were at least 50% self-sufficient in dry matter. As a consequence, in order to address the dry matter needs of livestock, forage needed to be imported. The data available did not allow us to estimate accurately the transport of fodder between departments. However, fodder is bulky and expensive to transport and the departments are relatively large, so we assumed that the amount transported between departments could be ignored.

The level of N content in crops or in livestock excretion can vary widely. The lowest and the highest values found in previous studies were recorded as the minimum and maximum. Reference values for N content were provided by national bodies, 46% by

Table 4 Source of the data used in CASSIS_N

Data type	Sources
Area, production	French Ministry of Agriculture: SSP (1940–2010)
Chemical fertilizer delivery	SSP, UNIFA
Atmospheric deposition	EMEP
N content in crops	ANSES (2013), SoeS (2013), COMIFER (2013), EEA (2001), Audouin (1991), Alvarez et al. (2014), Bach and Frede (2005), Bouwman et al. (2005), CORPEN (1988), Leip et al. (2011), Feedipedia (consulted the 25/11/14), comm. pers, CETIOM, comm. pers.CNTIP, Honda et al. (2005), comm pers. ITB, UNIFA (2008)
N content in livestock excretion	CORPEN (1988, 1999, 2001, 2003, 2006), Circular DERF/SDAGER/C2002-3013, JOFR, 2011
N loss from manure	EMEP-Corinair in CORPEN (2006) cited in Soes (2013), Gac et al. (2006)

COMIFER (*Comité Français d'Etude et de Développement de la Fertilisation Raisonnée*, 2013) and the remaining by ANSES (*Agence nationale de sécurité sanitaire de l'alimentation, de l'environnement et du travail*, 2013). Reference values for N content ranged from 0.5 (apples) to 56.5 kgN ton^{-1} (soya) (Suppl. 1).

Livestock farming practices changed between 1940 and 2010 (diet, housing, animal productivity) and those changes have influenced N excretion (Hou et al. 2016; Peyraud et al. 2012), and also N loss from organic fertilization (Peyraud et al. 2012; Reidy et al. 2008). In particular, higher milk yields have led to an increase in N excretion for dairy cows (CORPEN 1999). However, due to a lack of data, N excretion and N loss from manure to the atmosphere were assumed to be constant over time when calculating N balance (Bouraoui and Grizzetti 2011). The data available in France did not enable us to assess quantified data on changes in livestock practices at a departmental level between 1940 and 2010. Values for N excretion and N loss from manure were collected for a variety of situations corresponding to a large range of management practices and livestock characteristics (Table 2) assumed to include all those encountered in France over the past 70 years. The difference between the minimum and maximum value found reflected the degree of uncertainty in livestock practices. For example, departmental milk yield is considered to be between 4000 and 10,000 kg of milk per dairy cow per year. This range was chosen because below 4000 kg of milk per cow, milk yield no longer influences N excretion (personal communication UMR PEGASE). A milk yield of 10,000 kg per cow per year is the highest yield simulated in CORPEN (1999). Moreover, this yield was exceeded in only 0.2% of all the geographic entities during the whole period studied (SPP 1940–2010). N excretion was simulated for low milk yield (4000 kg per head), medium performance (6000 kg per head) and high performance (10,000 kg per head) and for various diets following Soes assumption (Soes 2013). These results were combined to obtain a minimum, an average and a maximum N excretion value (Table 2). This uncertainty on N excretion value for dairy cow was taken into account in N surplus uncertainty calculation (see 3.3). If N excretion values were not available, values of spreadable N in manure *i.e.* N excreted minus N loss to the atmosphere can be used to calculate the N excretion value if necessary (Table 2). Sheep, goat and horse N excretion were calculated from spreadable N assuming a 30% loss of N to the atmosphere.

Data for atmospheric N deposition were taken from the EMEP database (http://www.emep.int/mscw/index_mscw.html), which provided a 50 km × 50 km model of dry and humid N deposition. As the EMEP database covers less than half of our studied period, available data were averaged on a pro rata basis of the surface area of the department and were assumed to be the same throughout the studied period. The values ranged from 7 to 17 kgN ha^{-1},with a mean of 12 (±2) kgN ha^{-1}.

