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Research article

How much can changes in the agro-food system reduce agricultural nitrogen losses to the environment? Example of a temperate-Mediterranean gradient

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ABSTRACT

Ammonia (NH_3) volatilization, nitrous oxide (N_2O) emissions, and nitrate (NO_3^-) leaching from agriculture cause severe environmental hazards. Research studies and mitigation strategies have mostly focused on one of these nitrogen (N) losses at a time, often without an integrated view of the agro-food system. Yet, at the regional scale, N_2O, NH_3, and NO_3^- loss patterns reflect the structure of the whole agro-food system. Here, we analyzed at the resolution of NUTS2 administrative European Union (EU) regions, N fluxes through the agro-food systems of a Temperate-Mediterranean gradient (France, Spain, and Portugal) experiencing contrasting climate and soil conditions. We assessed the atmospheric and hydrological N emissions from soils and livestock systems. Expressed per ha agricultural land, NH_3 volatilization varied in the range 6.2–44.4 kg N ha^{-1} yr^{-1}, N_2O emission and NO_3 leaching 0.3–4.9 kg N ha^{-1} yr^{-1} and 5.4–154 kg N ha^{-1} yr^{-1} respectively. Overall, lowest N_2O emission was found in the Mediterranean regions, where NO_3^- leaching was greater. NH_3 volatilization in both temperate and Mediterranean regions roughly follows the distribution of livestock density. We showed that these losses are also closely correlated with the level of fertilization intensity and agriculture system specialization into either stockless crop farming or intensive livestock farming in each region. Moreover, we explored two possible future scenarios at the 2050 horizon: (1) a scenario based on the prescriptions of the EU-Farm-to-Fork (F2F) strategy, with 25% of organic farming, 10% of land set aside for biodiversity, 20% reduction in N fertilizers, and no diet change; and (2) a hypothetical agro-ecological (AE) scenario with generalized organic farming, reconnection of crop and livestock farming, and a healthier human diet with an increase in the share of vegetal protein to 65% (i.e., the Mediterranean diet). Results showed that the AE scenario, owing to its profound reconfiguration of the entire agro-food system would have the potential for much greater reductions in NH_3, N_2O, and NO_3^- emissions, namely, 60–81% reduction, while the F2F scenario would only reach 24–35% reduction of N losses.

1. Introduction

The Haber–Bosh process, industrialized in 1913 and initially used mainly for the production of explosives, was promoted after World War II for synthetizing nitrogen (N) fertilizers to increase agricultural production for an increasing global population (Erisman et al., 2008). In the meantime, beside mechanization of farming, overall modernization of daily life occurred based on cheap fossil energy and on expanding global food markets (Dyer and Desjardins, 2009). As a result, crop production increased more than threefold (Zhang et al., 2021). Moreover, with growing incomes, the proportion of animal products in the human diet increased, and became a sign of wellbeing. Consequently, livestock products in the diet rose by ~39% in Europe from the early 1960s to the 2010s (livestock density grew by 32% between these two periods), while the

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percentage of livestock protein ingestion from imported feed more than doubled at the expense of grass ingestion (Billen et al., 2021; https://www.fao.org/faostat/). Most of these changes occurred during 1960–1990, and since then only relatively minor adjustments were made to the established functioning of the agro-food system of Western Europe, which is regionally specialized with a strong dependence on industrial fertilizers and pesticides. This increase in agricultural N inputs, required for crop and livestock production, was followed by large losses of nitrate (NO_3^-) to the aquatic systems, and ammonia (NH_3) and nitrous oxide (N_2O) to the atmosphere (Galloway et al., 2003; Sutton et al., 2011; Velthof et al., 2014).

Two major agricultural reactive N species are lost to the atmosphere: ammonia (NH_3, a pollutant-forming harmful particulate matter and detrimental to biodiversity) and nitrous oxide (N_2O, the third major greenhouse gas [GHG] besides water vapor). NH_3 represents a large N loss, 94% originating from the agricultural sector of which 60% is attributed to manure management (EMEP/EEA, 2015). N_2O is a major GHG from European agriculture (43% of total agriculture GHG emissions for the former EU28 in 2019 (https://ec.europa.eu/eurostat/web/main/data/database), with manure management contributing 12% of total agricultural N_2O emissions. N_2O represents approximately 6.5% and 7% of total direct GHG emissions, respectively, in the world (https://ourworldindata.org) and in Europe (EEA, 2018, EMEP/EEA, 2015). Moreover, NO_3 contaminations of surface and groundwater, as well as an excess of N over other nutrients (phosphorus –P– and silica –Si–) at the coastal zone, are well-known environmental issues, although few studies directly link agricultural practices/systems to leached N in the form of NO_3^- (N–NO_3^-) at a regional scale (e.g., Desmit et al., 2018; Garnier et al., 2018; Aguilera et al., 2021; Cameira et al., 2021; Serra et al., 2021).

All reactive N losses are associated with significant environmental and health issues. Ammonia (NH_3) is a smelling (detected in the air at 5 ppm) and irritating gas. Manufactured NH_3 is largely used as fertilizer (e.g., 20, 30, 10% as urea for France, Spain, and Portugal on average during 2014–2019 (IFAstat, https://www.ifastat.org/databases), and also causes environmental damage when reacting with other chemicals (importantly NOx and SOx) to form secondary particles in the air (i.e., PM2.5) and acidifying soils (Krupa, 2003; Erisman et al., 2008; Sanz-Cobena et al., 2014). NH_3 also has deleterious effects on vegetation, both directly (e.g., metabolic) or indirectly (e.g., mycorrhizae reduction, insect pests proliferation), and tends to reduce biodiversity (Krupa, 2003). Following deposition, it can also be responsible for further NO_3^- contamination and N_2O emissions (Asman et al., 1998). N_2O GHG not only participates to global warming in the atmosphere of the Earth and has a role in the development of tropospheric ozone, negatively affecting human health (Wolfe and Patz, 2002) and the environment (e.g., vegetation: Wittig et al., 2009; Ainsworth et al., 2012; Li et al., 2017). In addition, N_2O stable in the troposphere, contributes to the destruction of the ozone layer in the stratosphere (Crutzen, 1970; Crutzen and Ehhalt, 1977; Bange, 2000; Ravishankara et al., 2009), thus causing dermatological problems linked to ultraviolet radiations (Henriksen et al., 1990; Young, 2009). Nitrate (NO_3^-), a very mobile ion, can be rapidly leached from soils polluting ground- and surface water in intensive agricultural areas. Recommended level for producing drinking water (50 mg NO_3^- l^{-1}, i.e., 11.3 mg N l^{-1}, EU-Nitrates Directive, 1991; EU-Water Framework Directive, 2000; WHO, 2007) is often exceeded. Moreover, the level of nitrate required for maintaining biodiversity in freshwater (~10 mg NO_3^- l^{-1}, i.e., ~2 mg N l^{-1}) is often exceeded as well (Camargo et al., 2005; James et al., 2005). Furthermore, many coastal zones are strongly eutrophicated, with harmful algal bloom development (Glibert, 2017; Garnier et al., 2021).

Scenario analysis can be a useful tool to gain insight on how the agro-food systems can be changed. Recently, several scenarios for the future of the European agro-food system (at 2050 horizon) have been designed and calculated in detail at NUTS2 resolution (Billen et al., 2022; Grizzetti et al., 2022). One scenario was constructed based on the recommendations of the EU-Farm-to-Fork (EU-F2F, 2020) and Biodiversity (BDS) strategies, which are part of the European Green Deal (EU-Green Deal, 2019). Another, called the "agro-ecological scenario" (AE), explored profound changes in the agro-food system (Billen et al., 2021, 2022), including human food consumption, which presently consists of excessive animal protein consumption. A healthier, more plant-based diet is indeed recommended by the FAO and WHO report (2019).

Apart from the pioneer work by Velthof et al. (2009, 2014), using the MITERRA-Europe model, and the studies by de Vries et al. (2021) and Schulte-Uebbing and de Vries (2021) based on INTEGRATOR model (derived from MITERRA –Europe), taking an integrated approach with both NH_3 N_2O and NO_3^- losses is still scarce. Here, we used the GRAFS approach (Billen et al., 2014), which additionally integrate the human diets in the analysis of the agro-food systems. We therefore aimed to jointly quantify NH_3 volatilization, N_2O emissions, and NO_3^- leaching from agriculture in France and the Iberian Peninsula (Spain and Portugal) at subnational scales (NUTS2) for the recent 2014–2019 period. These countries were chosen as they represent a wide variety of agricultural specificities among and within, as well as a climatic contrast between Temperate- Mediterranean climates (Billen et al., 2019).

Various mitigation measures, based on specific technological or agronomical practices, have been promoted to reduce the environmental losses of these reactive N compounds separately (e.g., NH_3: Sommer and Hutchings, 1995; N_2O: Rees et al., 2013; NO_3: Grizzetti et al., 2012; Cameira and Mota, 2017; Desmit et al., 2018; Garnier et al., 2018), but such measures are here explored comprehensively. Thus, we further applied the scenarios framework (Billen et al., 2022), aiming to show how, and to what extent, it would be possible to reduce agricultural N losses to the atmosphere and the hydrosphere, while feeding the population and improving the health of the environment and of humans.

2. Material and methods

2.1. Study area

France, Spain, and Portugal were chosen for their north-to-south gradient on the Atlantic coast of the EU, i.e., temperate to Mediterranean, with oceanic influence on the coasts. Agriculture has been important in the human activities and economy of these three countries and it has followed similar trends toward modernization and intensification; there was a time lag in this process for Spain and Portugal, which after authoritarian political regimes from World War II to the 1970s, joined, e.g., the EU and its Common Agricultural Policy (CAP) in 1986, later than France which was a signatory of the Treaty of Rome in 1957.

