Impact of changing agricultural management on the exceedance of empirical critical loads of nitrogen in terrestrial habitats of Southwestern Europe☆

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ARTICLE INFO	ABSTRACT
	Ammonia (NH₃) emissions from agricultural activities are one of the main sources of air pollution, a challenge for EU emission targets, and contribute significantly to nitrogen (N) deposition and eutrophication of sensitive ecosystems. This study used modelling to evaluate the mitigation of this eutrophication by improved fertilizer management techniques in the EU Interreg "SUDOE" region (Spain, Portugal, and southwestern France), comparing it with the current situation. The results showed that the implementation of improved fertilizer management -removing urea-based fertilizers and optimizing manure application-led to a 36 % reduction in NH₃ emissions and a 20 % decrease in total N deposition. Consequently, the area of habitats exceeding their critical nitrogen load dropped from 39 % to 22 %, representing a 43 % reduction in area at risk of eutrophication. However, spatial heterogeneity was considerable, driven by atmospheric transport, deposition patterns, and distribution of sensitive habitats. The majority of the most sensitive habitats experienced a reduction of their area at risk, particularly natural and semi-natural grasslands, and most of shrublands. However, some others, such as coastal dunes and certain Mediterranean shrublands, experienced smaller improvements. The Alpine region remained the most threatened region. This study highlights the importance of improved fertilizer management in achieving strategic environmental goals, and confirms that spatially explicit modelling and precautionary assessments using critical loads are useful tools to inform regionally adapted environmental policies, considering sensitivity of the ecosystems, deposition magnitude and pollution sources as key factors.

1. Introduction

Atmospheric deposition of nitrogen (N) pollutants causes negative impacts on terrestrial ecosystems. These impacts are associated with the direct exposure of vegetation to ammonia (NH₃), nitrogen oxides (NOₓ) and ozone (O₃), as well as eutrophication and acidification of ecosystems by N deposition, and often lead to increased susceptibility to drought, diseases and pests, losses of plant biodiversity, and disturbances of ecosystem functioning and services (De Vries, 2021; Bobbink et al., 2022; Clark et al., 2013). In response, the EU "Zero Pollution Action Plan" (EC, 2021), part of the European Green Deal, aims to reduce pollution to non-harmful levels by 2050, with a 2030 target of reducing by 25 % the area of ecosystems where air pollution threatens biodiversity.

These efforts align with the European Green Deal (EC, 2019), which integrates key strategies like the EU Farm to Fork Strategy (EC, 2020a), aiming to reduce nutrient losses by 50 % by 2030, and the EU Biodiversity Strategy for 2030 (EC, 2020b), which highlights air pollution's impact on ecosystems. Complementarily, the Gothenburg Protocol (UNECE, 2012), under the CLRTAP (UNECE, 1979), has long guided emission inventories, monitoring, and risk assessments to reduce eutrophication in Europe.

Building on this, the NEC Directive (2016/2284/EU; EC, 2016) sets binding targets for five major air pollutants, including NH_3, and obliges Member States to implement National Air Pollution Control Programmes (NAPCPs) that include measures to reduce emissions across relevant sectors. The NEC Directive is binding legislation, and Member States may ultimately face EU fines if they fail to comply. As noted in the EU Commission's Fourth Clean Air Outlook (EC, 2025), NH_3 emissions remain problematic, requiring further reductions. Moreover, the revised Ambient Air Quality Directive (EU) 2024/2881 acknowledges NH_3 as a pollutant of emerging concern and reinforces coordination with the NEC Directive and EMEP to support ecosystem-based targets.

The effectiveness of these strategies can be enhanced by performing spatial analysis based on risk-assessment methodologies that consider the sensitivity of ecosystems to N deposition impacts (De Vries et al., 2021, 2024). The critical loads (CLs) - quantitative estimate of N deposition below which no negative effects on sensitive receptors are expected - methodology developed in the framework of the LRTAP Convention (CLRTAP, 2023) is widely used for this purpose and supports spatially targeted policy decisions (Sutton et al., 2014). Recent updates of empirical CLs ($CL_{emp}N$) by Bobbink et al. (2022) provide thresholds based on observed ecological changes, allowing the identification of areas where deposition exceeds these limits and mitigation is urgently needed.

Over the recent decades, the European Union (EU) has made great efforts to abate airborne N pollution. NO_x emissions have been successfully reduced by 63 % from 1990 to 2021, mainly through reductions in road transport, public electricity and heat production, while NH_3 emissions only decreased by 32 % during the same period (EEA, 2023b). Moreover, NH_3 emission reduction has slowed down in recent decades, decreasing by only 16 % in the EU from 2005 to 2022—the smallest reduction among all pollutants (EEA, 2024). The agriculture sector is the principal source, responsible for 93 % of total NH_3 emissions in the EU, and, therefore, decreasing NH_3 emissions from agricultural activities remains the greatest challenge in meeting EU emission commitments and targets (EEA, 2024).

Geographically, not all countries have contributed equally to these (insufficient) NH_3 reductions. In particular, countries of the EU Interreg region of Southwestern Europe "SUDOE" territory (Spain, Portugal, and southwestern France) show lower than average reductions. For this period, France and Portugal reported reductions of 19 % and 16 % respectively, while Spain reported only 2 %. In addition, atmospheric NH_3 concentration may have even increased in these countries (along with the rest of the southwestern region of Europe) during the last decade (2008–2018; van Damme et al., 2021; and 2005–2022; EEA, 2024).

Giving this situation, increasing efforts to reduce emissions from agricultural activities are key in the conservation perspectives of European habitats, and particularly in some regions such as the SUDOE territory. There is a considerable mitigation potential in crop production strategies (Sanz-Cobena et al., 2014), but there is still a gap in the spatial assessment of the efficacy of these mitigation strategies on ecosystem conservation in Southwestern Europe. While various strategies have been studied and implemented to reduce ammonia emissions (de Vries et al., 2024), there is a lack of an integrated modelling approach at the regional scale, that is essential to understand spatially explicit environmental impacts and to support evidence-based decision-making (Carnell et al., 2017; Kros et al., 2024).

In this study, we use the CL methodology in combination with a quantitative scenario analysis to assess the potential of NH_3 emission mitigation in crop production to protect natural and semi-natural habitats from impacts of N deposition in most part of the EU Interreg region of Southwestern Europe "SUDOE": Spain, Portugal, and southwestern France (excluding Andorra and UK Gibraltar) This region faces considerable challenges in reaching agreed NH_3 reductions targets from 2030 onwards (EEA, 2023a). We estimated CL exceedances in natural and semi-natural habitats using a coupled set of models for agricultural N-emissions, atmospheric N transport, transformations and deposition. We assessed a scenario that represents current agricultural practices and an exploratory scenario integrating several well-known NH_3 emission mitigation options in crop production, such as the removal of urea-based fertilizers and the adoption of improved manure application methods.