Calculation of uncertainties in N surplus

Output uncertainties mainly result from basic uncertainty, that is to say, imperfect knowledge of reality (N content, magnitude of processes) and operational uncertainty, that is error in data (Oenema et al. 2003; Refsgaard et al. 2007). Therefore, uncertainty in

Table 5 Dry matter content in fodder

Root-tuber	% Dry matter	Notes	Reference
Beets	16		(1)
Carrots	18		(1)
Turnips	8		(1)
Suedes	15		(1)
Jerusalem artichokes	20		(1)
Parsnips	18	Like carrots	(1)
Celeriac	15	Like suedes	(1)
Cabbages	12		(2)
Pumpkins	10		(3)
Others	15	Mean of the others	This study

Sources in the table refer to: (1) Delteil (2012), (2) INRA (2007), (3) Duval (1995) model parameters was estimated according to the type of data (Oenema et al. 2003) and its availability. All parameters were assumed to follow normal distribution. When available, national survey data provided the average value and the standard deviation of the parameters (N content in crops). Otherwise, the standard deviation was estimated from minimum and maximum values in the literature (Table 6). The average values had been used to calculate N surplus time series now referred as 'base N surplus time series'. The influence of uncertainty brought by each variables of Eqs. (2)–(5) (N content in crops, number of livestock for example) and atmospheric deposition was tested by first setting all coefficient values used for the calculation of this variable to their minimum and then to their maximum range, while keeping the other variable at their base level. Variability was thus defined as the difference between the N surplus calculated with coefficients of a variable set to its maximum and that calculated when the considered item coefficients were set to their minimum values.

Output uncertainty was assumed to be a propagation of uncertainties associated with each parameter of the model. The imprecision in departmental N surplus calculation was obtained with a simple Monte Carlo simulation analysis. The model was then run 200 times with all the parameters' values selected randomly from their statistical distribution (mean, standard deviation and type of distribution). This resulted in 200 model outputs that could be analysed in terms of probability distribution and model performance (Loucks et al. 2005). The set of output results was tested for normality using the Chi square goodness-of-fit test ($p < 0.05$). Approximately 18% of the results did not follow normality, therefore imprecision was calculated as the average range between the ninth (E_9) and first (E_1) deciles of the 200 surplus values obtained for each year and each department.

Trend analysis of N surpluses over time and uncertainty influence

Significant trends in base N surplus time series, from now on referred to as 'base trends', were tested using Spearman's rho (ρ) ($p < 0.05$) (Yue et al. 2002). The correlation coefficient indicated the extent to which N surpluses and times were linked by a monotonic trend: higher absolute values of Spearman's ρ indicated stronger links between the variables. Spearman's ρ is a non-parametric test and therefore it does not assume statistical normality of results. It was applied to two different periods (1940–1991 and 1992–2010) corresponding to the date at which chemical fertilizer use changed (www.UNIFA.fr) and to the presumed impact of the Nitrates Directive (91/676/CEE).

The robustness of the trends was tested using output results obtained with simple Monte Carlo (MC) sample simulation. For one department, a set of 200 N surplus time series was constructed with values for each year selected randomly among the results obtained with simple Monte Carlo sample simulation (MC time series). Spearman's ρ ($p < 0.05$) was then performed for both selected periods for each of the 200 N surplus time series (MC trends). MC trends were then compared to base trends. The most robust trends were those for which there was the greatest number of MC trends equal to the base trends.

Table 6 Coefficient of variation (CV) used in the N soil surface N balance model

Category	CV (%) Median (range)
Mineral fertilizer	1.7
Deposition	1.7
Number of animals	1.7
N losses to the atmosphere	
From manure	15 (0.7–28.2)
From mineral fertilizer	6.9 (6.8–7.8)
Percentage of leguminous plants in meadows	8.3 (5.6–11.1)
N content in crops	20.3 (2.5–449)
N content in livestock excretion	13.3 (6.7–4000)

Results and discussion

The N exported when crops were harvested was the main factor among the seven variables included in the soil surface N balance (41%). Organic and chemical fertilizer inputs accounted for about the same percentage ($\sim$22% of the sum of absolute values of all items, Fig. 1a). In contrast, between 1985 and 2005 in 12 other European countries chemical fertilizers were reported to be the greatest anthropogenic N input (Bouraoui et al. 2011). However, the similar proportion of chemical and organic fertilizers at the national scale hides discrepancies between the 91 entities studied. Organic fertilizer was the main N input for 51 of them. Symbiotic fixation represented about 11% of the sum of absolute values of all N fluxes, followed by atmospheric deposition (3%).

