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Yield-scaled mitigation of ammonia emission from N fertilization: the Spanish case

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Abstract

Synthetic nitrogen (N) fertilizer and field application of livestock manure are the major sources of ammonia (NH_3) volatilization. This N loss may decrease crop productivity and subsequent deposition promotes environmental problems associated with soil acidification and eutrophication. Mitigation measures may have associated side effects such as decreased crop productivity (e.g. if N fertilizer application is reduced), or the release of other reactive N compounds (e.g. N_2O emissions if manure is incorporated). Here, we present a novel methodology to provide an integrated assessment of the best strategies to abate NH_3 from N applications to crops. Using scenario analyses, we assessed the potential of 11 mitigation measures to reduce NH_3 volatilization while accounting for their side effects on crop productivity, N use efficiency (NUE) and N surplus (used as an indicator of potential N losses by denitrification/nitrification and NO_3^- leaching/run-off). Spain, including its 48 provinces, was selected as a case study as it is the third major producer of agricultural goods in Europe, and also the European country with the highest increase in NH_3 emissions from 1990 to 2011. Mitigation scenarios comprised of individual measures and combinations of strategies were evaluated at a country- and regional level. Compared to the reference situation of standard practices for the year 2008, implementation of the most effective region-specific mitigation strategy led to 63% NH_3 mitigation at the country level. Implementation of a single strategy for all regions reduced NH_3 by 57% at the highest. Strategies that involved combining mitigation measures produced the largest NH_3 abatement in all cases, with an 80% reduction in some regions. Among the strategies analyzed, only suppression of urea application combined with manure incorporation and incorporation of N synthetic fertilizers other than urea showed a fully beneficial situation: yield-scaled NH_3 emissions were reduced by 82%, N surplus was reduced by 9%, NUE was increased by 19% and yield was around 98% that of the reference situation. This study shows that the adoption of viable measures may provide an opportunity for countries like Spain to meet the international agreements on NH_3 mitigation, while maintaining crop yields and increasing NUE.

S Online supplementary data available from stacks.iop.org/ERL/9/125005/mmedia

Keywords: NH_3 emission, fertilizers, mitigation scenarios, regional scale, N yields, nitrogen use efficiency

1. Introduction

Agriculture is the main sector responsible for ammonia (NH_3) emissions into the atmosphere, accounting for an estimated 94% of the total European emissions in 2011 (European Environment Agency (EEA) 2013). Synthetic nitrogen (N) fertilization and field application of livestock manure are the major sources (Bittman *et al* 2014). From an agronomic perspective, NH_3 emission from applied N fertilizers produces a substantial loss of the N resource for cropping systems. Additionally, volatilized NH_3 can cause soil acidification and eutrophication with subsequent loss of overall biodiversity. Moreover, NH_3 also leads to the formation of secondary particles with impacts on human health, and indirect emissions of nitrous oxide (N_2O) and N oxides following deposition (Asman *et al* 1998, Erisman *et al* 2008, García-Gómez *et al* 2014). Therefore, NH_3 volatilization from agriculture is considered an issue of major environmental and socioeconomic concern (van Grinsven *et al* 2013).

Mitigation strategies proposed for NH_3 emissions from fertilizer use focus on: 1) the type of fertilizer applied (total or partial substitution of urea, U, by other synthetic or organic fertilizers); 2) improved methods of fertilizer application such as incorporation, injection, and washing into soil either by rainfall or irrigation; 3) the use of additives that affect soil processes leading to reduced NH_3 volatilization (e.g. urease inhibitors); and 4) the use of polymer coatings on urea fertilizer (Bittman *et al* 2014). The vast majority of studies in which these strategies have been analyzed present two limitations: first, a lack of integrated and prioritized comparison among measures, which may lead to biased conclusions (Oenema *et al* 2009); second, some of these strategies may result in trade-offs in terms of lower crop yields and/or increased N losses in other chemical forms, for example nitrate (NO_3^-) and N_2O at the field scale, unless N fertilization rates are decreased to take into account the reduction in losses (with potential cost savings) and thus optimizing N management (e.g. increasing nitrogen use efficiency, NUE). These effects have rarely been tested at a regional scale (Stevens and Quinton 2009), thus a case-by-case analysis with regional implementations of mitigation options is instrumental to provide a more realistic approach for the achievement of national mitigation targets.

The process of intensification of the agricultural sector occurring in Europe over the last 50 years has produced an important increase in total agricultural production. In the case of Spain, which is the third largest agricultural producer in the European Union (EU), the increase has reached 82% (expressed as total weight), mainly during the period from 1960 to 1980, in which the net anthropogenic N inputs entering Spain increased threefold in parallel with the growth of the emissions of reactive N to the environment including the mentioned rise of NH_3 emitted Lassaletta *et al* 2014a). Significant reductions in NH_3 volatilization have been achieved during the last 20 years in some member countries of the EU (e.g. Denmark, The Netherlands, UK) through the implementation of environmental policies. In contrast, NH_3 emissions in Spain increased by 14% in the period from 1990 to 2011, ranking the country last in terms of target achievement (European Environment Agency (EEA) 2013). Due to the recent revision of the UNECE Gothenburg Protocol, the target for Spain is now slightly more restrictive, with a requirement to achieve a further 3% NH_3 reduction in the period from 2020 to 2029 compared with the 2005 national emission. Furthermore, more stringent targets (29% reduction compared with 2005) might be expected for 2030, associated with the proposed revision of the National Emissions Ceilings Directive, currently under discussion (European Commission 2013).

Based on the necessity for stakeholders and policymakers to use robust methods to assess NH_3 mitigation strategies for countries, the objective of the present work was to develop a country- and region-specific and novel methodology to estimate the NH_3 abatement potential of different N fertilizer-based measures. Spanish cropping systems were used as a case study. For this scenario testing, we also considered the potential trade-offs regarding N yields and NUE, as well as N surpluses, which may lead to losses of other reactive N forms from these systems. This exercise is intended to provide information about the most effective scenarios in order to optimize N resources (i.e. reducing NH_3 losses and N surplus, increasing or maintaining N yields, and increasing NUE) at the regional and ultimately national scales.