2. Material and methods

2.1. Ammonia emission scenarios

In the SUDOE territory, NH_3 emissions from agricultural activities represents 62 % of the N emitted to the atmosphere for the study period. The distribution of emissions by sectors varied across the territory (Fig. S1). Two contrasting scenarios were used in the present study to test the effect of changes in crop production practices on NH_3 emissions and on the resulting N deposition and subsequent CL exceedance: the *Baseline* scenario and the *Mitigation* scenario. The *Baseline* scenario represents current (2017–2018) management practices, while the *Mitigation* scenario includes several mitigation options for crop production that could be implemented in a near future (see Table S1 for details). The mitigation options selected align with those recommended in the UNECE guidance document on NH_3 abatement (Sutton et al., 2022) and the NEC Directive (Annex III, Part 2), which outlines measures to reduce NH_3 emissions from inorganic fertilizers and livestock manure.

In the *Mitigation* scenario, synthetic urea fertilizers (which account for 10–30 % of synthetic N fertilizers in the three countries) were removed without replacement, and the remaining synthetic fertilizers were fully substituted with calcium ammonium nitrate (CAN) fertilizers. The rationale behind this is that urea, although cheaper per kg N, leads to higher NH_3 volatilization losses due to increased soil pH following urea hydrolysis. Other approaches to mitigation of NH_3 emissions from urea, such as the use of low-emission application technology or urease inhibitors, are also interesting alternatives but were not considered in this study.

Additionally, in the *Mitigation* scenario, manure was incorporated into the soil shortly (<24 h) after application, while in the *Baseline* scenario a mix of practices was used. It should be noted that there is a range of other possible mitigation measures for NH_3 emissions related to livestock housing and manure treatment and storage and application (Bittman et al., 2014) that were not considered in the *Mitigation* scenario.

The scenarios were quantified in a territory where Mediterranean climate prevails, comprising the continental parts of Portugal and Spain, the Balearic Islands (Spain), and a part of southern France (Table 1; Fig. S2). As this territory covers almost all territory as defined by the EU Interreg-SUDOE programme for Southwestern Europe, it is referred to as the "SUDOE territory" throughout this paper.

2.2. Calculation and spatial distribution of ammonia emission

In the CLRTAP nomenclature, emissions from animal houses and manure storage form the category NFR K, while emissions from the application of synthetic fertilizer and manure, grazing excreta, and crop residue burning form the category NFR L. Ammonia emissions from NFR K were obtained from the national inventories submitted to the CLRTAP (EMEP, 2022a). Only NH_3 emissions from the NFR L were modified in the scenarios simulated in this study. Given the sensitivity of Mediterranean agricultural systems to climate change, all calculations were

Table 1
Statistics for total nitrogen deposition (kg N ha^{-1} y^{-1}), simulated by CHIMERE model, in the different biogeographical regions of SUDOE territory.
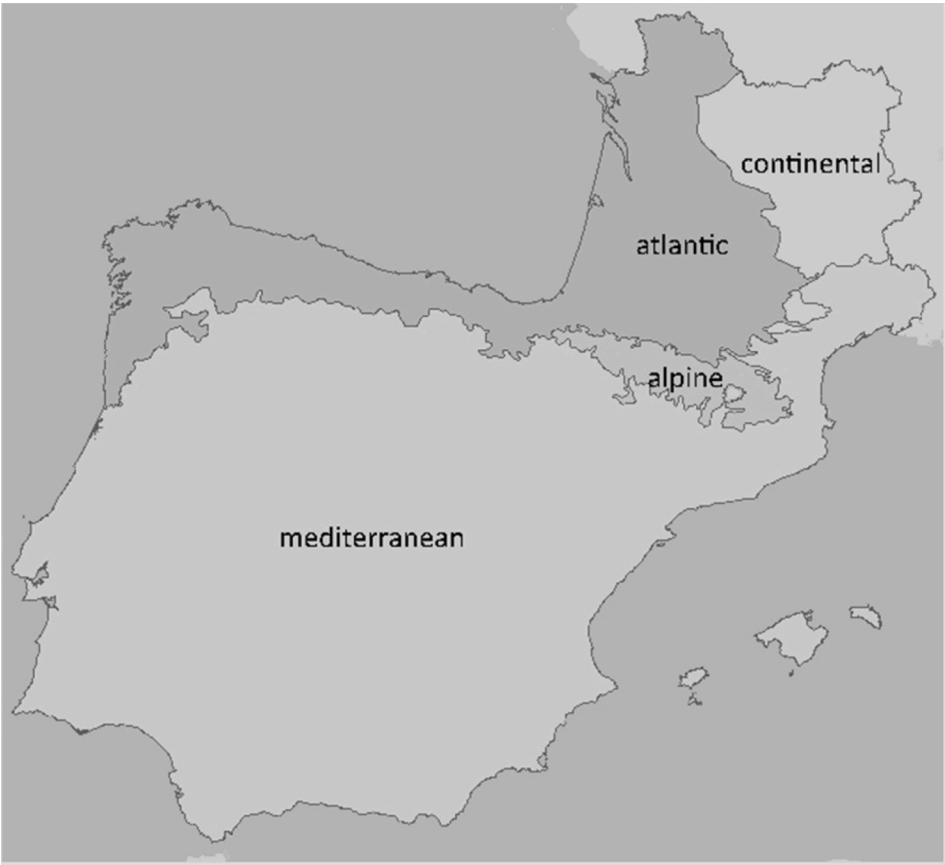
	Mediterranean		Atlantic	
	Baseline	*Mitigation*	*Baseline*	*Mitigation*
Mean	6.7	5.3	8.5	6.9
Std. Dev.	2.5	2.0	1.8	1.7
Median	5.9	4.6	8.4	6.5
Maximum	21.1	20.5	22.8	21.1
	Continental		Alpine	
	Baseline	*Mitigation*	*Baseline*	*Mitigation*
Mean	8.5	7.1	7.2	6.0
Std. Dev.	1.5	1.2	1.5	1.2
Median	8.5	7.1	7.0	5.8
Maximum	13.2	10.7	16.0	13.0

Statistics calculated with two-year averaged values resampled at 0.01° resolution.

conducted for a dry year (2017) and a wet year (2018).

2.2.1. Manure management and related emissions

Livestock N excretion was estimated by multiplying livestock populations in each NUTS2 region by country-specific excretion factors. Livestock population data were obtained from the 2016 Eurostat Farm Structure Survey (Eurostat, 2017) and aggregated to match the nomenclature of the Climate Convention (UNFCCC) inventories to the following categories: dairy cattle, non-dairy cattle, sheep, goats, swine, poultry, and horses. Excretion factors were obtained from the 2018 UNFCCC national inventories (2022 submissions; UNFCCC, 2022).

Livestock N excretion was distributed among three manure management systems: liquid manure, solid manure, and grazing excreta. This was done separately for each livestock category based on UNFCCC national inventory data, following Einarsson et al. (2021).

Ammonia emissions from livestock houses and manure storage were taken from CLRTAP national emission inventories. To ensure consistency with UNFCCC data, we used data for 2018 from the 2022 submissions to CLRTAP (EMEP, 2022a). The emission inventories specify emissions from livestock houses and manure storage (inventory category NFR K) separately for the same livestock categories. These emissions were assumed to remain constant across all scenarios.

