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ORIGINAL ARTICLE

How changes in diet and trade patterns have shaped the N cycle at the national scale: Spain (1961–2009)

Luis Lassaletta · Gilles Billen · Estela Romero · Josette Garnier · Eduardo Aguilera

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Abstract During the last five decades (1961–2009), Spain has experienced a considerable expansion in the nutrient cycle of its agricultural sector and, in particular, a threefold increase in anthropogenic reactive nitrogen inputs, from 536 Gg N year^{-1} in 1961–1965 to 1673 Gg N year^{-1} in 2005–2009. Import of feed (soybean, cereals, and cakes) from America and Europe to supply a growing livestock population constitutes the largest share of this increase, along with intensification of synthetic fertilizer use. While in the early 1960s, Spain was nearly self-sufficient in terms of food and feed supply, the net import of agricultural products presently equals domestic crop production, when expressed in terms of nitrogen content (ca. 650 Gg N year^{-1}). The most important driver of this shift appears to be the rapid change in domestic consumption patterns, which evolved from a typical Mediterranean diet to an animal-protein-rich diet similar to the North European and American diets. Besides livestock production mostly for national consumption, the Spanish agricultural system has specialized in vegetal products with low N content such as olive oil, wine, vegetables, and citrus fruit, which are for the most part exported. The nitrogen load exported outside the Spanish borders by rivers is very low (6.5 % of the total net N input). As a result of the high import and low export of reactive nitrogen, the Spanish mainland is suffering from considerable pollution by local emissions of reactive nitrogen forms to air and water.

Keywords Nitrogen cycle perturbation · Net anthropogenic nitrogen input (NANI) · Agriculture · Trade · Diet · Country scale

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L. Lassaletta (✉) · G. Billen · E. Romero · J. Garnier
CNRS/Université Pierre et Marie Curie, UMR Sisyphe, 4 Place Jussieu, 75005 Paris, France
e-mail: lassalet@bio.ucm.es

L. Lassaletta
Department of Ecology, Universidad Complutense de Madrid, c/ José Antonio Novais, 28040 Madrid, Spain

E. Romero
Instituto de Ciencias del Mar (CSIC), Pg. Marítim de la Barceloneta 37-49, 08003 Barcelona, Spain

E. Aguilera
Universidad Pablo de Olavide, Ctra. de Utrera km. 1, 41013 Sevilla, Spain

Introduction

The global alteration of the nitrogen (N) cycle leads to many effects on human and ecosystem health, biodiversity, and climate (Sutton et al. 2011). Food production is the main factor responsible for the conversion of N_2 to reactive nitrogen (N_r all nitrogen species except N_2), mostly through industrial fixation of N_2 (Haber–Bosch process) and the expansion of N-fixing crops (Galloway et al. 2008). The increasing use of synthetic fertilizers, together with other agricultural intensification practices, has resulted in a considerable increase in agricultural productivity during the last decades (Tilman et al. 2002). However, a large share of the increase in primary agricultural production is used as animal feed (Pelletier and Tyedmers 2010). Intensive livestock production contributes to several global and local environmental problems, such as deforestation, greenhouse gas (GHG) emissions, or water eutrophication (McAlpine et al. 2009; Aiking 2011; Weiss and Leip

2012). Regarding the global N cycle, due to the inefficient conversion of vegetal to animal protein, animal food production contributes to ca. 63 % of reactive N environmental losses (Pelletier and Tyedmers 2010) and 65 % of N_2O emissions (Steinfeld and Wassenaar 2007).

The growing trend in livestock production is related not only to an increase in the human population, but also to an increase in the share of animal products in the diet of several world regions (Kastner et al. 2012). Since the environmental emissions of N associated with the production of different food commodities are highly variable (Leach et al. 2012), the human diet is a crucial driver for the N cycle alteration. Recent research has explored the potential impact of a diet change in the emissions of N to the environment. Billen et al. (2012a), (2013) and Popp et al. (2010) have shown how a reduction in meat consumption in industrial countries could decrease water pollution and non-CO_2 GHG emissions, respectively. Even changes between different types of meat could effectively reduce nutrient flows in countries with intensive ruminant meat production (Bouwman et al. 2011). In Europe, where diet types are regionally highly variable but in general rich in animal proteins (de Boer et al. 2006), two-thirds of the total agricultural area is devoted to livestock production (Westhoek et al. 2011). This is, however, not enough to sustain the high European livestock densities, and 75 % of the protein-rich feed is imported, mainly from South America (Westhoek et al. 2011). Taking into account not only the GHG emission in the production of feed crops, but also those emissions associated with land use change, much of the GHG emissions associated with the EU livestock sector occur outside the EU (Weiss and Leip 2012).

During the last few decades, livestock has progressively lost its connection to land, and at the global scale, large territories have specialized either in crop or in meat and milk production (Naylor et al. 2005; Billen et al. 2010). As a result of this global specialization, trade of agricultural products has increased more than ten times during the past six decades, and it is forecasted to continue increasing together with many associated environmental problems, such as the increase in CO_2 emissions related to deforestation (Schmitz et al. 2012). When dealing with a regional territory such as a country or a watershed, the net commercial import of food and feed also has to be taken into account, in addition to the three types of inputs that account for the new anthropogenic N inputs (NANI) at the global scale (namely synthetic fertilizers, combustion of fossil fuels, and N-fixing crops) (Swaney et al. 2012). This balance between import and export of agricultural goods determines the “anthropogenic nitrogen autotrophic or heterotrophic character” of any territory according to its potential for commercial export or, on the contrary, to the need to import agricultural commodities as defined by Billen et al. (2010). In our globalized world, significant amounts of N embedded in agricultural products are internationally traded every year (Grote et al. 2005; Lassaletta et al. submitted), and meat production takes a large part in this international trade market (Galloway et al. 2007; Burke et al. 2009). Agricultural practices, diet, and import/export patterns thus have an important effect on the N cycle. Countries are geographical units with identifiable patterns and integrated statistics in terms of productivity and consumption issues. They are thus useful units to analyse the driving forces and leak in nutrient cycles (Senthilkumar et al. 2012; van Grinsven et al. 2012). The study of the NANI at the country level and the analysis of the exchanges throughout the world can be very useful to improve our understanding of the changing N cycle. Studies dealing with long-term country-level N cycling are scarce and, to our knowledge, only one paper focused on the USA has been published to date (Howarth et al. 2002).