Regional differences in relief, soil characteristics, and climate (Fig. 1) necessarily lead to major differences in the variety of farming systems and practices. France has large areas of lowlands favoring arable crop production, whereas such areas only exist in the southwest of Spain and Portugal but are less common and mostly located in the large river valleys (e.g., Ebro, Tagus, Guadalquivir, etc.) (Fig. 1a). Nevertheless, the upper large fertile plateaus of the Iberian Peninsula, where water deficit is less pronounced, are dedicated to agriculture. Regarding climate features, beside higher temperature and low rainfall in the south and a more temperate climate at the coast, the deficit of water is striking, especially in the center and south of Spain and Portugal, while the northwest of the Iberian Peninsula benefits from an oceanic regime (Fig. 1b).

France and Spain each have a surface area about 6 times that of Portugal (Tables 1SM and 2SM, Fig. 1). In France, urbanized areas are twice larger in proportion. Forest occupies 26%, 21.4%, and 17.9% of France, Spain, and Portugal, respectively (Fig. 1c; Table 2SM). The wide range in surface area and in population for each country overall leads to a population density that is rather similar between the three countries (121, 93, and 117 inhabitants km^{-2} for France, Spain, and Portugal, respectively) (see Table 2SM).

Utilized agriculture area (UAA, defined as the sum of arable cropland, permanent cropland, and permanent grassland) represents about

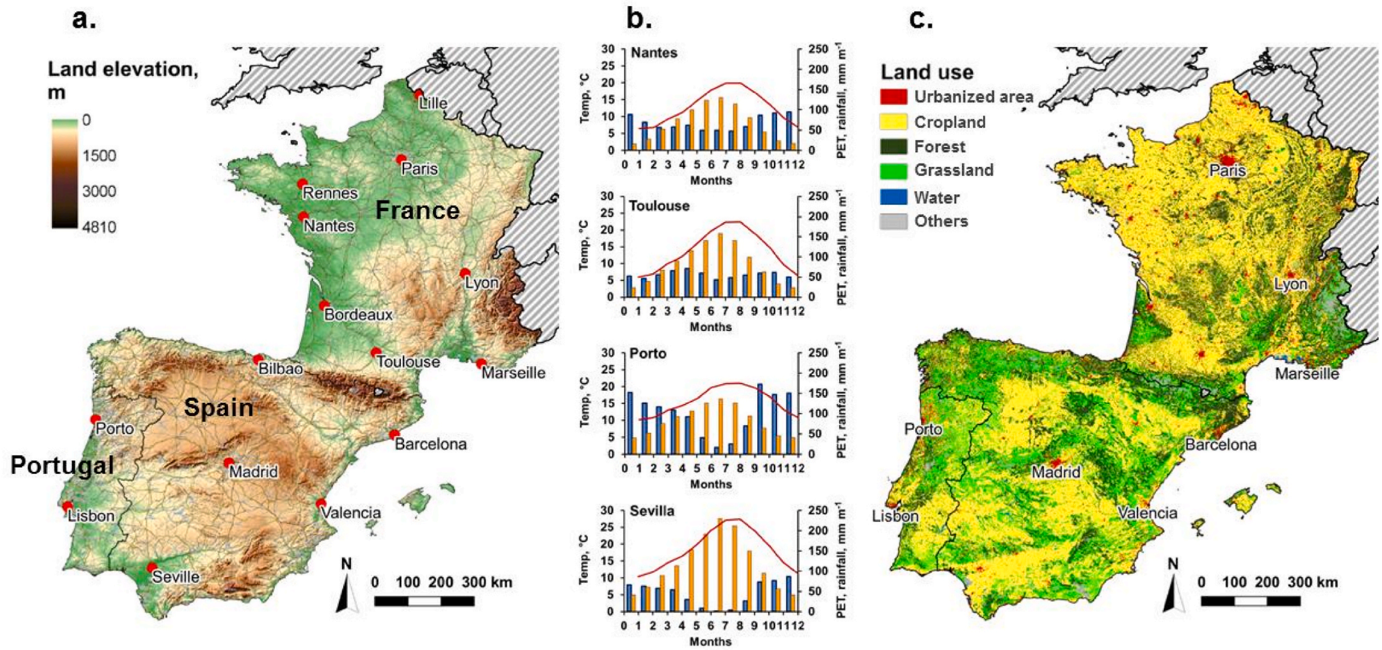

Fig. 1. a. Relief map of the study area. b. Meteorological characteristics of some cities illustrating the gradient in average monthly temperature (in °C, red line), in cumulative monthly rainfall (mm m^{-1}, blue bar chart), and in potential evapotranspiration (PET, mm m^{-1}, orange bar chart) for the period 1990–2018. The gap in the summer period between PET and rainfall histograms represents the water deficit. c. Map of main land use (Corine Land Cover, 2018). (Cropland includes arable cropland (with temporary grassland), permanent crops, and heterogeneous areas).

half of France and Spain (48.7% and 49.5%, respectively), but only 36.6% of Portugal (Table 2SM). Arable cropland as percentage of UAA (%UAA) is the highest in France (64.4%) followed by Spain (49.7%) and Portugal (23.7%), while the opposite is true for percentage of permanent crop land: Portugal > Spain > France (Table 2SM). Permanent grassland as percentage of UAA, which is close for France and Spain (32.0% and 30.8%), is much higher for Portugal (54.7%). Livestock density per hectare of UAA (LSU ha^{-1}) is the highest in France (0.67 LSU ha^{-1}), followed by Portugal (0.52 LSU ha^{-1}) and then by Spain (0.40 LSU ha^{-1}), thus within a factor of 1.7.

2.2. GRAFS approach

2.2.1. N flows and system typologies

Following the soil surface budget approach proposed by Oenema et al. (2003), we established a detailed accounting model for documenting N fluxes through agro-food systems. The resulting GRAFS approach (Generalized Representation of Agro-food Systems, Billen et al., 2013a) can be used at different scales, from small watersheds (Garnier et al., 2016) to countries (Lassaletta et al., 2014a, 2014b; Le Noë et al., 2017, 2018), Europe (Billen et al., 2021), and macroregions of the world (Billen et al., 2014; Lassaletta et al., 2016).

The GRAFS approach describes the agro-food system by considering four main components exchanging nutrient flows: cropland (arable and permanent crops), permanent grassland, livestock systems, and local population. The agro-food system is documented here for the period 2014–2019, at the territorial level of the EU administrative units NUTS2, in terms of N for (i) nutrient inputs to the soil (exogenous fertilization such as synthetic and/or organic fertilization and atmospheric deposition, as well as symbiotic fixation); (ii) the feed required for the existing livestock; (iii) the size of the human population, its dietary preferences, and its excreta; and (iv) food and feed imports/exports (Billen et al., 2018, 2021). These N flows link grassland and cropland productivity (from annual and perennial crops) to livestock feeding, and, finally, to human food. Detailed figures for these different components are presented in the Supplementary Material (Table 1SM).

Using the GRAFS approach to our study area, we also expand upon the approach used by Le Noë et al. (2018), which established a typology of the agro-food systems in France. We used very similar criteria, but urban systems were defined as those for which human food demand exceeds local food production (cropland production + livestock edible production), and thus they are structurally food importers, and they are superimposed on the other ones (see Fig. 2). This approach intends to describe the degree of coupling between crop and livestock farming, local production/consumption to shows regional differences (see Table 1SM for a detailed description of the typology defined).

2.2.2. Crop yields and N inputs to agricultural soils

Crop areas and yields were obtained at the required resolution from Eurostat. Some inconsistencies, particularly regarding fodder crops, temporary grassland and permanent grasslands were corrected as described in detail in Einarsson et al. (2021).

N-inputs to agricultural soils consist of synthetic N fertilizers application, manure inputs, atmospheric N deposition and symbiotic N fixation. Data on total inputs of *synthetic N fertilizers* at NUTS2 scale were collected from Eurostat, and their division between cropland (arable and permanent crops) and permanent grassland was based on national data (Einarsson et al., 2021). The fertilizer quantity allocated to cropland was further divided between arable and permanent crops using proportions provided at country-level from EFMA (today Fertilizers Europe) for the crop year 2005/2006. *Manure inputs* from ruminant and monogastric livestock were calculated according to a full analysis of the livestock management system (see below). *Atmospheric deposition* (including wet and dry, reduced and oxidized N) is calculated from EMEP data for the period 2014–2019. Symbiotic fixation by *legume crops* is calculated from N yield, using the relationship established by Anglade et al. (2015) and Lassaletta et al. (2014b). For permanent grassland, a share of 25% legumes was considered for unfertilized grassland, decreasing to half this value when N fertilization increases to 100 kg N ha^{-1} yr^{-1}.

2.2.3. N losses from livestock and crop farming

We referred to Misselbrook et al. (2004) and Sanz-Cobena et al.