N surplus results at the national and departmental scales

The average N surplus, determined from the base N surplus time series during the whole period and for the 90 departments and Corsica, was about 37 kgN ha UAA^{-1} with values ranging from -70 to $+187$ kgN ha UAA $^{-1}$ year^{-1}. The departmental N surplus means ranged from 10 to 86 kgN ha UAA^{-1} year^{-1}, with the lowest values found in the centre and the south east of the country and the highest in the north west (Fig. 2a). In 1940, only one N surplus was higher than 40 kgN ha UAA^{-1} year^{-1} with an average surplus of around 16 kgN ha UAA^{-1} year^{-1}. In 1991, the mean N surplus rose to 52 kgN ha UAA^{-1} year^{-1},with a greater spread of values either side of the mean. N surpluses covered a wider range in 2010 than in 1940 and 1991 but the mean surplus was lower ($\sim$34 kgN ha UAA^{-1} year^{-1}) (Fig. 2b).

Four departments showing different agricultural activities were characterised by different changes in N surplus. For department A, characterised by a UAA of mainly permanent grassland, the soil surface N balance was very close to equilibrium (Fig. 3a). By contrast, for a department where UAA represented almost as much cereal production as permanent grassland, or a majority of cereal production, N surpluses were higher and varied over time (Fig. 3b, c). The highest values and greatest variation in surpluses were found in departments where there were more livestock (Fig. 3d).

Uncertainties

Sensitivity analysis

The highest contribution to total uncertainties was crop production (39% of the total variability) (Fig. 1b), and it mainly originated from N content (Table 7). Organic fertilizer use contributed 37% of the total variability in N surpluses, with N losses to the atmosphere being the item that caused the most

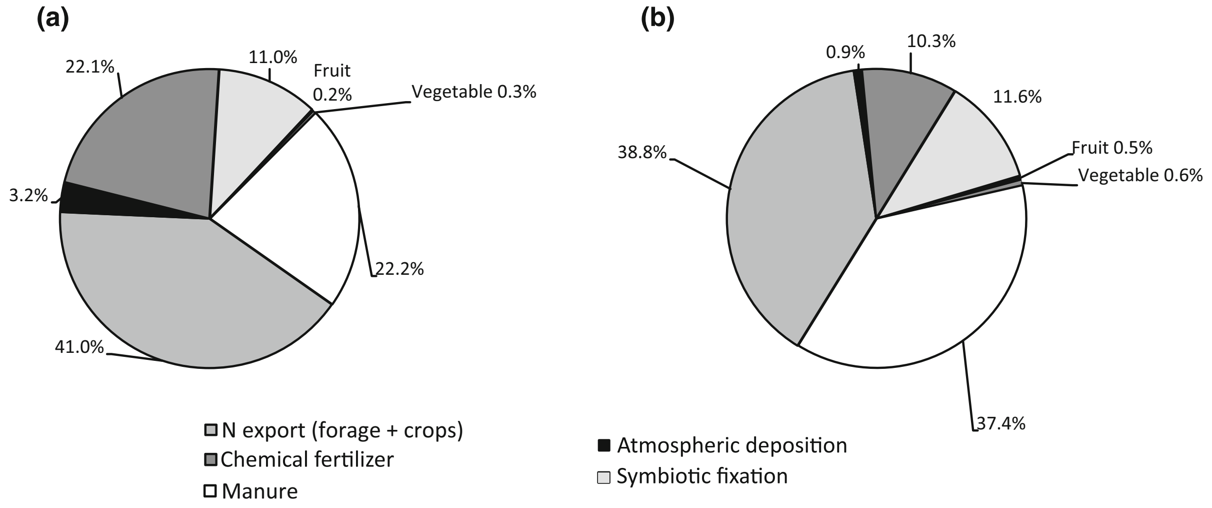

Fig. 1 **a** Contribution of each item to the soil surface N balance (mean value over the whole period studied from 1940 to 2010, % of the sum of absolute value of all items). **b** Percentage of the total variability contributed by each item (1940–2010)