Spain represents an interesting case study in that respect, as profound mutations have occurred during the last 50 years in this country regarding economic status and social behaviour, leading to considerable changes in its agro-food system (Carpintero 2005). In this paper, using the NANI accounting approach introduced by Howarth et al. (1996), we first describe those changes in terms of N fluxes at the national scale during the last five decades (1961–2009); we then show how changes in dietary patterns and international trade have exerted a major influence on the trends observed; we finally discuss the environmental consequences of the changing Spanish N cycling patterns.

Materials and methods

We calculated an overall N budget for Spain for the 1961–2009 period, using the net anthropogenic N input (NANI) concept (see Howarth et al. 1996, 2012; Swaney et al. 2012 for an overview of the NANI approach), adapted at the country level instead of the watershed level, the usual scale for this approach. To this purpose, we first estimated the total “new” anthropogenic N inputs entering the country, through synthetic fertilizers application, net atmospheric inputs, crop biological N fixation, and net import of food and feed. These budgets were calculated for all Spain including the Balearic and Canary Islands. Detailed description of the methodology is provided in Suppl. 1.

Yearly data on synthetic N fertilizer application, under different N forms, for the entire period were obtained from the International Fertilizer Industry Association (http://www.fertilizer.org/). To estimate net atmospheric inputs, we followed a slightly different approach from the procedure described by Howarth et al. (2006), estimating a net balance of atmospheric deposition and emission (Lassaletta et al. 2012). We obtained national figures of emission and deposition of oxidized and reduced compounds from EMEP (http://www.emep.int).

We estimated N fixation by the 24 Spanish N-fixing crops, including several legume species and sugar cane. We applied the relationship proposed by Lassaletta et al. (2012) calculating N fixation as the difference between total N biomass produced (including underground and aerial non-harvested parts) minus N applied as fertilizer. We also estimated N fixation in pastures and rice following MMARM (2010) and Herridge et al. (2008), respectively.

To estimate the annual net import of agricultural products, we used the yearly import–export quantities for Spain (1961–2009) provided by the Trade Module of the FAOSTAT database (for 572 commodities). The N content of every product was gathered from different sources (McDougall et al. 1993; FAO 2011; Asmala et al. 2011; USDA 2012; Lassaletta et al. 2012) and is provided in Suppl. 2.

Using the data from the detailed trade data matrix from the FAOSTAT database, we estimated and mapped the net N traded to Spain from every world country for the year 2007. We consider that any country has an anthropogenic N heterotrophic behaviour when the primary production, including crops and pastures, is not enough to meet the demands of the livestock and people of this territory and the import of products is needed. On the contrary, those countries with a surplus of primary production that is available for export are considered autotrophic.

We estimated the total yearly agricultural and animal N production of the 117 vegetable and 14 animal products produced in Spain (1961–2009) using the Production Module of the FAOSTAT database. We added to this total value the estimation of N production in Spanish meadows and pastures (MMARM 2010). We estimated the proportion of every product used for food or feed using the information provided in the food balance sheets of the FAOSTAT database. Manure N production was also estimated using the excretion factors provided by Bouraoui et al. (2011) for Spanish livestock. Diet characteristics, namely N available per capita and year and percentage of N of animal origin, were calculated for the 1961–2009 period from the food supply module of the FAOSTAT database.

The mean multi-annual N flow at the outlet of the main Spanish rivers was calculated using data on river flow and water quality for the 2000–2010 period obtained from several water authorities (Confederaciones Hidrográficas) in Spain, or calculated by a model (in the case of rivers to the Cantabric Sea; Project MARCE). We use the term "retention" to designate all the processes preventing nitrogen load from being transferred to the outlet of the drainage network, including definitive elimination as inert N_2 through denitrification processes, emission as reactive N gases, such as NH_3 or N_2O, and storage in soil, perennial vegetation, groundwater, or sediments (Lassaletta et al. 2012).

Historical changes

Many important changes related to the agro-food system have occurred in Spain during the last 50 years (Table 1). The total population has increased by 42 %, and a substantial rural depopulation process has occurred. The total agricultural area has slightly decreased (14 %) due to the abandonment of most marginal arable lands and pastures. In the past 50 years, Spanish agriculture has intensified as indicated by the considerable increase in some features such as mechanization, irrigation, fertilizer application, and energy invested in production factors. This intensification has produced an increase in the yields of many crops, particularly remarkable for maize and wheat. These changes, together with significant changes in the size and structure of farms or the expansion of permanent crops to sloping landscapes, have been associated with considerable environmental problems, such as soil erosion and salinization (Carpintero 2005; García-Ruiz 2010). In terms of energy, the recent Spanish agro-food system has been demonstrated to be highly inefficient (Infante Amate and González de Molina 2012).

Livestock numbers have considerably increased, particularly pigs and poultry. To supply feed to this livestock, the proportion of surface grain crops devoted to feed has increased from 30 to 70 % (Carpintero 2005). However, this is insufficient, and import of feed products has substantially increased, particularly for soybean and its derivate products. The fate of some products has also changed; e.g. in the early 1960s, 100 % of the wheat consumed in Spain was used for human food, whereas in the last decade, only 40 % is consumed by humans (Suppl. 3). The total import of agricultural products expressed in weight has increased ninefold and export 7.5-fold during this period, but expressed in financial terms, imports increased twofold and exports threefold. The contribution of the agricultural sector to the Spanish economy has decreased by 90 %.

