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PAPER

Assessment of nitrogen hotspots induced by cropping systems in the Bohai Rim region in China by integrating DNDC modelling and the reactive nitrogen spatial intensity (NrSI) framework

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Keywords: cropping systems, DNDC, reactive nitrogen, spatial intensity, Bohai Rim region

Supplementary material for this article is available online

Abstract

More than half of nitrogen (N) inputs to cropland are lost to the environment via denitrification, ammonia (NH_3) volatilization, nitrate leaching and surface runoff. Cropping systems are, therefore, a large contributor to reactive N (Nr, all species of N except N_2) losses. The Nr spatial intensity (NrSI) framework was developed to quantify the environmental burdens due to Nr losses on a per area basis. However, the current application of the NrSI framework is limited by the development of virtual N factors (VNFs, Nr released to the environment per unit of Nr consumed) for agricultural products and it could not differentiate pathways of Nr losses linked to consequences in various environmental media. As the Denitrification-Decomposition (DNDC) model is capable of tracking N fluxes across cropping systems and regions, we integrated the DNDC model and the NrSI framework to identify hotspots of Nr losses induced by cropping systems, and illustrate the approach with a case study for the Bohai Rim region (BR) in China. Altogether 29 types of cropping systems (i.e. 16 mono, 10 double and 3 triple cropping systems) in 429 counties were simulated for the N balance, Nr losses and the NrSI associated with crop production. Regarding the total Nr losses in the BR, 45% of the total N input was lost to the environment during crop production with NH_3 volatilization and nitrate leaching the two main pathways, making up 24% and 19% of the total N input, respectively. Shandong province was the biggest contributor of the total Nr losses (45.6%) among regions, and winter wheat-summer maize, triple vegetable and spring maize cropping systems were the top three contributors among various cropping systems. For Nr loss hotspots, there are substantial variations of NrSI across cropping systems (41–1024 kg N ha^{-1} y^{-1}) and counties (28–4782 kg N ha^{-1} y^{-1}). Beijing had the highest NrSI associated with crop production (307 kg N ha^{-1} y^{-1}) among regions, and vegetable systems had the highest NrSI of 355 kg N ha^{-1} y^{-1} among cropping systems. The application of this integrated method is useful to identify areas and/or cropping systems with particularly high Nr losses and NrSI to provide basic information for setting Nr mitigation priorities on a wide range of regions and cropping systems.

1. Introduction

Nitrogen (N) is an essential nutrient for plant growth, and its application in cropping systems has boosted crop yield to feed almost half of the world's population (Erisman *et al* 2008 , Galloway *et al* 2008). However, more than half of N inputs to cropland are lost to the environment via denitrification, ammonia (NH_3) volatilization, nitrate leaching and surface runoff (Erisman *et al* 2013, Lassaletta *et al* 2014,

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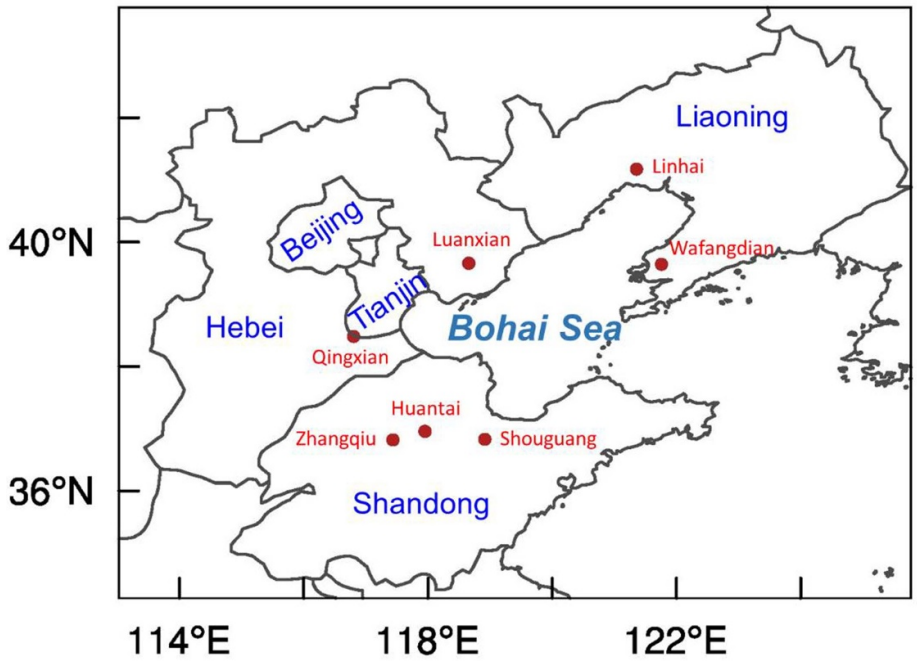

