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Nitrogen Flow Analysis in Spain: perspectives to increase sustainability

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Abstract

Nitrogen (N) is a macronutrient that, together with P and K, is vital for improving agricultural yields, but its excessive use in crop fertilisation and presence in treated wastewater and sludge are generating emissions both into the atmosphere and into natural water bodies, which leads to eutrophication events. The Haber–Bosch process is energy-intensive and it is the main chemical route to produce reactive nitrogen for the production of fertilisers. Furthermore, there is a strong dependence on imports of reactive nitrogen in Spain and Europe. For these reasons, it is necessary to propose sustainable alternatives that allow solving environmental and supply problems, in addition to proposing efficient management schemes that fit into the circular economy approach. In this context, a nitrogen flow analysis (NFA) was carried out for Spain with the year 2016 as reference. To assess some interactions and flows of N, specific sub-models were also considered for the agriculture and waste management systems. For the food and non-food flow systems, country-specific data were considered. The sectors covered were crop production (CP), animal production (AP), food processing (FP), non-food production (NF) and human consumption (HC). The results reveal a total annual import of 2142 kt N/y, of which 43% accumulated in stocks of soils and water bodies (913 kt N/y). The largest proportion of losses was associated with emissions from agriculture (724 kt N/y to water bodies and 132 kt N/y accumulated in soils), followed by industry emissions to the atmosphere (122 kt N/y). Wastewater treatment plants (WWTPs) received around 67 kt N/y, of which 26% was removed as biosolids and 20% of these biosolids were recovered to be used for fertilising applications. The 49 kt N/y discharged in the final treated effluent represented 79% of the total loss of reactive nitrogen to water bodies. In addition, an analysis of N-use efficiency and the actions required for its improvement in Spain, as well as the impact of the current diet on the N cycle, was carried out.

Keywords: Substance flow analysis; resource recovery; circular economy; sustainable resource management; reactive nitrogen.

1. Introduction

The human population has been growing continuously from the industrial revolution to the present time (e.g., an annual growth rate of 1.05%) (FAO, 2013). Therefore, to meet the growing demand, many agricultural procedures have changed drastically in the past century (Brodt and Ingels, 2011). In addition, the global trend of concentrating populations in dense urban nodes and the accumulation of livestock in nodes of intensive integration has led to a large flow of nitrogen-containing compounds from anthropogenic activities wasted in the environment in the form of gases, aqueous dissolved species and solid forms (Schlesinger, 2009). This acceleration of the nitrogen cycle is not only unsustainable at the level of environmental resources but is also very harmful to both natural ecosystems and humans (Stevens, 2019). Harmful effects of excess nitrogen (N) in the environment, i.e., atmosphere, water and soil, are biodiversity reduction, ecosystem degradation, climate change, photochemical smog and groundwater and drinking water pollution, which have been critically reviewed (Pan et al., 2019a).

Before the 20th century, the N cycle regulated itself along with the operation and functioning of natural ecosystems by biological N fixation (BNF), lightning N fixation (LNF), N deposition and denitrification (Luo et al., 2018). Since the development of the Haber–Bosch nitrogen fixation (HBNF) process, a considerable amount of reactive nitrogen (N_r), grouping all N species other than N_2, has been incorporated into terrestrial ecosystems (raising its value 14 times from 1890 to 2010) to ensure global food security and meet the food demands of approximately 48% of the world's population (FAO, 2022). Thus, N cycles in terrestrial and marine ecosystems have been greatly altered (Luo et al., 2018). In addition, inefficient use of fertilisers causes significant loss of nutrients (Sutton et al., 2013). Around 80% of N and 25–75% of phosphorus (P) are lost to the environment through run-off, leaching and off-gas emissions, causing environmental impacts such as eutrophication and global warming and leaving insufficient nutrients in the soil for crops (Jakrawatana et al., 2017; Sutton et al., 2013).

In contrast to other essential nutrients such as P, N is abundant in the atmosphere in the form of gas ($N_2(g)$). The problem lies in the fact that the conversion of inert N gas ($N_2(g)$) to its reactive forms is extremely energy-intensive and fossil fuel-dependent (Moomaw, 2002). Some studies point out that the Haber–Bosch process used for the production of synthetic N fertiliser entails 2.5% of the global fossil energy usage and implies the production of 4–8 tons of CO_2 eq. per ton of synthesised N fertiliser (Beckinghausen et al., 2020; Coppens et al., 2016a).

The impacts of N_r on natural ecosystems, (e.g., atmosphere, water bodies and soils) and human health ecosystems and the correlation between N_r and climate change have been critically reviewed (Erisman et al., 2013). The study concluded that although there is strong evidence for the cascade of N_r effects, better data are needed to quantify the components of the cascade to best support policy options. When talking about alterations in the natural N cycle due to anthropogenic activities, scientific research points to the popularisation of the application of inorganic fertilisers to agricultural soil as the main driver (Vitousek et al., 1997).

According to the FAOSTAT database on N fertilisers, in 2016, China was leading the market, especially in the production sector (Kahrl et al., 2010). This indicator agrees with the fact that Asia comprises a full 30% of the world's land area with 60% of the world's current population. Other regions such as America and Oceania are more reliant on external sources to meet the internal demand. Africa is an exceptional case. Due to historical, climate and economic reasons, some regions of the continent face scarcity of food and water, which has led to distinct challenges in the agricultural production field ("Land and environmental degradation and desertification in Africa," 2021). Europe shows particularly high rates of import and export of N fertilisers, although domestic production is sufficient for the agricultural needs of the region (Van Egmond et al., 2002). The estimated consumption of mineral N fertilisers in the EU-27 has remained around 10 million tons in the last 10 years, with fluctuations and a slight upward trend ("Eurostat," 2021). On the other hand, there is a decreasing trend with a period of stabilisation for the gross N balance in agricultural land in the EU-28 since 2004 ("Eurostat," 2021). The countries with the highest N fertiliser consumption are, in order, France and Germany. Following on the list, with similar amounts of around 1 million tons in 2017, stand Poland, the UK, Turkey and Spain ("Eurostat," 2021).

2. Literature review

Spain is an important exporter of agricultural products worldwide, but above all in the European market ("Spanish Agri-Food Exports Increase by 97% in Last Decade," 2019). In the early 1960s, Spain was almost self-sufficient in terms of food and feed supply. In the first stage of the 21st century, net imports of agricultural products equalled crop production expressed in terms of nitrogen content (650 Gg N/y) (Lassaletta et al., 2013).

This demonstrates a great dependence on external markets to satisfy the national demand for fertilisers ("Estadística de consumo de fertilizantes en la agricultura," 2021); with accumulation points along the N flows, these N-based compounds could be recovered, thus reducing the rapidly increasing import trend. Recovery of N from alternative sources (i.e., wastewater, manure or food waste) could serve not only as a national approximation to the circular economy approach but also as a way to reduce external dependency in case of price fluctuation and promote N_r recovery options from urban, industrial and agricultural cycles. Element flow analysis (EFA) has been applied to track nutrient flows and manage nutrients in several applications at a regional scale (Baroi et al., 2020; Cordell et al., 2009; WangShou et al., 2016; Wu et al., 2016). In addition, the management of nutrients along the supply chain has been evaluated and a new method of nutrient footprint has been introduced by Gronman et al. (2016) and others (van der Wiel et al., 2020; Xu et al., 2020; Zhang et al., 2021). A review of the state of the art identified a limited number of studies specifically focused on nitrogen, which are listed in Table 1.

Table 1 Summary of the most relevant Nitrogen Flow Analysis published in the last two decades

Element	Year	Area	Approach	Ref.
N	1998	Huizhou (China)	Urban	(Ma et al., 2007)
N	2000-2016	Beijing (China)	Urban	(Pan et al., 2019b)
N	2002	Illinois (USA)	Agricultural	(Singh et al., 2017)
N	2004-2014	France	Agricultural	(Billen et al., 2018)

N	2015	Scania (Sweden)	Regional	("Nitrogen flow in Scania - Epsilon Archive for Student Projects," 2015)
N	2010	Maeklong river (Thailand)	Regional	(Pharino et al., 2016)
N	2011	Bangkok (Thailand)	Urban	(Buathong et al., 2013)
N	2014	Thailand	Agricultural	(Suesatpanit, 2017)
N, P	2004-2007	Finland	National	(Antikainen et al., 2008)
N, P	2009	Flanders region (Belgium)	Reginal	(Coppens et al., 2016b)
N, P	2014	St. Eustatius (NL)	Agricultural and urban	(Firmansyah et al., 2017)
P	2012	Spain	National	(Álvarez et al., 2018)
N,P,K,Mg	2021	Okanagan (Canada)	Regional	(Harder et al., 2021)
N,P	2022	Sweden	National	(Sinha et al., 2022)
N	2021	China	Agricultural	(Jin et al., 2021)
N	2020	Xiamen (China)	Coastal City	(Li et al., 2020)
N	2021	Shanghai (China)	Food system	(Liao et al., 2021)
N	2001	Catalonia (Spain)	Regional	(Bartrolí et al., 2001)
N	2005	Catalonia (Spain)	Regional	(Bartrolí et al., 2005)
N	2012	Ebro River Basin (Spain)	Regional	(Lassaletta et al., 2012)
N	2013	Spain	National	(Lassaletta et al., 2013)

This research aims to perform a nitrogen flow analysis (NFA) to determine the feasibility of i) recovering this element from the several accumulation points within its material flow cycle or ii) defining actions to promote the reduction of its consumption. The geographic framework is the Spanish territory contained in the Iberian Peninsula excluding the Balearic and Canary Islands, as well as the Spanish cities in the African continent, Ceuta and Melilla. The year 2016 was selected as the reference year due to it being the latest date on which some of the official sources provided information. Nitrogen input and output data were gathered from public databases. STAN software was used as a tool to develop the NFA and an uncertainty analysis was also included. N recovery pathways were further discussed as a way to promote the transition to the circular economy approach. The results were compared with the few examples published for cities and regions, including the only two specific regional cases in Catalonia (NE Spain) (Bartrolí et al., 2001, 2005).