Total protein consumption in Spain has increased from 4.6 to 6.5 kg N cap^{-1} $year^{-1}$. The increase in the share of animal protein has been more intense, from 37 % in the 1960s to 65 % at the present time. Together with the population increase, this has resulted in a considerable absolute increase in the consumption of animal products. The current Spanish diet is very far from the so-called Mediterranean diet (de Boer et al. 2006), and Spain has

Table 1 Historical changes in Spain 1961–2009

	Variable	1961–1965	1981–1985	2005–2009	(%)
Population	Population (1000 inhab)	31418	38147	44560	42
	% Rural population	41	26	23	−43
Agricultural surface	Agricultural area (10^3 km^2)	330	309	283	−14
	Arable land (10^3 km^2)	161	156	126	−22
	Permanent crops (10^3 km^2)	46	49	48	5
	Pastures (10^3 km^2)	123	104	109	−12
Intensification indicators	Agricultural tractors (10^3 units)	111	591	1013	811
	Irrigated area % over agricultural land	10	15	22	117
	Fertilizers Tg N $year^{-1}$	0.37	0.86	0.88	139
	Energy in production factors (10^9 kcal ha^{-1})	70	204	268	285
Yields (ton ha^{-1})	Barley	1.4	2.0	2.7	94
	Grapes	2.5	3.4	5.3	112
	Maize	2.3	5.6	9.9	324
	Wheat	1.0	2.0	2.8	170
	Alfalfa for forage and silage	36.4	44.1	48.8	34
	Oranges	17.5	15.2	20.1	15
Livestock	Cattle (10^6 heads)	4	5	6	71
	Pigs (10^6 heads)	6	12	26	358
	Sheep (10^6 heads)	21	17	21	3
	Chickens (10^6 heads)	38	114	136	256
	Spanish grain surface devoted to feed (%)	31	45	70	126
Import of feed	Maize 10^3 Tn	845	4257	4929	483
	Soybeans 10^3 Tn	89	2675	2733	2982
	Soybean cakes 10^3 Tn	119	529	3405	2771
	Barley 10^3 Tn	429	548	999	133
Trade	Import 10^6 tonnes (420 commodities)	3	10	33	979
	Export 10^6 tonnes (421 commodities)	2	7	21	765
	Value import 10^9 $ (deflated)	14	11	26	91
	Value export 10^9 $ (deflated)	12	10	31	166
	Agriculture contribution GDP (%)	22.6	10.0	2.5	−89
Diet	Protein consumption (kgN cap^{-1} $year^{-1}$)	4.6	5.7	6.4	38
	Animal protein consumption (kgN cap^{-1} $year^{-1}$)	1.7	3.3	4.1	139
	Proportion animal protein (%)	37	58	64	73
Energy consumption	Total energy consumption (fossil fuels) kt cap^{-1} $year^{-1}$	0.7	2.5	2.7	259

now reached the same levels of protein ingestion as countries such as the USA and France. The percentage of animal protein in the diet is nowadays higher than the average value for Western European countries, and equivalent to the percentage found in the USA. This evolution has not been observed in other countries of the world. For instance, a decrease in both total protein consumption and the share of animal protein can be observed in the Democratic Republic of the Congo, and in China, there has been an increase but starting from much lower levels (Fig. 1). The diet changes observed in France over 200 years (Billen et al. 2012b) have occurred in Spain in only 50 years. Finally, the per capita consumption of fossil fuels has increased more than 2.5-fold.

The Spanish N budget

Evolution of NANI and its components

Total NANI inputs in Spain have tripled during the study period, from 536 Gg N $year^{-1}$ in 1961–1965 to 1673 Gg N $year^{-1}$ in 2005–2009 (Fig. 2; Suppl. 4). Compared to the country area (504,645 km^2), this means an input of 3314 kg N km^{-2} $year^{-1}$ in the most recent period. It is an intermediate value, higher than those estimated for the USA, Brazil, and the Baltic area (ca. 1900 kg N km^{-2} $year^{-1}$, Filoso et al. 2006; Swaney et al. 2012) but lower than the values for China and South Korea (5426 and ca. 8000 kg N km^{-2} $year^{-1}$; Ti et al. 2012 and

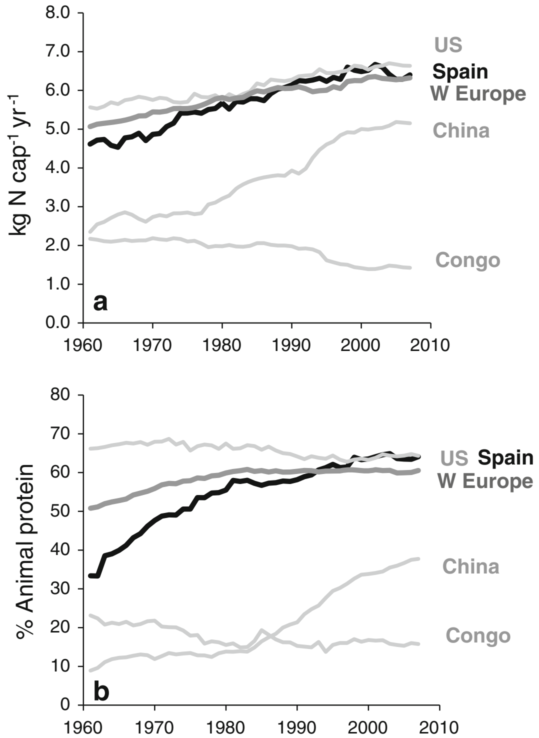


Fig. 1 Trajectories of the per capita protein ingestion (**a**) and the percentage of animal protein consumption in diets (**b**) for several world countries. The values correspond to the N supply per capita before subtracting food waste