2.2.2. Fertilizer application and grazing N input to land categories

Regional total use of synthetic N fertilizers was obtained from fertilizer consumption statistics at NUTS2 level (Eurostat, 2019). Data gaps in Portugal were filled using estimates from Serra et al. (2019). The manure N applied was calculated as described in the previous section. As mentioned above, in the *Mitigation* scenario, synthetic N input was modified by (1) removing all urea N input (10–30 % in the three countries) and (2) replacing the remaining synthetic N input by CAN. See Table S1 for details.

Following Einarsson et al. (2021), synthetic fertilizer inputs were allocated between different land use classes (arable land, permanent crops, and permanent grassland) within each Eurostat NUTS2 region, applying crop-specific fertilizer application rates to estimate the distribution. In Spain, crop-specific fertilizer application rates were obtained from the official Nitrogen Balance of Spanish Agriculture (MAPA, 2021). In France and Portugal, we used fertilizer rates from the latest IFA Fertilizer Use By Crop dataset (Ludemann et al., 2022). We multiplied the fertilizer rates by corresponding crop areas, aggregated the resulting fertilizer quantities to the three land use classes, and finally rescaled the results for each NUTS2 region to agree with the total regional fertilizer use according to Eurostat NUTS2 fertilizer use data.

Following Billen et al. (2021), we assumed that the applied manure N was distributed among the three land use classes in quantities proportional to the applied synthetic N fertilizer, and that grazing excreta was divided between arable land and permanent grassland in proportion to each NUTS2 region's areas of temporary and permanent grassland.

2.2.3. Emissions of NH₃ from applied synthetic fertilizer, manure and grazing

Ammonia emissions from grazing excreta were estimated using emission factors from the EMEP/EEA guidebook (Amon et al., 2019, see Table 4 9 therein). Emissions of NH_3 from applied manure and synthetic N fertilizers were estimated using the MANNER model (Misselbrook et al., 2004; Sanz-Cobena et al., 2014), an empirical model which considers the following variables: (i) fertilizer rate and type (manures and synthetic fertilizers), which were estimated as described above; (ii) organic fertilizer application method and timing (assumptions established based on data from Loyon, 2018; Foged, 2018); (ii) soil pH, which was obtained from the Harmonized Soil Database (FAO & IIASA, 2023); (iv) meteorological variables that strongly affect N volatilization, including monthly rainfall and temperature, which were obtained from the CRU TS dataset (Climatic Research Unit gridded Time Series; Harris et al., 2020) and wind speed.

2.2.4. Other field emissions of NH₃

In addition to NH_3 emissions from applied and grazing N inputs, relatively small quantities of NH_3 are emitted from field burning of crop residues. Given its minor contribution, we assumed these emissions to remain constant among both scenarios, using values from the CLRTAP inventories (EMEP, 2022a).

The sum of NH_3 emissions from application of manure and synthetic N fertilizer, grazing excreta, and crop residues constitute the whole emissions contained in the category NFR L of the CLRTAP inventory.

2.2.5. Spatial disaggregation of NH₃ emissions

The emissions assigned to the three land use classes used in the above calculations (arable land, permanent crops, permanent grassland) were spatially distributed within each NUTS2 region using the CLC2018 dataset and assuming the land-use share for the different classes shown in Table S2. Emissions from livestock houses and manure storage (category NFR K) technically do not occur on agricultural fields, but since manure is typically applied on fields close to storage facilities, we assumed housing and storage emissions to be distributed between land-use classes in a similar way to the application of stored manure. Ammonia emissions from burning of crop residues was assumed to occur on arable land. The results were finally aggregated to the 0.1-degree EMEP grid (EMEP, 2017), taking into account the fact that EMEP grid pixels on the border of two or more NUTS2 regions include NH_3 emissions from more than one NUTS2 region.

2.3. Atmospheric nitrogen deposition

The CHIMERE chemistry transport model (Menut et al., 2021) was used to estimate the total N deposition for the two scenarios studied. The CHIMERE model has been extensively used and evaluated in Europe (Pirovano et al., 2012) and, particularly, in Spain (Vivanco et al., 2009; García-Gomez et al., 2014). Model performances for estimating atmospheric concentration and deposition have been shown to be comparable to those of other air quality models applied in Europe (Bessagnet et al., 2016; Vivanco et al., 2018).

The meteorological data used by CHIMERE were adapted from the Integrated Forecasting System (IFS) simulations run by the European Centre for Medium-Range Weather Forecasts (ECMWF, www.ecmwf.int) for 2017 and 2018, obtained from the MARS archive at ECMWF through the access provided by AEMET for research projects. All the simulations used the same boundary conditions for the European domain, taken from LMDZ-INCA (Hauglustaine et al., 2004) and GOCART (Ginoux et al., 2001) global climatology models.

The CHIMERE model was applied to a domain containing Portugal, Spain and almost all of France with a spatial resolution of $0.1° \times 0.1°$, nested within a European domain ($0.2° \times 0.2°$). Emissions were taken from the EMEP emission database (EMEP, 2022b), except for NH_3 emissions from NFR L and NFR K, for which the emissions were calculated following the methodology described in the previous section. Annual simulations were performed for both 2017 and 2018, each using the two emission scenarios (*Baseline* and *Mitigation*). Annual N deposition estimates on a $0.1° \times 0.1°$ grid were obtained for use in the calculation of CL exceedances.

The modelled wet deposition of oxidised (WNOx) and reduced (WNHx) N, as well as their sum, were evaluated against measurements from the 14 EMEP monitoring stations within the SUDOE territory (12 in Spain and 2 in France). Model was assessed using the statistical metrics normalised mean bias (NMB), fraction of modelled estimates with a factor of two of the observed values (FAC2) and the Pearson correlation (r). Since there are no routine measurements of dry deposition of N within the modelling domain, model performance for dry deposition could not be evaluated.

2.4. Quantification of the area exceeding the empirical critical loads of nitrogen

The methodology developed under the Air Convention (CLRTAP) combines spatialized CLs (quantitative estimates of N deposition below which no negative effects are expected) with N deposition data to identify geographic areas and habitats at risk of threats due to eutrophication or acidification effects (CLRTAP, 2023). In this study, the critical levels for NH_3 or NO_x were not applied in risk evaluation.

The two-year average (2017–2018) of atmospheric deposition of total N (oxidised plus reduced forms) modelled with CHIMERE was used to evaluate the risk of N effects for the habitats in the SUDOE territory for both scenarios (*Baseline* and *Mitigation*). The empirical critical loads of nitrogen ($CL_{emp}N$; Bobbink et al., 2022) were applied to calculate critical load exceedances (CL_{exc}). Those CL_{exc} values were determined as the difference between total N deposition and $CL_{emp}N$, with a positive value indicating the occurrence of an exceedance and, therefore, a potential eutrophication or acidification risk for the habitat.

Following Vivanco et al. (2018), the *Ecosystem types of Europe v3.1* map (EEA, 2019) was selected as the receptor map to apply (Fig. S3). This map uses a 100 m × 100 m grid resolution and classifies habitats according to Level 2 of the 2012 EUNIS classification (EEA, 2021; 2022a). The *Review and revision of empirical loads of nitrogen for Europe* (Bobbink et al., 2022) established a range of values (lower and upper) for the CL of a great number of European habitats, most of them defined at level 3 of 2021/2022 EUNIS classification (Chytrý et al., 2020). $CL_{emp}N$ were established in the present study by aggregating the habitats studied in Bobbink et al. (2022) to EUNIS level-2 groups and selecting the minimum among the multiple lower limits of the $CL_{emp}N$

ranges aggregated for the corresponding EUNIS Level-3 category (Table 2). Marine, freshwater and anthropogenic habitats, and habitats lacking a defined $CL_{emp}N$ values were excluded from this assessment.

