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Global assessment of nitrogen losses and trade-offs with yields from





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HIGHLIGHTS

GRAPHICAL ABSTRACT

- We simulate N losses of major cereal crops by using a global crop model.
- N losses are focused on several main producers, where more attentions should be paid.
- NLI is a useful indicator for assessing trade-offs between N losses and yields.
- Mitigation scenarios show that N losses can be reduced without compromising yields.

N inputs, N losses, and yields relations (b) rice (a) maize (c) wheat 200 20 150 N losses (kg N ha⁻¹) Chins 100 100 rope (2 M 200 250 300 200 N inputs (kg N ha-1) N inputs (kg N ha⁻¹) N inputs (kg N ha⁻¹) 1.5 2.0 4.5 5.5 4.0 5.0 yields (t ha⁻¹)

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ABSTRACT

Agricultural application of reactive nitrogen (N) for fertilization is a cause of massive negative environmental problems on a global scale. However, spatially explicit and crop-specific information on global N losses into the environment and knowledge of trade-offs between N losses and crop yields are largely lacking. We use a crop growth model, Python-based Environmental Policy Integrated Climate (PEPIC), to determine global N losses from three major food crops: maize, rice, and wheat. Simulated total N losses into the environment (including water and atmosphere) are 44 Tg N yr^{-1} . Two thirds of these, or 29 Tg N yr^{-1} , are losses to water alone. Rice accounts for the highest N losses, followed by wheat and maize. The N loss intensity (NLI), defined as N losses pro unit of yield, is used to address trade-offs between N losses to produce the same amount of yields. Simulations of mitigation scenarios indicate that redistributing global N inputs and improving N management could significantly abate N losses and at the same time even increase yields without any additional total N inputs.

1. Introduction

Anthropogenic activities are the major driver of changes in the global nitrogen (N) cycle (Fowler et al., 2013). Terrestrial N flows resulting from anthropogenic activities have increased to >3.3-fold of those resulting from natural processes by 2010 (Fowler et al., 2013; Galloway et al., 2014). As a consequence, the global N cycle is now 3.5 times above what is considered as a safe threshold (Rockstrom et al., 2009). Agriculture is the largest consumer (63%) of annual terrestrial reactive N (Sutton et al., 2013). Global industrial N fertilizer application increased 9-fold from 1960 to 2010 (Ladha et al., 2016; Sutton et al., 2013), with N fertilizer inputs to croplands reaching 120 Tg N yr⁻¹ (Tg = 10^{12} g) at the end of that period (Fowler et al., 2013). This unprecedented increase in N flows was made possible by the development of industrial fixation of atmospheric N (Haber-Bosch process) and the associated mineral N fertilizer use (Galloway et al., 2008). The drivers of this development are the need to supply food for an increasing global population, dietary shift towards more meat and dairy products consumption, and growing biofuel demand (Foley et al., 2011). On the downside, this development is associated with increasing agricultural N losses into the environment. causing stratospheric ozone depletion, eutrophication and acidification of water and soil, as well as losses in the diversity of ecosystems (Babbin and Ward, 2013; Clark and Tilman, 2008; Conley et al., 2009; Davidson, 2009; Diaz and Rosenberg, 2008; Erisman et al., 2013; Foley et al., 2005; Foley et al., 2011; Guo et al., 2010; Liu et al., 2013b; Sutton et al., 2013). Many studies have found that N use efficiency (NUE, defined as the ratio of crop harvested N to total N inputs) is low in major food producing regions. On global average, it is only about 0.42 in 2010 (Zhang et al., 2015). Without emission reductions, global N losses are expected to further increase and reach levels higher than 150% of the 2010 values by 2050 (Bodirsky et al., 2014).

To control N emissions, it is important to quantify and identify the main pathways and major contribution regions of N emissions. Previous studies of N losses performed at a global scale were mainly based on mass balance methods. On this basis, Liu et al. (2010) found that about half of global total N inputs into croplands were lost to the environment. Bouwman et al. (2013) estimated that around 93 Tg N yr⁻¹ was lost from arable lands and 45 Tg N yr⁻¹ from grasslands. Lassaletta et al. (2014) investigated the relationship between crop yields and N inputs based on FAO (Food and Agriculture Organization of the United Nations) data from 124 countries and concluded that about 53% of N added to croplands was lost to the environment. Zhang et al. (2015) built a global N budget

database and the total N losses to the environment were estimated at about 100 Tg N yr⁻¹ in 2010. However, all of these studies require crop yields as data inputs to quantify harvested N. Consequently, interactions between N dynamics and crop growth cannot be represented, which are essential to explore the trade-offs of N losses and yield benefits for future N management. Mass balance method applies the same empirical equations to calculate N fluxes over a large scale without explicitly considering the spatial variability (e.g. site-, climate- and management-specific differences). Besides, most of these global N balance assessment studies focus on total N fluxes aggregated from different crops (and grasses) with much less attention on crop-specific disparity, which is important to guide N fertilization management, especially from the major cereal crop cultivations. Therefore, it is critical to explicitly investigate crop-specific N losses and the related trade-offs with yields in order to provide suggestions for controlling N emissions.