Figure 1. Map of the BR and the locations of seven field experiment sites.

Zhang *et al* 2015), making cropping systems a large contributor to reactive N (Nr, all species of N except N_2) losses (Rockstrom *et al* 2009, Bodirsky *et al* 2014). Increasing losses of Nr could result in a cascade of impacts on the environment and human health. For instance, NH_3 reacts readily with acid gases and aerosols to form particulate matter in the atmosphere and nitrate pollution of both groundwater and surface water risks human health and can lead to eutrophication (Erisman *et al* 2013, Galloway and Leach 2016).

To quantify the environmental burdens due to Nr losses, the Nr spatial intensity (NrSI) framework was developed to map the geographical locations of anthropogenic Nr losses for both agricultural and settled systems to inform management decisions and help mitigate Nr pollution for sustainable development (Liang *et al* 2018). For the calculations of NrSI of agricultural systems, a set of nation-/region-specific virtual N factors (VNFs, defined as the Nr released to the environment per unit of Nr consumed (Leach *et al* 2012)) for various food products are needed. Thus, it is not applicable to those countries/regions without VNFs. The current NrSI framework does not differentiate pathways of Nr losses, which should be considered because the loss route and form (e.g. nitrous oxide (N_2O) and NH_3 to the atmosphere, ammonium (NH_4^+) and nitrate to the ground and surface water) determine their environmental consequences (Erisman *et al* 2013). Therefore, it is necessary to develop an alternative approach to fill these gaps.

The primary objective of this study is to develop a new approach by integrating the Denitrification-Decomposition (DNDC) model and the NrSI framework to identify hotspots of Nr losses induced by various cropping systems and through different pathways on regional scale. One advantage of using the DNDC model to more accurately estimate the multiple pathways of Nr losses is that it can simulate the N biogeochemistry in agroecosystems with the consideration of effects of local soil properties, meteorological conditions and conventional farming practices (Li 2009). In contrast, previous studies mainly relied on N input/activity data, transformation/partitioning coefficients and emission factors (Guo *et al* 2012a, Wang *et al* 2018). For example, they used N loss factors for nitrate leaching, NH_3 and N_2O emissions during crop production to determine Nr losses, which may introduce uncertainties due to spatial variations among different agricultural regions. As the DNDC model is capable of tracking soil N dynamics, N gas fluxes and nitrate leaching across crops and their rotation regimes, it can help to identify Nr hotspots among various cropping systems within a specific region. In this study, we selected the Bohai Rim region (BR), a typical intensive crop production base in China, to illustrate the application of this new approach for identifying areas and/or cropping systems with particularly high Nr losses and NrSI to provide basic information for setting Nr mitigation priorities on a wide range of regions and cropping systems.