A large proportion of the unassessed area corresponded to habitat types without a defined $CL_{emp}N$, such as R7 *Sparsely wooded grasslands*; S7 *Spiny Mediterranean heaths*, and G4 *Mixed deciduous and coniferous woodland* (discontinued in the current EUNIS classification). To address this gap, the average of the $CL_{emp}N$ for T1 *Deciduous broadleaved forest* and T3 *Coniferous forest* was used as the $CL_{emp}N$ for category G4. For R7 and S7, $CL_{emp}N$ values were estimated based on their closest analogous habitat types, based on composition similarity at the genus level of the diagnostic species (defined in the EUNIS classification; Chytrý et al., 2020; Schaminée et al., 2019), using Sørensen Index (Santini et al., 2017; Sørensen, 1948) in RStudio (version 4.2.1; R Core Team, 2022). As a result, $CL_{emp}N$ of R7 *Sparsely wooded grasslands* was estimated as the average of *R1 Dry grasslands* and T2 *Broadleaved evergreen forests*, and the $CL_{emp}N$ of S7 *Spiny Mediterranean heaths* could be derived by considering either the $CL_{emp}N$ of R1 *Dry grasslands* or S6 *Garrigue*.

Deposition of total N from CHIMERE was resampled and projected at the Pan-European standard projection ETRS89 LAEA (EC, 2010) to ensure geographical alignment with the $CL_{emp}N$ gridded map. CL_{exc} values were then calculated for each grid cell. The extension of the exceedance area, along with the mean and standard deviation of the exceedance values were calculated for each habitat type. Additionally, for a more comprehensive description of the results, spatial statistics of emission, deposition and area at risk were also calculated at the NUTS3 regional level. All analysis were performed using ArcGIS Pro 3.1 (ESRI, Redlands CA, USA).

Table 2
Risk assessment data and results of the habitats localized in the SUDOE territory, averaged for both modelled years (the drier year 2017 and the wetter year 2018), for *Baseline* and *Mitigation* scenarios.

EUNIS code	EUNIS name	Evaluated area (km²)	$CL_{emp}N^a$ (kg N ha⁻¹ y⁻¹)	Altitude (m.a. s.l.)[b]	Area at risk (% of the evaluated area)		N deposition (kg N ha⁻¹ y⁻¹)[b]		CL_{exc} (kg N ha⁻¹ y⁻¹)[b]	
					Baseline	*Mitigation*	*Baseline*	*Mitigation*	*Baseline*	*Mitigation*
N1	Coastal dunes and sandy shores	339	5	23 ± 47	77 %	68 %	6.3 ± 2.0	5.8 ± 1.8	2.1 ± 1.6	1.7 ± 1.4
Q1	Raised and blanket bogs	18	5	1019 ± 410	98 %	93 %	7.9 ± 1.8	6.8 ± 1.7	3.0 ± 1.8	2.0 ± 1.6
Q2	Valley mires, poor fens and transition mires	7	5	1049 ± 449	98 %	97 %	7.9 ± 1.3	6.7 ± 1.1	3.0 ± 1.2	1.8 ± 1.0
Q4	Base-rich fens and calcareous spring mires	0.3	15	744 ± 342	0 %	0 %	5.9 ± 1.6	4.6 ± 1.2	–	–
R1	Dry grasslands	37835	5	828 ± 414	65 %	36 %	6.0 ± 1.8	4.9 ± 1.4	1.9 ± 1.7	1.4 ± 1.2
R2	Mesic grasslands	72173	10	437 ± 307	17 %	4 %	8.2 ± 2.1	6.7 ± 1.8	1.3 ± 1.6	1.8 ± 1.8
R3	Seasonally wet and wet grasslands	3800	10	454 ± 329	6 %	1 %	6.6 ± 2.0	5.5 ± 1.6	1.0 ± 0.8	1.1 ± 0.8
R4	Alpine and subalpine grasslands	2973	5	2031 ± 333	93 %	51 %	6.3 ± 1.2	5.2 ± 1.0	1.4 ± 1.1	0.9 ± 0.9
R7	Sparsely wooded grasslands	13394	7.5	427 ± 282	6 %	2 %	5.1 ± 1.4	4.2 ± 1.1	1.7 ± 2.2	2.9 ± 2.2
S2	Arctic, alpine and subalpine scrub	2168	5	2023 ± 348	74 %	44 %	5.9 ± 1.3	4.9 ± 1.1	1.4 ± 1.1	0.8 ± 0.8
S3	Temperate and Mediterranean-montane scrub	14029	5	900 ± 367	71 %	61 %	7.0 ± 2.6	6.1 ± 2.2	3.1 ± 2.3	2.5 ± 1.9
S4	Temperate shrub heathland	1807	5	1311 ± 393	83 %	69 %	6.8 ± 1.8	5.9 ± 1.6	2.3 ± 1.6	1.6 ± 1.4
S5	Maquis, arborescent matorral and thermo-Mediterranean scrub	22282	5	745 ± 487	66 %	41 %	6.5 ± 2.5	5.2 ± 2.0	2.6 ± 2.5	1.9 ± 2.1
S6	Garrigue	10002	5	603 ± 308	95 %	67 %	7.8 ± 2.2	5.9 ± 1.8	2.9 ± 2.2	1.7 ± 1.6
S7	Spiny Mediterranean heaths	10001	5	841 ± 356	71 %	37 %	6.2 ± 1.6	4.8 ± 1.2	1.9 ± 1.4	1.1 ± 1.0
T1	Deciduous broadleaved forest	153145	10	551 ± 366	10 %	4 %	7.0 ± 2.4	5.8 ± 2.0	1.8 ± 2.0	1.9 ± 1.7
T2	Broadleaved evergreen forest	27624	10	499 ± 265	14 %	4 %	7.6 ± 2.4	5.6 ± 1.7	1.5 ± 1.5	1.8 ± 1.2
T3	Coniferous forest	80108	5	778 ± 463	87 %	57 %	7.3 ± 2.6	6.0 ± 2.3	2.8 ± 2.6	2.2 ± 2.2
G4[c]	Mixed deciduous and coniferous woodland	20908	7.5	590 ± 427	41 %	18 %	7.4 ± 2.1	6.2 ± 1.9	1.8 ± 1.9	1.7 ± 1.7
		472613	7.9	634 ± 435	39 %	22 %	7.2 ± 2.4	5.8 ± 2.0	2.3 ± 2.2	2.0 ± 2.0

[a] Empirical critical load of nitrogen, based on Bobbink et al. (2022): minimum of the lower limits of all the $CL_{emp}N$ ranges of EUNIS level-3 habitats of at each EUNIS level-2 aggregation group.

[b] Mean ± standard deviation for the entire area (resolution of 1 ha) in which the habitat has been evaluated.