3. Methodology

EFA has been widely used to assess resource flows such as energy, water or minerals at any geographical scale (from global to local) (Graedel and Allenby, 2009) which is useful for providing relevant information to develop regional management strategies (van der Voet, 2015). The methodology was established to develop environmental management tools by assessing the technical (technosphere) and human (anthroposphere) metabolisms. It is based on a) mass balance, which enables a systematic assessment and tracking of flow materials (e.g., N) considering the inputs, transformations and the outputs within the system (van der Voet et al., 1995) and b) system analysis, consisting of a three-step procedure: (i) definition of the system, (ii) quantification of the overview of stocks and flows and (iii) interpretation of the results (Baccini and Brunner, 2012). To determine the unknown data, the algorithm follows a sequence of calculations defined by equations 1–4 (Cencic and Rechberger, 2008):

Balance equation: $\sum inputs = \sum outputs + change\ in\ stock$ (1)

Transfer coefficient equation: $output_x = transfer\ coef_{to\ output\ x} \cdot \sum inputs$ (2)

Stock equation: $stock_{Period\ i+1} = stock_{period\ i} + change\ in\ stock_{period\ i}$ (3)

Concentration equation: $mass_N = mass_{good} \cdot concentration$ (4)

The entire process is summarised in the general flow chart depicted in Figure S5 (Supplementary Material). The first step is to define the system and its boundaries. Then, all the flows involved need to be identified and quantified using the specific databases as detailed in the Supplementary Material (Table S2). The data were extracted from different databases and classified (listed in Tables S3–S8) to finally compose the list of all 29 flows characterised for Spain as summarised in the Supplementary Material (Table S9). This dataset is introduced in the model previously defined in STAN as defined by Cencic and

Rechberger (2008) to obtain the results of the mass balances and according to the definitions of flows, stocks, process, system, etc., collected in the Supplementary Material.

3.1 Studied area and boundaries

The NFA was developed for the Spanish territory, which is organised into 16 autonomous communities and excludes 3 other communities of the Balearic and Canary Islands, as well as two cities on the African continent (e.g., Ceuta and Melilla). The N flows (in kt N) in different forms were targeted and divided the NFA into several subsystems. This focused primarily on agriculture and food production systems, as N consumption as fertiliser accounts for 43% of the total N imported across national borders ("Productos fertilizantes," 2021), but it also considered other industrial uses such as nitrogen in fertilisers and chemicals. The flows associated with the recovery of N from wastewater and the application of urban and farming biosolids to agriculture were also considered in the analysis. The period selected was one year, 2016, which was the latest year for which accurate information regarding most flows could be gathered.

The model was developed using STAN 2.6, software developed by the Technische Universität Wien that allows the creation of graphical models with predefined components (processes, flows, system boundary, text fields), as well as the development of material flow analysis through mathematical-statistical tools such as data reconciliation and error propagation (Cencic and Rechberger, 2008).

Before the development of an NFA, a good understanding of the element of interest and its behaviour in the defined space and boundaries was required. To understand N in a Spanish context, different subsystems and processes were considered. As a starting point, the N cycle in a natural environment without human intervention was reviewed (Antikainen et al., 2008; Mengel et al., 2001). The formulation of this cycle is shown in Figure S1 (Supplementary Material). It is a relevant issue to identify all these compounds because they constitute large flows of N on a national scale. The final definition of the NFA will be based on the successful determination of these flows regardless of the weight contribution of N in the associated N compounds. Consequently, the main relevant processes to consider to estimate the N flows between systems are nitrogen fixation, ammonification, nitrification, denitrification and lightning, by which N is fixed from the atmosphere to the soil. In this sense, legumes play a key role in sustainable agricultural intensification by providing a source of edible protein for humans and livestock, making this family the second most cultivated crops on the planet after cereals. Therefore, the introduction of different species of legumes in cropping systems is of special relevance in order to: i) stabilise food production over time (Renard and Tilman, 2019), ii) contribute with nitrogen from BNF (Jensen et al., 2020; Peoples et al., 2009), reducing the environmental footprint of the N fertilisation practice (Jensen et al., 2012), iii) assist with the control of pests and diseases (Voisin et al., 2013) and iv) improve farming profitability (MacWilliam et al., 2014).

3.2 Model description

The final system comprises 29 flows and 6 subsystems. Regarding subsystems, the following were considered for calculations: agriculture, households, industry, transport, waste management and water bodies. The two most relevant stocks identified within these subsystems are agricultural land and water bodies.

Agriculture is the system with the greatest theoretical relevance in the NFA and comprises crops, livestock, forestry and pasturelands; the most important attribute of this subsystem is that it includes all the soil used for the aforementioned purposes. It should be noted that substances of this subsystem are in constant movement; therefore, it is necessary to approximate the quantification of certain flows. Households represent the majority of end-consumers of goods and services provided by the industrial sector. Consequently, the industry is a general category including all processes performed before the consumption phase except for transport, which has a subsystem on its own. Waste management is considered as an entity encompassing the largest urban waste management plants but also waste management within industries and agricultural waste management. Lastly, water bodies are all of those contained in the previously defined geographical boundaries, i.e., rivers, lakes, groundwater, etc.

Flows can be divided into those entering and exiting the system and those connecting two subsystems. The former are considered imports/inputs or exports/outputs depending on whether they enter or exit the geographical space; the latter are known as inner flows.

Imports and exports are key parts of the flow definition process. This NFA evaluates five groups of high N content goods: fertilisers, food, feed, fuel and non-food products. These substances are considered to enter or exit the established geographical boundaries. A scheme of imports and exports can be found in Figure S2 (Supplementary Material).

Non-food comprises listed chemicals in the global trade market containing significant amounts of N, mostly used for production purposes. Inner flows are more numerous and more difficult to classify. Based on their start and end points, they represent many unrelated substances that transport N. The most relevant inner flows in the model are the emissions to the atmosphere or water bodies in the specified area.

3.2.1 Auxiliary models for agriculture and waste management subsystems

Given the complexity of the flow quantification process, two auxiliary models were developed for agriculture and waste management subsystems to facilitate the build-up of the general model.

3.2.2 Agriculture model

The agriculture subsystem comprises three processes: i) crops, ii) livestock and iii) pastureland. Fishing and forestry were not considered due to lack of relevance compared to the three aforementioned processes. The inner flows accounted for are manure and fodder. The former comes from livestock and is applied to crops and pastureland; the latter follows the same path but the other way around. Out of the three processes, only soil (crops and pastureland that are considered a single process) appears as stock due to the fixation of N in the soil. Livestock animals do not appear as stock because, although they retain N in their bodies while they are alive, once they are slaughtered for human consumption, this N is transferred to humans in the form of protein, which appears as meat product leaving the boundaries of the subsystem. Imports and exports in the agriculture model are depicted in Figure S3 (Supplementary Material).

3.2.3 Waste management model

The waste management model comprises four end-of-life scenarios: i) wastewater treatment plants (WWTPs), ii) landfill, iii) waste treatment/composting and iv) incineration. Each scenario acts almost independently of the rest and many external flows, e.g., food waste, end up in end-of-life routes, distributed by fractions according to the waste management strategy defined in Spain. The only inner flow considered in this case is WWTP biosolids. It appears in WWTP and can be processed via incineration or used in cement industries as a substitute for coke and marginally sent for landfill, although this is banned by the EU regulation. The remaining flows are imports or exports from other subsystems as can be seen in Figure S4 (Supplementary Material).

3.3 Data collection

A key aspect of performing an EFA is the collection of the necessary data to quantify the flows of the system with a certain degree of reliability (Table S2). To fulfil this purpose, several sources of information were consulted (e.g., official statistical databases, published reports, surveys and interviews). The statistical data mainly contained the amounts of N compounds such as N-containing products, chemical fertilisers being applied to the field, crop harvests, milk production, sown crop areas, the number of livestock and the human population. Detailed information on how data were gathered and treated to fit the NFA can be found in the Supplementary Material (Section 3).

The availability of data for such a specific element (N) in the Spanish context is a challenge, especially in those systems where N concentration data are not collected or publicly accessible.

3.4 Data management and uncertainty assessment

One of the main concerns of EFA is the identification of potential errors and uncertainties. The diverse nature of sources and the varying quality and availability of data make NFA results inherently uncertain. In this work, the results have been cross-checked by using alternative estimates, comparing with values reported in the literature and making estimates of N mass balance when possible (Senthilkumar et al., 2012). In some cases, several estimates were made for the same point, and then these N flows were averaged (Bartrolí et al., 2005; Lassaletta et al., 2013).

Confidence ranges for NFA were obtained by using the HS approach developed by Seyhan (2009) and widely used in different EFA studies (Asmala and Saikku, 2010; Cooper and Carliell-Marquet, 2013; Seyhan, 2009). To evaluate the reliability of the results, the information used to quantify each flow was classified into four categories. Hedbrant and Sörme (2001) developed a method widely used for the assessment of uncertainty in EFA models. This method involves assigning uncertainty levels to various data sources, such as official statistics or values from the literature, and applying an interval to each level (Cooper and Carliell-Marquet, 2013). The intervals used in this study are summarised in Table S1 (Supplementary Material). Results, as incorporated into the STAN program, are depicted in Figure 1 along with the 95% confidence limits.

3.5 Assessment of N-use efficiency

N-use efficiency (NUE) is widely used as an indicator to assess N management in agriculture (Baligar et al., 2007; Reich et al., 2014). NUE is the ratio of the crop N uptake to the total N fertiliser input. It is also defined as the ratio between the N uptake of crops and the available N in the soil, which would include the N from applied fertiliser plus the residual mineral N in the soil. The Spanish NUE for the reference year was calculated according to European standards using Eq. 5:

$$NUE = \frac{Total\ plant\ N\ uptake}{N\ applied} \cdot 100 \qquad (5)$$

4. Results from the model

4.1 Main N flows in Spain

This section describes the results obtained for the NFA in Spain for the year 2016, intending to determine areas with an inefficient use of N, identifying the main losses and accumulations and estimating the dependence of Spain on imports of N-based chemicals and N-containing compounds. Figure 1 shows the results of the NFA using the STAN tool.