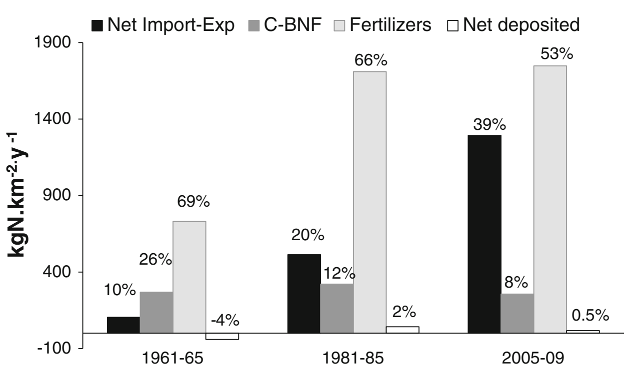


Fig. 2 Distribution of the components of NANI in three periods expressed in relation to the area. Every column shows the percentage contribution of every component to the total

Bashkin et al. 2002, respectively). The Spanish NANI value is in the highest quartile of the NANI estimates for the 3679 populated world catchments modelled in the GlobalNEWS model (Billen et al. 2010). NANI varies, however, among the different regions of the country; Lassaletta et al. (2012) have estimated, for instance, an input of new N of 5518 kg N km^{-2} year^{-1} for the Ebro catchment, and much higher values are reached in the most intensive agricultural areas.

Not only the total NANI has changed during the last five decades, but also the contribution of its four components has evolved (Fig. 2). The most remarkable feature has been the increase in the contribution of net N import in the form of food and feed, from only 10 % in the first period to ca. 40 % in the latest one. The absolute values of import and export have also significantly increased during the study period. The contribution of net import to the total NANI is very high when compared to some of the aforementioned countries, where this contribution is very low or even negative. Only in South Korea, a significant net N import has been observed, although it is much lower than in Spain (22 %). Fertilizer application increased threefold during the study period (Table 1); this rise is equivalent to that observed, on average, for Western European countries, where the highest increase occurred during the 1960–1990 period. In Spain, the highest application rate was observed in 1998 and was followed by a progressive reduction until today. The recent decline is probably related to a reduction in the fertilized surface together with an increase in the efficiency of the fertilizers, the rationalization of the doses, and the application of several environmental policies. In Spain, according to Steinfeld and Wassenaar (2007), 42 % of this synthetic fertilizer is applied to feed crops or pastures. The share of the different types of synthetic fertilizers has also changed. Urea is a very cheap fertilizer imported from Asia, and its use has progressively increased from 0.2 % in 1960 to 30 % in 2009 (Suppl. 5); this can produce specific environmental problems such as higher emissions of NH_3 that vary depending on climate and management (Sanz-Cobena et al. 2008).

Emissions of oxidized compounds have increased progressively reaching the maximum level (421 Gg N-NO_x) in the 1995–1999 period and slightly decreasing during the following years (Suppl. 6). Despite this decline, Spain was, together with the UK, the EU country that contributed the most to the emissions of NO_x in 2007 (EEA 2009). Approximately 60 % of these emissions correspond to transport while the rest results from industrial and domestic energy consumption. A large part (57 %) of these emissions, however, is not deposited in Spain but is atmospherically exported. Regarding reduced forms of N, 92 % of the emission is related to agricultural activities, and this emission largely exceeds deposition, showing that 45 % of it is also atmospherically exported outside Spain. As a result, the net balance of atmospheric input of Nr to Spain is very low and can even be negative (Fig. 2). This strongly contrasts with the situation of natural, low-populated regions such as northern Sweden, where deposition of oxidized compounds is often the major component of the NANI (Swaney et al. 2012).

The contribution of crop N biological fixation to Spanish agricultural systems (c-BNF) has progressively been reduced from 26 % in the 1960s to currently only 8 %, and this is mainly due to the increase in other NANI components, since the absolute variation of c-BNF is minor (Fig. 2). N fixation grew steadily during the first 20 years and reached its maximum in 1979 (176 Gg N year^{-1}). It declined afterwards to reach 126 Gg N year^{-1} in the last period. Alfalfa is the largest contributor to the total Spanish c-BNF, accounting for 72 % of the total in the last period (Suppl. 7). The clear increase in alfalfa N fixation during the first 20 years is due to an increase in yields (20 %) and also in surface area (52 %), and after this a gradual reduction in its agricultural surface area has caused a slight decline. The move to an agricultural model based on the application of external inputs has made farmers less reliant on biological N sources. In the 1961–1965 period, 15 % of arable land was devoted to N-fixing crops, whereas this proportion has now been reduced to 8 %.

The most remarkable historical change in Spanish NANI corresponds to the increase in the net N import in the form of food and feed. In the early 1960s, Spain was a country close to agricultural self-sufficiency; not only net, but also total imported and exported N were very low. During the following five decades, Spain has completely expanded its N cycle and now the Spanish agro-food system depends to a large extent on other countries' production. As a result, the net N import has increased by 1133 % in the 1961–2009 period. This increase is much higher than that observed in Europe. In the 1986–2009 period, net imports to Spain increased 109 %, whereas in Europe increased only 8 % (Lassaletta et al. submitted). The large difference between N import and export is not only due to a higher amount of imports over exports, but also due to the nature of the commodities (Table 2). The five most imported products are high protein content commodities mostly devoted to feed (cereals and soybean products); they account for 76 % of the total imported N. On the other hand, high-quality products with very low protein content such as citrus fruits, wine, tomatoes, and olive oil are exported.

Expressed in terms of N, pork is the most produced animal product, with the fastest increasing trend; the second is cow's milk, but production has remained constant over the last 30 years; poultry products (eggs and meat) also show an increasing trend (Suppl 8). Galloway et al. (2007) point out that the growth in production of non-ruminants relative to ruminants is due to a reduction in the price of feed grains together with higher conversion efficiencies. The increasing demand for imported feed can be observed in absolute terms but also in the proportion within the total N import; 85 % of protein imports are now used for feed compared to about 60 % in the 1960s (Suppl. 9). The current dependence on external production is such that the amount of proteins imported from abroad is now equivalent to that of the total production of Spanish crops (Fig. 3).