[c] This category has been discontinued in the current EUNIS classification.

3. Results

3.1. Ammonia emissions

The *Mitigation* scenario caused a large decrease in NH_3 emissions. For the entire SUDOE territory, the two-year average emission from synthetic fertilizer and manure application decreased by 68 % as compared to the *Baseline* scenario. This reduction, applied to the CHIMERE model, implied a reduction of 36 % in the total NH_3 emission in the SUDOE region (taking into account all emitting sectors).

The largest absolute reduction in NH_3 emission (comparing *Baseline* and *Mitigation* scenarios in Fig. 1) occurred in south-eastern Spain (Andalucía and Región de Murcia), followed by eastern Spain (Comunitat Valenciana, Aragón and Cataluña) and the Region of Poitou-Charentes in France (NUTS level-2 regions; Fig. S2). In relative terms, the largest emission reduction from these sources compared to the *Baseline* scenario occurred in the eastern half of Spain and Aquitaine, which mostly experienced a reduction of more than 70 %, especially in the south-eastern third of Spain (Andalucía, Región de Murcia and Comunitat Valenciana), where the reductions were larger than 80 % (note that in Fig. 1, these particular regional reductions are displayed in

the "Changes" panel as numerical labels rather than in the colour legend). However, the emission abatement reached no more than 60 % of the total NH_3 emissions in any NUTS3 region (Fig. 1, "Changes" panel, colour legend).

The changes in emissions between the dry and wet years were mostly small and varied by region. As a whole, the inter-annual differences resulted in an overall 5 % decrease in emissions in the wet year (2018) compared to the dry year (2017) for the entire SUDOE territory.

3.2. Nitrogen deposition

Fig. S4 and Table S3 compare modelled and observed wet deposition of reduced (WNHx), oxidised (WNOx) and total N (WNHx + WNOx) for the *Baseline* scenario. The model underestimated the mean wet deposition of WNHx and WNOx by less than 20 %, and mean bias of total N deposition was small (−6 % in 2017 and -10 % in 2018). The modelled estimates are within a factor of 2 of the observed values for all components at more than 75 % of the monitoring stations (FAC2 > 75 %). Pearson correlation coefficients (r) were all above 0.7, apart from the estimates of WNHx in 2017 (r = 0.40), due to the underestimation of deposition at two stations in NW Spain. Overall, model performance
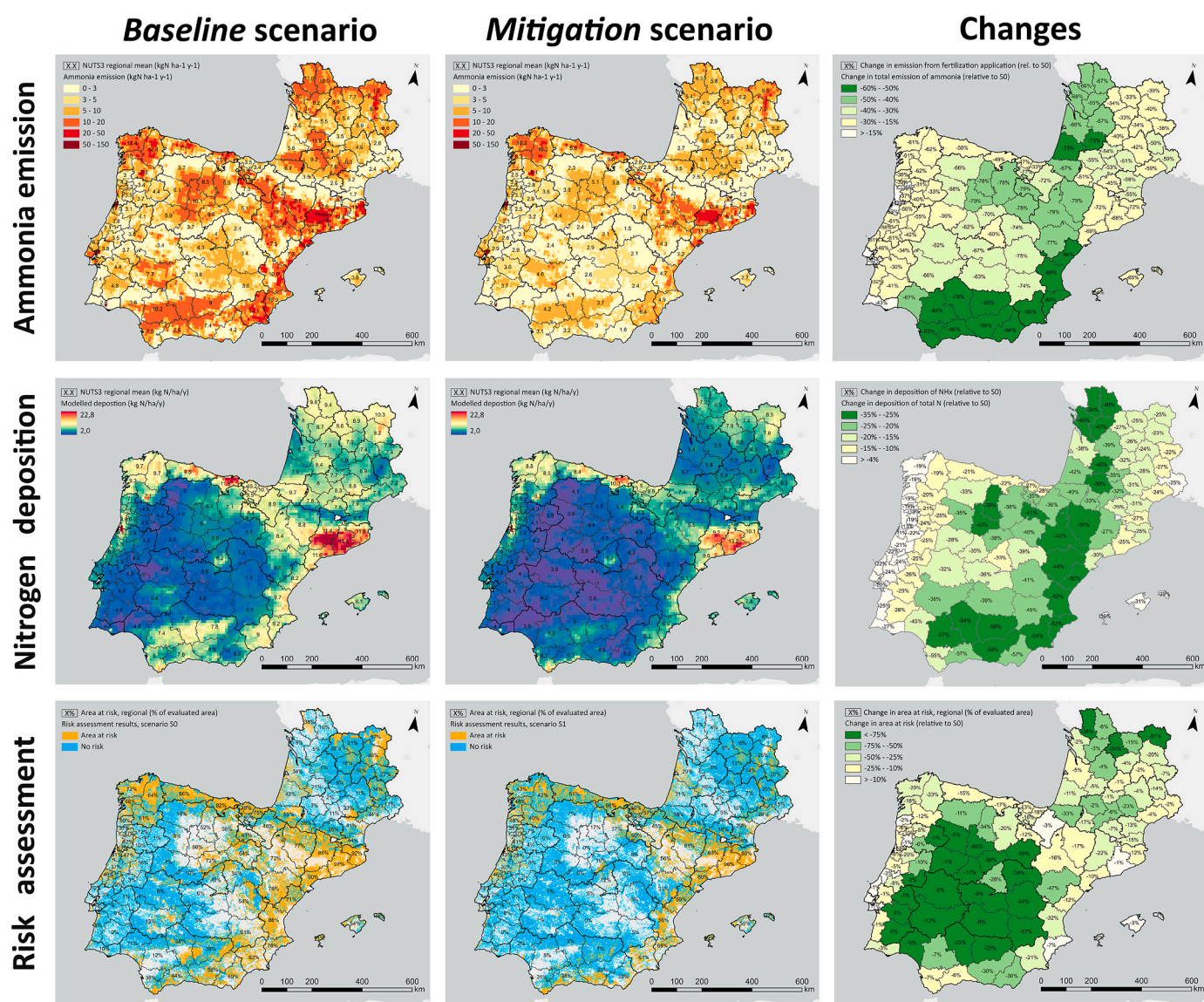

Fig. 1. Gridded and regionally averaged emission of ammonia and deposition of total nitrogen, and gridded and regionally aggregated area of habitats at risk of eutrophication, for the regions at level 3 of NUTS classification in the SUDOE territory.

compares favourably with prior studies on wet deposition in Europe, showing, for example, lower bias and higher FAC2 than most of the 14 models evaluated in Vivanco et al. (2018) for the year 2010. Similar levels of agreement were also found in Spain for the period 2005–2008, for CHIMERE and EMEP models (García-Gómez et al., 2014). Notably, agreement between modelled and measured values appears consistently poorer for reduced than oxidised N, highlighting a challenge in this matter, probably both for modelling and monitoring.