While biophysical crop growth models nowadays have the ability to account for site- and crop-specific interactions between plant growth and N turnover, only few studies so far have made use of this ability in assessing agricultural N losses on a large scale. Examples are the studies of van der Velde et al. (2009) who used the Environmental Policy Integrated Climate (EPIC) model (Williams, 1995; Williams et al., 1984) to estimate N losses through leaching for rapeseed cultivation in Europe; and the study of Del Grosso et al. (2009) who used the DAYCENT model to study global N losses from maize, soybean, and wheat cultures. In addition, the spatial resolution in the simulations by Del Grosso et al. (2009) is quite coarse (1.9 arcdeg), and no crop-specific information on N fertilizer use and N leaching is given. Another example is the study of Qiu et al. (2011) who applied the GIS-based DNDC (Denitrification-Decomposition) model to simulate N leaching from croplands at the county level in China, but did not give site- and crop-specific information on N losses. None of these studies include rice. Three major cereal crops, i.e. maize, rice, and wheat, together consume about 60% of global N fertilizer application (Ladha et al., 2005) and provide about 57% of the dietary calories produced by agriculture (Tilman et al., 2011). In order to identify the hotspots of N losses from crop cultivations, it is important to conduct a high spatial resolution assessment of N losses by focusing on these three major crops.

The concept of NUE is generally used in N management. Achieving high NUE is one of the major targets for modern agriculture (Conant et al., 2013; Cui et al., 2014; Lassaletta et al., 2014; Zhang et al., 2015). However, this concept cannot be directly used for N loss assessment due to soil N imbalance, either N accumulation or N depletion (Liu et al., 2010). For example, Liu et al. (2010) estimated NUE based on the

Table 1

Description of different nitrogen fertilization schedules and scenarios.

	Application time			Application rates					
	1st	2nd	3rd						
Schedule	25								
FixN1	3 days before planting			Applying N inputs once					
FixN2	3 days before planting	35 days after planting		One-second of N inputs for each time					
FixN3	3 days before planting	35 days after planting	65 days after planting	One-third of N inputs for each time					
Scenario	s								
FixN3E	3 days before planting	35 days after planting	65 days after planting	One-third of 122, 134, 100 kg N ha^{-1} for maize, rice, and wheat for each time ^a					
AutoN AutoNE	Dynamic Dynamic			Applying N when crop needs with a cap set at the current level of N inputs Applying N when crop needs with a maximum amount of 122, 134, 100 kg N ha ⁻¹ for maize, rice, and wheat ^a					

^a Global average nitrogen inputs for maize, rice, and wheat are 122, 134, and 100 kg N ha⁻¹, respectively.

ratio of crop harvested N to total N inputs to be 0.59 on global average, however the ratio of total N losses to total N inputs was 0.49 other than 0.41 based on their estimations. This imbalance stemmed from the difference between total N losses and total N inputs minus total crop harvested N. The former considers the soil N imbalance and the latter does not. Taking into consideration the soil N balance is important for understanding the global N budget and for N management. In low input countries, typically African countries, such as China, soil N accumulation may be significant (Zhou et al., 2016).

Here, we use PEPIC, a grid-based EPIC model developed in a Python environment, to determine global N losses from the cultivations of the three major crops at a high spatial resolution of 30 (about 50 km at the equator). The EPIC model adopts the Century model (Parton et al., 1994), which is widely used to simulate soil carbon and N dynamics (Bhattacharyya et al., 2010; Cong et al., 2014), to model carbon and N turnover (Izaurralde et al., 2006). Coupled with spatial analysis tools, EPIC has been widely applied to estimate impacts of agronomic practices and climate change on crop yields (Balkovič et al., 2014; Folberth et al., 2014; Liu et al., 2013a; among others). Due to its integration with Python, PEPIC can be easily applied at different spatial scales. It has been successfully applied to simulate global maize growth (Liu et al., 2016). In addition, its performance on simulating growth of the other two crops is also guite robust (Fig. S1). Given that N leaching to water is a major source of water pollution, we also consider the losses to water alone, in addition to total losses to the environment which includes water and atmosphere. To address the trade-offs between N losses and yields, we propose a concept as N loss intensity (NLI) which measures N losses per unit of yield. Finally, in order to demonstrate the feasibility of reducing N losses while still maintaining or increasing global production of the three target crops, the trade-offs between N losses and crop yields under three proposed N fertilization scenarios are also investigated.



Fig. 1. Global maps of nitrogen losses into the environment (Nt) and water (Nw) for maize (a, b), rice (c, d), and wheat (e, f).

2. Methods

2.1. Model and input data

The EPIC model (Williams, 1995; Williams et al., 1984) simulates crop growth and soil nutrient dynamics. In this study, the PEPIC model was used for the global simulation at a resolution of 30' (Liu et al., 2016). Default parameters of the EPIC model were used for global application, as it is difficult to adjust model parameters at specific regions on such a large scale. Additionally, the estimated N losses were compared with previous studies.

Soil N inputs considered in EPIC include fertilizer (N_{fer}) and manure (N_{man}) application, crop residue decomposition (N_{dec}), and rainfall deposition (N_{dep}). Soil N outputs are N up taking by crops (N_{up}), N losses into the atmosphere via volatilization of ammonia (N_{av}) and denitrification of nitrate (N_{ad}) and exports of dissolved or particle-bound N into water with soil erosion (N_{ws}), surface runoff (N_{wr}) and leaching (N_{wl}).