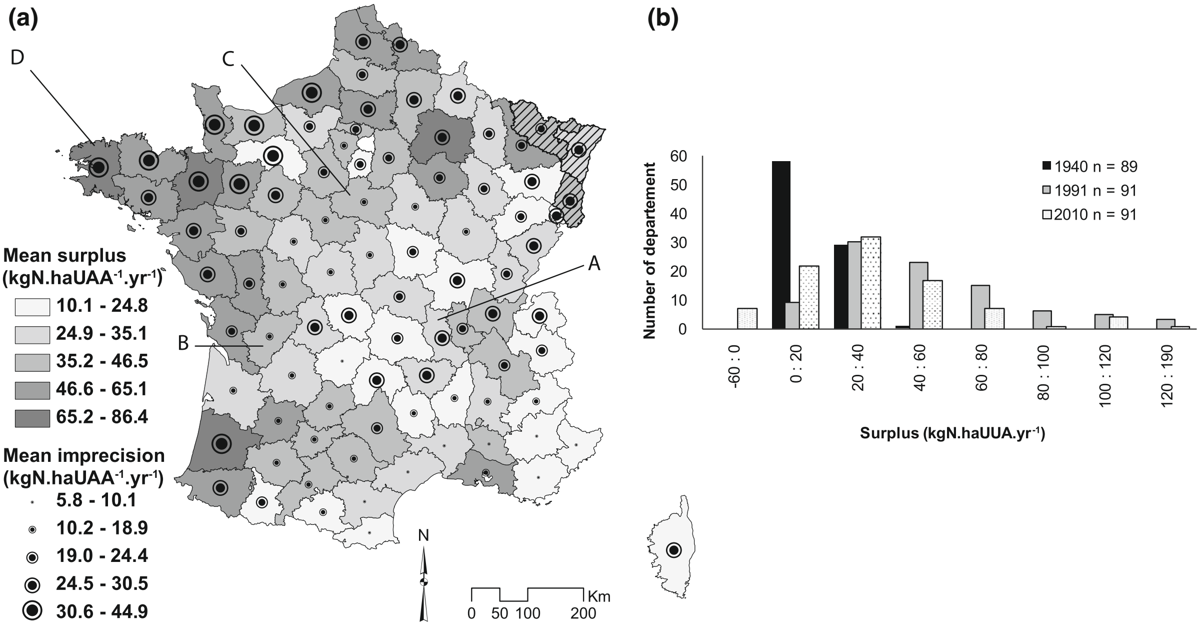

Fig. 2 **a** Mean departmental N surpluses and their associated imprecision (80% confidence interval) with *A*, *B*, *C* and *D* indicating four departments with different types of agriculture. Data were not available for 1940–1944 for hatched departments. **b** Departmental N surplus distribution for 1940, 1991 and 2010