2. Materials and methods

2.1. Bohai Rim region

The BR is located in north China around the Bohai Sea and includes two municipalities (Beijing and Tianjin) and three provinces (Hebei, Liaoning and Shandong) (figure 1). It covers an area of 520 000 km^2 (about 5.5% of the total land area in China) and is home to over 234 million people (amounting to 17.7% of the nation's total population). With ~15.3% of the total arable land in China, the BR is an important crop production base. The production of wheat, maize, peanut, cotton and vegetables made up 29.8%, 28.3%, 36.7%, 24.9% and 31.0% of the national total production in 2008 (the year of interest for this study) (NBSC 2009). Such a high productivity level in the BR relies on the intensive use of synthetic N fertilizer (Cui *et al* 2008), leading to high Nr losses and environmental risks (Yu and Mao 2002, Liu *et al* 2011, Huang *et al* 2013). The annual mean concentration of atmospheric NH_3 at agricultural sites in the BR was around 10 μg N m^{-3} in 2009, while the regional background NH_3 concentrations in China were 1.5–3.4 μg N m^{-3} (Shen *et al* 2009, Meng *et al* 2010, Liu *et al* 2011). High nitrate concentrations in the shallow groundwater were also observed in the BR, which is associated with the nitrate leaching from intensive cropping systems (Ju *et al* 2006). As reported by the Marine Environment Quality Bulletin of Bohai Sea (SOA 2009), the coastal waters in BR were also heavily contaminated, with inorganic N being one of the most important pollutants, which is largely derived from crop production (Huang *et al* 2013).

2.2. Model description and validation

The DNDC model is a process-oriented model that simulates carbon (C) and N biogeochemistry in agroecosystems (Li 2009). It was originally developed to model N_2O, carbon dioxide (CO_2) and N_2 emissions from agricultural soils in the United States (Li *et al* 1992a, 1992b), and has been applied and expanded by researchers worldwide in a range of countries and cropping systems (Giltrap *et al* 2010). The model is comprised of two components with six modules. The first component, containing the soil climate, crop growth and decomposition modules, simulates environmental variables such as temperature, moisture, pH, redox potential (Eh) and substrate concentration profiles driven by primary ecological factors. The second component, including the nitrification, denitrification and fermentation modules, simulates the biogeochemical production, consumption and emissions of CO_2, methane (CH_4), NH_3, nitric oxide (NO), N_2O as well as nitrate leaching from soil, driven by the variables generated by the first component. The detailed information of DNDC model can be found in Li *et al* (1992a, 1992b, 2004, 2006). In the past two decades, the DNDC model has been tested with observations from numerous field measurements of crop yields, soil physical and chemical conditions, soil C/N dynamics, C/N gas fluxes, and nitrate leaching across the major cropping systems in China, and applied successfully to conduct filed and regional simulations (Xie *et al* 2017). All the crop parameters used for the DNDC model were recalibrated for major crop varieties grown in China (Li *et al* 2017a).

To validate the DNDC model for cropping systems in the BR, field experiments were conducted during 2008 to 2009 for the seven typical cropping systems (Qiu *et al* 2012), i.e. winter wheat-open-field vegetable in Zhangqiu county, winter wheat-summer maize in Huantai county, greenhouse vegetables in Shouguang county, winter wheat-summer maize in Qingxian county, spring maize in Luanxian, Linghai and Wafangdian counties (figure 1, table S1). Specifically, the simulated soil temperature and moisture, daily net ecosystem exchanges of CO_2 and N_2O flux, crop yields, inorganic N content and nitrate leaching were validated against the observations of major cropping systems in the BR, and good agreements were found with statistically significant correlations (figures S1–S3) (available online at https://stacks.iop.org/ERL/15/105008/mmedia). The detailed information of model validation and simulation performance refer to previous work (Qiu *et al* 2012). Therefore, we assumed that the DNDC model would provide reliable simulation results to further analyze Nr losses and NrSI associated with crop production in the BR.

2.3. Input data for DNDC regional simulations

To conduct the DNDC simulation on regional scale, the region should be divided into polygons or grid cells with all the input data compiled in a Geographic Information System (GIS) database accompanied by a climate library for the target region. The GIS files include spatially differentiated information of location, climate file ID, soil properties, cropping systems and their areas, and farming management practices for each polygon/grid cell. The climate library contains the daily weather data (i.e. maximum and minimum air temperature, precipitation) (Li 2009). In this study, county was selected as the basic unit for the simulation, and the climate and soil conditions within one county were assumed uniform.