To close the soil N cycle, change of soil N stock (Δ N) is also considered. Soil N budget is expressed as following:

$$N_{fer} + N_{man} + N_{dec} + N_{dep} = N_{up} + N_{av} + N_{ad} + N_{ws} + N_{wr} + N_{wl} + \Delta N$$
(1)

In this study, we focused on N losses into the total and aquatic (water) environment:

$$N_t = N_{av} + N_{ad} + N_{ws} + N_{wr} + N_{wl} \tag{2}$$

$$N_w = N_{ws} + N_{wr} + N_{wl} \tag{3}$$

where N_t and N_w are N losses into the total and the aquatic (water) environment, respectively. All these fluxes are calculated in units of kg N ha⁻¹. In EPIC, N_{ad} is a function of soil temperature and water content, while N_{av} is calculated based on soil temperature and wind speed.



Fig. 2. Nitrogen inputs (N_{in}) and nitrogen losses into the environment (N_t) and water (N_w) for maize (a, b), rice (c, d), and wheat (e, f) at country level. Countries with the smallest areas (for a total of 1% of global total cropland areas of each crop) are discarded; different shapes represent different continents; sizes represent cropland areas for each country; colours present ratio of N losses $(N_t \text{ and } N_w)$ to N_{in} ; dashed blue vertical line represents the world average N_{in} for each crop; dashed green line represents linear regression between N_t and N_{in} (b, d, f); equation represents the linear relationship; R^2 is the coefficient of determination of equation. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

 N_{wr} and N_{wl} are calculated by the products of nitrate-N concentration and soil surface runoff and seepage water. N_{ws} is calculated by considering soil erosion, organic N concentration in the top soil layer, and enrichment ratio of N in sediment. It should be noted that, while N_w is N export from agricultural fields with water flows, this export may not end up entirely in water bodies. Also, a portion of N losses to water bodies can flow to the sea, imposing the environmental impacts on coastal waters instead of the areas where they are initially generated. However, quantifying N losses reaching the coastal waters requires hydrological models to simulate the transport of nutrient in the water channels, which is beyond the scope of this study. One of the main purposes of this study is to explore the possible N losses from the farmlands and identify the global hotspots of losses. We do not trace further the fate of the N that is lost to water bodies. More details about N routines in EPIC can be found in Williams (1995) and Izaurralde et al. (2006).

Crop-specific N inputs with application of mineral fertilizers and manure (N_{fer} + N_{man}, N_{in} hereafter) were obtained from EarthStat (http:// www.earthstat.org). Mineral N, phosphorus, and potassium fertilizer inputs are based on Mueller et al. (2012), while N, phosphorus, and potassium inputs from manure are based on West et al. (2014), which considers 36% of manure being volatilized before reaching croplands. These data are currently the most up-to-date crop-specific datasets on global agricultural N inputs. N deposition from precipitation was estimated by EPIC based on annual precipitation and N concentration in rainfall. The sources of other input datasets can be found in SI. As both land-use (1998-2002) and fertilizer data relate to the years around 2000, this also holds for the model outputs (averaging between 1998 and 2002). Simulation results are presented at four levels, i.e. grid, country, continent, and globe. Average N_{in}, N_t, N_w, and crop yields at country, continent, and global levels were calculated as area-weighted averages of each variable at the corresponding levels. Total N_{in}, total N_t, total N_w, and crop production were calculated by multiplying average values of N_{in}, N_t, N_w, and yields by corresponding cropland areas calculated by using Eqs. (4)-(7):

$$TN_{in} = N_{in} * A_i \tag{4}$$

 $TN_t = N_t * A_i \tag{5}$

$$TN_{w} = N_{w} * A_{i} \tag{6}$$

$$P = Y * A_i \tag{7}$$

where A_i is cropland areas at the country, continental, and global levels; TN_{in} [Gg N yr⁻¹], TN_t [Gg N yr⁻¹], and TN_w [Gg N yr⁻¹] are total N_{in} , N_t .

and N_w at each spatial level, respectively; P [Tg yr⁻¹] is the crop production and Y [t ha⁻¹] is the yield.

In order to reflect the trade-offs between N losses and yields, we used the NLI defined as:

$$NLI = N_t / Y \tag{8}$$

The NLI quantifies the N losses associated with the production of a unit mass (1 t) of yield. Lower values of NLI mean less N losses for producing the same amount of food.

2.2. Crop management parameters

The model was separately applied to rainfed and irrigated cultivations. For irrigated crops, we assumed that drip irrigation was applied when water stress exceeded 10% on a given day up to a maximum annual supply of 1000 mm. This is a common practice in crop modelling when there is no information on the actual irrigation schedules and the amounts of water applied over time in different regions (Balkovič et al., 2014; Folberth et al., 2012; Liu and Yang, 2010; Liu et al., 2016; Rosenzweig et al., 2014). After simulation of both rainfed and irrigated cultivations, aggregated outputs from both cultivation systems were calculated for each grid cell by using area-weight averaging (Liu et al., 2007). Based on Del Grosso et al. (2009), we assumed that 25% of the crop residues are left on field. Furthermore, we assumed that phosphorus and potassium fertilizers were applied immediately before planting (Balkovič et al., 2014), as we found no global-scale dataset on fertilization schedules. Tillage was implemented before planting. Previous studies proposed various schedules for N application. For example, Folberth et al. (2014) applied 1/3 of N inputs before planting, and the rest one month after germination in sub-Saharan Africa. Stehfest et al. (2007) applied equal amounts of N four times globally. Other studies, not aiming at N loss assessment, used an automatic application schedule (Balkovič et al., 2013; Folberth et al., 2012; Liu et al., 2007). In this study, we tested three different N fertilization schedules: FixN1, FixN2, and FixN3 (Table 1). While Nt predictions were quite similar for all three schedules (Fig. 5), we found that the predicted country-specific yields agreed best with the FAO reported yields for wheat simulation when we used the FixN3 schedule and that there were only minor differences to reported yields for maize and rice with this schedule (SI and Fig. S1). Therefore, we selected FixN3 as the baseline for the analysis of the three N mitigation scenarios: FixN3E, AutoN, and AutoNE as defined in Table 1. Briefly, FixN3E distributes world N inputs evenly using the FixN3 schedule. AutoN stands for automatic N fertilization with current value of N inputs, which means N application is dependent