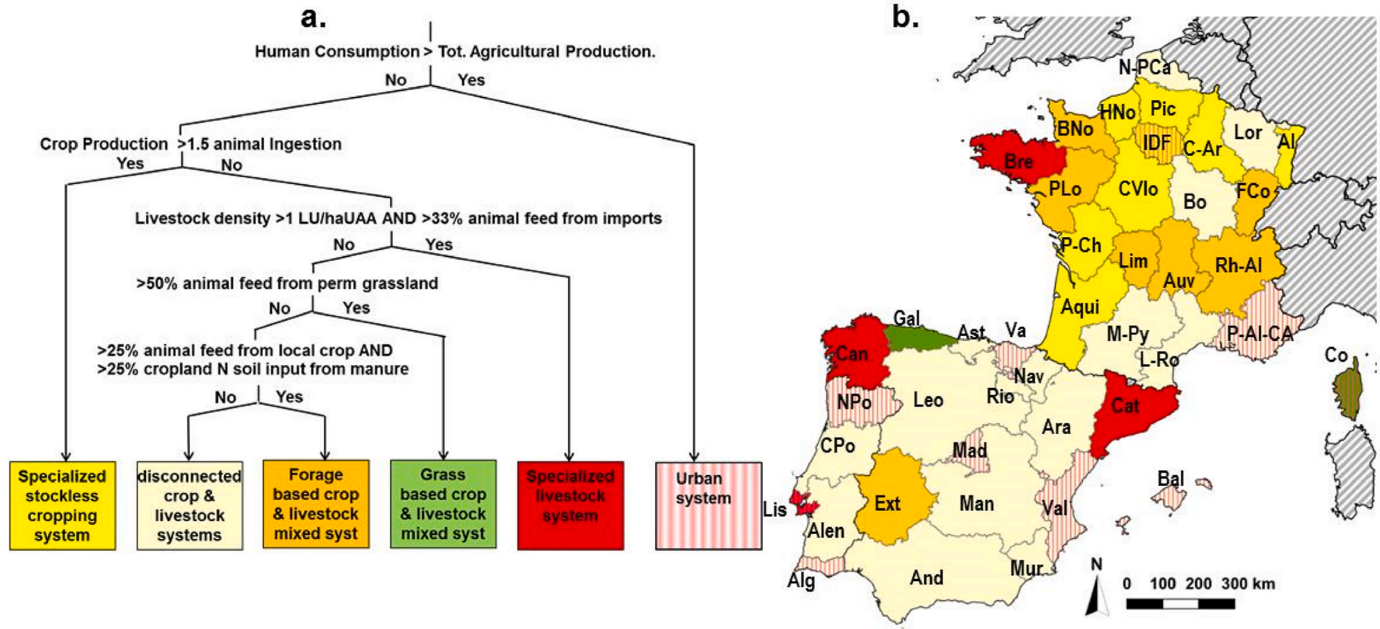

Fig. 2. Typology of the regional agro-food systems in the three countries. a. Decision tree representing the elaboration of the typology. b. Map of the different types of agricultural subnational regions. Pink hatched areas of the urban systems are also colored according to the other dominating system (specialized stockless, disconnected crop and livestock systems, grass-based crop, and livestock mixed system). Names of NUTS are abbreviated (see Table 5SM for complete definitions).

(2014) for the determination of NH_3 volatilization, and to Garnier et al. (2019) for the evaluation of N_2O emissions. Regarding N leaching to the hydrosphere, the soil surface surplus adjusted for NH_3 emissions and denitrification was used as a proxy (Anglade et al., 2015; Cameira et al., 2019).

Ammonia volatilization at application of synthetic fertilizers was estimated using the MANNER empirical model (Misselbrook et al., 2004; Sanz-Cobena et al., 2014), which considers the type of synthetic fertilizer (e.g., urea, ammonium nitrate, etc.), the application method and timing, as well as the soil pH. Monthly weather variables that significantly affect N volatilization process, such as rainfall, temperature, and wind conditions, are also considered. Effective fertilization is the difference between total input and NH_3 volatilization.

Estimating N losses at manure management and application required a complete description of the flow of N excretion from ruminants and monogastrics and of the application to the different agricultural land surfaces, in line with the procedure carried out at the national scale in the EuropeAgriDB project (Einarsson et al., 2021). The total amount of N excreted was calculated from livestock numbers by animal categories using country-specific emission coefficients compiled by the EU Livedate Project (Velthof, 2014). Manure was divided into different manure management systems, including direct excretion at grazing, according to data from national inventories (Einarsson et al., 2021). For ruminants, the share of direct excretion at grazing versus indoors is related to the degree of intensity of the livestock breeding systems, for which a good proxy is the conversion efficiency of ingested proteins into edible production, i.e., the amount of edible proteins obtained for one unit of ingested protein in feed (Fig. 1SM).

Nitrogen losses from manure storage were calculated according to Oenema et al. (2007) and Pardo et al. (2015). Country-specific partitioning of stored N manure between grass and non-grass areas was estimated based on Menzi (2002). Further details are provided by Einarsson et al. (2021). Finally, manure applied to grass and excreted at grazing was divided between permanent and temporary grassland in proportion to their areas. NH_3 volatilization at application of manure was then calculated using the MANNER model again, considering the liquid or solid form of the material applied. The required data were obtained from national reports (Loyon, 2018) and expert criteria.

The soil N surplus is calculated from the GRAFS approach by subtracting the N exported through harvest (of crops and possibly straw or other harvested residues) from the total effective N soil inputs (i.e., N inputs minus ammonia volatilization at application). Deducting NH_3 volatilization from the N soil balance (all fertilizer inputs minus harvest) represents a gross surplus (all terms in kg N ha^{-1} yr^{-1}).

$$\text{Gross Surplus} = \sum \text{N inputs} - \text{N harvest} - NH_3 \qquad [1]$$

We computed N_2O emissions per UAA (Em_{N2O}) based on established relationships with N exogenous inputs (N_{input}: organic and mineral, including atmospheric deposition), rainfall (Rain), and temperature (Temp) for arable land and grassland (i.e., UAA) (Garnier et al., 2019). The dataset includes data from studies performed on Mediterranean climate gathered by Cayuela et al. (2017) and temperate climate (see references in Garnier et al., 2019), if N_2O emissions were documented with the explicative variables chosen here. These studies document on annual N_2O emission data calculated from daily values representative of the whole year, as well as climatic and N management variables. Irrigation was not taken into account. The full dataset contains 208 and 138 data points for cropland and grassland, respectively. Because the relationships specific to arable land and grassland were not statistically different, equation [2] includes the 346 data points (Garnier et al., 2019).

$$Em_{N2O}\left(\text{kg N ha}^{-1}\ \text{yr}^{-1}\right) = \left[0.15 + 0.016 \text{ x } N_{input}\right] \text{x } (\text{Rain}/1000)^{1.2} \text{ x } 1.2 \qquad [2]$$

The parameters of the power function, determined by a systematic optimization procedure searching for the combination of parameter values providing the best fit of the calculated emissions to the observed N_2O emission values, were applied here. Denitrification during storage of manure represents a smaller pathway of N loss compared with NH_3 volatilization. Oenema et al. (2007) estimated that it represents approximately 7% of the annually stored N manure. The associated N_2O emission was estimated using a simple emission coefficient related to the N flow of manure storage, amounting to 0.009 kg N_2O (kg N)$^{-1}$ manure stored (i.e., 0.0057 kg N–N_2O (kg N)$^{-1}$ manure stored), as proposed from a meta-analysis by Pardo et al. (2015).

Denitrification was calculated from EmN_2O considering an N_2O/

(N_2+N_2O) ratio of 4 for agricultural soils (Schlesinger, 2009; Butterbach-Bahl et al., 2013).

$$\text{Denitrification}\ \left(\text{kg N ha}^{-1}\ \text{yr}^{-1}\right) = 4 \times \text{EmN}_2\text{O} \quad [3]$$

Finally, removing the value of denitrification (N_2 and N_2O) in the soils from the gross surplus provides a net surplus, which can be considered as the best proxy for leaching (Anglade et al., 2015), at least in the cases where the organic N pool in the soil is in equilibrium. Leaching concentration is estimated by considering that the annual net N surplus of arable cropland is diluted within the annual water flow percolating through the root zone, taken as the average total specific annual water drainage derived from the model LISFLOOD (de Roo et al., 2013; https://web.jrc.ec.europa.eu/policy-model-inventory/explore/models/model-lisflood/#model-outputs) for the period 2009–2018 (B. Grizzetti, pers. comm.). Net N surplus from permanent cropland and grassland is mostly stored in woody structures and soil organic matter respectively, with leaching occurring only for a net surplus higher than 80–100 kg N ha^{-1} yr^{-1} (Billen et al., 2013b).

2.3. Scenarios for reducing environmental losses at the 2050 horizon

We used the same procedures as explained above for calculating the agricultural N losses to the atmosphere and the hydrosphere in the F2F and AE scenarios at the 2050 horizon (Billen et al., 2022), (Table 3SM).

The F2F scenario promotes a reduction in synthetic fertilizer use of 20%, as well as an expansion of organic farming areas to 25% of the European agricultural land; moreover, in order to favour biodiversity, 10% of agricultural land is prescribed to be set aside and devoted to non-productive ecological infrastructures such as hedgerows. No change in human diet is prescribed in F2F (Table 3SM).

The AE scenario combines three levers: generalization of long and diversified organic rotations in arable cropland with complete ban of industrial fertilizers, reconnection of crop and livestock farming with no import of feed from outside the region, and changing the current human diet (Table 3SM). The AE scenario takes into account a more frugal and plant-based diet (5 kg N cap^{-1} yr^{-1} with 40% cereals, 10% grain legumes, 15% fruits and vegetables, and 35% animal proteins versus the current 60%). This amount of animal protein is, for example, in the range recommended by the so-called Mediterranean diet (Blas et al., 2019).

In both scenarios, the human population and its subnational distribution at the 2050 horizon are obtained from the "baseline projections" of Eurostat demographic prospects. Associated changes in food demand are also taken into account. Except for the 10% of land set aside in the F2F scenario, we did not consider any change in agricultural-related land cover (i.e., in arable land, permanent crops, and permanent grassland).

In the conventional cropland of the F2F scenario, the same crop mix as in the current situation is cultivated, but with a 20% reduction in the rate of synthetic fertilizer application. This means that the overall reduction in N fertilizers, including all UAA (i.e., also organic and set-aside land), would be much higher than the 20% goal of the F2F. Livestock numbers are sized to the feed resources, with a similar import of feed as in the current situation (i.e., 2014–2019). In the 25% of organic farming areas of the F2F, cropland is similar to the AE scenario.