2. Materials and methods

To assess NH_3 mitigation strategies over N fertilization of cropping systems and their potential trade-offs/co-benefits associated with crop yields and the loss of other N pollutants, we carried out a scenario testing as proposed by Oenema *et al* (2009). We developed a baseline scenario (BA) that intends to represent the current typical Spanish N application rates and practices for croplands and the resulting crop N yields. By combining estimated NH_3 emission factors (EFs) with data on fertilizer application rates, we were able to produce an emissions map for this BA scenario. Subsequently, 11 alternative management scenarios (table 1) were constructed to evaluate the efficacy of individual and combined technical measures and N application rates, and their corresponding emissions map was established. Finally, we related the NH_3 abatement effect of each scenario with its impact on crop N

Table 1. Description of the scenarios evaluated. 'NoU' refers to urea removal. In this scenario U is not applied thus N entering to crops decreased. 'Syn' refers to other synthetic N sources different than U. 'LowSyn' refers to a scenario where the rate of synthetic N fertilizers (others than U) is adjusted to the N harvested.

Name of scenario	Total N applied (Gg)	Urease Inhibitor	Mechanical incorporation of Urea (5 cm depth)	Incorporation of urea with irrigation	Urea removal	Manure Incorporation (10 cm depth)	Reduced application of N synthetic fertilizer	Incorporation of N synthetic fertilizer (others than Urea)
BA (Baseline)	1424	—	—	—	—	—	—	—
I. UI[a]	1424	X	—	—	—	—	—	—
II. U_{inc}	1424	—	X	—	—	—	—	—
III. U_{irrig}	1424	—	—	X	—	—	—	—
IV. M_{inc}	1424	—	—	—	—	X	—	—
V. $UI + M_{inc}$	1424	X	—	—	—	X	—	—
VI. $U_{inc} + M_{inc}$	1424	—	X	—	—	X	—	—
VII. NoU	1155	—	—	—	X	—	—	—
VIII. $NoU + Syn_{inc} + M_{inc}$	1155	—	—	—	X	—	—	X
IX. $NoU + LowSyn$	818	—	—	—	X	—	X	—
X. $NoU + LowSyn + M_{inc}$	818	—	—	—	X	X	X	—
XI. $NoU + LowSyn_{inc} + M_{inc}$	818	—	—	—	X	X	X	X

[a] Acronyms are explained in column heads.

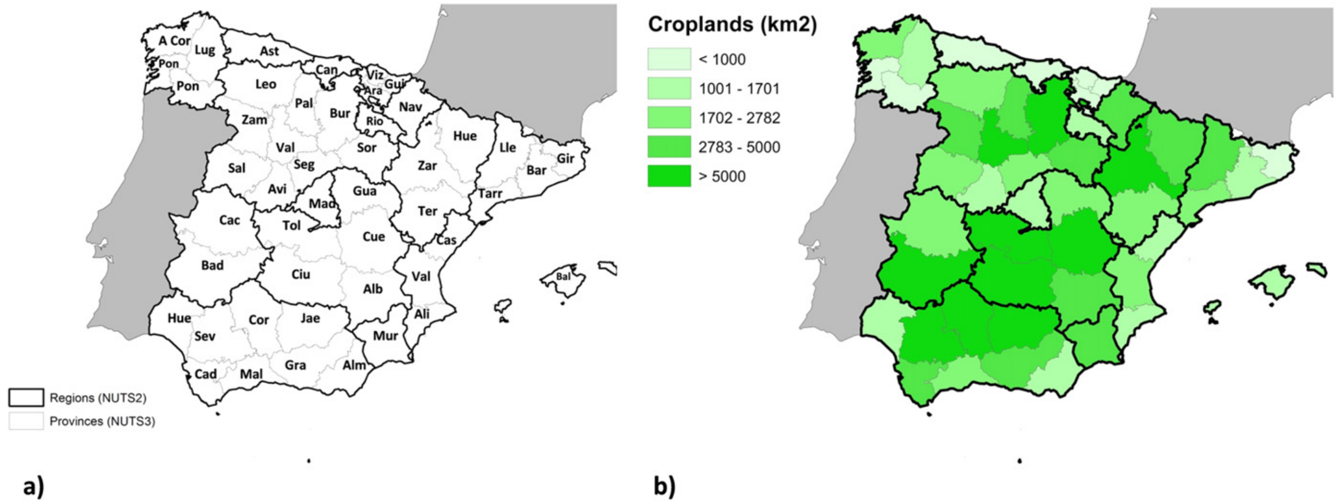

Figure 1. Map of Spain showing all provinces included in the study (a) and the distribution of national cropping area among provinces (b).

yield, potential trade-offs on other reactive N forms, and NUE. The latter was calculated as the ratio of yield to the total N input added to the cropping system, also described as partial factor productivity of applied nutrient index by Ussiri and Lal (2012).

2.1. Baseline scenario

The scale of the study was subregional (provinces; NUTS3 units in Eurostat; figure 1(a)). We established a detailed budget of the cropping systems for all the Spanish provinces using a soil surface N balance approach (e.g. Oenema *et al* 2003, van Grinsven *et al* 2012). This required estimating all the N inputs to the croplands (as explained below), including synthetic fertilizers (distinguishing between U and other synthetic fertilizers), animal manures, urban waste compost and sewage sludge, biological N fixation, and total atmospheric N deposition. The total N input after discounting NH_3 emissions in the crops was considered as effective fertilization (EFert). The difference between these effective agricultural inputs (EFert) and the outputs via harvested crops (including harvested straw) corresponds to the agricultural N surplus. We used this calculated N surplus as a proxy for other N environmental losses (through, for example, leaching and denitrification including possible associated N_2O) to assess which situation could potentially lead to less NH_3 loss while reducing trade-offs. The year 2008 was selected as the reference because it is the most recent year for which data from all N sources were available. We confirmed that it was a standard year in terms of rainfall and mean temperatures, which were only 10% higher (62 mm) and 0.7% lower (0.1 °C), respectively, than the 30-year average over all measurement sites (AEMET (Agencia Estatal de Meteorología) 2014).