In the *Baseline* scenario, the average N deposition for the entire SUDOE territory was 7.2 kg N ha^{-1} y^{-1} (4.1 and 3.1 kg N ha^{-1} y^{-1} of reduced and oxidised N, respectively). The *Mitigation* scenario lowered reduced N deposition by 1.5 kg N ha^{-1} y^{-1} in average for the entire territory, corresponding to a 36 % decrease in NH$_x$ deposition and a 20 % decrease in total N deposition relative to the *Baseline* scenario. Deposition of oxidised N remained virtually constant, changing its share in total deposition accordingly (and inversely) to NH$_x$ deposition reductions (Fig. S5).

Regional differences were evident, with the highest deposition rates observed in France and along the Spanish coastal perimeter. Specific hotspots were identified in northern and north-eastern Spain, particularly in Cataluña, where deposition averaged 12.5 kg N ha^{-1} y^{-1} in the *Baseline* scenario and decreased to 10.4 kg N ha^{-1} y^{-1} in the *Mitigation* scenario (Fig. 1). When analyzing by biogeographical regions (Atlantic, Continental, Alpine, and Mediterranean; Table 1), deposition rates reached the highest values in the Atlantic and Continental regions. The Mediterranean region exhibited the greatest variability (Table 1), with a northeast-to-southwest decreasing deposition gradient (Fig. 1). The relative changes in deposition were similar across regions.

Regarding the N deposition in the different habitat types (Table 2, Fig. S6), the highest simulated values in the *Baseline* scenario (above the average of 7.2 kg N ha^{-1} y^{-1}) were located in the following EUNIS habitat types: R2 *Mesic grasslands* > Q1 *Raised and blanket bogs* = Q2 *Valley mires, poor fens and transition mires* > S6 *Garrigue* > T2 *Broadleaved evergreen forests* > G4 *Mixed forests* > T3 *Coniferous forests*. All these habitats experienced substantial reductions in mean deposition under the *Mitigation* scenario, ranging from 1.1 to 2.0 kg N ha^{-1} y^{-1}. The lowest average deposition was found in the habitat type R7 *Sparsely wooded grasslands* (5.1 kg N ha^{-1} y^{-1}).

Interannual variability in total N deposition was also detected, with deposition in the wet year (2018) being, on average, 10.5 % higher than in the dry year (2017). This variation was spatially heterogeneous, with more pronounced increases in northern Spain, particularly in the Alpine region, whereas eastern Spain exhibited smaller increases or even slight declines.

3.3. Area exceeding the empirical critical loads of nitrogen

The area evaluated in this study (473,000 km^2) represented 61 % of the entire SUDOE territory. The CL$_{exc}$, calculated using the 2-year average of total N deposition, indicated that 39 % of this area in the *Baseline* scenario was at risk of eutrophication due to atmospheric N deposition for the simulated period (2017–2018). In the *Mitigation* scenario, the area at risk decreased to 22 % of the evaluated area, representing a 43 % reduction compared to the *Baseline* scenario (Table 2). The spatial distribution of CL$_{exc}$ combines the distribution of the high deposition areas and the most sensitive habitat types, resulting in a higher risk in the north and east of Spain, the northern half of Portugal, Aquitaine, and mountain areas of France and Mediterranean Spain, together with the entire Alpine region (Fig. 1). With the *Mitigation* scenario, the area at risk declined from 47 % to 27 % in Spain, meaning a 43 % of relative reduction relative to the *Baseline* scenario. The most noteworthy exceptions in the risk assessment improvement in Spain were the regions of Illes Balears, Barcelona, Alicante and Comunidad Foral de Navarra (Fig. 1), experiencing relative reductions of less than 10 %. In France, the area at risk decreased from 27 % to 14 %, representing a 47 % reduction of the area at risk (Fig. 1). In Portugal, there

was no major change.

The improvements of the *Mitigation* scenario varied across habitat types. All grassland and forest habitats (EUNIS groups R and T), except for T3 *Coniferous forest*, showed a larger-than-average decrease in their areas at risk. Conversely, some habitats that had a large proportion of their area at risk in the *Baseline* scenario exhibited none or small decreases in the *Mitigation* scenario. Those habitat types were N1 *Coastal dunes and sandy shores*, together with mires, bogs and fens (EUNIS group Q) and some Mediterranean shrublands (S3 and S4). Finally, some habitats obtained a large improvement, but still presented more than 50 % of their area at risk in the *Mitigation* scenario: R4 *Alpine and subalpine grasslands*, S7 *Spiny Mediterranean heaths (phrygana, hedgehog-heaths and related coastal cliff vegetation)* and T3 *Coniferous forests* (Table 2).

All the most sensitive habitats (CL$_{emp}$N of 5 kg N ha^{-1} y^{-1}) presented over 50 % of their evaluated area at risk in the *Baseline* scenario (Table 2), but most of them showed a great improvement in the *Mitigation* scenario, particularly natural and semi-natural grassland (EUNIS group R) and shrublands (EUNIS group S). T3 *Coniferous forests* represented the largest area of sensitive habitat types (aprox. 80,000 km^2 of area evaluated), followed by R1 *Dry grasslands* (38,000 km^2) and S5 *Maquis, arborescent matorral and thermo-Mediterranean scrub* (22,000 km^2). In the *Baseline* scenario, 87 %, 65 % and 66 % of these habitats were at risk, respectively. T3 showed still 57 % of its area at risk in the *Mitigation* scenario, which represented a relative decrease of 34 %; but R1 and S5 experienced a more substantial improvement, down to less than 50 % of the area at risk (relative decrease of 45 % and 38 %, respectively). Less sensitive habitats mostly were below the SUDOE average risk level (39 %) in the *Baseline* scenario, and also improved under the *Mitigation* scenario (Table 2).

The reduction in area at risk varied across biogeographical regions for all the EUNIS habitats (Table S4). The Alpine region resulted the most threatened one in both scenarios, despite experiencing a large decrease in its area at risk, from 64 % to 43 % of the evaluated area. The habitat type R1 *Dry grasslands*, which is predominantly found in the Mediterranean region (81 % of its total SUDOE area), experienced a substantial overall decrease in its area at risk, from 65 % to 36 % (Table 2), even with limited improvement in the rest of regions. Moreover, some habitats with limited or no overall improvement still exhibited notable reductions in specific biogeographical regions. For example, mires, bogs, and fens (EUNIS group Q) showed reductions in at-risk area only in the Alpine region. Other discrepancies between the general and regional results for some habitats are less noteworthy, considering the very small representation of some habitat types in particular regions.

4. Discussion

4.1. Emission and deposition variations between scenarios and years

The *Mitigation* scenario considered only the removal of urea-based fertilizers and the adoption of improved manure application methods that minimize contact between manure and the atmosphere. These measures led to a 36 % reduction in NH$_3$ emissions in the SUDOE territory, which would support the fulfilment of the commitments for NH$_3$ emissions abatement of the Annex II NEC Directive for 2030 onwards (ranging from France's 13 % to Spain's 16 % reductions of 2005 emissions). At present, none of those three countries have met this target. Moreover, the resulting 20 % decrease in atmospheric deposition of reactive N led to a 43 % reduction in the area at risk, as assessed using the CLRTAP critical load methodology.