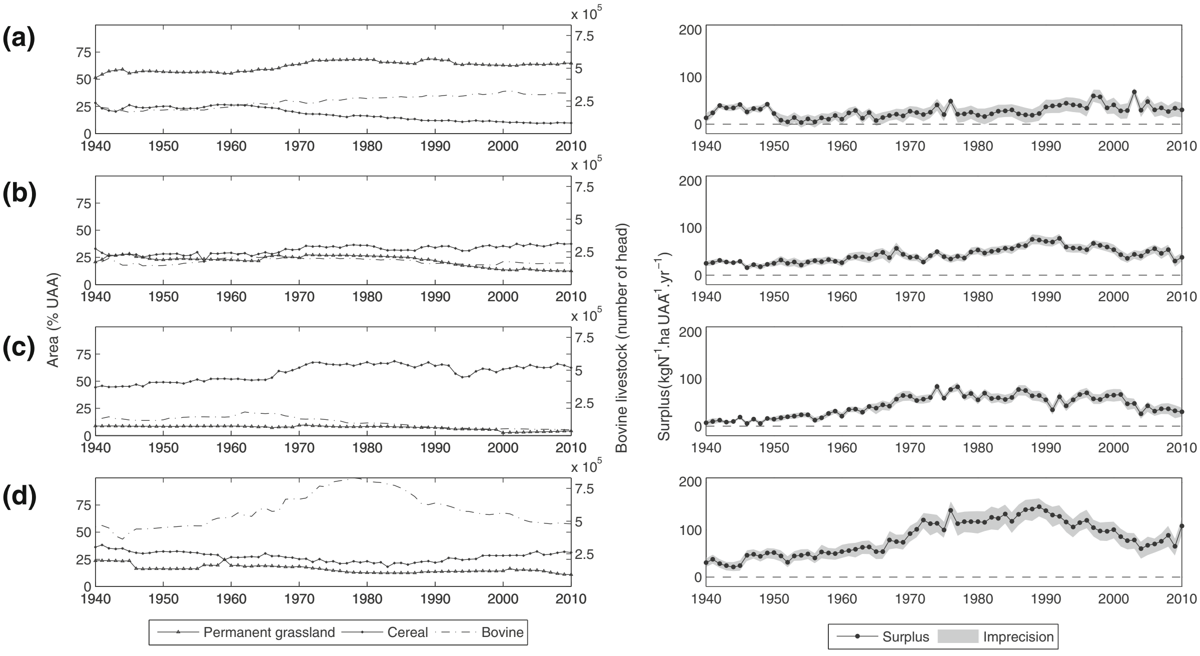

Fig. 3 Surplus time series with annual imprecision, for the four French departments indicated in Fig. 2a, characterized by different typical agricultural production systems: **a** department characterised by UUA used for permanent grasslands, **b** UAA used roughly equally for cereal and permanent grasslands, **c** UAA used mostly for cereal production, **d** department with the highest bovine livestock production

Table 7 Mean variability in surpluses caused by each item of the soil surface N balance (1940–2010) (kg N ha UAA^{-1} year^{-1})

Items	Variability produced by the item modification
N Export	
N content	44
Crop production	8
Manure	
Number of livestock	4
N excretion	25
N loss from manure	17
Chemical fertilizer	
Fertilizer delivery	10
N loss from chemical fertilizer	20
Fixation	
N content	16
Crop production	2
Coefficients (α, β, NHI, BGN)	26
Proportion of leguminous plants	5
Atmospheric deposition	1.2

Variability is defined as the difference between surpluses calculated with the coefficients of an item set to their maximum and to their minimum

variability. N fixation and chemical fertilizers contributed 12 and 10% of the variability respectively. Fruit and vegetable production, and atmospheric deposition together only accounted for 2% of the total variability.

Regarding chemical fertilizers, variations due to uncertainty in chemical fertilizer data (i.e. quantity of fertilizer delivered) were almost twofold smaller than the variation due to N loss into the atmosphere (Table 7). It was assumed that the quantity of fertilizer delivered in a department was entirely consumed within the same department during the same year. However, temporal and geographic permeability do exist (fertilizer stocks and exchange between departments), but this information was not available. To overcome this problem, models could be based on surveys of the amount of fertilizer used. This method has been used in other studies (NOPOLU, BASCULE see Table 1) but cannot be applied to such a small scale for the whole country or to long time series for the following three reasons. First, surveys are based on interviews of a sample of farmers which might not be representative (selection bias). Secondly, spatialized data for fertilizer practices regarding each type of crop are lacking. Due to their scarcity in the past, these surveys cannot be used to estimate past fertilizer use. Finally, dishonesty could introduce a bias which is difficult to evaluate (Payraudeau et al. 2007). Like chemical fertilizers, the uncertainty in N losses to the atmosphere was the main item that created variability for organic fertilizers, followed by the uncertainty in N content of organic fertilizer and then by the uncertainty in data (Table 7). Concerning organic and chemical fertilizers, the amount of fertilizers produced in one department was assumed to be totally used within that same department. On the one hand, some departments in western France like in Brittany are known to be in high N surplus situation because of the concentration of livestock breeding (i.e. Fig. 2a department D). On the other hand, the soils of some departments characterized by intensive cropping (i.e. Fig. 2a department B) are known to lack organic matter. However, any trade of manure between departments is unofficial and to our knowledge there is no quantified overview of this exchange (Aubert and Levasseur 2005). Moreover, manure transport is bulky and expensive and farmers tend to avoid it. There might be manure movement from high livestock areas to departments lacking organic matter, but this mainly involves dry dejections such as poultry manure. The latter has a very low N content, and thus would probably have a low impact on N surplus.

Imprecision in N surpluses: a Monte Carlo simulation analysis

Mean imprecision was determined using the 200 MC simulations in each of the 91 geographic entities and for each of the 71 years studied.