Total N applied as fertilizer from U and other synthetic fertilizers was calculated from the Spanish N balance (MARM (Ministerio de Medio Ambiente y medio Rural y Marino) 2008), considering all mineral N applied in each province and the corresponding N from U to the total mineral N ratio. Since there are some Spanish provinces where synthetic fertilizers are also applied to grasslands, we calculated this amount in a two-step process: 1) defining these regions based on MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente 2012) and 2) considering the percentage of synthetic N fertilizer applied in these provinces according to detailed application maps (Leip *et al* 2011). The result was deducted from the total synthetic N applied in each province. N inputs from organic fertilizer application were calculated as follows. Firstly, N excretion (Nexcr) was determined following the Spanish National Airborne Pollutant Emissions Inventory (MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2014) methodology, by multiplying livestock population and N excretion factors. The livestock population of each animal category (pigs, dairy cattle, non-dairy cattle, sheep, goats and poultry) for 2008 was obtained from the Agricultural Statistical Yearbook (MARM (Ministerio de Medio Ambiente y medio Rural y Marino) 2009) for each province. Nitrogen excretion rates (kg place^{-1} year^{-1}) for animal housing were derived for the base year from the National Inventory Report (MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2014). The N excreted by outdoor grazing animals was assumed to reach permanent grasslands (not studied herein) and also the croplands via temporary grasslands. The percentage of N corresponding to grazing animals was deducted from the total by applying the factor proposed by the National Inventories (UN 2006). Secondly, to obtain the amount of N available for field application, N lost during housing and manure storage as NH_3 and N_2O was subtracted from Nexcr using the factors proposed for Spain by Oenema *et al* (2007). We distributed the manure applied to crops and managed grasslands (not including natural rough grasslands) proportionally according to their share of surface area in each province, on the basis of the information on soil uses provided by the CORINE Land Cover project (2006). Nitrogen inputs from the agricultural application of urban waste compost were estimated based on production data at the regional scale (MAGRAMA (Ministerio de Agricultura, Alimentación

Table 2. N yield, NH_3 emitted and N surplus (Gg N yr^{-1}), and yield-scaled NH_3 emissions, yield-scaled N surplus, yield change with respect to the baseline scenario and nitrogen use efficiency (%) for the 11 mitigation scenarios and the baseline (BA).

	GgN yr^{-1} (Spain)				%		
Scenarios	Yield	NH_3	Surplus	NH_3/Yield	Surplus/Yield	Yield change	N.U.E.
BA	723	262	447	36	62	0	51
I. UI	730	222	472	30	65	1	51
II. U_{irrig}	720	252	452	35	63	0	51
III. U_{inc}	727	231	466	32	64	1	51
IV. M_{inc}	777	113	534	15	69	8	55
V. $UI + M_{inc}$	789	73	562	9	71	9	55
VI. $U_{inc} + M_{inc}$	787	82	555	10	71	9	55
VII. NoU	635	201	319	32	50	−12	55
VIII. $NoU + Syn_{inc} + M_{inc}$	705	42	408	6	58	−2	61
IX. NoU + LowSyn	482	191	145	40	30	−33	59
X. $NoU + LowSyn + M_{inc}$	562	42	213	8	38	−22	69
XI. $NoU + LowSyn_{inc} + M_{inc}$	565	37	215	7	38	−22	69

y Medio Ambiente) 2012). Nitrogen proportions of compost produced at biological treatment plants were assumed to be 1.8% for municipal solid waste and 2.2% for the organic fraction of municipal solid waste (MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2012). Similarly, N applied through sewage sludge addition was accounted for based on production data and transfers between different regions, considering an average N content of 4.7% (dry weight basis) (MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2012).

We estimated N fixation by the 18 Spanish N-fixing crops, including several legume species by applying the method proposed by Herridge *et al* (2008) that takes into account yield and belowground production. Spatialized information on atmospheric N deposition of reduced and oxidized N compounds is available from the EMEP 50×50 km grid. We estimated the total yearly crop production of the 103 annual and permanent crops produced in Spain in 2008 using the information of the Spanish Agricultural Yearbook (MARM (Ministerio de Medio Ambiente y medio Rural y Marino) 2009). This source also provides information on harvested straw for cereals and legumes. To assign the N content to every product we used the values gathered by Lassaletta *et al* (2014b).

In the absence of specific generalized NH_3 EFs (table 1, supplementary material) derived from the Spanish experiments, NH_3 EFs were obtained for different (1) organic fertilizer types (pig slurry, poultry slurry (PS) and farm yard manure (FYM), as kg NH_3-N kg^{-1} manure total ammonium N) and (2) synthetic fertilizers (kg NH_3-N kg^{-1} synthetic N), using the equations from the existing modeling approaches developed by Chambers *et al* (1999) and Misselbrook *et al* (2004), respectively. A fraction of the inorganic N is simulated to be rapidly lost as NH_3 emissions following the approach by Chambers *et al* (1999). The EFs are calculated considering the properties of the slurry, the environmental and soil conditions during manure application (temperature, rainfall, soil moisture) and management factors (e.g. the method of application and incorporation). Regarding mineral fertilization, the model simulates NH_3 volatilization using EFs developed by Misselbrook *et al* (2004), whereby a fraction of the N applied is volatilized depending on fertilizer type (e.g. urea, ammonium nitrate) as well as soil conditions. The proposed methodologies were compared to the ALFAM approach (Søgaard *et al* 2002; www.biocover.dk/alfam-model-dk). We chose the proposed methodology on the basis that their estimated NH_3 emissions are more sensitive to well known management and climatic factors than ALFAM and the approaches are simple enough for this type of exercise. Our proposed methodology to estimate NH_3 emissions differs from that proposed by MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2014, which uses default EMEP-CORINAIR emissions factors (European Environment Agency (EEA) 2013), which do not account for many different factors. It must be pointed out that the scope of this study is not to investigate the accuracy of estimated NH_3 emissions, which cannot be assessed due to lack of real measured data in Spain, but rather to assess the relative effect that management and climate factors exert on NH_3 emissions from fertilized crops, which account for 62% of national NH_3 emissions (MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2013a, European Environment Agency (EEA) 2013).