According to statistics derived from comparing modelled and measured values, CHIMERE model predicted wet N deposition acceptably for the SUDOE territory. Atmospheric NH$_3$ transport causes spatial heterogeneity in how deposition responds to emission reductions. In 'source' regions (deposition/emission ratio <0.5), such as Murcia, reductions in NH$_3$ emissions (−7.7 kg N ha^{-1} y^{-1} in this case) led to

smaller decreases in NH_x deposition (-2.9 kg N ha^{-1} y^{-1}). Conversely, 'receptor' regions (deposition/emission ratio >1; e.g., Languedoc-Roussillon, French Pyrenees, northern País Vasco), deposition decreased more than emission did (Fig. 2), since they benefited from emission reductions in the neighbouring regions. Additionally, in NO_x-dominated areas (e.g., Madrid, Barcelona, western Portugal or Gironde), NH_3 reductions had a smaller effect on total N deposition than in neighbouring regions with similar emission decreases (Fig. 2, S1).

Total N deposition in the SUDOE territory during the wet year (2018) was approximately 10 % higher than in the dry year (2017), despite NH_3 emissions being around 5 % lower. One reason is that wet scavenging is a more efficient process than dry deposition during periods of high rainfall. The influence of changes in precipitation on the effectiveness of N emission reduction in decreasing N deposition has been documented in previous studies, in which atmospheric modelling tools were also proved as highly valuable tools to include climate change modulation of other global change impacts (Theobald et al., 2019).

4.2. Methodology to quantify area exceeding the critical N load

In this study, the lower values of the $CL_{emp}N$ ranges were used for risk evaluation to ensure a precautionary assessment of eutrophication risk. Moreover, the low deposition of N (around 5 kg N ha^{-1} y^{-1}) in large areas of the Iberian Peninsula, together with the concerns about the reliability and completeness of $CL_{emp}N$ for Mediterranean habitats (UNECE, 2023) further justify this precautionary approach. Nonetheless, more experimental and gradient studies in the Mediterranean region are needed to further improve the robustness of CLempN, and to establish CLempN for habitats that have not yet been set.

At the time of the study, no EU-wide habitat map with EUNIS level-3 classification was available. Since most $CL_{emp}N$ values are defined at level-3 (Bobbink et al., 2022), we used the minimum lower limit of all level-3 values within each level-2 group (as described in section 2.4), ensuring a precautionary approach. This method may overestimate risk in some habitat groups (N1 *Coastal dunes and sandy shores*, R1 *Dry grasslands* and R3 *Seasonally wet and wet grasslands*), but helps prevent underestimation. Among them, N1 particularly deserved a more detailed classification for assessment, since it showed a large area at risk and small improvements in the *Mitigation* scenario.

This precautionary approach aligns with the biodiversity conservation goals of Europe, although its implementation ultimately requires balancing broad environmental benefits (including protection of human health), with technical and economic feasibility (Jacobs et al., 2016; Rodríguez et al., 2024). Policymakers must weigh science-based recommendations against any socio-economic constraint, taking into account the regional particularities that arise from this type of prospective evaluations.

Discrepancies were found in the representation of *Quercus ilex*, which forms both forests forests (T2 *Broadleaved evergreen forest*) and *dehesas* (R7 *Sparsely wooded grasslands*). According to the National Forest Inventory (NFI; MITECO, 2022), the receptor map (Ecosystem types of Europe v3.1 map; EEA, 2019) misclassified over half of these forests as T1 *Deciduous broadleaved forest*. However, since the same CL value (10 kg N ha^{-1} y^{-1}) was applied to T1 and T2, and the distribution in northeastern Spain was accurate, the risk estimates remain robust. Improving receptor maps through national surveys and adopting standardized classifications, such as EUNIS level-3 or the Annex I classification of the Habitats Directive (EEC, 1992), would enhance the consistency and reliability of $CL_{emp}N$ application for biodiversity conservation.

4.3. Results for area exceeding the critical N load

The *Mitigation* scenario implied a substantial improvement in the prospect of terrestrial ecosystem conservation in the SUDOE territory that would contribute to meet the target of reducing the area where air pollution threatens biodiversity by 25 % in 2030, compared to 2005 levels (EC, 2021). However, the spatial distribution of results revealed significant heterogeneity across the SUDOE region in terms of the percentage of area at risk, depending on the N deposition distribution and its reduction, and on the different sensitivity of the habitats.

The Atlantic and Alpine biogeographical regions showed the highest percentage of area at risk, with limited efficacy of the *Mitigation* scenario. The Alpine region (Pyrenees mountain range), which 52 % of its territory is included in the Natura 2000 network and previous studied proved at risk of eutrophication by N deposition (Boutin et al., 2017; Camarero and Catalan, 2012; Camarero and Aniz, 2010; García-Gómez et al., 2014, 2017), showed an average $CL_{emp}N$ of 6.8 ± 2.3 (SD) kg N

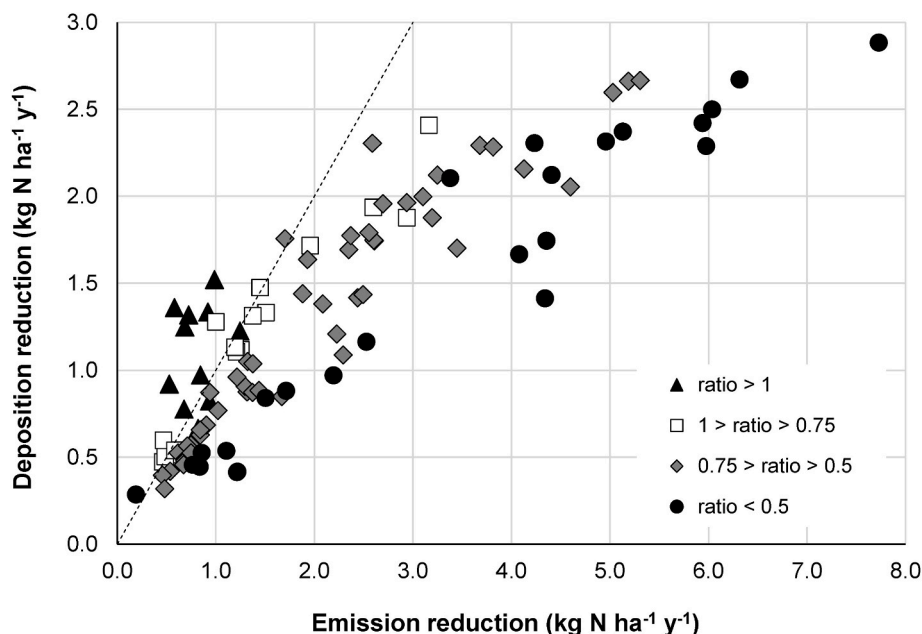**Fig. 2.** Relationship between reduction in emission and reduction in deposition in absolute terms (kg N ha^{-1} y^{-1}). Data of the different NUTS3 regions are grouped by the deposition-to-emission ratios in *Curent* scenario (symbols). Line 1:1 is shown (dashed line).

ha^{-1} y^{-1}, making it the most sensitive region. Its proximity to high NH$_3$ emission areas of Cataluña and Aragón increased its vulnerability. A similar, less extreme, case occurred at *Les Landes de Gascogne* (wooded area across the regions of Gironde, Landes and Lot-et-Garonne), with low CL$_{emp}$N and high emission of NH$_3$ and NO$_x$ in neighbouring regions (particularly in Landes and Gironde –from the city of Burdeaux-, respectively; Fig. S1). In contrast, high risk in the Atlantic region was mainly due to high NH$_3$ emissions in the region combined with high rates of wet deposition.