In the AE scenario, crop rotations are similar to those currently used by organic farms in the different regions (Billen et al., 2021), with a large contribution of grain and forage legumes and no input of industrial fertilizers. The yield is calculated from the total soil N input using the same yield–fertilization relationships as for current conventional arable rotations. This relationship is defined by a yield parameter (Ymax, see Lassaletta et al., 2014b) which is considered only dependent on the pedo-climatic conditions in each region (see Table 1SM for Ymax values). As in the F2F scenario, livestock numbers in each region are sized to the feed resources, here restricted to local production of grass and fodder crops including forage legumes (for ruminants) and cereals in surplus over human local requirements as well as food spills (for monogastrics). A fixed value of 20% was considered for the feed conversion efficiency into edible products by monogastrics. For ruminants, the conversion efficiency was derived from the relationship with time spent outdoors empirically found in current livestock systems as shown in Fig. 1SM, assuming that ruminants spend 79%, 51%, and 60% of the time outdoors in Portugal, Spain, and France, respectively. Monogastrics are considered to spend all their time indoors in the three countries, although extensive pig farming (outdoors) can represent (e.g., in Spain) about 10% of production. The AE scenario also considers the recycling of 25% of human excreta to market gardening and cropping systems (Esculier, 2018; Billen et al., 2021). Arable land thus received all available manure stock (animal and human) remaining after manure management losses and distribution to permanent crops and permanent grassland. Permanent grasslands are also fertilized by symbiotic N fixation and manure excreted outdoors. All land types receive atmospheric deposition. The same factors of losses at storage and application as in the current situation were considered in the scenarios (see Table 1SM).

3. Results

3.1. Current agricultural N flows and system typology

An overview of the main N inputs used in the study area and of the share of monogastric animals in the total livestock population is given in Table 2SM. At the country scale, fertilizers are applied on French UAA at rates twice those of Spain and Portugal (Table 4SM). Livestock density is the highest in France (see Table 2SM), with the lowest proportion of monogastrics (20%), followed by Portugal (28%) and Spain (45%) (Table 4SM).

On the basis of the GRAFS analysis of the three countries, briefly described in the previous sections and extensively presented in Table 1SM, the typology for all NUTS2 typically shows more *specialized cereal cropping systems*, and *forage based-crop and livestock systems* in the temperate conditions of the northern two thirds of France, while *disconnected crop and livestock systems* are mostly found in the south of France and Iberian Peninsula (Fig. 2).

Specialized intensive livestock farming systems are characterized by a high livestock density combined with a large share of imported feed to meet animal nutrition; in these systems, livestock farming is loosely connected to regional cropping systems. *Mixed crop and livestock systems* have a high degree of coupling between crop and livestock farming activities because (i) manure provides a relatively high proportion of cropland soil fertilization, and (ii) local agricultural production provides a high share of animal nutrition. In *grass-based systems*, permanent grassland provides at least half the animal feed, while in *forage-based systems*, local cropland produces a significant share of animal nutrition. In *disconnected crop and livestock systems*, crop and livestock farming both co-exist but without strong connections in terms of manure used by crops and local feed products in livestock feeding. *Specialized stockless cropping systems* refer to agro-food systems where crop production is much more important in terms of material flow than livestock farming (Fig. 2).

Urban systems are mostly represented in regions with densely populated, often touristy cities (regions including Paris and Marseille and Corsica in France; Madrid, Bilbao, and Valencia in Spain; Porto and Faro in Portugal; Fig. 2). These newly defined urban systems are superimposed on the other ones, e.g., the Regions of Corsica [Co], Ile-de-France [IDF], Comunitad de Madrid [Mad] and Norte Portugal [NPo] being additionally and respectively, a grass based crop and livestock mixed system, a specialized stockless cropping system, and two disconnected crop and livestock systems (Fig. 2).

France can be distinguished by its intensive livestock farming (Bretagne [Bre]), large cereal production areas (Center-Val de Loire [CVLo], Champagne-Ardennes [C–Ar], and Picardie [Pic] in the north of France, and Aquitaine region [Aqui], in the southwest), as well as forage-based crop and livestock mixed systems in the east/center and the region around Bretagne (see also Le Noë et al., 2017, 2018). Disconnected crop and livestock systems characterizing Mediterranean regions are often linked to permanent crops, such as olive, vineyards, orchard crops, etc., which represent 4% in France, but about 20% in both Spain and Portugal, together with greenhouse vegetable production (Fig. 2).

In Spain two NUTS2 regions are specialized in livestock systems (Catalonia [Cat] and Cantabria [Can]) and one in Portugal (Área Metropolitana de Lisboa [Lis]), while grassland-based crop and livestock systems only dominate in the regions of Galicia ([Gal] northern Spain). Typically, specialized livestock systems are located on the coastal regions where the presence of ports enables the easy import of feedstuff, and they are not necessarily found where the percentage of grasslands in UAA is the highest (Fig. 2SM a, b). Monogastrics are mostly found in the Centro Portugal and Alentejo regions [CPo and Alen] of Portugal, in Aragon [Ara], Region de Murcia [Mur], and Castilla y Leon and Catalonia [Leo and Cat] in Spain, as well as in Bretagne [Bre] in France (Fig. 2SM b).

3.2. Spatial distribution of current agricultural N flows

The application of fertilizers and manure inputs to cropland are an apt illustration of the distribution and intensity of cropping and livestock breeding, which are at the origin of NH_3 volatilization and N_2O emissions. Logically in the three countries, synthetic N fertilizers are mostly applied where manure is not available, and *vice versa*, since these synthetic fertilizers are generally applied to cereal systems, permanent crops, and greenhouse vegetable production. The spatial distribution of manure input to cropland is associated with the livestock density distribution (Fig. 2SM c). Interestingly, in some NUTS2 regions, manure and synthetic fertilizers seem to be applied simultaneously, mostly in specialized livestock systems or mixed systems, which also produce some feed (e.g., Bre and Basse Normandie [BNo] in France, Cat and Gal in Spain, and the small Lis region in Portugal; Figs. 2SM and 3SM).

The share of imported feed in the total ingestion of livestock typically reflects the region where livestock is intensively raised. The regions importing the most feed are those of Bretagne and the two thirds of the Iberian Peninsula (northeast–southwest regions) where monogastrics represent about half of the livestock (Spanish east Mediterranean coast, and north Portugal in particular) (Fig. 3SM d).

Total fertilization rates of arable cropland are 202, 113, and 120 kg N ha^{-1} yr^{-1}, in France, Spain, and Portugal, respectively. Corresponding rates in permanent crops are 51, 71, 47 kg N ha^{-1} yr^{-1} representing 26%, 70% and 55% respectively of the total cropland fertilization (arable + permanent); this illustrates the importance of fertilization of permanent crops in Spain, and less so in Portugal with a comparable percentage of surface area (see Table 4SM). In France, the low proportion of fertilization to permanent crops is linked to their relatively small cultivation area, which is restricted to the south (see Fig. 2, *Disconnected crop and livestock systems*). Total inputs to grassland are 122, 61, 69 kg N ha^{-1} yr^{-1} in France, Spain and Portugal respectively, much higher in France due to mineral synthetic application, in addition to manure input and symbiotic fixation which were similar in the three countries (See Table 1SM).

In order to analyze each category of the typology, they have been characterized on average for temperate and Mediterranean regions separately, i.e., a total of 21 and 22 regions, respectively (Table 1). The temperature is higher by 0.5–1.8 °C in Mediterranean regions compared to temperate ones, while rainfall is lower in the former by 7–50%. Interestingly, some systems are poorly represented, such as specialized livestock systems (two regions in each climate type) and grass-based crop and livestock mixed systems (one region for each), while specialized cropping systems comprise eight regions in the temperate area but none under Mediterranean climate. Forage-based crop and livestock mixed systems were more representative of temperate areas than of those with Mediterranean climate (five vs. two), with the opposite being found for disconnected crop and livestock systems, with a much lower number in temperate areas (six vs. 16).

Typically, intensive livestock systems show a high total fertilization and a high proportion of arable land in UAA that receives about 50% of manure. Livestock density is the highest for this system (>1 LSU ha^{-1}), and is dominated by milk-producing ruminants in temperate regions (62%) and monogastrics (71%) in Mediterranean areas (Table 1). Specialized cropping systems have a low livestock density and hence low manure available but, conversely, they have high total fertilization rates (220 kg N ha^{-1} yr^{-1}).

Grassland systems, which only represent 1.7% of the total UAA surface area, are dominated by grasslands (~90%) and ruminants (~97%), but the small proportion of arable land receive the highest total fertilization with more than 50% as manure. Forage-based crop and livestock mixed systems as well as disconnected crop and livestock

Table 1
Average characteristics in the different classes of typology for temperate (T) and Mediterranean (M) climates of the study domain: mean temperature; rainfall; utilized agriculture area (UAA) total surface area in Mha; percentage of arable cropland and of permanent grassland area in UAA; total fertilizers to UAA and to arable cropland (kg N ha^{-1} yr^{-1}) and percentage of manure input to arable cropland in its total fertilization; livestock density in livestock unit (LSU) per ha UAA; percentage of monogastrics and ruminants in the total livestock (see Fig. 4SM for the distribution of climate zones). NA: not applicable.

Systems	Climate	Mean temp	Rainfall	UAA total area	Arable cropland in UAA	Perm. grassland in UAA	Total fertilizer input to UAA	Total fertilizer input to arable cropland	Manure input to arable cropland	Livestock density	Monogastrics	Ruminants
		°C	mm yr^{-1}	Mha	%	%	kg N ha^{-1} yr^{-1}	kg N ha^{-1} yr^{-1}	%	LSU ha_{UAA}^{-1}	%	%
Intensive livestock	T	12.2	1150	2.5	65	31	206	223	55	1.5	38	62
Intensive livestock	M	13.2	556	1.3	47	29	153	216	47	1.2	71	29
Grass based	T	11.9	1028	1.1	6	92	122	465	61	0.8	1	99
Grass based	M	13.7	722	0.9	5	87	60	296	60	0.4	3	97
Forage based	T	11.1	891	6.2	54	45	169	191	38	1.0	13	87
Forage based	M	11.1	891	4.0	33	57	88	146	34	0.5	15	85
Disconnected	T	12.2	880	5.0	45	42	152	208	29	0.6	19	81
Disconnected	M	13.7	560	25.6	48	32	87	123	22	0.3	36	64
Special. stockless	T	12.0	770	9.6	78	18	195	217	10	0.4	17	83
Special. stockless	M	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

systems represent 73% of the total surface UAA, with lower livestock density in Mediterranean regions, a dominance of ruminants, a relatively well-balanced percentage of arable land and grassland, and a total UAA fertilization rate of 87–169 kg N ha^{-1} yr^{-1}, i.e., ~15% less than for the other systems (Table 1).