2.2. Mitigation scenarios

The different mitigation scenarios used in this study are summarized in table 1. The mitigation strategies were selected based on the conclusions from the UNECE Task Force on Reactive Nitrogen (TFRN), recently published by Bittman *et al* (2014). Based on expert assessment, we selected those considered most feasible to be implemented in Spain. The scenarios evaluated can be grouped as U management, improved application of manure, and reduction of synthetic N fertilizer.

Mitigation factors (MFs) for U incorporation, as well as for U irrigation (U_{irrig}) were derived from Bouwman *et al* (2002). The MFs for urease inhibitors (UIs) were taken from the available literature (table 2, supplementary material). Although there are several UIs available on the market, we used the MF for NBPT (N-butyl-thiophosporic triamide), the most widely used UI (Sanz-Cobena *et al* 2014). Soil pH has been considered to be an influencing factor over the effectiveness of NBPT (Linquist *et al* 2013). We calculated three different MFs for NBPT corresponding to the three pH types described in Abalos *et al* (2014), and then assigned the surface proportion for each pH class in all provinces, using the HWSD 1.2 (Harmonized World Soil Database) viewer (FAO/IIASA/ISRIC/ISSCAS/JRC 2012). Mitigation factors for manure incorporation (M_{inc}) were calculated using the same procedure as for NH_3 EFs, being sensitive to temperature, rainfall and soil moisture (see section 2.1). The estimated excess N fertilizer inputs to Spanish crops led us to study the effect of a controlled fertilization based on suppression of U application in all provinces (i.e. NoU scenarios, table 1) and the reduction of the application rate of the remaining synthetic fertilizers (i.e. LowSyn scenarios, table 1).

To assess the change in yield resulting from changes in N inputs to soil associated with the different scenarios we used the following approach. In agreement with the views developed by Lassaletta *et al* (2014c), we considered that the cropping system of each region can be characterized by a one-parameter hyperbolic relationship between yield and the effective total N input to the soil, both integrated over all arable land and the duration of a crop rotation cycle:

$$Y = Y\max \cdot \mathrm{EFert}/(\mathrm{EFert} + Y\max) \qquad (1)$$

where Y is the long term integrated annual yield; EFert is the effective fertilization rate calculated as the sum of total inputs of N to the soil by symbiotic fixation, atmospheric deposition, manure and synthetic fertilizer application, the net of the amount lost through NH_3 emission; and Ymax is a region-specific parameter representing the yield value reached at saturating fertilization. We first determined the Ymax value of this relationship using the calculated data on Y and EFert for each Spanish province in the BA scenario (Y, Ymax, EFert are expressed in kg N ha^{-1} yr^{-1}):

$$Y\max = Y \cdot \mathrm{EFert}/(\mathrm{EFert} - Y) \qquad (2)$$

For each scenario, for which a change in EFert can be predicted because of changes in the N inputs to the soil and/or in the NH_3 emissions, the resulting Y is recalculated using relation (1). We then used this approach to calculate a new surplus (the difference between EFert and Y) which can be considered as a proxy for other environmental losses of N via leaching and denitrification. It is important to clarify that Y corresponds to estimated yields that integrate several crops in rotation, in the particular conditions of the given region, and without significant changes with respect to the practices (besides fertilization) that characterize the current cropping systems. Indeed, such changes could produce a shift to another value of Ymax (Bodirsky and Müller 2014, Lassaletta *et al* 2014c). The latter parameter should therefore not be considered as the maximum regional potential yield in order to estimate yield gaps.

Finally, dividing the NH_3 emissions or the surplus by the total output (Y), we calculated the yield-scaled NH_3 emissions and surplus.

N in slurries is usually highly mineralized and therefore its soil dynamics is very similar to that of mineral fertilizers. N in solid manures is only partially mineralized the first year of application, but in long-term managed systems the cumulative residual effect of previous applications would compensate for a large share of this reduced short-term availability (Gutser *et al* 2005).

3. Results

3.1. Baseline scenario

The estimated annual N input to the Spanish cropping systems was twice as high as the amount of N exported with harvest (i.e. a total excess of N amounting to 711 Gg N yr^{-1}) (table 2). Of the total N applied (i.e. ~1400 Gg N), 74 and 15% come from synthetic N sources and manures, respectively (figures 2(b), (c)). From a regional perspective, most of the provinces presented excess N application to crops (>150 kg N ha^{-1} yr^{-1}) (figure 2(a)), especially those with high animal density in the north of the country. One-half of the manure N from non-grassland–based livestock production was applied in ten provinces (out of 48) at rates averaging 96 kg N ha^{-1} yr^{-1} (figure 2(c)). Pig slurry and poultry manure were respectively applied for 33 and 12% of the manure-fertilized crop surface area. The remaining fraction was considered to be applied as FYM. Synthetic N fertilizers were applied in all regions, but at particularly high rates (>170 kg N ha^{-1} yr^{-1}) in seven provinces of the northern part of the country (figure 2(b)). Urea was applied in all regions except Cantabria (Can) (northwestern Spain, figure 1(a)), and it is the most frequently used form of synthetic N fertilizer, together with calcium ammonium nitrate (c. 29% for both). Nineteen of the 48 provinces, scattered across the whole country and representing 43% of the national cropping area, accounted for 68% of the total urea-N used in croplands. In sum, the highest total inputs were mostly distributed in the north of Spain and in some provinces of the periphery of the country (figure 2(a)).

High N application and availability in croplands were associated with large NH_3 losses in the BA (262 Gg NH_3-N for 2008, representing 18.5 kg NH_3-N ha^{-1} yr^{-1} and 18% of the applied N on average and a surplus of 447 Gg N yr^{-1}, table 2). Hot spots of NH_3 volatilization were located in regions with intensive non-grassland–based livestock production systems and fertilized crops such as Lleida (Lle) and Murcia (Mur) (i.e. 6.2 and 3% of the total NH_3 emission, respectively) (figures 1(a), 2(d)). In contrast, provinces of the Castilla–Leon region such as Soria (Sor), Burgos (Bur) and Palencia (Pal) and of the Castilla–La Mancha region, such as Cuenca (Cue) and Ciudad Real (Ciu); as well as some provinces in the north (e.g. Gipuzcoa (Gui), Vizcaya (Viz), and Asturias (Ast)) showed the lowest emissions from fertilized crops, mostly due to low organic fertilization (Castilla–León