Input data for the DNDC model included: **1) meteorological variables**, including daily maximum and minimum air temperature, and daily precipitation. They were obtained from 82 weather stations of China Meteorological Administration (http://data.cma.cn/en). For those counties without national weather stations, they shared the meteorological data from the nearest one. **2) soil properties**, such as soil bulk density, clay fraction, soil organic C content and pH were derived from the Second National Soil Survey of China, and China Soil Records

(The National Soil Survey Office 1993). **3) land use by crops**, i.e. cropping systems and their areas. The county-level data of arable land, planting areas for all crops and their productivity were from Ministry of Agriculture and Rural Affairs, China (table S2). To determine the areas for each cropping system (table S3), we reanalyzed the original crop area and merged the census data with a prioritized list of most possible cropping systems on county scale in the BR (table S4) (Qiu *et al* 2003, Li *et al* 2017a). **4) farming management practices**, e.g. sowing and harvest date, fertilization application time, irrigation and tilling method. These were obtained primarily from field surveys. When conducting the field survey, the BR was divided into seven sub-regions based on variations in planting regimes, soil types and climate conditions etc (table S5). Typical counties were selected to conduct the field surveys and represent each sub-region. **5) crop physiological/phenology parameters**, i.e. maximum yield, biomass partitioning into and C/N ratios of grain, shoot and root, cumulative thermal degree days to maturity (TDD), water requirement, and N fixation index (see SI for detailed information). The crop parameter database was developed for 23 major crops in China (Li *et al* 2017a). **6) fertilizer and livestock data**. The amount of fertilizer consumption and number of different types of livestock in each county were obtained from the Ministry of Agriculture and Rural Affairs, China. The livestock category considered in this study included beef cattle, pig and sheep based on the data availability on county level. We then determined the amount of manure produced in each county based on animal excreta parameter and associated N content (Wang *et al* 2006), and assumed that in each county, 20% of the manure produced was evenly applied to the cropland (Qiu *et al* 2008), i.e. each county had the same N input from manure application on a per area basis (equation S1). The DNDC model also included N input via atmospheric N deposition and mineralization, and from crop residue (the straw return ratios were set between 25%–60% in the BR (table S6)) (see SI for more detailed information). Input files for model simulation and main parameters in each file were provided in the SI (table S7).

Based on the above input data, the DNDC simulations were run for each cropping system in each county. Altogether, 29 types of cropping systems (figure 4) in 429 counties were simulated for the year 2008. Winter wheat-summer maize, triple vegetable and spring maize were the major cropping systems in the BR. The three major crops (wheat, maize and vegetable) made up 74.2% of the total planting area and 92.1% of the total crop productivity in the BR in 2008 (table S2). The year 2008 was selected for approach illustration because there were sufficient field measurements for model validation, comprehensive field surveys of farming practices for model input and high quality of the county-level statistic datasets. Synthetic N fertilizer is the key driver of Nr losses (NH_3 volatilization, nitrate leaching and N_2O emission) (Bouwman *et al* 2002, Behera *et al* 2013, Li *et al* 2014), and the crop planting area, total synthetic N input, and per area synthetic N application rate did not change much from 2008 to 2015 in the BR (tables S8–10). Therefore, the 2008 results from this study were useful to identify historical Nr hotspots and to compare with current hotspot after the implementation of the national 'Zero-growth Action Plan' for synthetic fertilizer use since 2015 (MARA 2015).

2.4. Calculations of NrSI associated with crop production

The NrSI estimates the intensity of the Nr losses on a per area basis, expressed in the unit of kg N ha^{-1} y^{-1}. In this study, NrSI associated with crop production was calculated based on the DNDC simulation results for each county/province and cropping system, and Nr losses via four pathways were considered during crop production—N_2O emission, NO emission, NH_3 volatilization and nitrate leaching. The NrSI associated with crop production for a specific county/-province i and for a specific cropping system j was calculated as follows:

$$NrSI_i = \left(\sum_{j=1}^{n} Nr_{ij}\right) \times \left(\sum_{j=1}^{n} S_{ij}\right)^{-1}$$

$$NrSI_j = \left(\sum_{i=1}^{m} Nr_{ji}\right) \times \left(\sum_{i=1}^{m} S_{ji}\right)^{-1}$$

where Nr_{ij} and Nr_{ji} is the Nr losses from cropping system j in county i, in the unit of kg N; S_{ij} and S_{ji} is the planting area for cropping system j in county i, in the unit of ha; m is the total number of counties in the BR; n is the total types of cropping system in the BR.