The average of the departmental imprecision for the whole period ranged from 6 to 45 kgN ha UAA^{-1} year^{-1} (Fig. 2a). The average departmental imprecision was 21 kgN ha UAA^{-1} year^{-1}. The departmental imprecision appeared to be spatially organized (Fig. 2a). In fact, imprecision was mainly linked to N export and to a lesser extent to organic fertilisation (Fig. 4). Departments with the greatest imprecision were those with higher N export and greater livestock production (Figs. 3, 5).

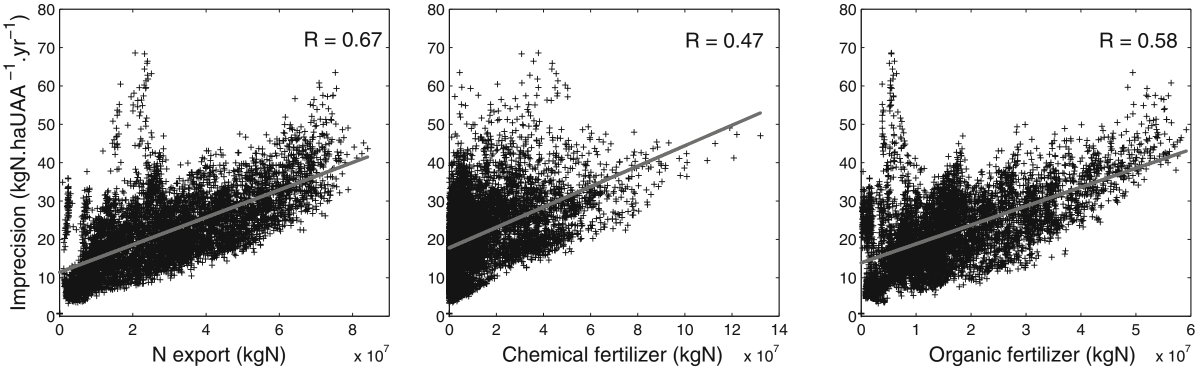

Fig. 4 Relation between imprecision and three items of the soil surface N balance, N export, and chemical and organic fertilizers

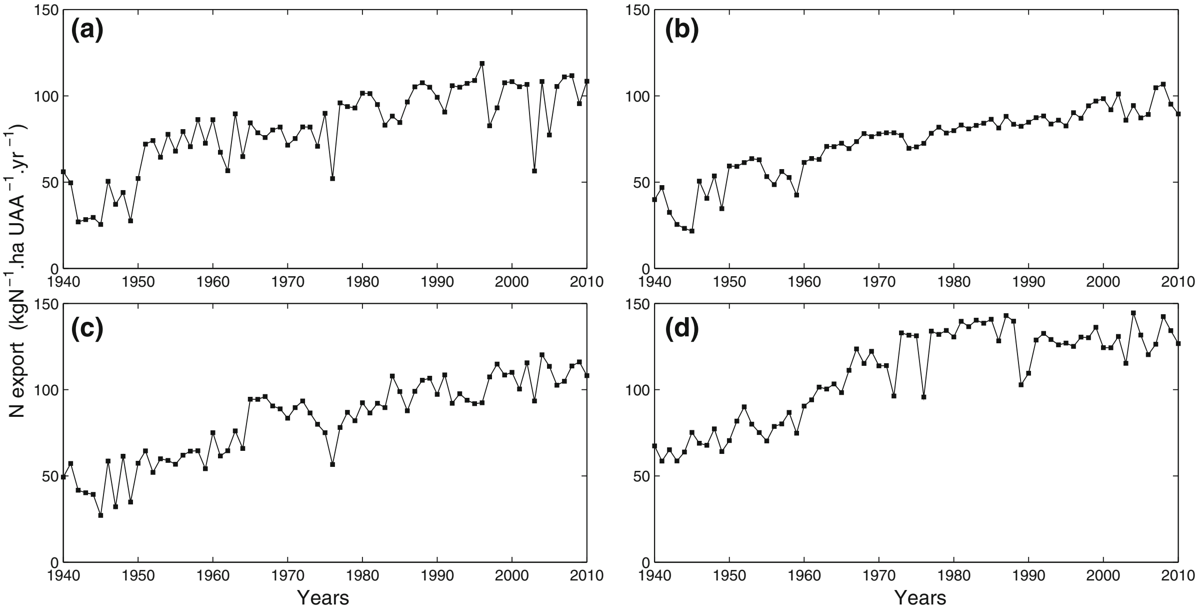

Fig. 5 N export for four departments (for the four French departments indicated in Fig. 3)