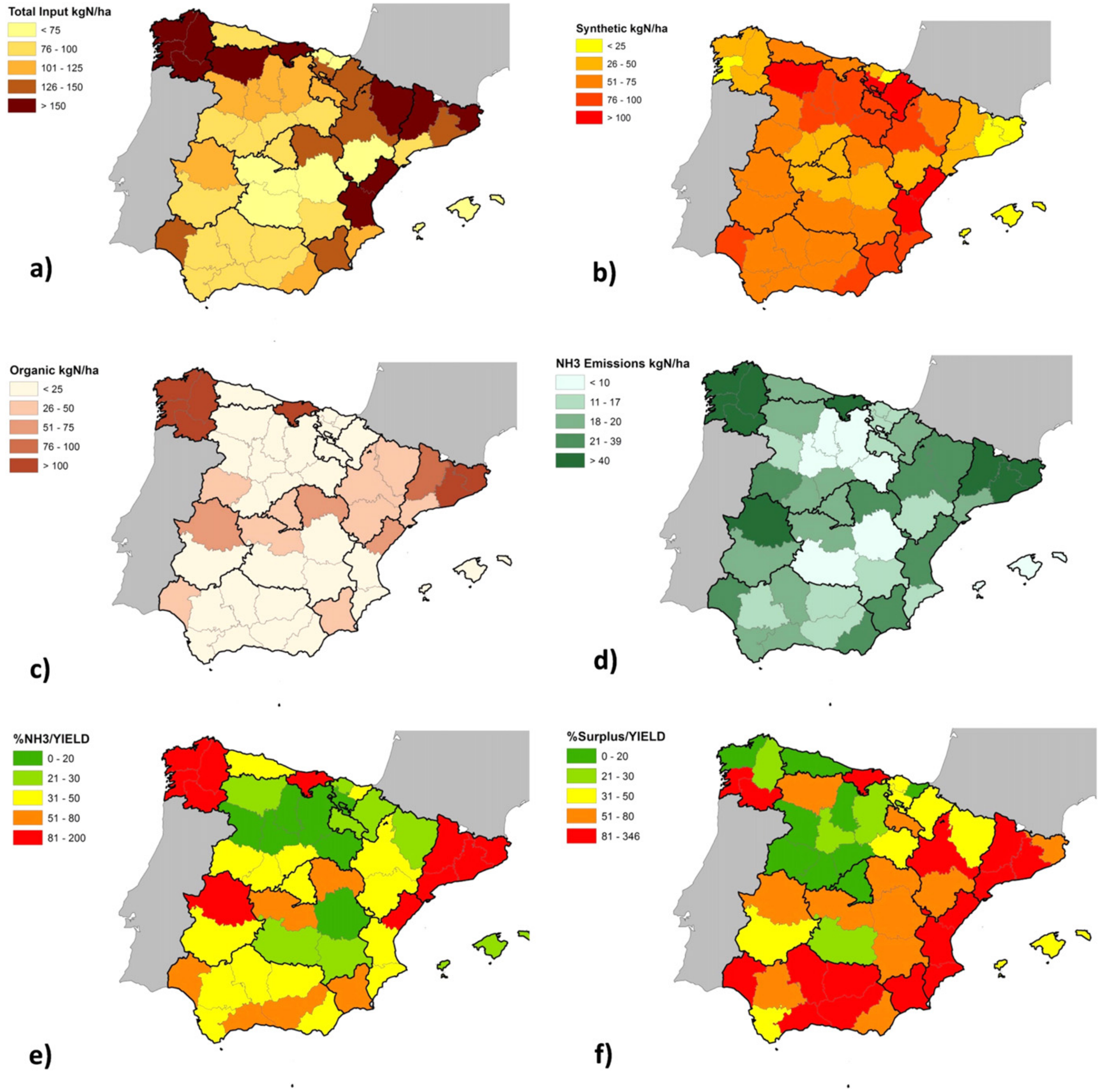

Figure 2. Maps of Spain showing total N entering to cropping soils (kg N ha^{-1} yr^{-1}) (a); N applied as synthetic fertilizers (kg N ha^{-1} yr^{-1}) (b); N applied as organic fertilizers (kg N ha^{-1} yr^{-1}) (c); NH_3 emission (kg NH_3 -N ha^{-1} yr^{-1}) (d); yield-scaled NH_3 emissions (%) (e) and yield-scaled surplus (%) (f) in each of the provinces analyzed.

and Castilla–La Mancha) and the dominance of grasslands in the north of Spain (figures 1(a), 2(c), (d)).

3.2. Mitigation scenarios

3.2.1. NH_3 emissions, yield, surplus and NUE. All mitigation scenarios showed a reduction of total NH_3 emission compared to BA (table 2). The best U management strategy in terms of NH_3 mitigated at a national scale was the use of urease inhibitors (UIs (I), see table 1) with 15% mitigation. Total emissions were reduced by 12% and 4% for U_{inc} (II) and U_{irrig} (III), respectively (table 2). For organic fertilizers, improved application (M_{inc}, IV) led to the highest reduction in total emissions when all N was assumed to be applied to crops (57% reduction). The combination of technical mitigation approaches for both synthetic fertilizers and manures had a high NH_3-abating effectiveness (scenarios V and VI, table 2). The UI + M_{inc} scenario (V) showed a 72% reduction compared to BA, and a similar mitigation of 69% was found for U_{inc} + M_{inc} (VI). Decreasing the rate of N in the NoU (VII) and NoU + LowSyn (IX) scenarios (table 1) lowered total NH_3 emissions by 23% and 27%, respectively, still referring to BA (table 2).

Looking at the effectiveness of the different strategies in decreasing NH_3 emissions for each province, we observed that the combination of technical strategies and controlled fertilization produced the highest mitigation. The NoU + $LowSyn_{inc}$ + M_{inc} scenario (VIII) reduced NH_3 volatilization in all regions (⩾77% mitigation in all cases, averaging 84%). On the other hand, the most effective individual strategies to abate NH_3 emissions were M_{inc} (IV) and NoU (VII) in 79% and 21% of the provinces, respectively (table 3, supplementary material). Assuming the implementation of the most effective individual measure for each province enhanced the mitigation of NH_3 losses the most, and led to a 63% reduction in the national emission.