Asmala et al. (2011) have recently shown how European Baltic countries are also currently net importers of nitrogen. We studied the trajectories of the net N traded in several European countries for the years 1960 and 2007 (Fig. 4). France is the only country that has evolved towards an autotrophic character, i.e. exporting more than it imports, although the UK and Germany show a similar trend. The Netherlands is clearly the country with the highest increase in the per capita net N import. Spain, along

Table 2 Characteristics of the commodities exported and imported to Spain in three periods expressed in weight and in nitrogen

	1961–1965			1981–1985			2005–2009		
	Commodity (% N)	10^3 tons	Gg N	Commodity (% N)	10^3 tons	Gg N	Commodity (% N)	10^3 tons	Gg N
Export	Oranges (0.18)	947	1.7	Oranges (0.18)	792	1.4	Tangerines (0.18)	1526	2.7
	Tomatoes (0.14)	205	0.3	Tangerines (0.18)	679	1.2	Wine (0.00)	1458	0.0
	Wine (0.00)	186	0.0	Wine (0.00)	570	0.0	Oranges (0.18)	1314	2.4
	Potatoes (0.25)	121	0.3	Soybean oil (0.0)	411	0.0	Tomatoes (0.14)	912	1.3
	Bananas (0.11)	106	0.1	Soybean cake (7.36)	380	27.9	Olive oil (0.00)	588	0.0
Import	Maize (1.52)	845	13	Maize (1.52)	4257	65	Wheat (1.95)	5436	106
	Wheat (1.95)	463	9	Soybeans (6.08)	2675	163	Maize (1.52)	4929	75
	Barley (1.76)	429	8	Sorghum (1.61)	610	10	Soybean cake (7.36)	3405	251
	Potatoes (0.26)	164	0.4	Barley (1.76)	548	10	Soybeans (6.08)	2733	166
	Sugar Ref (0.00)	146	0	Soybean cake (7.36)	529	39	Barley (1.76)	999	18
Total N	Total export	178 Commodities	7		309 Commodities	65		393 Commodities	160
	Total import	213 Commodities	60		312 Commodities	324		400 Commodities	812
		Net N input import–export	53			259			652

Export and import commodities correspond to the most traded at each time. The lower part presents the result of the total calculations including all commodities

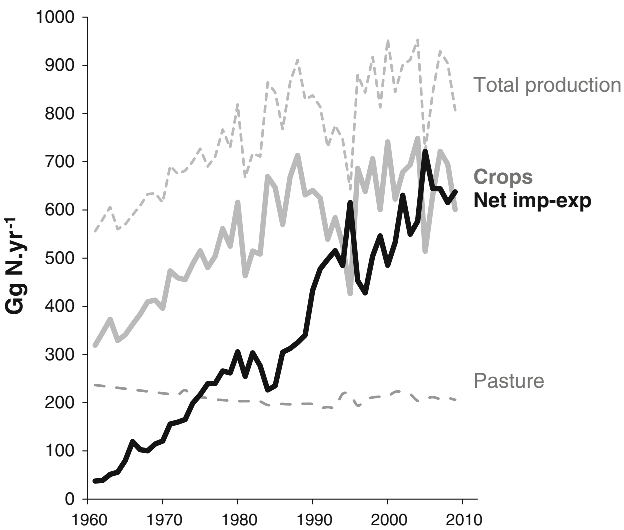


Fig. 3 Spanish production of crops and pastures compared with net N import in the form of food and feed. Crop production includes information on 117 cultivated species and pasture production on four grassland types

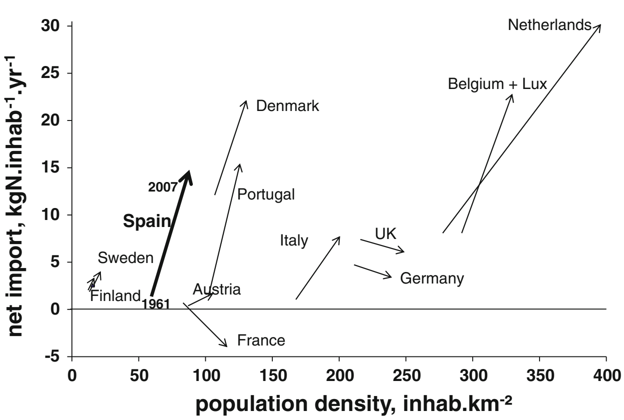


Fig. 4 Evolution of the net import associated with international trade in several European countries for the 1960–2007 period. The beginning of the arrow corresponds to the 1960 situation and the end to the 2007 situation. Points above 0 mean heterotrophic character and below 0 autotrophic character

with Belgium, Portugal, and Denmark, shows the highest heterotrophic increase over the last five decades. Countries such as the Netherlands, Denmark, and Belgium are important net exporters of animal products, but this production is, however, sustained by the imports of large amounts of feed.

Figure 5 shows Spain's net N export or import to all world countries for the year 2007. Portugal is one of the few countries importing significant amounts of N from Spain (12 Gg N year^{-1}); Italy, Turkey, Saudi Arabia, and the Maghreb are also importing from Spain, but small amounts. Most other countries are net exporters of N to Spain, the most significant ones being Argentina, Brazil, the USA, and France (295, 214, 69, and 40 Gg N year^{-1}, respectively); 52 % of the net N input to Spain corresponds to soy products for feed; 89 % of soybean cakes come from Argentina, and 82 % of soybeans come from Brazil.

The Spanish hydrologic budget

The riverine flux of N leaving the Spanish territory has been estimated for the 2000–2010 period at 114 Gg N year^{-1}, i.e. 225 kg N km^{-2} year^{-1}. Considering that the average NANI for the same period was 3314 kg N km^{-2} year^{-1}, this means that 6.5 % of the total net N input to the Spanish territory is exported by the rivers, the remaining 93.5 % being retained or eliminated inside the country. This is a very high retention value when compared with US or European temperate catchments, which retain 25 % and 50–80 %, respectively (Howarth et al. 2002; Billen et al. 2011). Lassaletta et al. (2012) found similar high retention values that were related to the highly regulated hydrology characteristic of dry Mediterranean catchments. In the northern Spanish catchments with a temperate climate, such as the Miño Basin, the N export is higher (520 kg N km^{-2} year^{-1}), while in some catchments of the middle-southern part of the Iberian Peninsula such as the Segura, Júcar, Guadiana and Tagus, river N exports are lower (0.5, 22, 89, and 151 kg N km^{-2} year^{-1}, respectively) (Fig. 6). The latter catchments have been recently classified as "streams under water stress" by the Water Exploitation Index (EEA 2012). We do not have information on riverine N fluxes for the 1960s; however, since water in these catchments was not yet so highly regulated, the proportion of N retained was probably lower at that time.