3. Results and discussion

3.1. N balance in the BR

In 2008, the total N input (N from atmospheric deposition, fixation, mineralization, synthetic fertilizer, animal manure and crop residues) into the croplands was 8.1 Tg N y^{-1} in the BR (tables S11 and S13). Synthetic N fertilizer was the largest N input to the croplands, amounting to 5.1 Tg N y^{-1}, which accounted for 22% of the national total input (NBSC 2009). The average application rate of synthetic N fertilizer for cropland in the BR was 311 kg N ha^{-1} y^{-1} (table 1), significantly higher than the national average of 233 kg N ha^{-1} y^{-1} (Yan *et al* 2014b). N from organic fertilizer (including animal manure and crop residues) was the second largest N input to the croplands (about 2.4 Tg N y^{-1}) with a mean application

Table 1. Cropland N balance on a per area basis in the BR in 2008 (Unit: kg N ha^{-1} y^{-1}).

	This Study						Guo *et al* 2012a
Region	Beijing	Tianjin	Hebei	Liaoning	Shandong	The BR	The BR
Input	658.6	447.5	468.9	418.8	553.5	497.6	606.2
Synthetic fertilizer	369.1	282.0	287.9	254.6	353.4	310.6	481.8
Manure	196.8	97.8	106.4	114.1	116.3	113.1	34.3
Crop residue	45.4	33.6	34.4	10.7	40.3	32.4	23.5
N deposition	10.3	12.1	14.3	6.5	13.5	12.3	28.0
N fixation	7.0	4.5	4.8	8.7	3.9	5.2	21.7
N mineralization	29.9	17.6	21.1	24.2	26.2	23.9	–
Others (i.e. seeds, irrigation water)	–	–	–	–	–	–	17.0
Output	522.1	361.0	377.1	346.1	439.9	399.7	337.4
N uptake by crops[a]	204.4	168.6	161.3	102.3	192.9	164.0	176.7
Nitrate leaching[b]	147.4	72.2	81.7	130.1	85.1	93.4	38.2
NH_3 volatilization	151.4	110.6	116.6	79.4	140.3	119.8	113.8
N_2O emission	5.1	0.8	4.3	16.7	3.9	6.5	
NO emission	2.8	1.8	3.0	2.9	4.9	3.8	8.7
N_2 emission	11.0	7.0	10.2	14.8	12.7	12.2	
Net N balance	136.5	86.5	91.8	72.7	113.6	97.9	268.9[c]

[a]N uptake by crops refers to N uptake by whole crop plant.
[b]Nitrate leaching in this study refers to nitrate leached out of a 0–50 cm soil profile.
[c]This value was calculated based on information provided in Guo *et al* (2012b).

rate of 145 kg N ha^{-1} y^{-1}, which is nearly three times as much as the national average (~50 kg N ha^{-1} y^{-1}) (Yan *et al* 2014b). Atmospheric deposition, biological fixation and mineralization made up the rest N inputs of 0.7 Tg N y^{-1}. The total N output (crop uptake, gas emissions via N_2O, NO, N_2, NH_3, and nitrate leaching) was 6.5 Tg N y^{-1}, resulting in a N surplus of 1.6 Tg N y^{-1} in the cropland in the BR. Among the N output, about 2.7 Tg N y^{-1} was taken up by crops, accounting for 33% of the total N input. A large amount of Nr (45% of the total N input) was lost to the environment during crop production with NH_3 volatilization and nitrate leaching the two main pathways, making up 24% and 19% of the total N input, respectively (table S11). The NH_3 volatilization in the BR amounted to 2.0 Tg N y^{-1} with an average emission intensity of 120 kg N ha^{-1} y^{-1} for croplands, which contributed 24% of the national total NH_3 emitted during crop production (Xu *et al* 2016). Nitrate leaching totaled 1.5 Tg N y^{-1} with an average intensity of 93 kg N ha^{-1} y^{-1}. N output via other gas emissions (i.e. N_2O, NO and N_2) was about 0.4 Tg N y^{-1}. Notably, the total amount of N_2O emission in the BR accounted for 22% of the national total N_2O emission during crop production (Zhou *et al* 2014) with a mean emission intensity of 6.5 kg N ha^{-1} y^{-1} (table 1).