3.3. N losses from agriculture: NH_3 volatilization, N_2O emissions, and N leaching

Here, NH_3 volatilization (at fertilizer application and manure storage) represents about 90% of the sum of reactive N losses to the atmosphere (calculated as NH_3 volatilization + N_2O emission, although nitric oxide emission –NO- is possibly similar to N_2O, Smith et al., 1997), for the different regions in France, Spain, and Portugal. N_2O emission thus represents about 10% of the total atmospheric reactive N emissions (Table 2). Dinitrogen (N_2) issued from denitrification, although inert, represents a loss of N of 28%, 19%, and 22% of the total atmospheric N losses for France, Spain, and Portugal, respectively. Expressed relatively to synthetic fertilizers and manure applied to UAA, i.e., exogenous fertilizers provided by the farmers, atmospheric losses are higher in Portugal (24%) and Spain (20%) than in France (17%), mostly because of higher NH_3 volatilization rates (Table 2).

At subnational scales, hotspots of both NH_3 volatilization (>40 kg N–NH_3 $ha_{UAA}{}^{-1}$ yr^{-1}) and N_2O emissions (>4 kg N–N_2O $ha_{UAA}{}^{-1}$ yr^{-1}) often coincide with the specialized livestock systems, in Bretagne (France), in Catalonia and Galicia (Spain), and in Lisbon (Portugal) (Fig. 3; see also Fig. 2). In other agro-food systems, either defined as forage-dominated mixed crop and livestock or in those where crop and livestock are disconnected, NH_3 volatilization is logically rather high, and ranges from 20 to 40 kg N–NH_3 ha^{-1}_{UAA} yr^{-1}. Conversely, N_2O emissions dominate in specialized stockless cropping systems where soils are mainly fertilized with synthetic products, but also in the disconnected crop and livestock systems (Fig. 3a and b). Whereas most of the French regions (except at the French Mediterranean coast) are relatively high N atmospheric emitters (as N–N_2O and/or N–NH_3), the highest N atmospheric losses are mostly distributed around the Iberian Peninsula on the Mediterranean coast of eastern Spain, and at the Portuguese and Spanish Atlantic coast (Fig. 3a and b). Regarding N losses by arable land through leaching, which we derived from the corresponding N net arable land surplus (Fig. 3c), the average concentrations were the highest in Spain (50 N–NO_3 mg L^{-1}) and the lowest in Portugal and France (respectively, 20 and 15 mg N l^{-1}), far above the concentration that is allowed for aquifers, and drinking water, namely 11.3 mg N–NO_3 L^{-1} (50 mg NO_3 L^{-1}; see EU-Nitrate Directive, 1991; EU-Ground Water Directive, 2006) (Fig. 3d). As mentioned above, net surplus from arable land is more likely to be leached, grassland and permanent crops leaching occurring for higher net surplus, and permanent crop surface areas being relatively lower in proportion (Fig. 5SM, Table 2SM).

Overall, N_2O emissions averaged for the regions belonging to a specific system are systematically lower for areas with a Mediterranean climate, which is also true for NH_3 volatilization except for disconnected systems (Table 3). However, N leaching concentrations were mostly lower in temperate areas, owing to the dilution of the surplus in a larger leached water flux. When plotting N_2O emission against NH_3 volatilization, we found a concomitant increase, except for intensive livestock systems in the region of Bretagne [Bre], Galicia [Gal], AM Lisbon [Lisb], and Catalonia [Cat], for which NH_3 increases more (Fig. 6SM). Similarly, NH_3 volatilization is relatively higher in the disconnected Spanish Murcia [Mur] region and two French regions, Pays de Loire (PLo) and Basse-Normandie [BNo], where livestock density is in the higher range of the forage system to which they belong (Fig. 6SM a). Leaching was overall well related to the net surplus, but the intensive livestock system in Catalonia [Cat] leads to high leaching, while the grass-dominated system of Asturias [Ast] shows lower leaching (Fig. 6SM b).

3.4. Farm-to-Fork and agro-ecological scenarios: comparison with the reference

We explored two scenarios (F2F and AE) to assess the impact of systemic changes in the agro-food systems in terms of environmental N losses and food requirements of the European population at the 2050 horizon. While the F2F scenario depicts the continuation of the current agro-food system, although some measures intend to reduce intensification and maintain biodiversity, the more radical but not prescriptive AE scenario takes the option of completely banishing feed imports and the use of industrial N fertilizers.

On the whole, compared to the figures in the reference period 2014–2019, livestock would only show a minor reduction for the F2F scenario (by ca. 10–15% for France, Spain, and Portugal) while it would be reduced by approximately 70% in Spain and Portugal and by 35% in France for the AE scenario. Total N inputs to soils would decrease strongly in France, Spain, and Portugal (38%, 63%, and 52%, respectively) for AE, while the reduction would be about 3 times less with F2F (Table 4).

Applying the same criteria as those used for classifying the current regions in terms of the typology described in Fig. 2 to the results of the F2F and AE scenarios shows that the former scenario would not suppress the current regional specialization, while the latter would result in all subnational regions becoming crop and livestock mixed systems (Fig. 4).

Accordingly, both gaseous losses to the atmosphere (NH_3 and N_2O) and leaching nitrate losses from arable land would be substantially more reduced in the AE than in the F2F scenario (Table 4).

These differences were similar when the regions were considered (Fig. 5). Clearly, a profound change in the agro-food system, as formulated in the AE scenario, would be required to obtain a systematic decrease in environmental N losses in all regions, while the F2F scenario

Table 2
NH_3 volatilization and N_2O emissions from agriculture in France, Spain, and Portugal. Inert N lost from denitrification is shown for comparison with the reactive losses as well as net surplus (in Gg N yr^{-1} and kg N ha^{-1} yr^{-1} for UAA). Total atmospheric losses are provided as a percentage of exogenous intended inputs. (All values determined from the methodology described in section 3.1.; Table 1SM).

	NH_3 volatilization	N_2O emissions	N_2 from denitrif.	Total N inputs[a]	Atmos. losses	N net surplus	
	Gg N yr^{-1}	Gg N yr^{-1}	Gg N yr^{-1}	Gg N yr^{-1}	%	Gg N yr^{-1}	
	kg N $ha^{-1}yr^{-1}$	*kg N $ha^{-1}yr^{-1}$*	*kg N $ha^{-1}yr^{-1}$*	*kg N $ha^{-1}yr^{-1}$*		*kg N $ha^{-1}yr^{-1}$*	
France	491	64.7	214	4576	17	1144	
	18	*2.4*	*8.0*	*171*		*43*	*(50)*[b]
Spain	339	21.9	83	2191	20	1047	
	14	*0.9*	*3.4*	*89*		*42*	*(54)*[b]
Portugal	43	3.8	13	247	24	103	
	13	*1.2*	*4.1*	*76*		*32*	*(50)*[b]

[a] Synthetic fertilizers + manure + Atm deposition + N fixation In Italics are the values for ha UAA (arable cropland + permanent cropland + grassland).
[b] Between brackets are the values per ha of arable cropland, considered as a proxy for leaching.

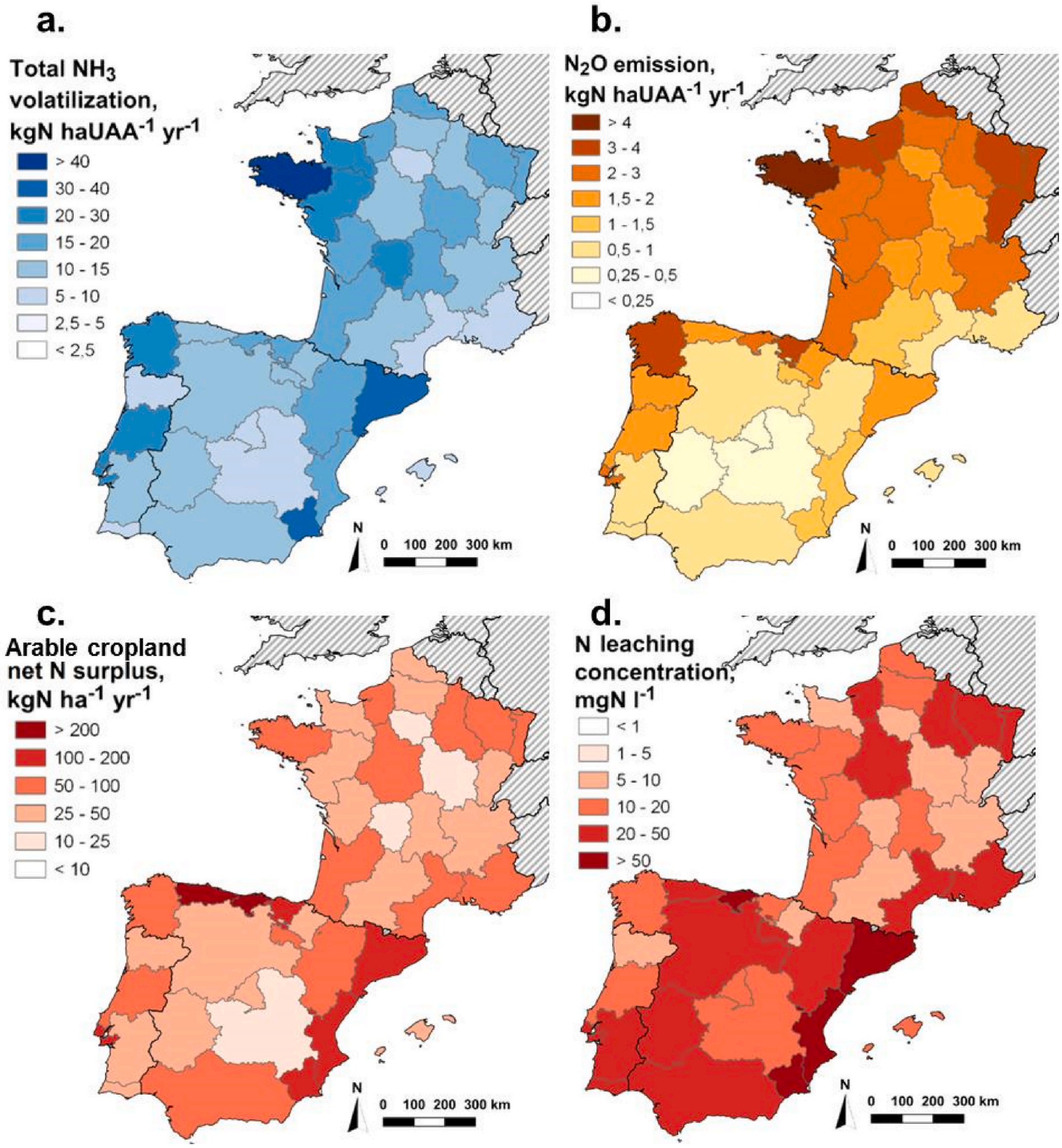

Fig. 3. Regional N losses in the three countries (average for 2014–2019). a. NH_3 volatilization and b. N_2O emissions, expressed in kg N per ha and year for utilized agricultural area (UAA); c. net surplus from arable cropland. d. N leaching concentration from arable cropland, in mg N L^{-1}.