The Spanish hydrologic agro-food system

Figure 7 summarizes the N transfers through the Spanish hydrologic agro-food system for three periods. It illustrates the functional links between arable land, livestock, domestic human consumption, and the hydrosystem. The major changes revealed by our data for the 1960s to the 1980s period are the increase in the net import of feed and the use of synthetic fertilizers. Livestock numbers increased during this period together with its productivity, but animals were still raised for the most part on domestic grass and feed production. The second period, from the mid-1980s to the present, shows the greatest increase in livestock numbers, now more than 50 % fed by imported feed, which doubled in magnitude. Export of vegetal products increased during the whole period from the 1960s, but remained at a low value because of the low N content of the products concerned. Because of the current high animal protein diet, Spanish domestic consumption of

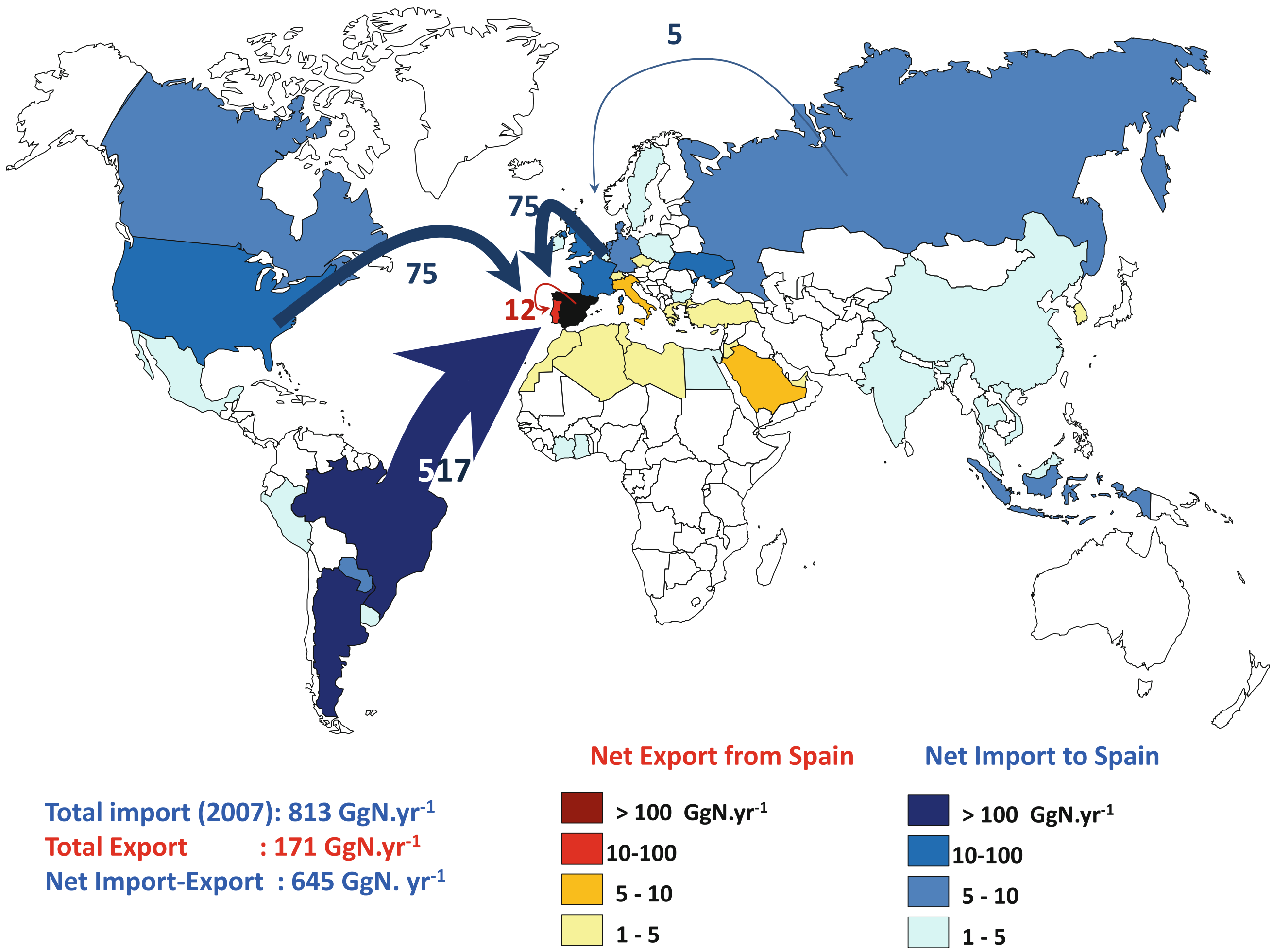


Fig. 5 Net N traded from Spain to world countries

animal products (132 Gg N year^{-1}) far exceeds their export (34 Gg N year^{-1}, mainly beef and pork). Since 29 Gg N year^{-1} (mainly dairy products) are imported, only 5 Gg N year^{-1} are finally net exported. N agricultural surpluses increased substantially during the first period and stabilized during the second period, in contrast to the situation observed in Switzerland where surpluses were reduced 27 % during the same time period (Spiess 2011). In 2010, cereals used for bioethanol production (wheat, maize, and barley) accounted for ca. 2 % of feed import and 1 % of national production. Since almost all N contained in energy crops is finally used for feed, these data have not been included in the diagram.