The total amount of Nr losses associated with crop production in the BR was 3.6 Tg N y^{-1} with an area of 16.3 million ha of croplands (table S12). Among the four Nr loss pathways, NH_3 volatilization and nitrate leaching were dominant forms, accounting for 53.6% and 41.8%, respectively; while N_2O and NO emission only made up 2.9% and 1.7% of the total on-farm Nr losses. The highest Nr losses were in Shandong province with a proportion of 45.6% to the total Nr losses in the BR, followed by Hebei province (30.6%) and Liaoning province (20.4%). Beijing and Tianjin had a similar share with a percentage of 1.6% and 1.8%, respectively. For the different cropping systems, 60.5% of the total Nr losses were derived from three major contributors, i.e. winter wheat-summer maize double cropping system (29.4%), triple vegetable cropping system (17.3%) and spring maize mono cropping system (13.8%) (table S13), with planted area accounted for 31.9%, 3.8% and 16.3% to the total cropland in the BR, respectively (table S3).

Overfertilization and poor nutrient management practices are the major reasons for the low N use efficiency and high Nr losses in China (Gu *et al* 2015, Wang *et al* 2018), especially for the regions with intensive agricultural activities. In Shandong province, the average N application rate was 470 kg N ha^{-1} in 2008 (table 1), which was more than doubled the threshold of safe N fertilization rate (225 kg N ha^{-1}) set by developed counties to avoid soil and water pollution (Cai *et al* 2018). The overfertilization of N is even more common in greenhouse vegetable production. The results show the N application rate for triple vegetable system averaged 1360 kg N ha^{-1} y^{-1} in the BR in 2008, much higher than that required by vegetables.

The per area based N balance in the BR of our results was compared to a previous study that used a farmland N balance model (Guo *et al* 2012a) (table 1). Results from both studies showed high N input, Nr losses and N surplus in the cropland in the BR. Per area based total N input and synthetic N fertilizer application rate were lower in our study compared with the results of Guo *et al* (2012b) due primarily to the difference in data source. We applied the most reliable county-level dataset obtained