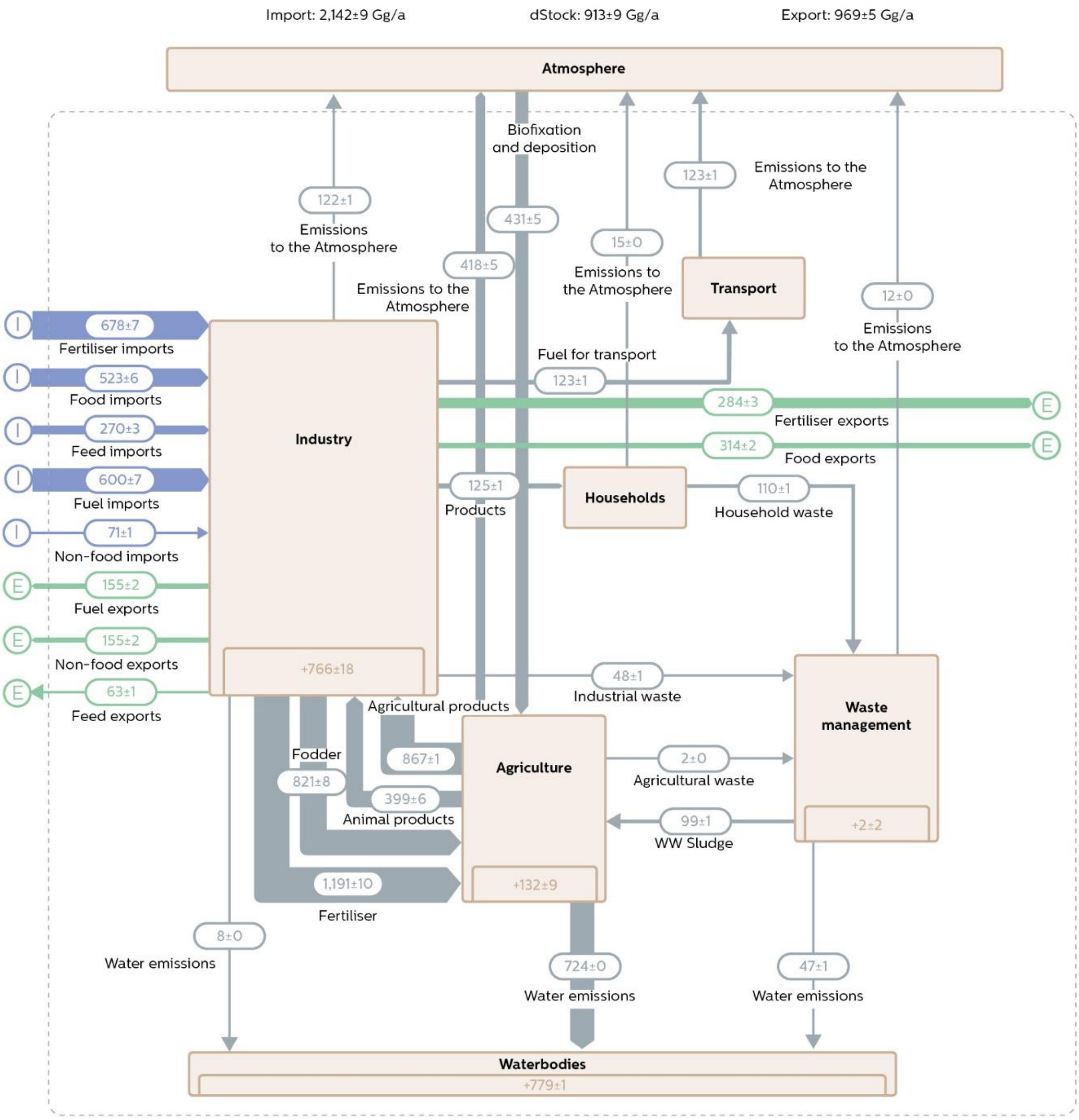

Figure 1 Nitrogen Flow Analysis for Spain with 2016 as the reference year

In Figure 1, the boxes represent the main processes and where stocks involving N are located, while the connecting arrows represent the main N flows. The N flows are presented in the Sankey format, in which the width of the arrow is shown proportional to the size of each flow. The quantity of each N flow (expressed in kt N/y) accompanied by the uncertainty is included in the circle along each arrow. A list of all the streams is summarised in Supplementary Material Table S2.

The NFA results point out that Spain in 2016 had a budget of 913 kt N/y (dStock) when only accounting for the accumulation in that year. This accumulation can be found in agriculture (132 kt N/y), water bodies (779 kt N/y) and dumpsters (2 kt N/y). N accumulates in soil, forests and in groundwater and landfills. There is a clear need for international products with a high N content. Among imports, fertiliser was the largest flow, closely followed by fuels and food. Exports appear to have minimal influence on the national N budget, with the most relevant export flow being that of fertiliser with 284 kt N/y.

From Figure 1, it can be seen that the greatest interaction between subsystems involves industry and agriculture. Industry is the subsystem with the most linked flows; however, agriculture concentrates the largest flows of N, the flow of fertiliser from industry to agriculture (1191 kt N/y) being one of the most remarkable. Since the amount of fertiliser that the industry produces is higher than the imports, it means that the industry is converting part of the agricultural products into fertilisers.

In 2016, 2142 kt N entered the system, while 971 kt N left the geographical boundaries. The strong relationship between the food industry and the anthropogenic intervention in the N cycle is clear as depicted in Figure S6 (Supplementary Material), which indicates that fertiliser, food and feed account for 89% of the annual N flows entering Spain's boundaries. When it comes to exports, the proportions change. The three aforementioned groups of goods represent 62% of the exported N. The most remarkable distinction between exports and imports is the amount of non-food N that is traded to other countries in comparison to the amount acquired by Spain. This is mainly because Spain exports more than twice the amount of N it imports.

When it comes to emissions to the atmosphere, a detailed table is provided in the Supplementary Material (Table S8) including the contribution of NO_2, NH_3 and N_2O to the different emissions to the atmosphere. It is noteworthy that the main N emissions to the atmosphere within industry and transport processes exceed 90% due to NO_2 emissions. These emissions come from the N content in the fossil fuels and industrial N_2 fixation through high-temperature combustion.

4.2 NFA for the agriculture subsystem

To have a broader understanding of the behaviour of N in the Spanish system, two auxiliary NFA models were developed, one of which focuses on agriculture and livestock as a theoretical framework. The review of the state of the art indicates that agriculture could be one of the most important subsystems when evaluating N flows in a given region (Senthilkumar et al., 2012). However, each territory has different weaknesses and strengths when it comes to nutrient management.

A diagram with the main results of the NFA for the agriculture subsystem is shown in Figure 2. The largest import of N to the system comes from fertiliser applied to agricultural soils.

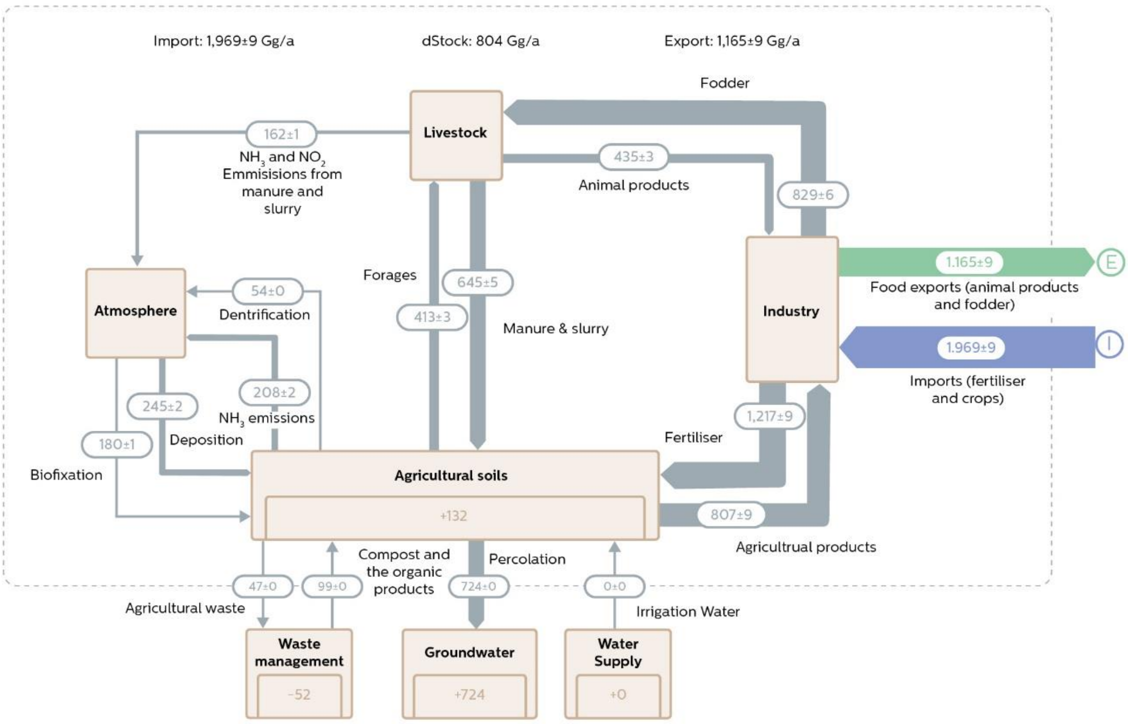

Figure 2. NFA for the agriculture subsystem for Spain with 2016 as the reference year

As can be seen in Figure 2, the imports, which are formed by different types of crops and fertilisers, are sent to their respective processes. The fertilisers go to the agricultural soils to produce forage, which is later combined with the fodder produced by the industry to feed livestock. The value of biofixation by leguminous crops, which accounts for 9% of N imports, is noteworthy.

Regardless, the accumulation is balanced by the fact that plant products accumulate a portion of N and this amount leaves the system once the crop is ready for harvest. Livestock fodder was another major nitrogen import for this subsystem. Livestock was not considered a stock system since once animals are slaughtered for meat production; the accumulated N is transferred to the consumer. A significant flow of N links livestock and cropland, given that manure is used as an organic fertiliser. Pastureland shows a higher stock, mainly attributed to the fact that the values of atmospheric deposition and biological fixation are similar to those assigned to cropland, but the transfer of nutrients from land to plant products is significantly lower when there is no agricultural activity.

As a final balance, in the subsystem defined as agriculture and livestock, imports and exports add up to 1969 kt N and 1165 kt N, respectively. Consequently, the stock of N in agricultural land and water bodies amounted to 132 kt N and 724 kt N in 2016.

4.3 NFA for the waste management subsystem

The second subsystem assessed was waste management. It consists of four main processes (end-of-life scenarios) in Spain (Figure 3). Composting seems to be the most widespread waste treatment process with the highest N flows, especially due to the high proportion of food waste in Spain biologically treated for reuse as fertiliser.