Yield was reduced by suppressing U application (~12% reduction for NoU, VII) and combining NoU with controlled synthetic fertilization (~33% reduction for NoU + LowSyn, IX) (table 2). The combination of U suppression with incorporation of synthetic fertilizers and manure (NoU + Syn_{inc} + M_{inc}, VIII) resulted in yields 22% lower than the BA scenario. Overall, the surplus was increased for all scenarios increasing yield and reduced for those resulting in decreased yield (table 2), according to the shape of the yield–fertilization relationship used to calculate the yield. All individual measures increased N surplus uniformly in all provinces, M_{inc} being the strategy that increased it the most (with an average increase of 19%). The combination of strategies resulted in decreasing N surplus only when NoU was combined with other mitigation practices (scenarios VII to XI). However, our regionalized analysis allowed us to notice that this effect was not uniform among provinces since there were some regions where N surplus increased even in these scenarios (data not shown).

Nitrogen use efficiency showed very little effect from the technical mitigation scenarios (table 2). Nevertheless, it was increased in the scenarios involving manure incorporation or controlled application of fertilizers (scenarios IV–XI). Interestingly, by avoiding a significant loss of reactive N such as NH_3, these measures resulted in a higher yield with the same N input. Nitrogen use efficiency was also increased in all scenarios involving reduction of N inputs at the expense of lower yields, again in agreement with the yield–fertilization relationship, which implies higher efficiency at a lower fertilization rate.

3.2.2. Yield-scaled NH_3 emissions and yield-scaled N surplus. Scaling the results of NH_3 abatement to N yield (Gg NH_3-N Gg N yield^{-1}, %) revealed a similar trend, despite a change in the degree of success for the different strategies evaluated (table 2). Although crop yields decreased when limiting the N applied to crops, this reduction was even larger for NH_3 emissions in most cases and consequently, yield-scaled emissions generally decreased (table 2). The lowest yield-scaled NH_3 emissions were achieved for NoU + Syn_{inc} + M_{inc} (VIII) and NoU + $LowSyn_{inc}$ + M_{inc} (XI) (table 2). An improved application of manures performed better than any of the strategies focused on U management (table 2).

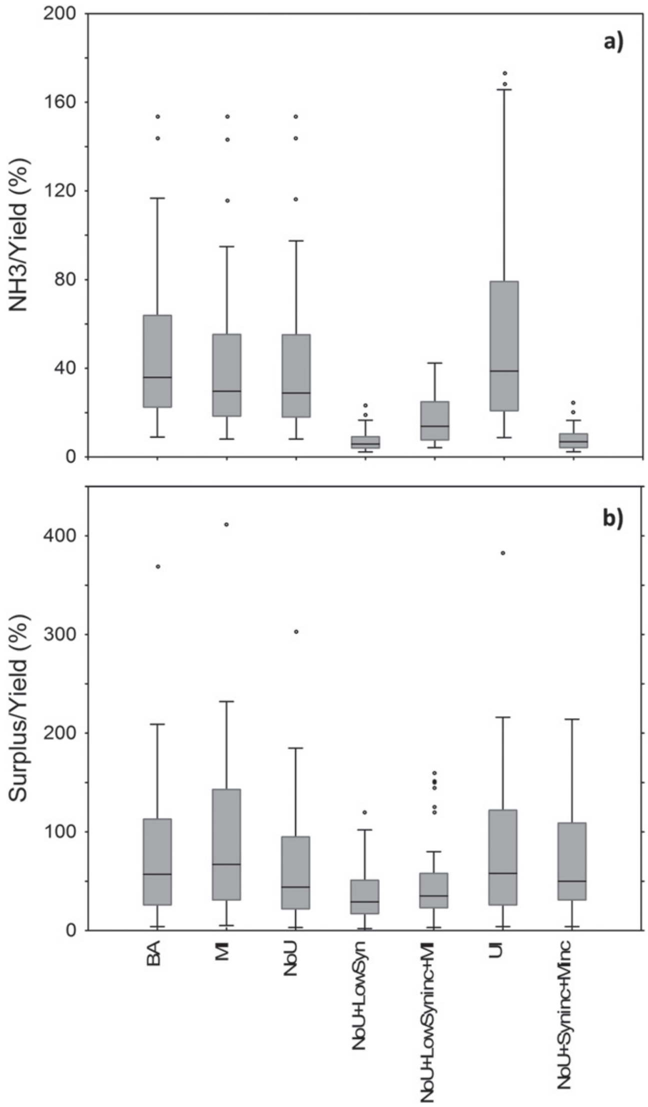

Figure 3. Yield-scaled NH_3 emissions (%) (a) and yield-scaled surplus (%) (b) for 6 of the 11 scenarios tested at each province: M_{inc}; NoU; NoU + LowSyn; NoU + $LowSyn_{inc}$ + M_{inc}; UI; NoU + Syn_{inc} + M_{inc}. Vertical bars indicate standard error of the mean.

From a regional perspective, scenarios X and XI (i.e. NoU + LowSyn + M_{inc} and NoU + $LowSyn_{inc}$ + M_{inc}) showed the largest yield-scaled NH_3 mitigation with a uniform effect in all provinces (77% and 81% mitigation, respectively). In contrast, NoU + $LowSyn_{inc}$ was the only combination of measures showing increased yield-scaled NH_3 emissions in more than half of the provinces (i.e. 66%; table 4, supplementary material). Differences between regions were also observed for strategies focused on individual measures (e.g. UI, M_{inc}, and NoU) (figure 3(a)). In a hypothetical scenario based on implementing the most effective individual strategy in each province, yield-scaled emission was reduced 67% (table 4, supplementary material).

Mitigation of NH_3 also had an effect on yield-scaled N surplus (Gg N-surplus Gg N-yield^{-1}, %) in the soil (table 2). All technical strategies (i.e. UI, M_{inc}) increased yield-scaled N surpluses (average 8% increase), whereas those measures based on suppression of U application and controlled fertilization (scenarios VII–XI) reduced yield-scaled N surpluses (average 31% reduction) (figure 3(b)).