Discussion and conclusion

During the last 50 years, a significant change in the N cycle of Spain has occurred, with a threefold increase in NANI from 526 Gg N year^{-1} in the 1961–1965 period to 1673 Gg N year^{-1} in the most recent years (2005–2009). Food and feed import now accounts for ca. 40 % of the total NANI compared to 10 % in the early 1960s. In particular, it should be noted that nowadays imported agricultural products equal the total N production of domestic Spanish crops. The main part of the N import corresponds to soybean feed products, mainly originating from Brazil and Argentina. The Spanish N cycle is currently mostly driven by livestock production, 80 % of which is consumed domestically. The increase in population per capita protein consumption and the share of animal protein in the diet that have occurred over the last five decades are therefore the main factors responsible for the changes observed.

Besides livestock farming, Spanish agriculture has progressively specialized in the production of low N content commodities for export, and the N losses resulting from this production therefore remain in Spain. Further, only 114 Gg N year^{-1}, i.e. 6.5 % of total NANI, is transferred outside of the country's borders by the river flow, so that an exceptionally high proportion of the N that enters Spain annually is retained or eliminated inside the country (93.5 %), with several problems in terms of freshwater

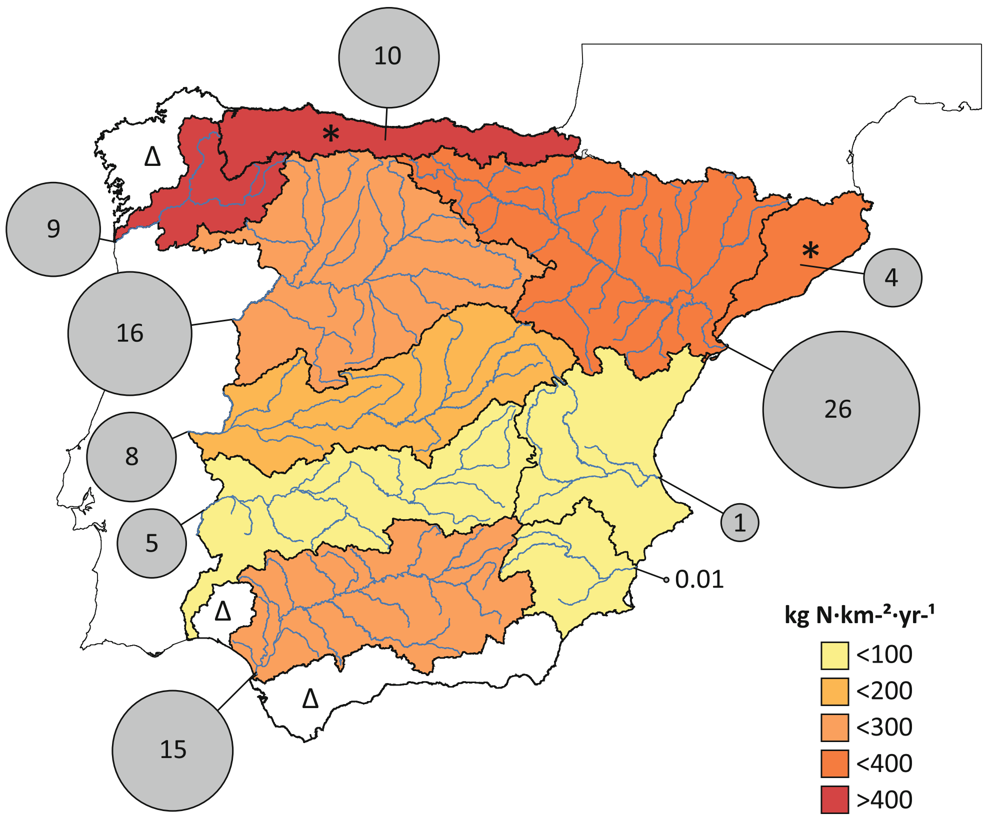
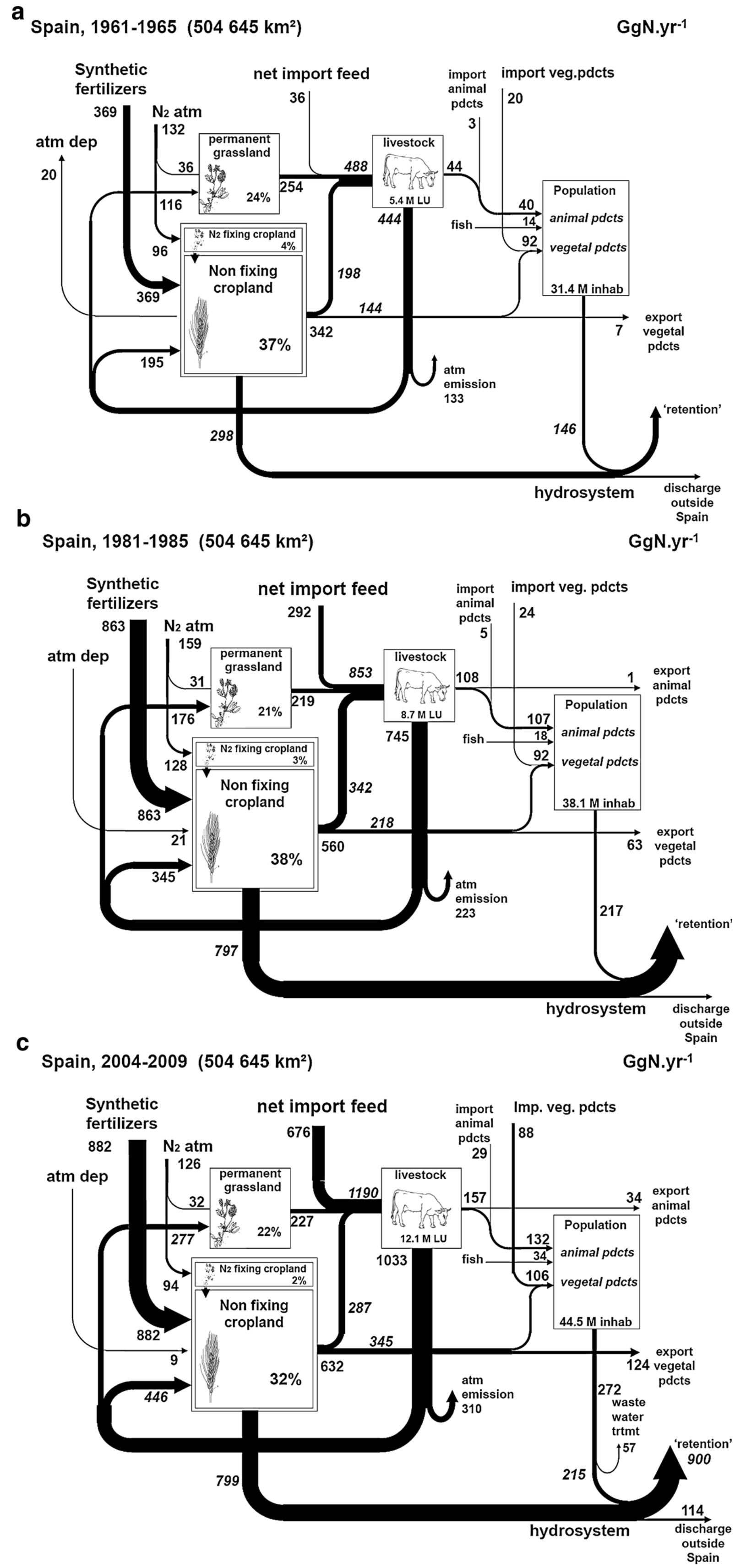