Table 2

Total nitrogen losses and crop production globally, continentally, and in the top 10 producing countries for maize, rice, and wheat in 2000. TN_{in}, TN_t, and TN_w are total nitrogen inputs (only including mineral fertilizer and manure), total nitrogen losses into the environment, and total nitrogen losses into water (in Gg N yr⁻¹), respectively; P is crop production (in Tg yr⁻¹).

Maize					Rice					Wheat				
Regions	TN _{in}	TNt	TN_{w}	Р	Regions	TN _{in}	TNt	TNw	Р	Regions	TN _{in}	TNt	TNw	Р
Global	17,816	12,414	6777	780	Global	20,332	17,129	13,531	677	Global	20,859	14,253	8583	517
Africa	1001	852	322	54	Africa	204	245	136	17	Africa	559	298	120	15
Asia	6363	4364	2728	190	Asia	19,492	16,432	13,107	624	Asia	12,899	8560	5893	232
Europe	2223	1629	721	115	Europe	37	26	11	3	Europe	3598	2893	1542	130
N. America	7013	4386	2267	347	N. America	299	153	91	12	N. America	2704	1714	658	95
Oceania	7	5	3	0	Oceania	2	2	0	0	Oceania	618	304	69	20
S. America	1209	1179	737	74	S. America	298	271	186	21	S. America	483	484	301	25
USA	5649	3286	1540	289	India	5126	3660	2729	188	China	6002	3197	2057	118
China	5048	3184	2002	128	China	8744	7533	6345	178	India	3594	3100	2636	36
Brazil	851	817	527	44	Indonesia	1271	1270	980	61	USA	2054	1324	576	65
Mexico	958	731	450	40	Thailand	713	491	296	46	Russia	408	643	134	39
India	381	318	175	19	Bangladesh	1209	1224	1007	45	Australia	610	298	64	20
Russia	57	174	27	9	Vietnam	831	715	557	30	Canada	580	340	48	29
Nigeria	88	92	39	8	Myanmar	152	353	289	16	Kazakhstan	41	261	7	16
Argentina	146	164	79	19	Philippines	267	91	31	10	Turkey	771	517	285	20
Ukraine	91	174	17	8	Pakistan	199	225	171	14	Pakistan	1190	600	502	6
France	694	379	246	27	Brazil	140	121	77	11	Argentina	348	251	117	19

on crop demands before reaching the current application rates. AutoN can be also used as baseline of efficient N fertilization, as the needed amount of N is applied at the time when it is needed. AutoNE stands for automatic N fertilization, but puts limits on the maximum application amounts evenly with global average N_{in} of maize, rice, and wheat. It should be noted that in all different N fertilization schedules and scenarios, we did not consider different types of N fertilizers due to unavailable data. This simplification has been widely used in large-scale crop modelling (Balkovič et al., 2014; Folberth et al., 2014; Rosenzweig et al., 2014; van der Velde et al., 2009).

2.3. Uncertainty analysis

We applied the Latin Hypercube Sampling (LHS) method to explore model uncertainties derived from model parameters (Mckay et al., 1979). LHS first divides the parameters into indicated number of segments. Then the parameter segments are randomized, and finally a random sample is chosen in each segment. It is more efficient than Monte Carlo (Mckay et al., 1979) and also used in the SWAT-CUP software (Abbaspour, 2011) to calibrate SWAT (Soil and Water Assessment Tool) model parameters (Abbaspour et al., 2007; Schuol et al., 2008; Yang et al., 2008). Parameters associated with N, phosphorus, and carbon routines in EPIC and their possible ranges for uncertainty analysis were carefully selected based on Della Peruta et al. (2014) and Wang et al. (2012) (Table S2). In this study, we considered 100 parameter segments for each crop based on the LHS method.