4. Discussion

Our results showed that the country is not homogeneous in terms of N application (figures 2(a)–(c). Sutton *et al* (2013) introduced the distinction between too-little N and too-much N regions and countries. Even though Spain as a whole could be seen as a too-much N country, there are provinces differing in terms of N application excess, N accumulation and management, and thus NH_3 emissions (figure 2). Therefore, as recently proposed by Bustamante *et al* (2014) for greenhouse gas (GHG) emissions in agriculture and forestry, the effectiveness of any mitigation strategies should be assessed at the regional scale, on a case-by-case basis, as we did here for NH_3. In this study, we also proposed and linked productivity (e.g. N yields) and environmental (i.e. NH_3 emissions, N surplus) indicators as a necessary stage to effectively assess the implementation of any NH_3 mitigation measure at regional and ultimately national scales.

4.1. Effective yield-scaled NH_3 mitigation scenarios

Combining NH_3 abatement and crop productivity has pivotal implications for effective guiding of EU mitigation policy. This approach, firstly introduced by Van Groenigen *et al* (2010) for N_2O emissions at plot scale, allows identification of strategies needed to achieve dual goals of ensuring food security while protecting natural resources and the environment (Pittelkow *et al* 2013), through optimum fertilizer N management (Mosier *et al* 2006). Scaling emissions to yields might be helpful when assessing any mitigation policy potentially acceptable by farmers. However, since farmers will not be willing to implement any measure negatively affecting crop yields, information on yield-scaled emissions have to be used in combination with yield data.

According to our study, only four scenarios (M_{inc}, $UI + M_{inc}$, $U_{inc} + M_{inc}$ and $NoU + Syn_{inc} + M_{inc}$, table 2) led to a significant reduction in yield-scaled NH_3 emissions (i.e. 50% or higher mitigation respecting to BA) while maintaining (i.e. ⩾95% BA yield) or even enhancing N yields.

An improved application of organic fertilizers (M_{inc}, IV) was the measure showing the greatest yield-scaled NH_3 abatement at the country scale (57% with respect to BA, table 2). Incorporation of manure has previously been shown to have a high NH_3 abatement potential due to a reduction in the contact surface between the fertilizer and the atmosphere (Sommer and Hutchings 2001). It must be pointed out that the strategies considered in this study refer to land application of manure; thus NH_3 emissions from previous stages such as housing and storage are not included. In any case, since for this study it was assumed that all manure is applied as fertilizer, modifying the application strategy (i.e. manure incorporation) should not affect the previous management of this resource in terms of NH_3 emissions at the housing or storage stages.

Although it was three times less effective than incorporation of manures, the use of UIs (I) decreased yield-scaled NH_3 emissions (and increased yields) more than incorporating U either by washing or mechanically (II and III) in all provinces (table 2, table 4 supplementary material). This is in agreement with Abalos *et al* (2014), whose recent meta-analysis showed a significant crop yield increase when applying these fertilizer technologies. However, it was only when combining UIs with M_{inc} that both yield-scaled NH_3 emissions were substantially abated (i.e. 75%) and yields increased 9%, compared with BA. Of the most promising scenarios in terms of yield-scaled NH_3 abatement, only combining NoU with incorporation of all other sources of N fertilizer (i.e. other synthetic and manures, scenario VIII), was a fully beneficial situation, since yield-scaled emission decreased 82% and yields remained almost the same as the BA. Scenarios IX, X and XI, although they reduced yield-scaled NH_3 emissions the most, also decreased yields between 22% and 33% which can be hardly assumed by farmers. These results show that reducing environmental pollution might compete with food and feed production without any effort to change the animal/human diet (van Grinsven *et al* 2013).

4.2. Trade-offs of the mitigation scenarios

Potential trade-offs at the field scale, through the release of other N reactive forms (e.g. N_2O, NO_3^-), must also be addressed when assessing any N loss mitigation strategy. N surpluses in soils were increased by all measures excluding those involving NoU and NoU plus other strategies, thus enhancing the risk of trade-offs in the form of N pollution at the field scale (table 2). In our case, the calculated N surpluses of the most effective yield-scaled mitigation measures (table 2) could result in emissions of N_2O ranging from −0.39 to 1.15 Gg N_2O-N y^{-1} compared to the BA and according to the IPCC emission factor of 1%. In a more realistic approach, considering the proportion of rainfed and irrigated areas in Spain (78.4% and 21.6%, respectively) and the corresponding EFs proposed by Aguilera *et al* (2013) for Mediterranean conditions (0.08% for rainfed crops and 0.84%, as an average value between 1.01% for high-water irrigation treatments and 0.66% for drip-irrigated crops), the indirect N_2O emissions might range between −0.10 and 0.28 Gg N_2O-N y^{-1}, again compared to our BA.

In the particular case of manure, U, and other N synthetic fertilizers incorporation at seeding, NO_3^- losses can be triggered by leaching during high rainfall seasons or irrigation periods (Quemada *et al* 2013) and also enhance N_2O emissions when compared to surface applications (Pain *et al* 1989). These N losses could be minimized by synchronizing N application rates with crop demand through split applications thus avoiding excess of N (Rees *et al* 2013). As for mechanical incorporation, washing U could favor the presence of reactive N under conditions of higher soil moisture, due to favorable soil conditions for denitrification processes and subsequent high N_2O emissions (Wrage *et al* 2001). In cases of high NO_3^- content of soils previously formed through nitrification (Sánchez-Martín *et al* 2010), increased irrigation or rainfall could lead to most N losses as NO_3^- leaching. According to the European Nitrate Directive, 50% of Spanish aquifers have been declared Nitrate Vulnerable Zones, which is particularly serious in a country where the scarcity of water resources is significant. Mitigation scenarios which lead to the lowest surpluses should play a significant role for these areas where manure and synthetic fertilizer application is limited to 170 kg N ha^{-1} yr^{-1}.

In this study, both N surplus and yield-scaled N surplus were increased in a majority of scenarios, but decreased for those where the combination of measures and controlled synthetic fertilization were implemented (tables 1, 2; figure 3(b)). In addition to discussed trade-offs such as N_2O and NO_3^- losses, we should take into account that, with the present spatial concentration of livestock, NoU scenarios may involve the transport of large amounts of manure over long distances, thus increasing fuel consumption and the GHG net balance of the cropping areas (Robertson and Grace 2004), in spite of lower N_2O emissions associated with organic fertilizers than those issued from synthetic fertilizers, in Mediterranean areas at least (Aguilera *et al* 2013).