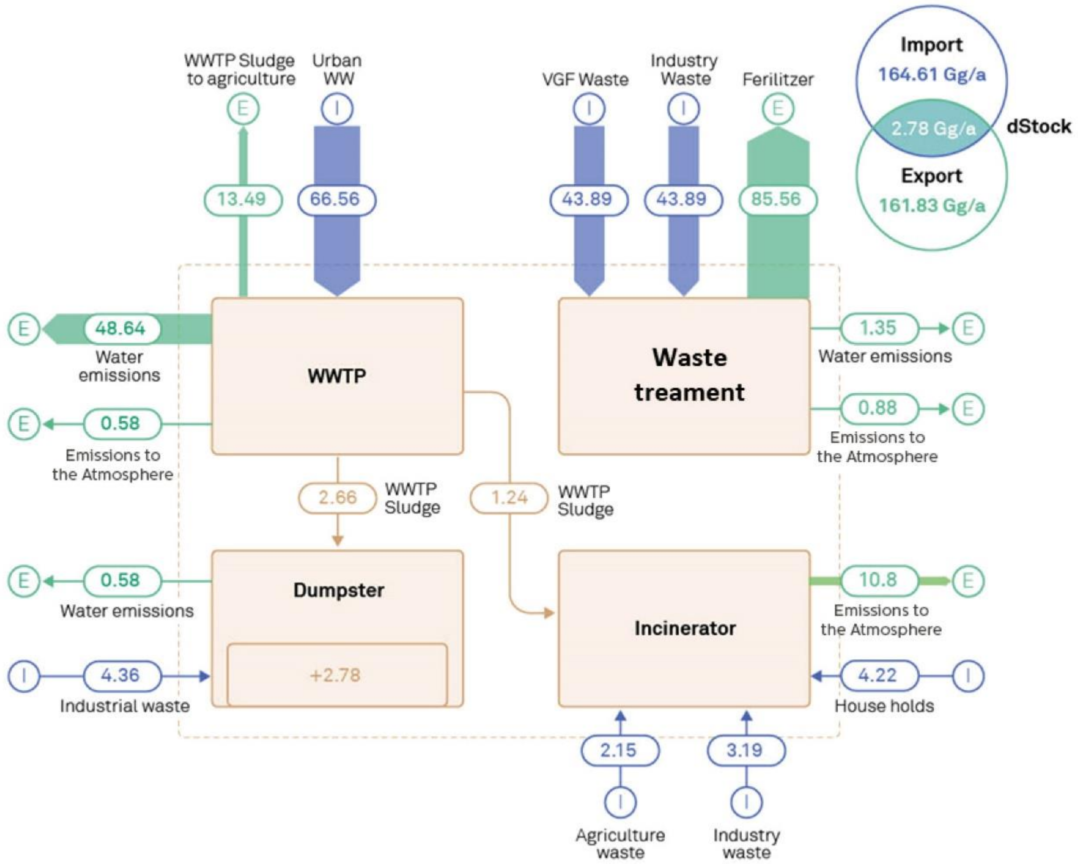

Figure 3. *NFA for Waste management subsystem in Spain with 2016 as the reference year*

The second most important process is that of WWTPs, which treat urban wastewater and, in some cases, such as in large metropolitan areas, mix urban and industrial wastewater. This wastewater usually has a high N content, mainly due to the contribution of the human excretion of urea (Hanson and Lee, 1971). WWTPs are one of the most recurrent secondary sources identified to implement material recovery strategies, especially for nutrient cycles (e.g., N and P) (Bernal et al., 2016; Guaya et al., 2016; Lebuf et al., 2012). WWTP effluents have also shown great potential, which is due to the large volume of treated water that contains a relatively low concentration of total N of approximately 15–60 g N/m^3. Consequently, efforts to reduce the concentration of N in waterworks that already carry out tertiary treatment are nowadays a costly management option. In addition, the volume of biosolids generated is less than that of the treated water, while the N concentration is up to 6–7 times greater than that of water, about 70–80 g N/kg. The fact that more than 75% of the biosolids is valorised in agriculture is having a great positive impact since 86 kt N is recovered as organic fertilisers. However, there are some doubts about the capacity of the crops to use them efficiently, as is the case for agricultural waste (Loyon, 2017), and future options are to promote the recovery of reactive nitrogen as NH_3(l) or liquid fertilisers (Vecino et al., 2019). Finally, landfills and incineration are widely used waste management options in Spain, but they do not imply particularly high N flows, apart from emissions to the atmosphere derived from waste incineration, which amount to 11 kt N.

It has been difficult to quantify the specific contribution of the agricultural sector to the N cycle; wastewater generated on farms, especially those producing pigs, follows management routes where it is spread on agricultural soils to benefit from the C, N, P and K contents. The continued intensification of livestock farming systems is increasing their total environmental impact, resulting in increased emissions of NH_3(g), greenhouse gases (GHG) and odours that derive from the housing, storage and application of manure and slurry in the field. In a recent study in Europe, it was estimated that animal manure is contributing up to 65% of total anthropogenic NH_3, 40% of N_2O and 10% of CH_4 emissions (Hou et al., 2017). Therefore, recovery of NH_3 from agricultural waste will be a priority and an opportunity to reduce Spanish ammonia imports. While the recovery of P from wastewater is mandated by regulation in countries such as Germany and Switzerland, regulation is expected to move forward to promote the recovery of nitrogen in the form of any reactive type of N. Research efforts are directed at recovering ammonium salts to be used as fertilisers and efforts are also directed to recover NH_3 (Vecino et al., 2019).

5. Discussion of results

Calculation of the net anthropogenic nitrogen input (NANI) was described by Lassaletta et al. (2013) for the agri-food system. The authors reported that the NANI in 2009 for Spain was 1673 kt N/y. In addition, the study provided the evolution of this value since 1961. Thus, taking into account the growth rate for this period and calculating the corresponding value for 2016, which would correspond to the period studied in this work, a NANI of approximately 1835 kt N/y would be expected.

In this study, first the total 'new' anthropogenic N input to the country, through the application of synthetic fertilisers (1191 kt N/y), net atmospheric inputs (245 kt N/y), BNF by crops (180 kt N/y) and net import of food and feed (479 kt N/y), was estimated, finally representing a NANI of 2095 kt N/y for Spain, 14% higher than expected by the growth trend of the data reported by Lassaletta (2013).

The current results were compared with those reported for other reference cases in Spain. Bartrolí et al. (2005) developed a study with the same compartments as defined in the present work, but only for Catalonia. Lassaletta et al. (2013) evaluated the agri-food compartment but for the entire Spanish territory.

Initially, the main N stocks were compared. Subsequently, the N loss flows (denitrification, NH_3 emissions, N discharged to water bodies) were analysed with respect to the total N inputs reported in these works for one year (426 kt N/y for Bartrolí et al. (2005); 1673 kt N/y for Lassaletta et al. (2013); and 2090 kt N/y in this study). Finally, other relevant flows were compared with total N inputs. All these results are collected in Table 2[1].

Table 2. Comparison of percentages of stock and flows with studies based on the Spanish territory (Bartrolí et al., (2005 and Lassaletta et al., (2013))

<table>
<tr><th></th><th>This study</th><th>Bartrolí et al., (2005)</th><th>Lassaletta et al., (2013)</th></tr>
<tr><td>Overall stock</td><td>53%</td><td>63%</td><td>54%</td></tr>
<tr><td>Stock in soils</td><td>10%</td><td>7%</td><td>5%</td></tr>
<tr><td>Stock in water bodies</td><td>37%</td><td>23%</td><td>59%</td></tr>
<tr><td>Denitrification</td><td>3%</td><td>7%</td><td rowspan="2">19%</td></tr>
<tr><td>NH_3 emissions (soil and livestock)</td><td>18%</td><td>13%</td></tr>
<tr><td>Water bodies</td><td>37%</td><td>23%</td><td>64%</td></tr>
<tr><td>Food exports</td><td>15%</td><td>37%</td><td>9%</td></tr>
<tr><td>Fertilizer imports to soil</td><td>55%</td><td>14%</td><td>53%</td></tr>
<tr><td>Atmospheric deposition</td><td>12%</td><td>3%</td><td>1%</td></tr>
<tr><td>Biofixation</td><td>9%</td><td>4%</td><td>15%</td></tr>
</table>

[1] These percentages are calculated by dividing the value of the flow by the total N input reported by the authors.

All these results indicate that overall N stocks within the country have been consistently reported to be around 50–60% of total N inputs. Furthermore, Lassaletta et al. (2013) reported that only 6.5% of N is transported out of the country through river flows, which allows N stocks to be achieved in soils and water bodies of between 50–60% of total N inputs. Other significant differences that can be found with the previous models are those related to food exports and the contribution of fertilisers. In this case, Bartrolí et al. (2005) reported different values but this is due to the fact that their study only considered the autonomous community of Catalonia, which is the fourth in terms of agriculture production ("Agricultura: valor de la producción por región en España," 2020).

Measures must be taken not only to improve N efficiency at the national level, but also to meet the SDGs in accordance with the 2030 Agenda for Sustainable Development. The areas that can contribute to a considerable reduction of N in the atmosphere are road transport (passenger cars), electricity and heating production, heavy-duty vehicles and buses. Some actions that could reduce these contributions are i) the reduction of NO_x emissions with the application of limitation policies aimed at the industry and transport sectors; ii) a paradigm shift in the electricity and heating sector; and iii) a change in the urban mobility model. In this sense, the penetration of electric vehicles in the market in the coming decades could make an important contribution to reduce N emissions.

5.1 Linking agriculture and waste subsystems

Analysis of the waste subsystem has shown its potential to produce fertilisers from waste products. In that case, 86 kt of N is valorised as a biofertiliser from industrial and domestic waste. However, in the case of WWTPs, there are no such synergies, although there is the same potential. This is due to several legal barriers that until now do not recognise recovered products like fertilisers, maintaining their status as waste.

Nevertheless, the European Commission has adopted a delegated act to include sewage sludge among the authorised input materials for fertiliser sold across the EU, paving the way for further investment in P recovery from sludge. The measure extends the Fertilisers Regulation adopted by the Commission in June 2019, which left the issue of sludge-based nutrients open. This was considered a temporary setback by those hoping for a breakthrough in the nutrient recovery market ("European Sustainable Phosphorus Platform - News Archive," 2022).

The provision approving sludge-sourced fertilisers has been added to the regulation, which will come into force in 2022. According to Brussels-based water lobby group EurEau, the new text allows P to be recovered from sludge as struvite (phosphate salts) and from incinerated sludge ash. The fertiliser thus obtained can be sold across borders within the EU Single Market ("Status of the Regulation (EU)," 2022).