3. Results

3.1. Nitrogen loss assessment

The simulations identified some regions with particularly high N losses (Fig. 1), to which more attentions should be paid. As Fig. S2 suggests, the high losses in these regions are mainly related to high N inputs, and also Fig. 2 shows a clear trend that N losses increase with increasing N inputs. For maize cultivation, the model predicted particularly high levels of N_t in the eastern parts of China, South Korea, Japan, Indonesia, western Europe, the northeastern parts of the USA, and southern Mexico (Fig. 1a). Predicted N_w show similar spatial patterns as N_t (Fig. 1b). The USA and China are two major maize producers and also produced quite high Nt, especially in China (Fig. 2a). Consequently, these two countries together accounted for 52% of global TN_t for maize (Table 2). On the other hand, Nt is also relatively high for some countries with low N_{in} (e.g. <50 kg N ha⁻¹), especially in Africa. In some cases, the simulations indicate that the ratio of N_t to N_{in} was even higher than 1. Such high ratios in combination with low total N inputs suggest that soil N depletion should be considered a major factor. The relationship between N_w and N_{in} is not as clear as that between N_t and N_{in} , as shown in Fig. 2 the linear relationship and coefficient of determination (R^2) . We found that the total volume of growing season precipitation (GSP) and irrigation water also affects N losses, with low volume of GSP and irrigation tending to have low values of N losses at the country level (Fig. S3). These effects can also be observed in different climate regions. For example, Nt of maize is highest where the total volume of GSP and irrigation water is higher than 600 mm in temperate regions, although N_{in} is not highest in these regions (Fig. S4). The global TN_t and TN_w were calculated to be 12,414 and 6777 Gg N yr⁻¹ (Gg = 10^9 g) (Table 2). Asia and North America produced the highest total N losses. Together they accounted for 71% and 74% of the global TN_t and TN_w , respectively. While predicted total environmental N losses were similar in Asia and North America, with North America having much higher maize vields (Table 2).

For rice cultivation, high levels of N_t and N_w were found in China, South Korea, Japan, Vietnam, Bangladesh, and Indonesia (Figs. 1c and d). India and China have the largest rice cultivation areas. Together the two countries accounted for two thirds of global total N losses, both to the total environment as well as to water alone (Table 2). As Fig. 2 shows, the higher N losses in China are related to correspondingly higher inputs of N fertilizers compared to India. The total volume of GSP and irrigation water for rice cultivation is much higher than that for maize and wheat cultivation, particularly for wheat (Fig. S3). This difference may partially explain the higher N losses for rice in addition to its high N inputs than the other two crops. At the same time, high levels of total GSP and irrigation are associated with high levels of N losses from rice cultivation in various climate regions (Fig. S4). The predicted global TN_t and TN_w are 17,129 and 13,531 Gg N yr⁻¹ from rice cultivation (Table 2). Asia on the whole produced about 92% of the global rice harvests in 2000, and also played an overwhelming role in contributing to total N losses. It alone accounted for 96% of the global



Fig. 3. Intensity of nitrogen losses (NLI) into the environment in relation to yields against nitrogen inputs ($N_{\rm in}$) at country level. Countries with the smallest areas (for a total of 1% of global total cropland areas of each crop) are discarded; shapes represent different continents; sizes represent cropland areas for each country; colours present crop yields; FAO reported yields are used for China for rice cultivation and for India and Pakistan for wheat cultivation because of their underestimated yields, more details see SI; dashed vertical line represents the world average $N_{\rm in}$ for each crop.

total N released from rice fields into the total environment and for 97% of the global N released form rice fields into water.

For wheat cultivation, high level of N losses were predicted for large parts of southeastern China, northern India, South Korea, Japan, Thailand, southeastern USA, and central Europe (Figs. 1e and f). India and China are also the largest wheat producers and together contributed 44% and 55% of global TN_t and TN_w for wheat, respectively (Figs. 2e and f). In contrast, the USA and Russia, another two major wheat producers, produced much less N losses, but also applied much less N inputs, especially Russia. The global TN_t and TN_w were 14,253 and 8584 Gg N yr⁻¹ (Table 2). Asia produced 45% of global wheat harvests, but contributed 60% and 69% of global TN_t and TN_w , respectively. Following Asia, Europe presented the second highest total N losses, accounting for 20% and 18% of global TN_t and TN_w , respectively.

3.2. Intensity of nitrogen losses in relation to yields

Generally, yields increase with N inputs as long as N is limiting crop plant growth, but with increasing N inputs also N losses increase (Figs. 2-3). In order to demonstrate the complex relationship among N_{in}, yields, and N_t more clearly, we use the concept of NLI. The NLI presents guite high variation, indicating some countries perform better, in terms of low N losses, to produce the same amount of yields, while the other countries do not (Fig. 3). For maize cultivation, NLI is around 8-30 kg N t⁻¹ (Fig. 3a). High N inputs can be associated with high NLI, as in the case of China, as well as with low NLI, as in the case of the USA, where N is obviously more efficiently used to produce high yield and thus less N is wasted. Vice versa, also low N inputs can be associated with quite high NLI due to low yields and relatively high N loses compared to N inputs. For example, the variation of NLI in Africa is between 6 and 45 kg N t⁻¹ despite very small N inputs. For rice cultivation, predicted NLI were around 10–40 kg N t^{-1} (Fig. 3b). China has NLI about 30 kg N t⁻¹, while India around 20 kg N t⁻¹. For wheat cultivation, NLI is higher compared with the other two crops, around 10-50 kg N t⁻¹ (Fig. 3c). Among the major wheat producers, India has the highest NLI, followed by China, the USA, and Russia.