Due to the significant environmental implications of the N pollution established for Europe (Sutton *et al* 2011), optimum fertilizer application rates from a social welfare perspective should be far lower than the recommended rates from a narrow agronomic perspective (van Grinsven *et al* 2013). In the present study, the highest optimization of N resources was found in the NoU+Syn_{inc} + M_{inc} scenario (X), almost reaching a fully beneficial situation in terms of yield-scaled NH_3 and surplus, yields, and NUE (table 2). However, the best NH_3 mitigation practice at the country level is that in which the most effective strategy for each province is implemented (63% mitigation of yield-scaled NH_3, 64% decrease in N surplus and 9% increase in yields).

4.3. Uncertainties of the study and applicability of the proposed mitigation measures

4.3.1. Baseline scenario. For the BA scenario we assumed that split-application of fertilizers was not carried out in any case. This assumption would have lowered total emissions in the BA since a split-application may enhance the synchrony between N application and crop needs.

All fertilizers were assumed to be surface-applied, which conversely may result in an overestimation of NH_3 emissions for BA even though U and ammonium nitrate (the most used fertilizers among farmers in 60% of the cases) are usually applied at dressing, when incorporation is not possible. Urea incorporation with irrigation water is only carried out in irrigated and fertigated crops (during summer, mainly fruit trees and maize, which account for 2.1% and 7.1% of total Spanish cropping area). The incorporation of synthetic N fertilizers via rainfall water when rainfall events are forecasted is also a common practice among Spanish farmers when possible. Similarly to the case of U, animal manure is often applied before seeding and then incorporated by tillage, thus posing a serious risk of NH_3 volatilization immediately following application. Finally, we also assumed that no urease inhibitors are used by Spanish farmers, which is generally the case since they generally prefer other options that may provide similar benefits without incurring the additional cost of these products.

The methodology followed aimed to determine which NH_3 emission factors might lead to uncertainties in the results. Assumptions made in terms of meteorological and land conditions during application were based on expert judgment due to difficulties in gathering real data on activity data (e.g. manure and mineral fertilizer management). This lack of information also impairs the numerical calculation of the uncertainty associated with each emission factor. Nevertheless, it is important to consider that this uncertainty on the EFs could lead to an over- or underestimation of total emissions, but this effect does not have an impact when comparing scenarios.

4.3.2. Mitigation scenarios. A general assumption of 100% applicability was considered for all the mitigation measures assessed. However, the applicability of these strategies is always limited by the fertilization strategy and the specific characteristics of Spanish agroecosystems. For example, mechanical incorporation of fertilizer at top-dressing is infrequent among farmers due to the heavy equipment needed for its incorporation, which can damage the root system of growing crops and also involves cost and management constraints. Also, we assumed that all the manure was incorporated in the first 2 h by injection technique (i.e. 10 cm depth), which may be difficult in some soils. Further, all generated manure was supposed to be applied to croplands, which is not the current situation as farms and croplands are separated by long distances due to the associated costs of manure transportation.

Irrigated agriculture in Spain only accounts for 21.6% of the cultivated area (MAGRAMA (Ministerio de Agricultura, Alimentación y Medio Ambiente) 2013b), which prevents urea incorporated with irrigation water from being completely adopted. Another example relates to UIs, whose applicability relies on the proven crop yield improvements (Abalos *et al* 2014) and cost savings by avoiding top-dressing applications. Therefore, current acceptance of UIs by Spanish farmers is far from 100% assumption. Besides, reduced application of N synthetic fertilizers is expected to increase due to higher professional formation of Spanish farmers and new regulations about integrated production and environmental impacts of agriculture.

As a whole, this study allowed us to assess in a realistic manner the implementation of NH_3 mitigation measures on a regional basis. Although this methodology could be used for other countries and mitigation scenarios leading to different results, in the case of Spain, the most promising set of measures (NoU + Syn_{inc} + M_{inc}) led to a decrease of c. 76% (63% for the application of the most effective individual measure at each province) with an assumed applicability of

100% while preserving N yields and also decreasing the N surplus. According to expert judgment about the applicability of NoU + Syn_{inc} + M_{inc}, a low estimate of 50% could be assumed meaning that NH_3 mitigation from N fertilizing would drop to 38% (32%), which is not negligible in terms of achievement of emission targets and may allow the reduction of national emission below the ceiling.

5. Conclusions

Nitrogen fertilization in Spain is heterogeneous and thus NH_3 mitigation options must be assessed on a regional basis to account for within-country variability. We presented a novel and country-specific methodology to assess and propose the best strategies to abate NH_3 from N fertilizer application to crops. This was carried out by considering the side effects on crop productivity (i.e. on N yields), N use efficiency (NUE), and trade-offs (i.e. N surplus, used as an indicator of potential N losses by denitrification/nitrification and NO_3^- leaching/run-off). Such approach allowed us to show that implementing the most effective mitigation strategy for each province may lead to 63% NH_3 mitigation at the country level, whereas implementation of a single strategy for all regions would mitigate NH_3 by 57% at the highest. Strategies that involved combining mitigation measures produced the largest abatement in all cases (c. 80% reduction in some provinces). This study shows that the adoption of certain viable measures at a regional scale provides an opportunity for countries like Spain, where agricultural policies are transferred to regional administrations, to meet the international agreements on NH_3 mitigation, while supporting crop yields and increasing NUE. From the eleven scenarios tested, only four reduced yield-scaled emissions (i.e. 50% or higher mitigation with respect to the baseline, BA) while maintaining (i.e. ⩾95% BA yield), or even enhancing N yields. Among them, only one (combining NoU with incorporation of all other sources of N fertilizer, scenario VIII) showed a fully beneficial situation, since yield-scaled emission decreased 82%, reduced N surplus 9% and yields remained almost the same as the BA. Therefore, this approach can be seen as a tool to detect which are the most promising mitigation measures in terms of farmers' acceptance and environmental protection.

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