This procedure has demonstrated the feasibility of obtaining fertiliser status for a recovered product, including struvite within the recovered materials that can be used in agriculture and thereby granting it the CE marking. The same procedure should be followed for other recovered nitrogen-based fertilisers, such as ammonium. In this sense, if struvite and ammonium salts are considered potential alternative sources of N, then WWTPs can be considered resource recovery facilities (Bolzonella et al., 2017; Lebuf et al., 2012; Vaneeckhaute et al., 2013; Verstraete et al., 2009).

Figure 3 shows that each year 49 kt N is discharged into water bodies, which represents 73% of the input flow and could be potentially recovered. In recent years, several technologies have been developed to recover nutrients from wastewater. The most common is the crystallisation of struvite, which allows for the recovery of both N and P or even potassium (K-struvite) (Vaneeckhaute et al., 2017). Stripping and absorption are used to recover ammonia, mainly as ammonium sulphate, but they are not implemented in WWTPs due to energy requirements and combinations of ion exchange with membranes to obtain ammonium salts (nitrate, sulphate or phosphate) because of their potential use as fertilisers. However, all these technologies require a high concentration of nutrients to be economically recovered. In a WWTP, these concentrations can only be found in the anaerobic digestion centrates, which can range from 500 to 1500 mg $N\text{-}NH_4/L$. The N present in these streams usually accounts for 10–20% of the total N in the influent of the WWTP (Vaneeckhaute et al., 2016). Considering N losses of 5% in the sludge, there remains a total of 5–10% of the total N in the influent that could be valorised as fertiliser, which accounts for 3–7 kt N/y. Considering the flows represented in Figure 3, the amount of N recoverable from the WWTPs represents from 0.2% to 0.5% of the total N produced as fertilisers in Spain. Although it may not seem huge in terms of global impact, it must be taken into account that this percentage range would allow for a fertiliser-producing company such as Fertiberia (www.fertiberia.es) to manage all the production and distribution of recovered products from WWTPs. In addition, several advantages associated with the use of recycled nutrients should be considered: i) a reduction in operating costs for WWTPs associated with less maintenance due to uncontrolled precipitation of struvite, ii) lower energy consumption in the aeration of the biologic reactor due to lower nitrogen load, iii) lower cost of sludge transport due to improved sludge dewaterability and iv) reduced carbon footprint for recovered fertilisers compared to conventional ones following the Haber–Bosch process (Basosi et al., 2014).

5.2 Assessment of NUE in Spain and actions to increase efficiency

Data from the agriculture NFA were used to determine the Spanish NUE value according to Eq. 5. This value considered the N uptake by the plant as the sum of N in plant products and fodder (1296 kt N) and applied N as the sum of the application of fertiliser, seeds and manure in crops, as well as N fixed to the soil through BNF or atmospheric deposition (1610 kt N), giving an NUE value of 71%. Regarding agricultural practices, the EU Nitrogen Expert Panel presented the possible objectives for the optimisation of N management in the *NUE Indicator Report* (Figure 4).

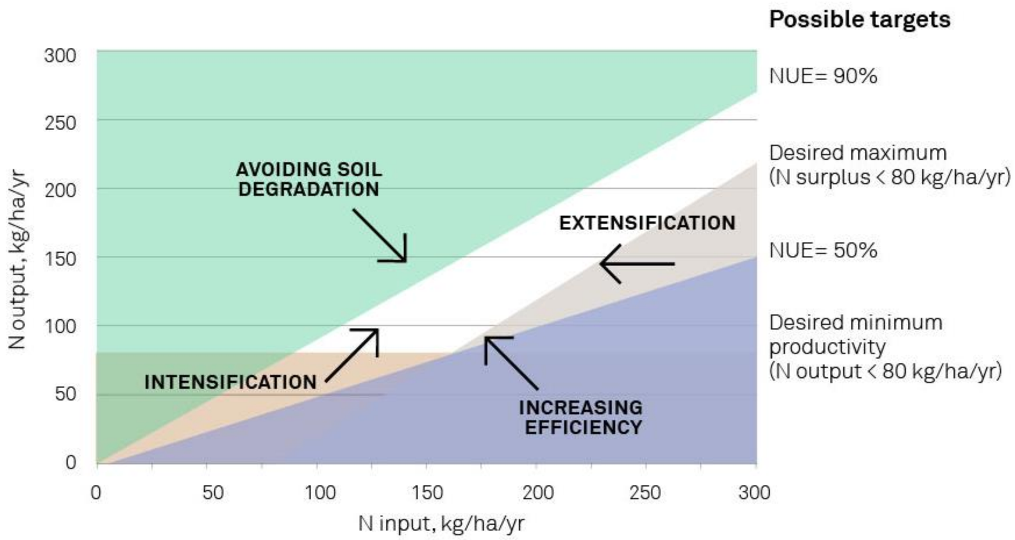

Figure 4. Evaluation of N outputs (kgN/y.ha) as a function of N inputs (kg(ha.h) under potential target indicators as a function of main directions of change in the N use efficiency (Nitrogen Expert Panel, ("Homepage | EU Nitrogen Expert Panel," 2014)*)*

The most desirable scenario for a country is to have an NUE between 50% and 90% (Panhwar et al., 2019). Lower values indicate inefficient use of N that can lead to externalities. On other hand, NUEs above 90% should be avoided to reduce the risk of soil mining. The Spanish NUE value for 2016 is within the established range; however, it being closer to the upper limit indicates that the efficiency could be improved but taking into account the risks of surpassing the 90% limit.

Although much of the work must be done at the farm scale, important policies need to be implemented at the national and multi-national scales, e.g., facilitating technology transfer and promoting agricultural innovation (Zhang et al., 2015). Additionally, improving NUE should be adopted as one of the SDG indicators used alongside crop yield and perhaps other soil health parameters to measure the sustainability of the agricultural sector. Countries should be encouraged to collect data on their N management in crop and livestock production. These data should be used to trace pathways of the three indices of agricultural N pollution, agricultural efficiency and food security targets (N_{sur}, NUE and N_{yield}). In the case of Spain, an Action Plan was presented through the *Integrated National Energy and Climate Plan 2021–2030* published in January 2020 ("National energy and climate plans | European Commission," 2021), where the Spanish government presented several measures related to N management in the country, such as i) the introduction of leguminous plants into crop rotation systems to improve N levels in the soil, its structure and fertility. Subsequent crops would require less nitrogenous fertiliser. At the current level, leguminous plants contribute 245 kt N yearly, which already represents 25% of the N contributed by inorganic fertilisers. Further increase in the quantity of N fixed by these crops would result in a lower inorganic requirement, not only contributing to reduce the carbon footprint associated with the use of inorganic fertilisers but also reducing external dependencies. Also, ii) the production of organic fertiliser using pig and cattle manure in areas with a high concentration of livestock was recommended. In addition, there was iii) the National Plan for Emissions Reduction, with the replacement of fossil fuels with renewable energies for electricity production. In this case, if there is a 20% reduction in fuel imports it would directly reduce industry emissions to the atmosphere, mainly due to the reduction in NO_x production.

5.2.1 NUE in food: a transition towards a sustainable diet

Quantification of the flows in the development of the NFA together with the state of the art indicate that the food industry is an important factor in the N cycle (Socolow, 1999). In the specific case of Spain, food waste is the indicator that places the country behind the rest of the EU member states and that could be key to taking a step forward in terms of the efficiency of N. For instance, according to the European food waste levels reported in 2016 ("Estimates of European food waste levels | FAO," 2016), Spain was listed as one of the countries providing data of insufficient quality; therefore, up-to-date information of relevant quality must be collected to identify sources of food loss and act to reduce the waste generated in this sector. It is important to highlight the need to establish policies for the progressive reduction of food waste as well as to promote the recirculation of all those by-products of the food production industry, which could lead to greater efficiency in the use of N.

In terms of livestock production, changes in feed composition can increase NUE in animals destined for human consumption without affecting the quality of human digestible protein in meat products (Cowling et al., 2001). The increasing use of synthetic fertilisers, together with other practices of agricultural intensification, has resulted in a considerable increase in agricultural productivity during recent decades (Tilman et al., 2002). However, a large part of the increase in primary agricultural production is used as animal feed (Pelletier and Tyedmers, 2010). A recent study, *Nitrogen on the Table* ("Nitrogen on the table: the influence of food choices on nitrogen emissions and the European environment | PBL Netherlands Environmental Assessment Agency," 2016), indicated that current average per capita protein intake in the EU was about 70% higher than that recommended by the World Health Organization (WHO). Spain also stands out as one of the countries with the highest meat consumption in the EU-27. This can be seen as an opportunity to experience a notable reduction of N pollution by pursuing a redistribution of the land dedicated to food production by limiting the average meat consumption in the national diet.

Using biophysical models and methods, the large-scale consequences of replacing 25–50% of animal-based foods with plant-based foods on a dietary energy basis have been calculated, assuming corresponding changes in production in the EU (Westhoek et al., 2014). The results showed that cutting consumption of meat, dairy and eggs in half would achieve a 40% reduction in N emissions, 25–40% reduction in GHG emissions and 23% less per capita use of cropland for food production.

In this sense, a sensitivity analysis was carried out to assess the impact of dietary changes on the overall NFA model. Assuming a 50% reduction in meat consumption implies a 50% increase in the N consumed through plant-based foods to compensate for the protein consumed through animal products, as can be seen in Figure 5.