Regarding the regions with the lowest improvements in the *Mitigation* scenario (i.e., NUTS3 regions where the reduction in their area at risk was lower than 10 %), in some cases it may be partly due to the proposed measures being insufficient to significantly reduce NH$_3$ deposition, as observed in Comunidad Foral de Navarra and Barcelona. One probable explanation to this is that NO$_x$ plays a more significant role than NH$_3$, as in the case of Illes Balears and most of Portugal. Other regions with important deposition decrease showed larger decreases in their area at risk, yet still showed less improvement than neighbouring regions with similar reductions in deposition. This is the case for southern and eastern regions of Spain, which may be explained by very low CL$_{emp}$N values (indicating a high abundance of sensitive habitats) and the agglomeration of high-emitting regions ("sources" regions), which emission reductions favoured more other regions (the "receptor" ones) than themselves.

Regarding the most sensitive habitats (those with the lowest CL$_{emp}$N; i.e. 5 kg N ha^{-1} y^{-1}), some minority habitats (Q1 and Q2) remained largely unprotected under the *Mitigation* scenario. Moreover, 37 % of the overall SUDOE territory (covered by sensitive habitats R1, S3, S5, S6, S7 and T3) remained moderately unprotected (36 %–67 % at risk under *Mitigation* scenario), which contributes to the point that, even achieving the strategic targets at European scale, large areas might remain unprotected. Among the sensitive habitats, T3 *Coniferous forests*, that provide multiple ecosystem services (Torres et al., 2021; Schaubroeck et al., 2016), are well distributed throughout the SUDOE territory and represented the largest share of the total area at risk (44 % and 37 in *Baseline* and *Mitigation* scenarios, respectively). Other habitats, like S3 *Temperate and Mediterranean-montane scrub* (the scrub habitat with the lowest improvement), were affected unevenly by mitigation measures, improving in the Mediterranean but not in the Atlantic region. This pointed again to the elevated deposition of N in the Atlantic region as a major threat that needs more ambitious mitigation plans than those proposed here for agricultural practices. Alternatively, R4 *Alpine and subalpine grasslands*, one of the most threatened habitats by both N deposition and climate change (García-Gómez et al., 2014; Pérez-García et al., 2013), experienced an above-average improvement.

4.4. Societal costs and benefits of mitigation

Ammonia emissions entail significant societal costs. Van Grinsven et al. (2013) estimated that NH$_3$ accounted for 46 % of total nitrogen pollution costs in the EU27 in 2008 (€75–485 billion/year), mainly due to health impacts from PM2.5 formation (€12/kg NH$_3$-N) and biodiversity loss from nitrogen deposition (€3/kg NH$_3$-N). Recent studies updated the latter to €13 (van Grinsven et al., accepted), and De Vries et al. (2025) estimated a total marginal cost of €35/kg NH$_3$-N for all impacts in 2021. These high damage costs suggest strong societal benefits from reducing NH$_3$ emissions.

Given the high marginal damage costs of NH$_3$, Van Grinsven et al. (2013) demonstrated that reducing manure N use by 50 % in winter wheat would produce societal benefits that exceed the costs from potential lower yields. In our baseline, NH$_3$-N losses are 5–11 % for synthetic fertilizers and 8.5 % for manure, yet the societal costs from these emissions likely outweigh the benefits of increased yields, especially considering the low market prices of urea and CAN (around €1.3/kg N) compared to marginal pollution costs (€3/kg N at 8 % emission rate). Thus, each additional kg of N must yield at least €1.7 in crop value to be

justified. However, this condition holds primarily under low total N inputs; at higher levels, crop yield response diminishes, as shown by Van Grinsven et al. (2013, 2022).

Regarding fertilizers costs alone, according to Spanish National statistics for 2017 (MAPA, 2024), a first approximation at national scale showed that switching from urea to CAN could imply partial increase of 33 %. Switching from NPK to CAN could imply up to a 7 % of partial increase. The total cost of the mitigation measure varied between saving 14 % and increasing 10 % the overall cost of fertilizers to farmers at national level.

Furthermore, the low cost of manure incorporation represents another important factor. Although the exact mitigation costs per kilogram of avoided NH$_3$-N emissions have not been calculated in this study, they are likely to be lower than the corresponding marginal pollution damage costs. Rapid incorporation—carried out within 4–24 h after application—can reduce emissions by 30–65 %, with estimated implementation costs ranging from €0 to €1.5 per kilogram of abated nitrogen (Bittman et al., 2014). However, the associated costs for fuel, labour, and equipment must be borne by farmers, who currently receive no compensation. These costs also vary significantly depending on farm size (Bittman et al., 2014).

5. Conclusions

Nitrogen deposition currently poses a threat for conservation of many natural and semi-natural habitats in the SUDOE region. The results demonstrate the potential effectiveness of removing urea-based fertilizers and improving manure application in substantially reducing NH$_3$ emissions and the environmental impacts they cause. The areas at risk due to exceedance of critical deposition loads can be reduced by 43 % with these mitigation techniques, highlighting the importance of agricultural mitigation measures in the cropping system to achieve the 2030 goal of ecosystem protection in the framework of the European conservation strategy.

The response of N deposition and eutrophication risks to the emission reductions is spatially very heterogeneous, with some regions benefitting from the emission reductions in neighbouring regions. Strategies to abate N emissions should take into account the national-to-local spatial heterogeneity regarding emission hotspots, share of reduced and oxidised forms, atmospheric transport and chemistry of pollutants, and sensitivity of ecosystems to N and their associated environmental, cultural and social values.

The development and use of integrated modelling systems to evaluate mitigation measures is demonstrated as a valuable tool to support spatially differentiated and transboundary policy decisions for the urgent improved protection of natural and semi-natural habitats in a global change framework. The presented modelling system could straightforwardly be extended to include additional mitigation measures (particularly in the livestock system) and cover a larger geographical area. The use of receptor maps based on national biodiversity survey data would be advisable. Further studies on economic feasibility of implementing emission mitigation measures and societal cost-benefits analyses would be needed to supplement these type of research.

CRediT authorship contribution statement

Héctor García-Gómez: Writing – original draft, Visualization, Methodology, Formal analysis, Conceptualization. **Rasmus Einarsson:** Writing – original draft, Software, Methodology, Formal analysis, Conceptualization. **Mark Theobald:** Writing – original draft, Formal analysis. **Eduardo Aguilera:** Writing – review & editing, Software, Methodology. **Tania Carrasco-Molina:** Visualization, Software, Formal analysis. **Victoria Gil:** Software. **Benjamín S. Gimeno:** Writing – review & editing. **Coralina Hernández:** Formal analysis. **Luis Lassaletta:** Writing – review & editing, Software, Methodology. **Isaura Rábago:** Writing – review & editing, Funding acquisition. **Hans J.M. van**

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2025.126867.

Data availability

Data could be made available on request under certain conditions, depending on the intended use, which could include a commitment to referring or an invitation to participate in potential publications

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