would only lead to reduced losses in the currently most intensive regions. From the point of view of environmental protection, the F2F scenario can only be considered as a first step toward achieving environmental targets, but it is not able to halve N agricultural losses as recommended by the Colombo Declaration in 2022 (https://www.inms.international/colombo-declaration/colombo-declaration; Leip et al., 2022). Overall, N losses, currently amounting 3920 Gg N yr^{-1} for the three countries, would be reduced to 2544 Gg N yr^{-1} in the F2F scenarios and to 829 Gg N yr^{-1} in the AE scenario (cf. graphical abstract).

4. Discussion

The concept of N cascade states that the acceleration in the introduction of anthropogenic reactive N to the biosphere since the modernization and intensification of agriculture during the 1950s–1990s has led to increased environmental losses and transformation of N, but also to accumulation in some compartments (specifically the atmosphere and hydrosphere), which has had many environmental impacts (Galloway et al., 2003; Sutton et al., 2011).

4.1. Strong specialization of the regions

After a long period of self-subsistence with an agricultural system dominated by integrated crops and livestock farming, increasing agricultural production that was exported on the international market was observed in European countries (Bouwman et al., 2005, 2017). The strong specialization at subnational regions as observed for these west EU countries is at the origin of most of the adverse effects on the environment and is a typical result of the development of the market economy (Timmer, 1997; Klasen et al., 2016).

The typology highlights particularly well the specialized regions, i.e., livestock-dominated areas, on the one hand, and crop-dominated areas, on the other hand. According to Barrantes et al. (2009), this intensification of livestock systems has occurred in the extensive and

Table 3
Average losses in the different typology classes for temperate (T) and Mediterranean (M) climates of the study domain. N_2O total emission (kg N ha^{-1} yr^{-1}), NH_3 volatilization (kg N ha^{-1} yr^{-1}), net surplus values of arable cropland (kg N ha^{-1} yr^{-1}) from which leaching (mg N–NO_3 L^{-1}) is derived, diluted within the annual leaching water flux (see text).

Systems	Climate	N_2O total emissions	NH_3 total volat.	Net N surplus of arable cropland	Leaching conc.
		kg N ha^{-1} yr^{-1}	kg N ha^{-1} yr^{-1}	kg N ha^{-1} yr^{-1}	mg N L^{-1}
Intensive livestock	T	4.1	32.7	67.9	12.5
Intensive livestock	M	1.7	31.5	114.3	65.5
Grass based	T	1.9	14.8	240.0	38.6
Grass based	M	0.5	11.0	126.0	35.0
Forage based	T	2.4	21.9	33.4	9.2
Forage based	M	1.3	13.7	41.7	18.6
Disconnected	T	2.2	16.4	84.6	18.8
Disconnected	M	0.9	12.3	57.5	58.5
Special. stockless	T	2.6	15.3	61.4	19.2
Special. stockless	M	NA	NA	NA	NA

Table 4
Percentage of reduction in livestock density and total fertilization, of NH_3 volatilization, N_2O emission and N leaching concentration for France, Spain, and Portugal in the agro-ecological (AE) and Farm-to-Fork (F2F) scenarios, compared to the reference situation (2014–2019).

	France		Spain		Portugal	
	Percentage of reduction compared to the reference					
	F2F	AE	F2F	AE	F2F	AE
Livestock density	9	35	15	73	14	74
Total fertilization	13	38	21	63	17	52
NH_3 volatilization	33	47	41	80	32	77
N_2O emission	26	58	27	63	23	60
N leaching	29	63	28	94	16	84

semi-extensive ruminant livestock systems of Mediterranean countries and more generally where the climate and soil contexts were favorable. As shown by the typology, this is the case for Catalonia and Galicia in Spain, for which these extensive ruminant livestock systems have more or less evolved into monogastric breeding, (representing 73% and 26%, respectively) or dairy systems fueled by imported feed (e.g., Galicia). Bretagne also belongs to the specialized livestock typological class, with ~50% of monogastric breeding. It is the main region of meat and milk production in France, generating a significant number of jobs in the industrial sector (ca. 55,000) (Deschamps et al., 2016). However, Bretagne imports large quantities of protein crops (particularly soybean meal) from South America (Le Noë et al., 2016). Similarly, feed import dependence is high in Catalonia and Galicia (81% and 57%, respectively). Importantly, such specialized systems remain more vulnerable than diversified systems in the context of a crisis (economic, price volatility, environmental, drought period, etc.) (Lebacq et al., 2015).

Regarding stockless cropping systems — observed in the Parisian basin, in the middle Loire alluvial plain, and in the southwest of France — none of this type appears in the Iberian Peninsula; stockless cropping systems are indeed favored by lowland areas with loamy soils and temperate climate, whether they are drained (in the Parisian basin, wheat is the dominant crop) or irrigated (in the southwest of France, maize is the dominant crop). These systems are mostly fertilized with synthetic N fertilizers at an average rate exceeding 150 kg ha^{-1} yr^{-1}, a value among the greatest of the three countries. Interviews from 10 conventional farmers for 6 years over a typical 2–3-year rotation in the Parisian basin led to an average synthetic N inputs of 152.2 kg ha^{-1} yr^{-1} ($n = 132$, SD = 57.5 kg N ha^{-1} yr^{-1}, unpublished data; see also Benoit et al., 2016). Typically, in these intensive cropping systems with low livestock density, manure input to arable land is low and the transportation of manure from a livestock-specialized region (e.g., Bretagne), 300–500 km away from cropping areas, cannot be manageable with sustainability e.g., transportation costs (Flotats et al., 2009); therefore industrial synthetic fertilizers — which, however, are not manufactured locally (French production N fertilizers hardly covers 34% of the N needs of French agriculture, UNIFA, 2022) — are still preferably used despite their price increasing considerably and steadily since January 2021 (World Bank, https://blogs.worldbank.org/opendata/fertilizer-prices-expected-remain-higher-longer).

However, even though livestock and cropping systems can coexist in the same region, disconnection can occur, which is especially the case in most of Spain and Portugal, but also in some regions of France. In these

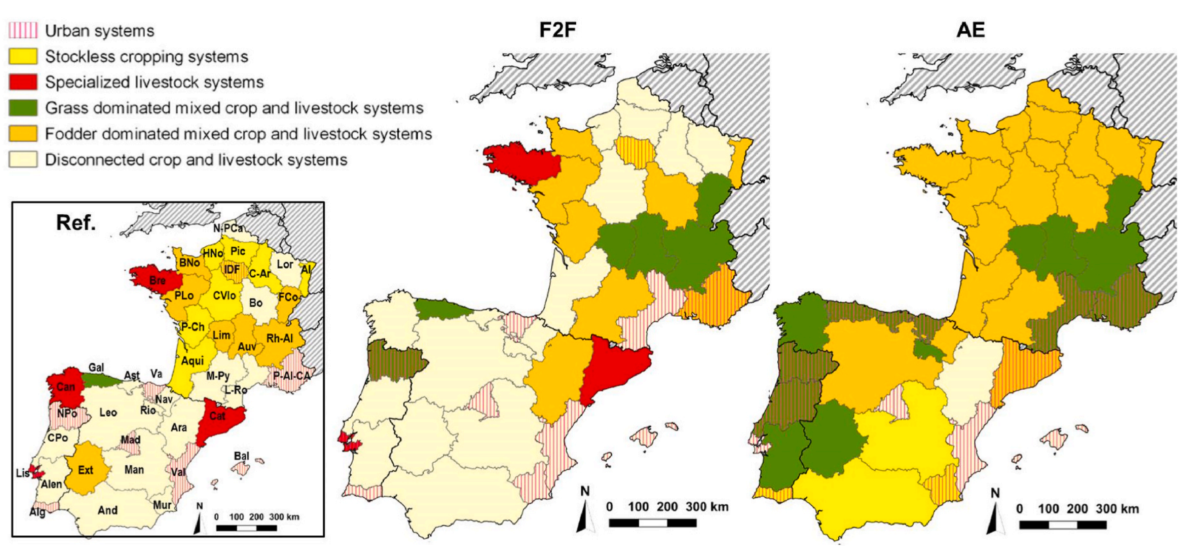

Fig. 4. Typology of the agro-food systems in the F2F and AE scenarios. The reference (Ref.) situation is provided for comparison. Pink hatched areas of the urban systems are also colored according to the other dominating system (specialized stockless, disconnected crop and livestock systems, grass-based crop, and livestock mixed system). Names of NUTS on the Ref. map (left) are abbreviated (see Table 5SM for complete definitions).