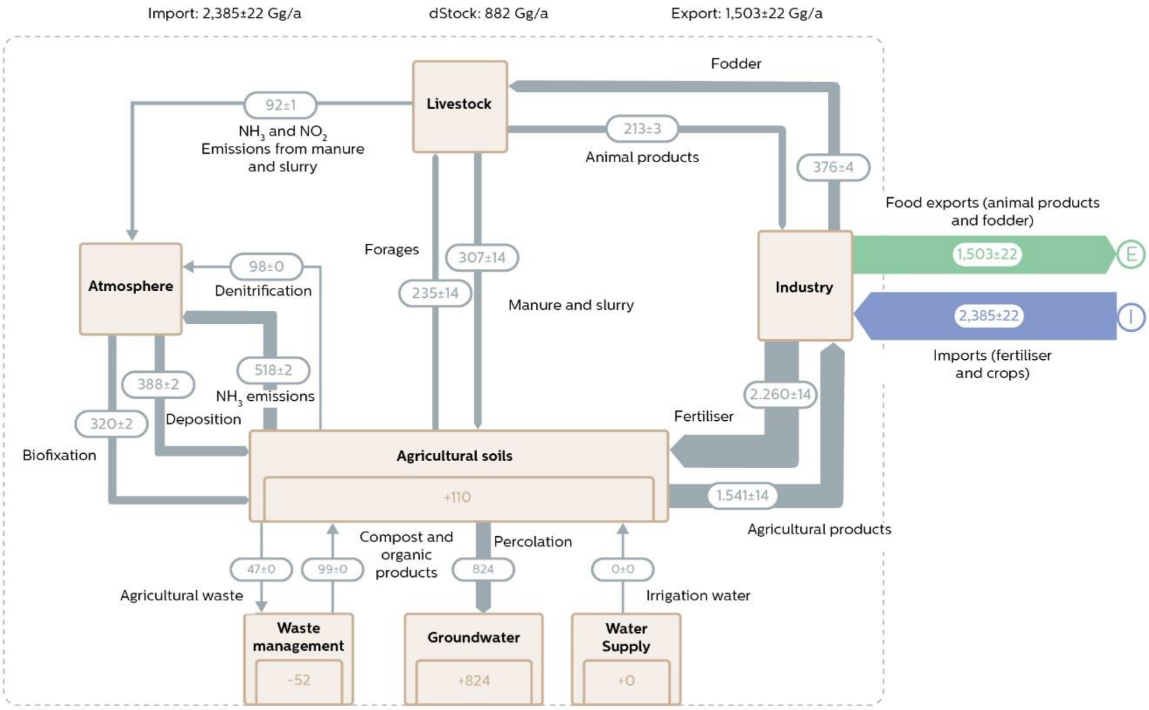

Figure 5. Impact of the hypothesis of 50% reduction in meat consumption on agricultural land

Imports into the system through food would remain constant since no distinction was made in the flow between plant and animal products. Therefore, the study focused on the subsystem of agriculture and livestock. Applying a 50% reduction to all integrated flows in livestock and a 50% increase in all flows connected to agriculture, the results indicate that the stock in the agricultural soil would decrease by 22 kt N/y, a 16% reduction of N stock compared to the current model. Taking into account the increasing trend of NANI reported by Lassaletta et al. (2013), this model could help reduce N imports to the system, and therefore the accumulation of N in the soils.

A large EU-level initiative to reduce food waste at the individual level is being promoted ("EU actions against food waste," 2021) in addition to other initiatives through the EU Green Deal Action Plan with programmes such as Farm to Fork ("Farm to Fork Strategy," 2022). However, communication is a key aspect to promoting significant changes in the habits of citizens and even more in the change of mentality towards certain aspects of their lifestyle. Nitrogen flows are strongly linked to consumption habits and agricultural practices that must be managed in such a way that the greatest benefit is obtained both for the productivity of the land and to minimise the consequences of the social and environmental problems that arise globally.

6. Conclusions

In this study, an NFA has been carried out within the Spanish territory with the data collected in 2016. The lack of updated data and, in some cases, the variability of the information were the main challenges to developing this analysis at a national level. Despite the availability of various reliable sources of information for the quantification of N flows related to agriculture, a detailed comparison has been made between the most relevant studies published in the Spanish territory to verify the credibility of the data obtained in this study.

This NFA has been developed taking into account not only agricultural-related flows but also those linked to industry and waste management, among others. Food consumption habits, heavily linked to the trends in agricultural production, have been proven to be important in the N surplus reduction process.

In this study, the total contribution of anthropogenic N input (NANI) to the country has been estimated, considering the application of synthetic fertilisers (1191 kt N/y), the net atmospheric inputs (245 kt N/y), BNF by crops (180 kt N/y) and the net food and feed imports (479 kt N/y), providing a NANI of 2095 kt N/y for Spain, 14% higher than expected by trend of growth from the data reported by Lassaletta et al. (2013).

Regarding waste management, prioritising biological treatment as the main end-of-life scenario for compostable goods would be the best strategy to reduce the wasting of N to the environment, specifically in the atmosphere, water or soil. In this way, avoiding incineration as an end-of-life scenario, 11 kt N per year could be prevented from reaching the atmosphere.

According to the results, it is recommended to increase the circular strategy within the territory. This could be achieved by recovering nutrients present in wastewater and transforming them into fertilisers. Taking into account that Spain reports an N stock between 40% and 60% (913 kt N/a), actions should focus on reducing N losses in water bodies in the agricultural sector, which represent 724 kt N per year. Furthermore, considering the agricultural system, there is an annual accumulation of 132 kt N per year in soils that also needs to be addressed. It is important to improve soil N management to increase the organic N in soil and thus maintain the C/N ratio above 10, which is a sign of soil organic matter accumulation. However, it is also important to reduce the risk of nitrate accumulation and leaching, in order to comply with Council Directive 91/676/EEC on the protection of waters against pollution caused by nitrates from agricultural sources. This directive establishes a limit of 170 kg N/ha·y for livestock manure in vulnerable zones. In 2009, the overall value in Spain was 33.14 kg N/ha·y according to Lassaletta et al. (2013). In this study, a value of 41.5 kg N/ha·y was estimated for 2016, which corresponds to an increase of 25% in only 7 years.

Finally, a revision of dietary patterns in Spain showed that is possible to reduce nitrogen stored in soils by 16% when considering 50% less consumption of animal products.

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7. References

Agricultura: valor de la producción por región en España [WWW Document], 2020. URL https://es.statista.com/estadisticas/1219154/agricultura-valor-de-la-produccion-en-espana-por-region/ (accessed 8.11.22).

Eurostat [WWW Document], 2021. URL https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Agri-environmental_indicator_-_gross_nitrogen_balance&oldid=546526 (accessed 8.14.22).

Álvarez, J., Roca, M., Valderrama, C., Cortina, J.L., 2018. A Phosphorous Flow Analysis in Spain. Science of The Total Environment 612, 995–1006. https://doi.org/10.1016/J.SCITOTENV.2017.08.299

Antikainen, R., Haapanen, R., Lemola, R., Nousiainen, J.I., Rekolainen, S., 2008. Nitrogen and phosphorus flows in the Finnish agricultural and forest sectors, 1910-2000. Water Air Soil Pollut 194, 163–177. https://doi.org/10.1007/S11270-008-9705-0

Asmala, E., Saikku, L., 2010. Closing a Loop: Substance Flow Analysis of Nitrogen and Phosphorus in the Rainbow Trout Production and Domestic Consumption System in Finland. AMBIO 2010 39:2 39, 126–135. https://doi.org/10.1007/S13280-010-0024-5

Baccini, P., Brunner, P.H., 2012. Metabolism of the Anthroposphere: Analysis, Evaluation, Design, second edition. ed, Journal of Industrial Ecology. John Wiley & Sons, Ltd, Cambridge. https://doi.org/10.1111/J.1530-9290.2012.00558.X

Baligar, V.C., Fageria, N.K., He, Z.L., 2007. NUTRIENT USE EFFICIENCY IN PLANTS. https://doi.org/10.1081/CSS-100104098 32, 921–950. https://doi.org/10.1081/CSS-100104098

Baroi, A.R., Chowdhury, R.B., Roy, B.B., Sujauddin, M., 2020. Sustainability assessment of phosphorus in the waste management system of Bangladesh using substance flow analysis. J Clean Prod 273. https://doi.org/10.1016/j.jclepro.2020.122865

Bartrolí, J., Martin, M.J., Rigola, M., 2005. The nitrogen balance for Catalan agriculture soils and livestock sectors, J. Agricultural Resources, Governance and Ecology.

Bartrolí, J., Martin, M.J., Rigola, M., 2001. Issues in system boundary definition for substance flow analysis: the case of nitrogen cycle management in Catalonia. ScientificWorldJournal. https://doi.org/10.1100/tsw.2001.260

Basosi, R., Fierro, A., Jez, S., 2014. Mineral Nitrogen Fertilizers: Environmental Impact of Production and Use.

Beckinghausen, A., Odlare, M., Thorin, E., Schwede, S., 2020. From removal to recovery: An evaluation of nitrogen recovery techniques from wastewater. Appl Energy 263, 114616. https://doi.org/10.1016/j.apenergy.2020.114616

Bernal, E.E.L., Maya, C., Valderrama, C., Cortina, J.L., 2016. Valorization of ammonia concentrates from treated urban wastewater using liquid – liquid membrane contactors. Chemical Engineering Journal 302, 641–649. https://doi.org/10.1016/j.cej.2016.05.094

Billen, G., le Noë, J., Garnier, J., 2018. Two contrasted future scenarios for the French agro-food system. Science of The Total Environment 637–638, 695–705. https://doi.org/10.1016/J.SCITOTENV.2018.05.043

Bolzonella, D., Fatone, F., Gottardo, M., Frison, N., 2017. Nutrients recovery from anaerobic digestate of agro-waste: Techno-economic assessment of full scale applications. J Environ Manage 216. https://doi.org/10.1016/j.jenvman.2017.08.026

Brodt, F., Ingels, C., 2011. Sustainable Agriculture | Learn Science at Scitable. Environmental science 3, 17.

Buathong, T., Boontanon, S.K., Boontanon, N., Surinkul, N., Harada, H., Fujii, S., 2013. Nitrogen Flow Analysis in Bangkok City, Thailand: Area Zoning and Questionnaire Investigation Approach☆. undefined 17, 586–595. https://doi.org/10.1016/J.PROENV.2013.02.074

Cencic, O., Rechberger, H., 2008. Material flow analysis with Software STAN. Journal of Environmental Engineering and Management 18, 3–7.