Fig. 6 N export from the main Spanish catchments. *Grey circles* show the riverine flux of N leaving the Spanish borders for 2000–2010 expressed in Gg N $year^{-1}$. The colour of the catchments represents the N export in units of mass per unit area. (*Asterisk*) These values correspond to the addition of the main rivers in this part of the territory. In Catalonia, four rivers (Ter, Tordera, Besòs, and Llobregat). In the Cantabric area, 18 rivers (Nalón, Navia, Sella, Deva, Saja, Nervión among some others). (*Delta*) No data could be obtained for these areas, and N export was estimated assuming the mean export value of similar geographical and climatological regions. See Materials and Methods section for details

quality and atmospheric pollution. This exceptionally high retention of the NANI within the country produces lower impacts in the sea, resulting in fewer coastal eutrophication problems than in Northern European seas (Romero et al. 2013). However, increasing nitrate concentrations associated with agricultural activities have been observed on many Spanish streams from headwaters to the river mouth (Arauzo et al. 2008; Lassaletta et al. 2009, 2010; Monteagudo et al. 2012). Aquifers have also been highly affected by nitrates (Arauzo et al. 2008, 2011). Indeed, 17 % of the Spanish land surface area has been declared nitrate vulnerable zones (ca. 50 % of the aquifers). Given that in Spain 80 % of drinking water comes from aquifers (Grizzetti et al. 2011), the loss of this resource entails enormous consequences in terms of water availability for human consumption. Overexploitation of water resources together with climate change could increase the effects of water regulation practices in some Spanish regions (Fleskens et al. 2012), exacerbating this situation in a near future (Fleskens et al. 2012; Stigter et al. 2012). Negative consequences of the atmospheric emission of reactive nitrogen such as biodiversity loss, N_2O emissions, or atmospheric pollution have also been reported for Spain (Maté et al. 2010; Ariño et al. 2011; Aguilera et al. 2013).

The low nitrogen export to the sea observed in several Mediterranean rivers (Lassaletta et al. 2012; Romero et al. 2013) is related not only to a lower N input into the agricultural land than that observed in temperate European catchments (Grizzetti et al. 2012; Leip et al. 2011) but also to a higher N retention inside the Mediterranean catchments that is related to the characteristic water management in this less humid area (Lassaletta et al. 2012; Bartoli et al. 2012; Castaldelli et al. 2013). These Mediterranean landscapes have a high density of irrigation channels and dams that produce very high nitrogen retention in the continental system. In this paper, we have not estimated the value of N retention during the first two periods because the information required was unavailable. However, since in Spain the irrigated area has been progressively expanded, we hypothesize that the proportion of N retained in the continental part was lower during the first decades analysed in this study than nowadays.

Fig. 7 Major nitrogen fluxes through the Spanish agro-food system at three moments in the last half century (1961–1965, 1981–1985, 2004–2009). The width of the *arrows* is roughly proportional to the N fluxes, expressed in Gg N year^{-1}. *Figures* in italics are calculated by difference. All others are independent estimations, explaining some minor discrepancies in the fluxes when calculated from different sources (agricultural statistics and trade statistics, see Materials and Methods). The overall picture provided is, however, completely coherent for each of the three periods



In addition to the local effects of the Spanish N cycle perturbation, corresponding to the environmental externalities of the agricultural production remaining in the country, a high amount of virtual N is also being emitted by Spain in the countries specialized in feed production for export to Spain (Burke et al. 2009). Although Spain is certainly quite different from Japan, for which Galloway et al. (2007) have estimated that only 32 % of N release associated with the Japanese food production system is generated in Japan, a significant share of the environmental N losses associated with the Spanish production system occurs in other countries, particularly soybean cultivation in South America (Filoso et al. 2006; Smaling et al. 2008).

In addition to the delocalization of environmental burdens, the reliance on imported external production of inputs, together with the high dependence on fossil fuel-based inputs such as mineral N fertilizers, implies a high vulnerability of the Spanish agro-food system to market fluctuations in a context of peak oil and increasing resource scarcity. As we have seen, however, the majority of this dependency does not occur to satisfy human needs but to maintain a very high animal protein consumption in the diet which is actually above WHO recommendations (WHO 2007). Therefore, a change in diet towards a return to the traditional Mediterranean diet, with a lower animal protein content, would be a win–win situation implying the reduction in N cycle perturbation within and outside Spain, the reduction in the vulnerability of the agro-food sector, and positive public health outcomes.

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