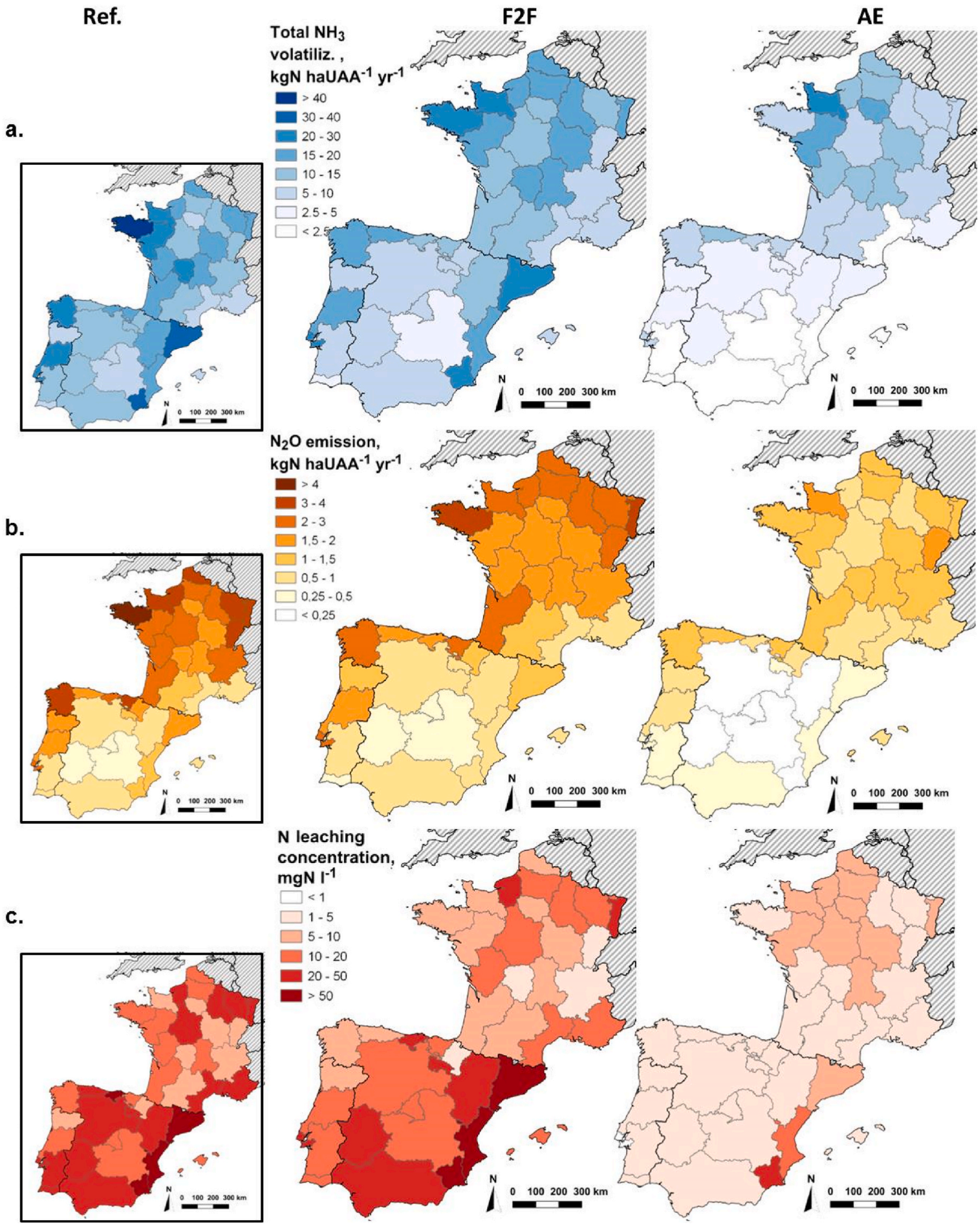

Fig. 5. Atmospheric and hydrological N losses at the regional scale of the three countries for reference (2014–2019) and the F2F and agro-ecological scenarios. a. NH_3 volatilization. b. N_2O emissions, both expressed in kg N per ha utilized agricultural area (UAA) and per year. c. N leaching concentrations, in mg N L^{-1}.

disconnected crop and livestock systems, livestock is rather low and comprises the second higher percentage of monogastrics (after the livestock intensive systems), so that manure represents hardly 25% of the crop fertilization (see Table 1). The crops of these disconnected systems include the highest proportion of permanent types (e.g., olive trees, vineyards) or vegetable and fruit gardening, often irrigated and in greenhouses, rather intensive agricultural practices, requiring thus 75% of mineral fertilizers (Lassaletta et al., 2021; Aguilera et al., 2021; Sanz-Cobena et al., 2023).

4.2. N losses to the environment

Our results show that NH_3 agricultural volatilization represents a significant proportion of total N inputs to UAA, from a few percent to more than half of the inputs, on average of 11% (France), 15% (Spain), and 17% (Portugal) loss. This proportion is similar to the 14% reported at the global scale and is a cause for lowering N use efficiency (Bouwman et al., 2002). Interestingly, the proportion of NH_3 volatilization in total losses is higher in the two Mediterranean countries, due to higher temperatures and drier soils following surface applications of fertilizer according to the MANNER model (Bittman et al., 2014; Buijsman et al., 1987; Sommer and Hutchings, 1995), which is exacerbated in certain regions with large confined pig farms (e.g., Aragón). The MANNER model also accounts for the types of fertilizers used (urea-based) and their method of application (liquid manure, alkalinity of the solution, no incorporation in soils, etc.) can promote NH_3 volatilization. The proportion of urea utilized in agriculture (10%, 30%, 20%, respectively, in Portugal, Spain, and France), together with the amount of other synthetic nitrate-based fertilizers (e.g., ammonium nitrate and sulfate, calcium ammonium nitrate (46%, 29%, 38%) and N solution (8%, 9%, 31%) have indeed different propensity to volatilization (IFAstat). The use of urease inhibitors can, however, reduce ammonia emissions from urea-based fertilizers (Sanz-Cobena et al., 2008, 2014; Abalos et al., 2014). Yet, despite synthetic fertilizer input to arable land represents a higher proportion (49%, 55%, and 61%) than manure (40%, 26%, and 21%) in Portugal, Spain, and France, respectively, manure application had a proportionally bigger role in NH_3 volatilization, albeit variable (80%, 47%, and 51%, respectively, for the three countries, Table 1SM).

Nitrous oxide fluxes have often been reported to be associated with synthetic fertilizer applications (Bouwman, 1996; Skiba et al., 1996; Smith et al., 1997), but manure and other organic fertilizers also contribute to N_2O emissions (Aguilera et al., 2013; Cameira et al., 2020). Although agriculture is responsible for 79%, 81%, and 92% of total N_2O emissions in Portugal, Spain, and France, respectively (83% for Europe, EU-Eurostat data, 2022), N_2O emissions account for only a rather low proportion of the total UAA fertilization (1.5% and 1.4% for France and Portugal and 1% for Spain). These proportions might seem a little high when compared to the emission factors (EF) estimated for Mediterranean countries between 0.27% and 0.91% by Cayuela et al. (2017) and Cameira et al. (2021), below the default IPCC EF of 1% (IPCC, Intergovernmental Panel on Climate Change, 2006, Tier 1). However, we show large variations between regions of 0.9–2.1% for France, 0.4–2.4% for Spain, and 1.2–1.9% for Portugal, a regional approach also taken into account in the 2019 IPCC refinement report (IPCC, Intergovernmental Panel on Climate Change, 2019), updated through a disaggregation of EFs by dry or wet areas and by fertilizer type (see also Hergoualc'h et al., 2021). While NH_3 volatilization is a physical-chemical process and N_2O emissions are mediated by microorganisms, the processes are influenced by similar drivers such as soil and crop management factors (e.g., crops, soil mineral N, pH, water content and oxygen, carbon availability, C:N ratios) and environmental factors (temperature, rainfall, drying/wetting vs. freezing/thawing) (Röver et al., 1998; Vilain et al., 2010; Lesschen et al., 2011; Saggar et al., 2013).

Due to the high mobility of NO_3^- and despite the Nitrates Directive (EU-Nitrates Directive, 1991), the massive use of N fertilizers in agriculture has led to regular increase in surface water NO_3^- concentrations, as shown by long-term NO_3^- trends in large agricultural watersheds of the study domain (e.g., the Seine: Garnier et al., 2020; the Loire: Minaudo et al., 2015), but also in many other places around the world (e.g., the Mississippi: Turner et al., 1998; Alexander et al., 2008; the Yellow River: Xia et al., 2002; Liu et al., 2008). In France, many wells for drinking water have been closed due to nitrate contamination or to both NO_3^- and pesticide contamination at an average rate of 43 wells yr^{-1}, i.e., more than 1000 closures since 1994 (Garnier et al., 2023 under review; Source: https://ades.eaufrance.fr/Recherche). In Portugal, pollution mainly occurs in the Tagus Nitrate Vulnerable Zone (Nistor, 2020; Cordovil et al., 2018; Cameira et al., 2021; Serra et al., 2021), while it also occurs in several locations in Spain aggravated by the low aquifer recharge, mainly in the semi-arid areas that have increased in the context of climate change (Arrate et al., 1997; Pérez-Martín et al., 2016). Very recently, however, nitric contamination seems to have stabilized in some places, although this can be due to (i) best agricultural practices, mostly in regions where the residence times of aquifers are relatively low (e.g., 5–10 years) but also (ii) to implementation of stringent N treatment in wastewater plants.

The high NO_3^- leaching concentration in Spain (about 50 mg N l^{-1}), which is 2–3 times higher than in Portugal and France despite similar net surplus per unit area in the three countries, is the result of water deficit in Spain (see Fig. 1); this leaching concentration is indeed derived from the net surplus diluted in a much lower average annual runoff together with large areas devoted to intensive and highly irrigated agriculture. Although water management (e.g., construction of dams, diversion canals) has led to a notable N retention, including N denitrification in several ecosystems of the drainage basin (and not only in soils, as represented in Table 2), the remaining NO_3^- can reach surface water and aquifers (Romero et al., 2016; Pérez-Martín et al., 2016). Whereas several EU Directives (EU-Nitrates Directive, 1991; EU-Water Framework Directive, 2000; EU-Groundwater Directive, 2006) aimed at reducing or maintaining nitrate concentrations below 50 mg NO_3^- l^{-1} (i.e., 11.3 mg N l^{-1}), the leaching concentrations as defined in our study are well above this threshold.