Cooper, J., Carliell-Marquet, C., 2013. A substance flow analysis of phosphorus in the UK food production and consumption system. Resour Conserv Recycl 74, 82–100. https://doi.org/10.1016/J.RESCONREC.2013.03.001

Coppens, J., Meers, E., Boon, N., Buysse, J., Vlaeminck, S.E., 2016a. Follow the N and P road: High-resolution nutrient flow analysis of the Flanders region as precursor for sustainable resource management. Resour Conserv Recycl 115, 9–21. https://doi.org/10.1016/j.resconrec.2016.08.006

Coppens, J., Meers, E., Boon, N., Buysse, J., Vlaeminck, S.E., 2016b. Follow the N and P road: High-resolution nutrient flow analysis of the Flanders region as precursor for sustainable resource management. Resour Conserv Recycl 115, 9–21. https://doi.org/10.1016/J.RESCONREC.2016.08.006

Cordell, D., Drangert, J.O., White, S., 2009. The story of phosphorus: Global food security and food for thought. Global Environmental Change 19, 292–305. https://doi.org/10.1016/J.GLOENVCHA.2008.10.009

Cowling, E., Galloway, J., Furiness, C., Barber, M., Bresser, T., Cassman, K., Erisman, J.W., Haeuber, R., Howarth, R., Melillo, J., Moomaw, W., Mosier, A., Sanders, K., Seitzinger, S., Smeulders, S., Socolow, R., Walters, D., West, F., Zhu, Z., 2001. Optimizing Nitrogen Management in Food and Energy Production and Environmental Protection: Summary Statement from the Second International Nitrogen Conference. ScientificWorldJournal 1, 320976. https://doi.org/10.1100/tsw.2001.481

Erisman, J.W., Galloway, J.N., Seitzinger, S., Bleeker, A., Dise, N.B., Petrescu, A.M.R., Leach, A.M., Vries, W. de, 2013. Consequences of human modification of the global nitrogen cycle. Philosophical Transactions of the Royal Society B: Biological Sciences 368. https://doi.org/10.1098/RSTB.2013.0116

Estadística de consumo de fertilizantes en la agricultura [WWW Document], 2021. URL https://www.mapa.gob.es/es/estadistica/temas/estadisticas-agrarias/agricultura/estadisticas-medios-produccion/fertilizantes.aspx (accessed 8.18.21).

Estimates of European food waste levels | FAO [WWW Document], 2016. URL http://www.fao.org/family-farming/detail/es/c/412647/ (accessed 8.24.21).

EU actions against food waste [WWW Document], 2021. URL https://ec.europa.eu/food/safety/food-waste/eu-actions-against-food-waste_en (accessed 8.24.21).

European Sustainable Phosphorus Platform - News Archive [WWW Document], 2022. URL https://www.phosphorusplatform.eu/scope-in-print/news#_Toc108595806 (accessed 8.13.22).

Eurostat [WWW Document], 2021. URL https://ec.europa.eu/eurostat/statistics-explained/index.php?title=Agri-environmental_indicator_-_mineral_fertiliser_consumption (accessed 8.18.21).

FAO, 2022. The future of food and agriculture | FAO | Food and Agriculture Organization of the United Nations [WWW Document]. URL http://www.fao.org/publications/fofa/en/ (accessed 8.17.21).

FAO, 2013. The state of the world's land and water resources for food and agriculture: Managing systems at risk, The State of the World's Land and Water Resources for Food and Agriculture: Managing Systems at Risk. https://doi.org/10.4324/9780203142837

Farm to Fork Strategy [WWW Document], 2022. URL https://ec.europa.eu/food/horizontal-topics/farm-fork-strategy_en (accessed 8.24.21).

Firmansyah, I., Spiller, M., de Ruijter, F.J., Carsjens, G.J., Zeeman, G., 2017. Assessment of nitrogen and phosphorus flows in agricultural and urban systems in a small island under limited data availability. undefined 574, 1521–1532. https://doi.org/10.1016/J.SCITOTENV.2016.08.159

Graedel, T.E., Allenby, B.R., 2009. Industrial ecology and sustainable engineering.

Grönman, K., Ypyä, J., Virtanen, Y., Kurppa, S., Soukka, R., Seuri, P., Finér, A., Linnanen, L., 2016. Nutrient footprint as a tool to evaluate the nutrient balance of a food chain. J Clean Prod 112, 2429–2440. https://doi.org/https://doi.org/10.1016/j.jclepro.2015.09.129

Guaya, D., Hermassi, M., Valderrama, C., Farran, A., Cortina, J.L., 2016. Recovery of ammonium and phosphate from treated urban wastewater by using potassium clinoptilolite impregnated hydrated metal oxides as N-P-K fertilizer. J Environ Chem Eng 4, 3519–3526. https://doi.org/10.1016/j.jece.2016.07.031

Hanson, A.M., Lee, G.F., 1971. Forms of Organic Nitrogen in Domestic Wastewater. J Water Pollut Control Fed 43, 2271–2279.

Harder, R., Giampietro, M., Mullinix, K., Smukler, S., 2021. Assessing the circularity of nutrient flows related to the food system in the Okanagan bioregion, BC Canada. Resour Conserv Recycl 174. https://doi.org/10.1016/j.resconrec.2021.105842

Hedbrant, J., Sörme, L., 2001. Data Vagueness and Uncertainties in Urban Heavy-Metal Data Collection. Water, Air and Soil Pollution: Focus 2001 1:3 1, 43–53. https://doi.org/10.1023/A:1017591718463

Homepage | EU Nitrogen Expert Panel [WWW Document], 2014. URL http://www.eunep.com/ (accessed 8.23.21).

Hou, Y., Velthof, G.L., Lesschen, J.P., Staritsky, I.G., Oenema, O., 2017. Nutrient Recovery and Emissions of Ammonia, Nitrous Oxide, and Methane from Animal Manure in Europe: Effects of Manure Treatment Technologies. Environ Sci Technol 51, 375–383. https://doi.org/10.1021/acs.est.6b04524

Jakrawatana, N., Ngammuangtueng, P., Gheewala, S.H., 2017. Linking substance flow analysis and soil and water assessment tool for nutrient management. J Clean Prod 142, 1158–1168. https://doi.org/10.1016/j.jclepro.2016.07.185

Jensen, E.S., Carlsson, G., Hauggaard-Nielsen, H., 2020. Intercropping of grain legumes and cereals improves the use of soil N resources and reduces the requirement for synthetic fertilizer N: A global-scale analysis. Agron Sustain Dev 40. https://doi.org/10.1007/S13593-020-0607-X

Jensen, E.S., Peoples, M.B., Boddey, R.M., Gresshoff, P.M., Henrik, H.N., Alves, B.J.R., Morrison, M.J., 2012. Legumes for mitigation of climate change and the provision of feedstock for biofuels and biorefineries. A review. Agron Sustain Dev 32, 329–364. https://doi.org/10.1007/S13593-011-0056-7

Jin, X., Zhang, N., Zhao, Z., Bai, Z., Ma, L., 2021. Nitrogen budgets of contrasting crop-livestock systems in China. Environmental Pollution 288. https://doi.org/10.1016/j.envpol.2021.117633

Kahrl, F., Li, Y., Su, Y., Tennigkeit, T., Wilkes, A., Xu, J., 2010. Greenhouse gas emissions from nitrogen fertilizer use in China. Environ Sci Policy 13, 688–694. https://doi.org/10.1016/J.ENVSCI.2010.07.006

Land and environmental degradation and desertification in Africa [WWW Document], 2021. URL http://www.fao.org/3/x5318E/x5318e01.htm (accessed 8.17.21).

Lassaletta, L., Billen, G., Romero, E., Garnier, J., Aguilera, E., 2013. How changes in diet and trade patterns have shaped the N cycle at the national scale: Spain (1961–2009). Regional Environmental Change 2013 14:2 14, 785–797. https://doi.org/10.1007/S10113-013-0536-1

Lassaletta, L., Romero, E., Billen, G., Garnier, J., García-Gómez, H., Rovira, J. v., 2012. Spatialized N budgets in a large agricultural Mediterranean watershed: High loading and low transfer. Biogeosciences 9, 57–70. https://doi.org/10.5194/BG-9-57-2012

Lebuf, V, Accoe, F., Vaneeckhaute, C., Meers, E., Michels, E., Ghekiere, G., 2012. Nutrient recovery from digestates: techniques and end-products.

Lebuf, Viooltje, Accoe, F., Vaneeckhaute, C., Meers, E., Michels, E., Ghekiere, G., 2012. Nutrient recovery from digestates: techniques and end-products. Venice 2012 : fourth international symposium on energy from biomass and waste 18. https://doi.org/March 15, 2014

Li, Y., Cui, S., Gao, B., Tang, J., Huang, W., Huang, Y., 2020. Modeling nitrogen flow in a coastal city—A case study of Xiamen in 2015. Science of the Total Environment 735. https://doi.org/10.1016/j.scitotenv.2020.139294

Liao, C., Xia, Y., Wu, D., 2021. Nitrogen flows associated with food production and consumption system of Shanghai. Environmental Pollution 279. https://doi.org/10.1016/j.envpol.2021.116906

Loyon, L., 2017. Overview of manure treatment in France. Waste Management 61, 516–520. https://doi.org/https://doi.org/10.1016/j.wasman.2016.11.040

Luo, Z., Hu, S., Chen, D., Zhu, B., 2018. From Production to Consumption: A Coupled Human-Environmental Nitrogen Flow Analysis in China. Environ Sci Technol 52, 2025–2035. https://doi.org/10.1021/ACS.EST.7B03471

Ma, X., Wang, Z., Yin, Z., Koenig, A., 2007. Nitrogen Flow Analysis in Huizhou, South China. Environmental Management 2007 41:3 41, 378–388. https://doi.org/10.1007/S00267-007-9053-7

MacWilliam, S., Wismer, M., Kulshreshtha, S., 2014. Life cycle and economic assessment of Western Canadian pulse systems: The inclusion of pulses in crop rotations. Agric Syst 123, 43–53. https://doi.org/10.1016/J.AGSY.2013.08.009

Mengel, K., Kirby, E.A., Kosegarten, H., Appel, T., 2001. Principales of Plants Nutrition 849.

Moomaw, W.R., 2002. Energy, Industry and Nitrogen: Strategies for Decreasing Reactive Nitrogen Emissions. Ambio 31, 184–189.

National energy and climate plans | European Commission [WWW Document], 2021. URL https://ec.europa.eu/info/energy-climate-change-environment/implementation-eu-countries/energy-and-climate-governance-and-reporting/national-energy-and-climate-plans_en (accessed 8.24.21).

Nitrogen flow in Scania - Epsilon Archive for Student Projects [WWW Document], 2015. URL https://stud.epsilon.slu.se/8129/ (accessed 2.9.22).

Nitrogen on the table: the influence of food choices on nitrogen emissions and the European environment | PBL Netherlands Environmental Assessment Agency [WWW Document], 2016. URL https://www.pbl.nl/en/publications/nitrogen-on-the-table-the-influence-of-food-choices-on-nitrogen-emissions-and-the-european-environment (accessed 8.24.21).