3.3. Mitigation scenarios

Based on the FixN3E mitigation scenario, which evenly distributes the globally available N fertilizers, the global production of maize, rice, and wheat could increase by 29, 62, and 45 Tg yr^{-1} without any additional N inputs (Table 3). At the same time, it would lead to significant decreases in total N_t and N_w, especially for rice and wheat cultivations according to our simulations. Maize yields and environmental N losses are predicted to decline in some European countries and the USA, while only Nt but not yield would decrease in China (Fig. 4a). Large increases in maize yields but also in N losses would be expected in South America and Africa (Table 3). Some countries such as Brazil would be expected to show a substantial increase in yield with only a negligible increase Nt compared to a large increase in Nin (from 75 to 122 kg N ha^{-1}). For rice, a major decrease in N losses would be expected for China, while yield could be maintained at today's level (Fig. 4b). For wheat, China would see a large decrease in Nt with a small decrease in yield, while India would only experience a decrease in Nt (Fig. 4c). In the contrary, Russia shows a high potential to increase yield (about 1 t ha⁻¹) with a minor increase in N_t.

In the AutoN mitigation scenario, crop yields would not increase significantly (at least for maize and rice), but as much less N inputs would be needed (Figs. 4d–f), it would result in a significant reduction in global N losses with more efficient N fertilization (Table 3). For maize and rice cultivation, China shows a dominant decrease in N_t (Figs. 4d and e). For wheat, China and India would see large decreases in N_t, while many countries in Europe would decrease N_t and simultaneously increase wheat yields (Fig. 4f).

Compared with the FixN3E scenario, the impacts on yields are similar for maize and rice cultivations by adopting the AutoNE mitigation scenario with more efficient N management (Figs. 4g and h). Further decreases of N_t in China and the USA from maize cultivation and in China from rice cultivation are predicted. As for wheat, the influence is more positive with more countries moving into a condition where increasing yields and decreasing average N_t happen simultaneously, especially in Europe (Fig. 4i). Besides, the increases of N_t are less significant in the rising N losses countries under the FixN3E scenario; while the decreases of N_t in China and India would be further enhanced.

3.4. Uncertainties

Generally, the range of uncertainty for N_t derived from parameters is relatively small for maize, rice, and wheat either globally, continentally, or for the top 10 producing countries (Fig. 5). The results calculated by using model default parameters are very close

Table 3

Differences of total nitrogen inputs $(TN_{in}, in Gg N yr^{-1})$, total nitrogen losses into the environment $(TN_{in}, in Gg N yr^{-1})$ and into water $(TN_{w}, in Gg N yr^{-1})$, and crop production (P, in Tg yr^{-1}) between mitigation scenarios and base scenario.

Region	Scenarios	Maize				Rice				Wheat				
		TN _{in}	TNt	TNw	Р	TN _{in}	TNt	TNw	Р	TN _{in}	TNt	TNw	Р	
Global	FixN3E	0	-411	-627	29	0	-1586	-1892	62	0	-1723	-2158	45	
	AutoN	-1625	-1898	-1418	7	-2045	-2011	-1708	3	-4008	-4432	-4015	42	
	AutoNE	-1134	-2182	-1801	46	-1026	-2505	-2397	64	-2471	-5351	-5148	95	
Africa	FixN3E	1675	959	679	19	628	353	299	11	199	28	-2	4	
	AutoN	-152	-100	-62	1	-24	-12	-2	0	-113	- 78	-63	0	
	AutoNE	1269	555	357	20	558	290	253	11	161	- 38	-46	5	
Asia	FixN3E	-1120	-992	-989	6	-957	-2080	-2290	42	-2521	-2177	-2158	4	
	AutoN	-1210	-1123	-966	1	-2020	-1986	-1709	3	-3795	-3096	-2950	7	
	AutoNE	-1473	-1431	-1247	8	-1895	-2886	-2721	42	-4822	-4357	-4091	17	
Europe	FixN3E	448	167	69	-1	36	18	13	1	804	139	-32	12	
*	AutoN	-88	-213	-139	2	0	-3	-1	0	-26	-780	-674	23	
	AutoNE	195	-234	-238	2	31	9	7	1	764	-694	-704	36	
N. America	FixN3E	-1901	-913	-629	-21	-42	-33	-27	0	628	108	-14	9	
	AutoN	-148	- 385	-205	1	-1	-1	6	0	-70	-374	-273	9	
	AutoNE	-1955	-1263	-796	-12	-48	-43	- 32	0	558	-274	-269	18	
Oceania	FixN3E	4	0	-1	0	16	7	4	0	534	134	41	7	
	AutoN	0	-1	0	0	0	0	0	0	-2	-57	-25	2	
	AutoNE	3	-1	-1	0	14	5	3	0	513	31	-11	10	
S. America	FixN3E	893	369	244	25	319	149	109	8	356	44	8	8	
	AutoN	-26	-77	-45	2	1	-9	-1	0	-1	-46	-30	1	
	AutoNE	827	191	124	27	314	119	93	8	355	-19	-26	10	

to the median values for all the three crops, which, in another way and to some degree, reflect the reasonable estimations of N losses by using the constructed simulation framework in this study. Furthermore, differences in N losses between simulations comparing the three fertilization schedules, i.e. FixN1, FixN2, and FixN3, were also small (Fig. 5).

4. Discussion

4.1. Comparisons with other studies

We compared our results with previous studies, which investigated large-scale N losses from the whole agricultural sector, to check the reliability of our simulations. The ratio of N_t to N_{in} ranged between 0.76 and 0.85 in such studies (Bouwman et al., 2009; Bouwman et al., 2013; Liu et al., 2010; Mekonnen and Hoekstra, 2015; Sutton et al., 2013) (Table 4). The ratio found in our study is just slightly below this range. No legume crops were considered in this study as other studies did; this might contribute to the slight difference. Generally, even though much less soil N excess over crop plant demand can be expected to occur in legume crops compared to the major cereal crops investigating here, some N losses do also occur from legume crop cultivations. On

the other hand, the ratio of N_w to N_{in} is 0.49 in our study, which is within the range of 0.22–0.55 as reported by these previous studies. The reported variation of N_w/N_t is quite high (0.26–0.70), implying the large uncertainties to separate N_w and N losses into atmosphere from $N_t.$ Our estimation is also within this range. These comparisons thus indicate that our simulations produced plausible results.