Whatever the fate of agricultural N — whether lost in the atmosphere as NH_3 or N_2O and even as inert N_2 or leached as e.g., NO_3^- in hydrosystems — this N represents not only a decrease in N use efficiency (Bouwman et al., 2002), a burden for the environment and for human health, but also an economic waste for farmers, from 35% to 52% of the N inputs to soils, i.e., manure plus synthetic fertilizers.

4.3. Toward an ecological transition based on an integrated view of the agro-food system: barriers or motivations to change

The AE scenario explored here in the Temperate-Mediterranean gradient, considers three major levers of the agro-food system: human population and diet, livestock sized to local feed resources only, crop fertilization based on legume N fixation and manure from livestock, as well as recycled human excreta) (Billen et al., 2018, 2021). It clearly leads to large reductions in hydrological and atmospheric N losses; this was already shown at smaller geographical scales with similar scenarios (France, and Seine and Loire watersheds for NO_3^- and N_2O: Garnier et al., 2019, Garnier et al., 2020; Garnier et al., 2018; Spain provinces for NH_3 and/or N_2O: Sanz-Cobena et al., 2014; Sanz-Cobena et al., 2023) and at the European scale for scenarios testing the Nitrates Directives (Velthof et al., 2014) and deriving boundaries for N losses and N inputs (de Vries et al., 2021). Such an AE ambitious scenario has been proved feasible biogeochemically and able to ensure Europe's food autonomy (Billen et al., 2021, 2022), while we showed here the important benefit it would bring for the environment. People would have access to better air and water quality, but also a healthier diet with more vegetal proteins, while causing less global warming and, overall, improving human as well as animal welfare. Such a scenario is typically in line with Smil (2000, 2002), who already advocated that reducing N losses and modifying human diet, with less animal proteins, would support food for the growing world population.

The official F2F European strategy invites people to consume a diet in line with dietary recommendations (HLPE, 2017), but has no clear goal of dietary change. No specific recoupling of livestock and crop production is hypothesized in this scenario. Yet, several studies have shown the value of reconnection for improving feed and fertilizer autonomy, as well as pest regulation, and hence encourage the generalization of sustainable agriculture with reduced N losses (Ryschawy et al., 2012; Bonaudo et al., 2014; Regan et al., 2017). The F2F strategy prescribed 25% of agricultural area (at least) to be under organic farming, thus necessarily driving a reduction in the use of synthetic fertilizers. Despite a further abatement of synthetic fertilizers on the conventional surface areas by 20%, the 11.3 mgN l^{-1} threshold was only barely achieved for France (10.8 mgN l^{-1}), while leaching concentrations would remain far from the target in Spain (36.9 mgN l^{-1}) and closer to the target for Portugal (15.9 mgN l^{-1}). Our F2F scenario indicates a risk that the EU-F2F strategy (2020) would not fully meet all its ambitions, especially in terms of an effective reduction of 50% of N losses to the environment, as the reduction in atmospheric losses is also modest. To reach its targets without requiring large additional food imports to the EU, the EU F2F strategy should clearly encourage consumers to change their dietary habits, including reducing their consumption of animal products, a diet that is fully aligned with the Mediterranean diet recommendations (Leip et al., 2022). Further, in addition to favoring biodiversity with organic farming (Smith et al., 2011), the EU F2F strategy, including some biodiversity strategy goals, requires the promotion of at least 10% of dedicated land set aside specifically for increasing biodiversity. Overall, the Green Deal and F2F strategy can represent a step further toward sustainable agriculture, a fair and healthy diet, increased biodiversity, and reduced environmental damages, but is at serious risk of bending under pressure from detractors, such as those that have appeared with the Russian attack of Ukraine (Aubert et al., 2022).

As previously stated by Smith et al. (2013), the need to feed a growing population and to limit climate changes (including a healthier diet and water resource protection) is still today, 10 years later, one of the greatest challenge our society faces.

4.4. Strengths and weaknesses of our approach

The strength of our approach is that it integrates a thorough analysis of the agro-food systems at the scale of 43 subnational regions in a large domain with a north–south distance of 2000 km in a climatic gradient from temperate to Mediterranean. Whereas key input data related to N fertilization and to crop and livestock production are available at a subnational scale, N losses are generally only provided at a national scale and are mostly based on emission factors that are not always adapted to subnational specificities (e.g., Cayuela et al., 2017 for N_2O; Sanz-Cobena et al., 2014 for NH_3 emissions). Ammonia volatilization and N_2O emissions have been measured for several decades in many places, mostly at plot scale, so that emission factors have been periodically revised (IPCC, Intergovernmental Panel on Climate Change, 2006, 2019). Here, we used empirical modeling tools based on subnational statistics of N fertilizer application and N manure management, as well as subnational climatic variables, to calculate regional N losses.

When comparing for France and Iberian Peninsula the results obtained at a same NUTS2 resolution for Europe by Velthof et al. (2014) and de Vries et al. (2021), we observe coherent spatial patterns and annual ranges for NH_3 volatilization and N_2O emissions in France, although we generally find higher fluxes for the Iberian Peninsula. Results are more difficult to compare for denitrification and leaching. Indeed, soil denitrification is difficult to directly measure on field and is often derived from other measurements, i.e., N_2O emission here and difference between N soils surplus and leaching in Velthof et al. (2014) and de Vries et al. (2021).

The major weaknesses of our approach could be related to the fact that our data sources have a yearly resolution (average for 2014–2019) without documenting the hot moments in the year associated with, e.g., fertilizer applications, rainfall, and temperature, etc. (Saggar et al., 2013, and ref. in). These hot moments are however integrated in the yearly average losses by the GRAFS approach. The relatively large spatial scale NUTS2 used may prevent highlighting local N hotspots, particularly NH_3, N_2O emissions and/or N leaching to surface- and groundwater (Serra et al., 2019; Lassaletta et al., 2012; Benoit et al., 2016). Therefore, in complement to the empirical GRAFS approach integrating all N fluxes and assessing responses of the whole agro-food system, other mechanistic tools applied at fully distributed spatial scale, such as DAYCENT, DNDC models (Smith et al., 2008; Li et al., 2019; Lutz et al., 2019) should be used to simulate peaks of N emissions which often represent a large proportion of the annual fluxes; these models would help refining mitigating emissions.

Furthermore, many soil characteristics, reported as driving factors for gaseous losses, are not considered here (Saggar et al., 2013). Additionally, N incorporation in the soil organic matter pool, or N release from soil organic matter when there is a net mineralization, are not considered; soil organic matter was assumed to be at steady state (van Grinsven et al., 2022). Irrigation was not taken into account, although it is known to greatly affect N_2O emissions (Cayuela et al., 2017). Yet, it is well developed in Mediterranean regions, where it has enabled the generalization of disconnected crop and livestock systems, especially for maize grain to feed poultry and pigs in Spain, both having increased by a factor of 3–4 since the 1960s (Lassaletta et al., 2014a).

There is a continued need for more knowledge on factors controlling N losses to the atmosphere and the hydrosphere, in order to possibly determine mechanistic relationships and specific model parametrizations, particularly in countries with distinct agro-climatic regions, and specifically in Portugal, Spain, and France. Such process-based modeling approaches could also account for seasonal variations, including the peaks in emissions occurring at specific occasions.

5. Conclusions

The GRAFS approach offers an integrated view of the agro-food system, while quantifying annual N losses to the atmosphere and to the hydrosphere. This approach is applicable at various scales, from local to continental or global scales. Here, we adopted a subnational resolution (NUTS2), which clearly identifies a variety of agricultural systems. In the north-to-south gradient from temperate to Mediterranean climate, we found a dominance of specialized stockless cropping systems, together with mixed systems in France (forage and livestock based), while disconnected crop and livestock systems were seen in the Mediterranean climate of the Iberian Peninsula. Intensive livestock systems are present in all three countries, associated with a specialization of agriculture. With a total average fertilization rate in the order of $171 > 89 > 76$ kg N ha^{-1} yr^{-1} for France, Spain, and Portugal, respectively, we found the highest atmospheric losses of NH_3 and N_2O for France (13.7 and 2.4 kg N ha^{-1} yr^{-1}), followed by Portugal (13.3 and 1.2 kg N ha^{-1} yr^{-1}), and finally Spain (9.4 and 0.9 kg N ha^{-1} yr^{-1}). However, the highest leaching NO_3^- concentration was found for Spain (~50 mg N l^{-1}), followed by Portugal (~20 mg N l^{-1}) and France (~15 mg N l^{-1}). Compared to the reference situation (on average for the period 2014–2019), the two scenarios explored would lead to reductions in N loss. By far, the biggest reductions are seen in the AE scenario, for both atmospheric and hydrosphere losses, with emission reductions in the range of 47–80% for NH_3 volatilization, 58–63% for N_2O emission, and 63–94% for NO_3^- leaching concentrations, i.e., 1.9, 2.4, and 3.6 times higher for NH_3, N_2O, and NO_3^-, respectively, than the F2F scenario.

The results of these two scenarios confirm the need for profound structural changes in the agrofood system, including tight connections between the distribution, consumption, and production systems. The EU F2F strategy could be more efficient if revisited from a dietary perspective, beyond its current general recommendations.

Credit author statement

JG: Conceptualization, Methodology; Writing – original draft; GB: Conceptualization; Methodology; Writing – review & editing; EA, LL, RE, ASC: Methodology; Writing – review & editing; JS, RC, CC: Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary figures, tables and data to this article can be found online at https://doi.org/10.1016/j.jenvman.2023.117732 and at https://zenodo.org/record/7457183#.Y6BF-u2ZPDC.

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