Temporal trends of N surplus

Reference trends of surplus over time were statistically identified over two periods: 1940–1991, and 1992–2010. Statistical analysis of the outputs of the soil surface N balance model determined only with reference values revealed significant trends during the two periods. The main trend during the first period was a surplus which increased over time (for 82% of the area), indicating an increase in diffuse N pressure (Fig. 6). During the second period, 8% of the area still showed a trend of increasing N surplus. However, more than 90% of the area presented a stable or decreasing N surplus. Hence, over the whole period, the main pattern was an increasing N surplus over time within the first period, followed by a decrease (47%) or an increase and then a stable N surplus over time (30%). This change in trends between the two periods can be interpreted as a consequence of the Nitrates Directive. This Directive has played a major role in European legislation and introduced a limit on fertilizer use and aimed to balance N input (mineral, manure, reactive nitrogen from the stock in the soil). Since 1990, many other European countries (e.g.

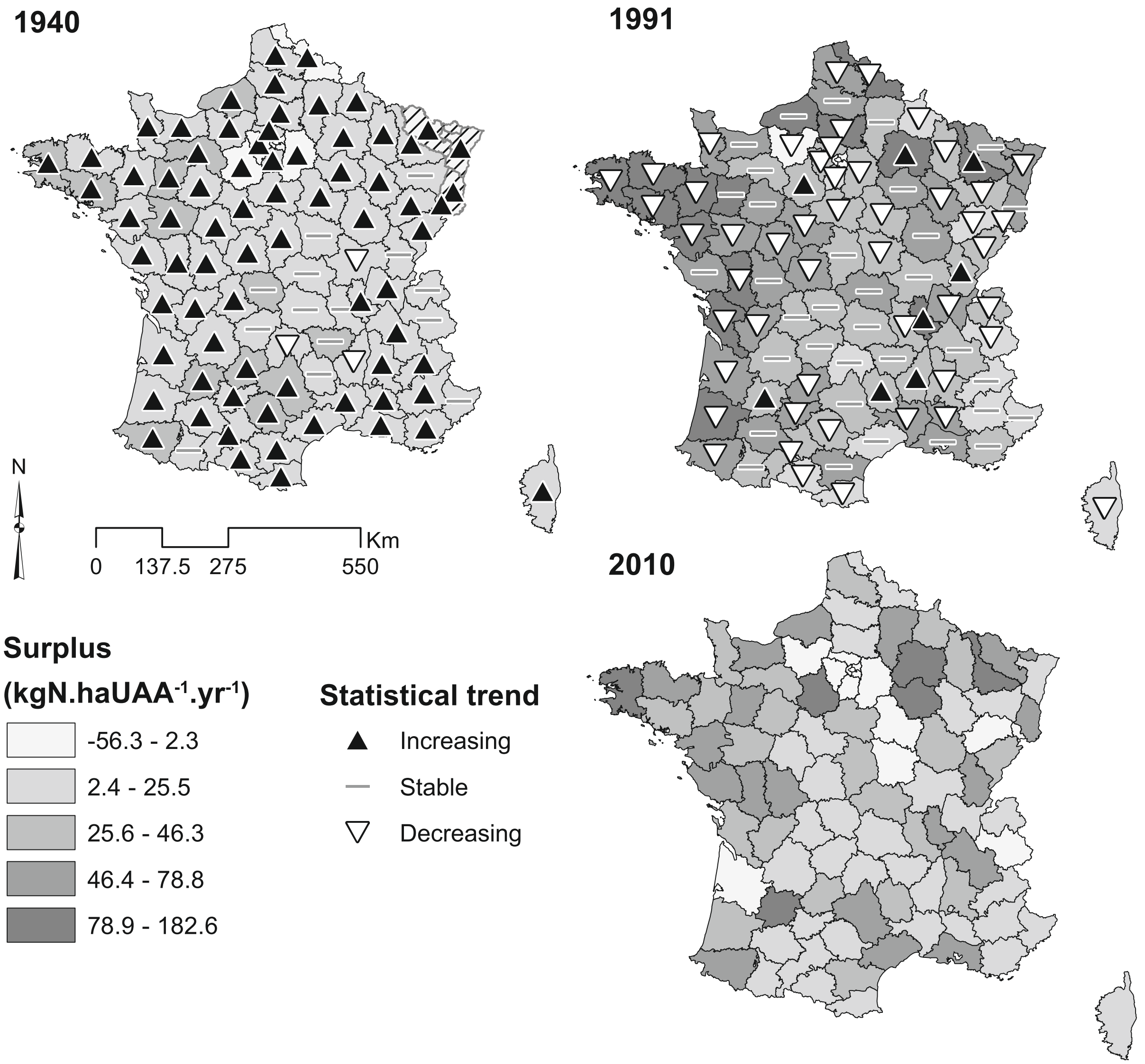

Fig. 6 Statistical trends in departmental N surplus since 1940 in France for two periods: 1940–1991 and 1992–2010. Data were not available for 1940–1944 for hatched departments

Germany, Italy and Portugal) have shown a trend of decreasing N surplus (OECD Compendium of Agric-environmental Indicators 2013).

However, the analysis of uncertainties raised the issue of some possible variations in the output model. Although Spearman's ρ test is a powerful tool to detect trends in time series, its power depends on the amount of variation within a time series (Yue et al. 2002). Strong variations within data can hide the magnitude of the trend, decreasing the robustness of the test and preventing detection of temporal trends. Therefore, the influence of imprecision in trend detection was tested with the outputs of the Monte Carlo simulation analysis (MC trends). Based on the 200 series of surpluses for each of the 90 departments tested, we found that only 13.0% of trends changed compared to the reference trends. The greatest changes were a switch from a significant trend (downward or upward) to a non-significant trend, that was considered stable (7.2 and 4.3% of the 36,400 simulated trends respectively, Table 8). During the first period, the change was mainly from increasing to stable trends, while in the second period, the changes were mostly from decreasing to stable trends. This clearly suggests

Table 8 Estimation of robustness of departmental trends in N pressure despite uncertainty in soil surface N balance

Types of switch	Percentage
No trend switch[a]	87.0
Trend change[a]	13.0
Trend no longer significant[a]	11.5