It should be noted that the difference between 1 and N_t/N_{in} in Table 4 cannot be directly compared with the previously reported NUE, e.g. in Lassaletta et al. (2014) and Zhang et al. (2015). This is because N_{in} in this table only includes N_{fer} and N_{man} , while NUE was generally estimated by considering the whole N inputs (including N_{fer} , N_{man} , N_{dep} , N_{dec} , etc.). Another reason is that N_t estimated in this study also takes into account the changes of soil N stock, whereas the calculation of NUE only focuses on crop N_{up} .

4.2. Environmental impacts associated with high nitrogen losses

Nitrogen losses are associated with significant environmental impacts, especially in the high emissions regions (Erisman et al., 2013). Our study identified four hotspots of N losses: China, India, eastern parts of the USA, and Central Europe (Fig. 1). Considerable environmental consequences have already been detected in these regions. Because



Fig. 4. Differences of yields and nitrogen losses into the environment (N_t) between different mitigation scenarios and base scenario for maize (a, d, g), rice (b, e, h), and wheat (c, f, i). Countries with the smallest areas (for a total of 1% of global total cropland areas of each crop) are discarded; sizes represent cropland areas for each country; colours represent different continents.



Fig. 5. Uncertainties of nitrogen losses into the environment (N_t) globally, continentally, and for the top 10 producing countries of maize (a), rice (b), and wheat (c). Lines from top to bottom are 95th, 75th, 50th, 25th, and 5th percentiles, respectively. Red cycles represent results with default parameters by using FixN3 fertilization schedule; green pluses represent results with default parameters by using FixN1 fertilization schedule. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of much higher N inputs than crop demands, China has experienced significant N deposition enhancement (Liu et al., 2013b), which has led to substantial soil acidification in major croplands (Guo et al., 2010). It was reported that N imbalance also resulted in significant nitrate accumulation in Chinese croplands, even at soil depth bellow 4 m (Zhou et al., 2016). Downstream, large amounts of N have been discharged into coastal water bodies and caused severe eutrophication (Tong et al., 2015). Eastern parts of the USA and India experienced similar N deposition patterns as China (Erisman et al., 2013). Furthermore, Diaz and Rosenberg (2008) identified 415 eutrophic and hypoxic coastal water systems around the world mostly located in the northern Gulf of Mexico, Chesapeake Bay, and Baltic and North Seas. The eutrophic conditions in these regions were mainly due to N losses in the Mississippi river basin and respective catchment in the eastern parts of the USA and central Europe. These regions were detected in our study as high N loss regions.

4.3. Assessment of nitrogen loss conditions

Generally, our results show three patterns of N loss conditions: a) high N inputs associated with high N losses due to N overuse, thus decreasing N_{in} is required; b) low N inputs associated with low yields due to insufficient availability of N inputs, thus increasing N_{in} is needed but appropriate fertilization management should be considered when intensifying these croplands; c) high yields associated with low NLI, indicating that the right amount of N inputs is applied. We will focus on several major crop producers to discuss these patterns below.

China showed the largest overuse of N for all the three crops, especially for rice cultivation (Fig. 4). Decreasing its N inputs for the three crops to the respective world averages would not affect yields, except for a small decrease in wheat yield. This action in China alone would avoid about 7.5 Tg N yr⁻¹ of environmental N losses from the three crops. This finding is consistent with Mueller et al. (2012), who also found that China has a particularly high potential to reduce N_{in}. Similar as China, also India shows a high potential to decrease N inputs for wheat cultivation and thus to reduce environmental N losses.

Many countries in Africa and South America, on the other hand, should increase N inputs to increase yields. In our simulations, food production in Africa increased by between 30 and 65% when N inputs were increased to the world average (Table 3). But also N losses increased. Therefore, appropriate N fertilization is needed when increasing N inputs. For example, yield was predicted to increase by 1.0 t ha⁻¹ and N_t by 21 kg N ha⁻¹ in the FixN3E scenario for maize production in Brazil, while yield increased by 1.2 t ha⁻¹ and N_t by only 11 kg N ha⁻¹ in the AutoNE scenario (Figs. 4a and g). Similarly in Russia for wheat cultivation, an increase of yield rising from 1.1 to 1.2 t ha⁻¹ is obtained with increase of N_t declining from 27 to 16 kg N ha⁻¹ when N fertilization scenario changes from FixN3E to AutoNE (Figs. 4c and i).

In the USA, maize cultivation belongs to the third pattern of N loss conditions (Fig. 4). The current average N application rate in the USA in maize cultivation is 178 kg N ha⁻¹. In the AutoN scenario, we identified a value of 174 kg N ha⁻¹, which is only slightly lower than the actual value. In addition, N_t differs only slightly between FixN3 and AutoN. This suggests that in average the N input amounts applied in the USA in maize cultivation are just matching the demand of the crop. This may also partially explain why we found much better performance for the USA in maize cultivation than for China in terms of NLI (Fig. 3). Although the USA and China are the major N loss contributors of maize cultivation, with appropriate N fertilization and higher yields, the NLI obtained for the USA was only 46% of that for China.