Pan, J., Ding, N., Yang, J., 2019a. Changes of urban nitrogen metabolism in the Beijing megacity of China, 2000–2016. Science of The Total Environment 666, 1048–1057. https://doi.org/10.1016/J.SCITOTENV.2019.02.315

Pan, J., Ding, N., Yang, J., 2019b. Science of the Total Environment Changes of urban nitrogen metabolism in the Beijing megacity of. Science of the Total Environment 666, 1048–1057. https://doi.org/10.1016/j.scitotenv.2019.02.315

Panhwar, Q.A., Ali, A., Naher, U.A., Memon, M.Y., 2019. Fertilizer Management Strategies for Enhancing Nutrient Use Efficiency and Sustainable Wheat Production. Organic Farming: Global Perspectives and Methods 17–39. https://doi.org/10.1016/B978-0-12-813272-2.00002-1

Pelletier, N., Tyedmers, P., 2010. Forecasting potential global environmental costs of livestock production 2000-2050. Proc Natl Acad Sci U S A 107, 18371–18374. https://doi.org/10.1073/pnas.1004659107

Peoples, M.B., Brockwell, J., Herridge, D.F., Rochester, I.J., Alves, B.J.R., Urquiaga, S., Boddey, R.M., Dakora, F.D., Bhattarai, S., Maskey, S.L., Sampet, C., Rerkasem, B., Khan, D.F., Hauggaard-Nielsen, H., Jensen, E.S., 2009. The contributions of nitrogen-fixing crop legumes to the productivity of agricultural systems. Symbiosis 2009 48:1 48, 1–17. https://doi.org/10.1007/BF03179980

Pharino, C., Sailamai, N., Kamphaengthong, P.L., 2016. Material Flow Analysis of Nitrogen in Maeklong River Basin in Ratchaburi and Samut Songkhram Province, Thailand. Water Conservation Science and Engineering 1, 167–175. https://doi.org/10.1007/S41101-016-0011-1

Productos fertilizantes [WWW Document], 2021. URL https://www.mapa.gob.es/es/agricultura/temas/medios-de-produccion/productos-fertilizantes/ (accessed 8.19.21).

Reich, M., Aghajanzadeh, T., de Kok, L.J., 2014. Physiological Basis of Plant Nutrient Use Efficiency – Concepts, Opportunities and Challenges for Its Improvement 1–27. https://doi.org/10.1007/978-3-319-10635-9_1

Renard, D., Tilman, D., 2019. National food production stabilized by crop diversity. Nature 2019 571:7764 571, 257–260. https://doi.org/10.1038/s41586-019-1316-y

Schlesinger, W.H., 2009. On the fate of anthropogenic nitrogen. Proceedings of the National Academy of Sciences 106, 203–208. https://doi.org/10.1073/PNAS.0810193105

Senthilkumar, K., Nesme, T., Mollier, A., Pellerin, S., 2012. Regional-scale phosphorus flows and budgets within France: The importance of agricultural production systems. Nutr Cycl Agroecosyst 92, 145–159. https://doi.org/10.1007/S10705-011-9478-5

Seyhan, D., 2009. Country-scale phosphorus balancing as a base for resources conservation. Resour Conserv Recycl 53, 698–709. https://doi.org/10.1016/J.RESCONREC.2009.05.001

Singh, S., Compton, J.E., Hawkins, T.R., Sobota, D.J., Cooter, E.J., 2017. A Nitrogen Physical Input-Output Table (PIOT) model for Illinois. Ecol Modell 360, 194–203. https://doi.org/10.1016/J.ECOLMODEL.2017.06.015

Sinha, R., Thomas, J.B.E., Strand, Söderqvist, T., Stadmark, J., Franzen, F., Ingmansson, I., Gröndahl, F., Hasselström, L., 2022. Quantifying nutrient recovery by element flow analysis: Harvest and use of seven marine biomasses to close N and P loops. Resour Conserv Recycl 178. https://doi.org/10.1016/j.resconrec.2021.106031

Socolow, R.H., 1999. Nitrogen management and the future of food: Lessons from the management of energy and carbon. Proc Natl Acad Sci U S A 96, 6001–6008. https://doi.org/10.1073/PNAS.96.11.6001/ASSET/FA4B2334-A807-43C8-8801-3BDEFAE3D148/ASSETS/GRAPHIC/PQ1090896001.JPEG

Spanish Agri-Food Exports Increase by 97% in Last Decade [WWW Document], 2019. URL https://www.foodswinesfromspain.com/spanishfoodwine/global/food/news/new-detail/spanish-agri-food-exports-2018.html (accessed 8.18.21).

Status of the Regulation (EU) [WWW Document], 2022. URL https://www.staphyt.com/2022/02/25/point-sur-la-reglementation-ue-n-2019-1009-relative-aux-matieres-fertilisantes-et-aux-biostimulants/ (accessed 8.13.22).

Stevens, C.J., 2019. Nitrogen in the environment. Science (1979) 363, 578 LP – 580. https://doi.org/10.1126/science.aav8215

Suesatpanit, T., 2017. Substance Flow Analysis of Nitrogen in Food Production and Consumption System of Thailand. undefined.

Sutton, M.A., Bleeker, A., de Vries, W., Bekunda, M., Reis, S., Howard, C.M., Grizzetti, B., Erisman, J.W., van Grinsven, H.J.M., Abrol, Y.P., Adhya, T.K., Billen, G., Davidson, E.A., Datta, A., Diaz, R., Liu, X.J., Oenema, O., Palm, C., Raghuram, N., Scholz, R.W., Sims, T., Westhoek, H., Zhang, F.S., 2013. Our Nutrient World - The challenge to produce more food and energy with less pollution.

Tilman, D., Cassman, K.G., Matson, P.A., Naylor, R., Polasky, S., 2002. Agricultural sustainability and intensive production practices. Nature 418, 671–677. https://doi.org/10.1038/nature01014

van der Voet, E., 2015. Substance flow analysis methodology. A Handbook of Industrial Ecology. https://doi.org/10.4337/9781843765479.00018

van der Voet, E., Kleijn, R., van Oers, L., Heijungs, R., Huele, R., Mulder, P., 1995. Substance flows through the economy and environment of a region. Environmental Science and Pollution Research 1995 2:2 2, 90–96. https://doi.org/10.1007/BF02986723

van der Wiel, B.Z., Weijma, J., van Middelaar, C.E., Kleinke, M., Buisman, C.J.N., Wichern, F., 2020. Restoring nutrient circularity: A review of nutrient stock and flow analyses of local agro-food-waste systems. Resour Conserv Recycl 160. https://doi.org/10.1016/j.resconrec.2020.104901

Van Egmond, K., Bresser, T., Bouwman, L., 2002. The European nitrogen case. Ambio 31, 72–78. https://doi.org/10.1579/0044-7447-31.2.72

Vaneeckhaute, C., Lebuf, V., Michels, E., Belia, E., Vanrolleghem, P.A., Tack, F.M.G., Meers, E., 2017. Nutrient Recovery from Digestate: Systematic Technology Review and Product Classification. Waste Biomass Valorization. https://doi.org/10.1007/s12649-016-9642-x

Vaneeckhaute, C., Lebuf, V., Michels, E., Belia, E., Vanrolleghem, P.A., Tack, F.M.G., Meers, E., 2016. Nutrient Recovery from Digestate: Systematic Technology

Review and Product Classification. Waste and Biomass Valorization 2016 8:1 8, 21–40. https://doi.org/10.1007/S12649-016-9642-X

Vaneeckhaute, C., Meers, E., Michels, E., Ghekiere, G., Accoe, F., Tack, F.M.G., 2013. Closing the nutrient cycle by using bio-digestion waste derivatives as synthetic fertilizer substitutes: A field experiment. Biomass Bioenergy 55, 175–189. https://doi.org/10.1016/j.biombioe.2013.01.032

Vecino, X., Reig, M., Bhushan, B., Gibert, O., Valderrama, C., Cortina, J.L., 2019. Liquid fertilizer production by ammonia recovery from treated ammonia-rich regenerated streams using liquid-liquid membrane contactors. Chemical Engineering Journal 360, 890–899. https://doi.org/https://doi.org/10.1016/j.cej.2018.12.004

Verstraete, W., van de Caveye, P., Diamantis, V., 2009. Maximum use of resources present in domestic "used water." Bioresour Technol 100, 5537–5545. https://doi.org/10.1016/J.BIORTECH.2009.05.047

Vitousek, P.M., Aber, J.D., Howarth, R.W., Likens, G.E., Matson, P.A., Schindler, D.W., Schlesinger, W.H., Tilman, D.G., 1997. Technical Report: Human Alteration of the Global Nitrogen Cycle: Sources and Consequences. Ecological Applications 7, 737. https://doi.org/10.2307/2269431

Voisin, A.S., Guéguen, J., Huyghe, C., Jeuffroy, M.H., Magrini, M.B., Meynard, J.M., Mougel, C., Pellerin, S., Pelzer, E., 2013. Legumes for feed, food, biomaterials and bioenergy in Europe: a review. Agronomy for Sustainable Development 2013 34:2 34, 361–380. https://doi.org/10.1007/S13593-013-0189-Y

WangShou, Z., XuYong, L., Swaney, D.P., XinZhong, D., 2016. Does food demand and rapid urbanization growth accelerate regional nitrogen inputs? J Clean Prod 112, 1401–1409.

Westhoek, H., Lesschen, J.P., Rood, T., Wagner, S., de Marco, A., Murphy-Bokern, D., Leip, A., van Grinsven, H., Sutton, M.A., Oenema, O., 2014. Food choices, health and environment: Effects of cutting Europe's meat and dairy intake. Global Environmental Change 26, 196–205. https://doi.org/10.1016/J.GLOENVCHA.2014.02.004

Wu, J., Franzén, D., Malmström, M.E., 2016. Nutrient flows following changes in source strengths, land use and climate in an urban catchment, Råcksta Träsk in Stockholm, Sweden. Ecol Modell 338, 69–77. https://doi.org/10.1016/j.ecolmodel.2016.08.001

Xu, R., Cai, Y., Wang, X., Li, C., Liu, Q., Yang, Z., 2020. Agricultural nitrogen flow in a reservoir watershed and its implications for water pollution mitigation. J Clean Prod 267. https://doi.org/10.1016/j.jclepro.2020.122034

Zhang, X., Davidson, E.A., Mauzerall, D.L., Searchinger, T.D., Dumas, P., Shen, Y., 2015. Managing nitrogen for sustainable development. Nature 2015 528:7580 528, 51–59. https://doi.org/10.1038/nature15743

Zhang, X., Zhang, Y., Wang, Y., Fath, B.D., 2021. Research progress and hotspot analysis for reactive nitrogen flows in macroscopic systems based on a CiteSpace analysis. Ecol Modell 443. https://doi.org/10.1016/j.ecolmodel.2021.109456