Trend no longer significant in first period[b]	7.6
Including Increasing to Stable	6.2
Trend no longer significant in second period[b]	15.4
Including Increasing to Stable	2.4
Switch from decrease to stable over the two periods[a]	7.2
Switch from increase to stable over the two periods[a]	4.3
Switch from stable to increase or decrease[a]	1.5
Switch from significant trend to another[a]	0
Total (36,400 trends)	100

Trends in N diffuse pressure at departmental level calculated with reference values for all parameters (base trends) were compared with trends observed in surplus times series constituted with Monte Carlo outputs (MC trends).The more the trend is conserved between reference trends and MC trends, the more robust the trend is considered

[a] N = 36,400

[b] N = 18,200

that even when taking into account imprecision, diffuse N pressure in France has remained stable or decreased since 1991.

These N surpluses and their associated uncertainties assessed over a long time period are essential for modelling past and present N pressure at a subnational scale. Adapting these results to a suitable scale (large catchments) and comparing this diffuse N pressure time series to N concentration in rivers could provide valuable information about N transfer, in particular, its transit time from soil to river networks and its retention time in river basins.

Conclusion

N surpluses were assessed over a 71-year period with yearly results between 1940 and 2010 in France. National mean N surpluses calculated for the whole area rose from 16 to 52 kgN ha UAA^{-1} year^{-1} between 1940 and 1991, and decreased to 34 kgN ha UAA^{-1} year^{-1} in 2010. This change in N surpluses has been found in other European countries. However, national trends in N pressure hid discrepancies between the different departments and the mean department surpluses ranged from 10 to 86 kgN ha UAA^{-1} year^{-1}. The N surpluses obtained in this study were characterized by a large variability, mainly due to uncertainties in N content in crops and in N excreted by livestock, but also in the estimation of symbiotic fixation. The imprecision, defined here as an 80% confidence interval in departmental N surpluses, showed a spatial organization due to its strong correlation with organic fertilizer use and N export. This departmental imprecision ranged from 6 to 45 kgN ha UAA^{-1} year^{-1}.

The model revealed an upward trend in surplus values between 1940 and 1991 for 82% of the studied area and a downward or stable trend for more than 90% of the area between 1991 and 2010. Imprecision did not modify the statistical trend for most departments (86%). In particular, diffuse N pressure remained stable or decreased in most of the area under study, probably as a consequence of the Nitrates Directive.

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