4.4. Current progress

DUE to limitations in available input data, our results relate to the situation around the year 2000. How this situation has developed since the year 2000 can be inferred from the trend in global total N fertilizer consumption. Generally, global total N consumption increased by 25% from 2002 to 2013 (Fig. S5), implying an increase in total N losses. In particular, total N fertilizer consumption in India increased by >60% in this period. Considering that N losses were already high in 2000 according to our simulation, the need to reduce N pollution appears to be even more urgent today. During the same period, the total N fertilizer consumption in China has only increased by 10%. This is mainly related to China's effort to find ways to improve its nutrient management. For example, an integrated soil-crop system management was introduced in China to produce more grains with less fertilizer use at lower environmental costs (Chen et al., 2014; Chen et al., 2011; Ju et al., 2009). The success achieved in China may be extended to other N overuse countries and countries planning to increase N fertilizer inputs (Zhang et al., 2013). Total N consumption in Brazil and Russia, which showed high potential to improve yields by increasing N inputs in our study, almost doubled in this period. Similar increasing trends were also observed in some other insufficient-N countries, e.g. Nigeria, Paraguay, Ukraine, etc. In Kenya, for example, maize yield almost tripled from 0.8 ton ha^{-1} in 2005 to 2.2 ton ha^{-1} in 2007, after farmers were subsidized to buy 100 kg fertilizer per farm in 2005 (Sanchez, 2010). With the development of better N management under the pressure to give more attentions to environmental protection, N inputs decreased in many western European countries (Fig. S5), and N losses were considerably reduced without compromising yields (Sutton et al., 2011; Velthof et al., 2014). However, their potential to reduce N_{in} without yield losses is not fully exhausted yet. For example, Van Grinsven et al. (2013) performed a cost-benefit assessment and concluded that N application could be further lowered on average by around 50 kg N ha⁻¹ in northwestern Europe.

4.5. Limitations

Because of the unavailability of data on fertilizer application timing, assumptions have been made for the fertilizer application schedules in the model simulation. As we do not know how well these assumptions match reality, there will be some errors in the results, although the overall differences of N_t among the three N fertilization schedules are small. We only considered uncertainties derived from the possible ranges of model parameters. The impacts of cross- and spatial-corrections of these model parameters could also be important (Kros et al., 2012). Besides, uncertainties in other management practices (e.g. planting and harvesting dates, residue management, tillage, etc.), N inputs, and soil inputs were not addressed. These factors may also play important roles in N fluxes (Molina-Herrera et al., 2016). Estimating these uncertainties is out of the scope in this study, but should be the subject of future studies.

5. Conclusions

In this study, we applied a spatially explicit crop model, PEPIC, to quantify N losses and to explore the trade-offs with yields from three major cereal crop cultivations, i.e. maize, rice, and wheat, on a global scale with high spatial resolution. Without requiring yields as input data, this method can be used to determine the N losses and yields relations under different N mitigation scenarios, which is the major advantage of large-scale crop modelling for assessing N losses compared to empirical mass balances.

Table 4

Comparison of estimated global total nitrogen losses into the environment $(TN_{tr} in Tg N yr^{-1})$ and water $(TN_{wr} in Tg N yr^{-1})$, and ratio of total nitrogen losses to total nitrogen inputs $(TN_{inr} in Tg N yr^{-1})$ with results from previous studies.

	Bouwman et al. (2009)	Bouwman et al. (2013)	Liu et al. (2010)	Mekonnen and Hoekstra (2015)	Sutton et al. (2013)	Current study
Time period	2000	2000	2000	2002-2010	2000-2010	2000
TN _{in} (Tg N yr ⁻¹) ^a	184	175	85	134	177	59
TN _t (Tg N yr ⁻¹)	157	138	67	109	135	44
TN _w (Tg N yr ⁻¹)	41	57	47	53	95	29
TN _t /TN _{in}	0.85	0.79	0.79	0.81	0.76	0.74
TNw/TNin	0.22	0.33	0.55	0.40	0.54	0.49
TN _w /TN _t	0.26	0.41	0.70	0.49	0.70	0.66

^a Only nitrogen inputs from fertilizer and manure are considered here.

Global total N losses were 44 Tg N yr $^{-1}$ for the three crops, with rice contributing the most. These losses were concentrated in a few regions, for example in China and the USA for maize cultivation, and China and India for rice and wheat cultivations. This concentration of N losses calls for more attention to N management in these countries. With the simultaneous consideration of N losses and yields, we were able to assess trade-offs between them using an N loss index: NLI. The NLI showed high variations among different countries, indicating diverse performance in terms of N losses associated with the production of the same amount of yield. This variation suggests that there is still considerable potential to improve the efficiency of N use in cereal production in many countries without compromising yields. The analysis of mitigation scenarios also shows that N losses can be significantly reduced and yields at the same time increased by transferring N from currently high application countries to countries with low application. Furthermore, there is also still much potential to increase yields by using more efficient N fertilization schemes in low N application countries. The findings of this study are useful for policy makers to guide better N management and reduce N emissions.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx. doi.org/10.1016/j.scitotenv.2016.08.093.

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