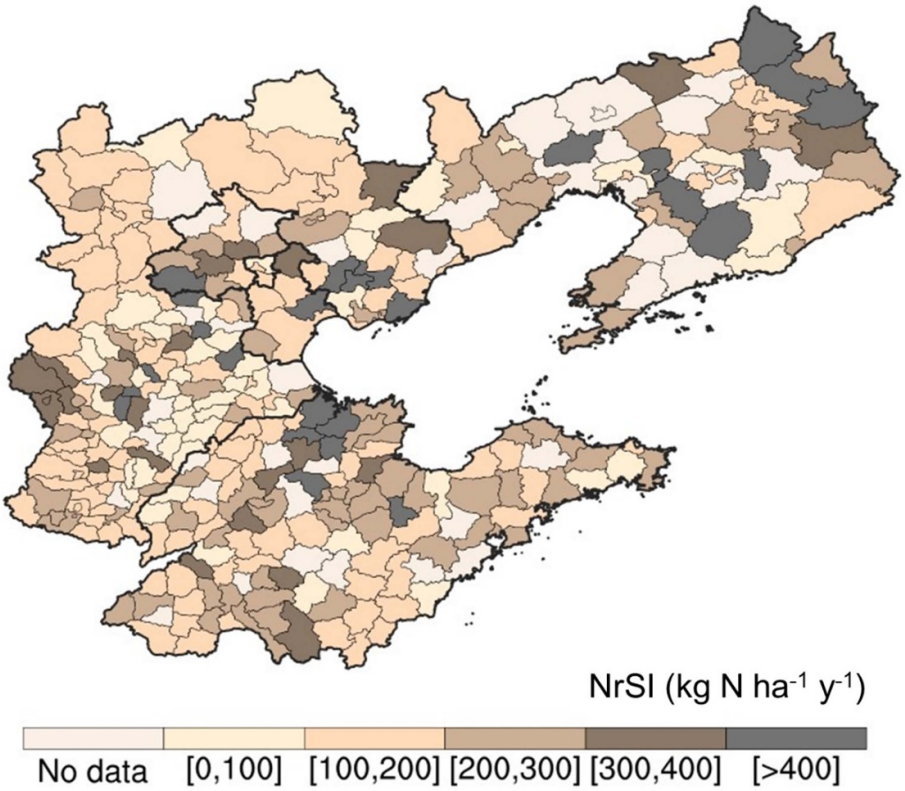

Figure 2. Spatial distribution of NrSI associated with crop production in the BR in 2008. For all 429 counties calculated in this study, 363 counties' cropland NrSI exceeded 96 kg N ha^{-1} y^{-1} (threshold for Nr hotspots in China defined by Wang *et al* (2018)).

from the Ministry of Agriculture and Rural Affairs, China. The total amount of synthetic N fertilizer used in the BR in 2008 was 5.1 million tons in this study. While it was reported to be 9.1 million tons in Guo *et al* (2012b), which is more than doubled that recorded on provincial level by the National Bureau of Statistics that reported a value of 4.1 million tons. Different estimation methods could be another possible reason for the differences in the results. Our estimated N taken up by crops, and NH_3 volatilization are comparable to those by Guo *et al* (2012b), but we found higher nitrate leaching and lower N surplus rates. A likely explanation is that the DNDC model simulated the nitrate leached out of a 0–50 cm soil profile in this study, so nitrate deeper in the soil profile was included in the Nr loss via leaching rather than N surplus in the soil.

3.2. Spatial variations of cropland NrSI

The average cropland NrSI in the BR was 224 kg N ha^{-1} y^{-1}. Although contributing the least to the total cropland Nr losses in the BR, Beijing had the highest NrSI of 307 kg N ha^{-1} y^{-1} due to its relatively small cropland area (the least among the five provinces) but high proportion of vegetable area (~17% of the total planting area in Beijing). Tianjin had the lowest NrSI of 185 kg N ha^{-1} y^{-1}. Hebei, Liaoning, and Shandong had similar cropland NrSI (206, 229 and 234 kg N ha^{-1} y^{-1} respectively). Compared to the provincial level, the cropland NrSI of 429 counties varied greatly, ranging from 28 kg N ha^{-1} y^{-1} to 4782 kg N ha^{-1} y^{-1} (figure 2). Based on a national analysis, Wang *et al* (2018) grouped all counties in China into four groups based on the average per area Nr losses, and defined the top 25% as the hotspots, corresponding to a critical value of 96 kg N ha^{-1} y^{-1}. Their results showed that the Nr hotspots contributed to 52% of the national Nr losses with only 9% of the total area in China in 2012. The BR was part of the 9%. Based on this critical value, more than 360 counties, accounting for 84% of the croplands in the BR, were categorized as hotspot of Nr losses. Such a high NrSI in the BR has caused serious environmental pollution, including haze events (Ma *et al* 2014, Miao and Liu 2019), surface- and ground-water nitrate contamination (Gu *et al* 2013), and high eutrophication potential for the Bohai Sea. Note that Nr losses via surface runoff were not taken into account in this study, which may underestimate the cropland NrSI in the BR.

NH_3 volatilization and nitrate leaching were the two major Nr loss pathways from cropland in the BR. Beijing and Shandong province suffered more serious NH_3 pollution with mean emission intensities of 151 kg N ha^{-1} y^{-1} and 140 kg N ha^{-1} y^{-1} from cropland (figures 3(a) and 3(c)). The NH_3

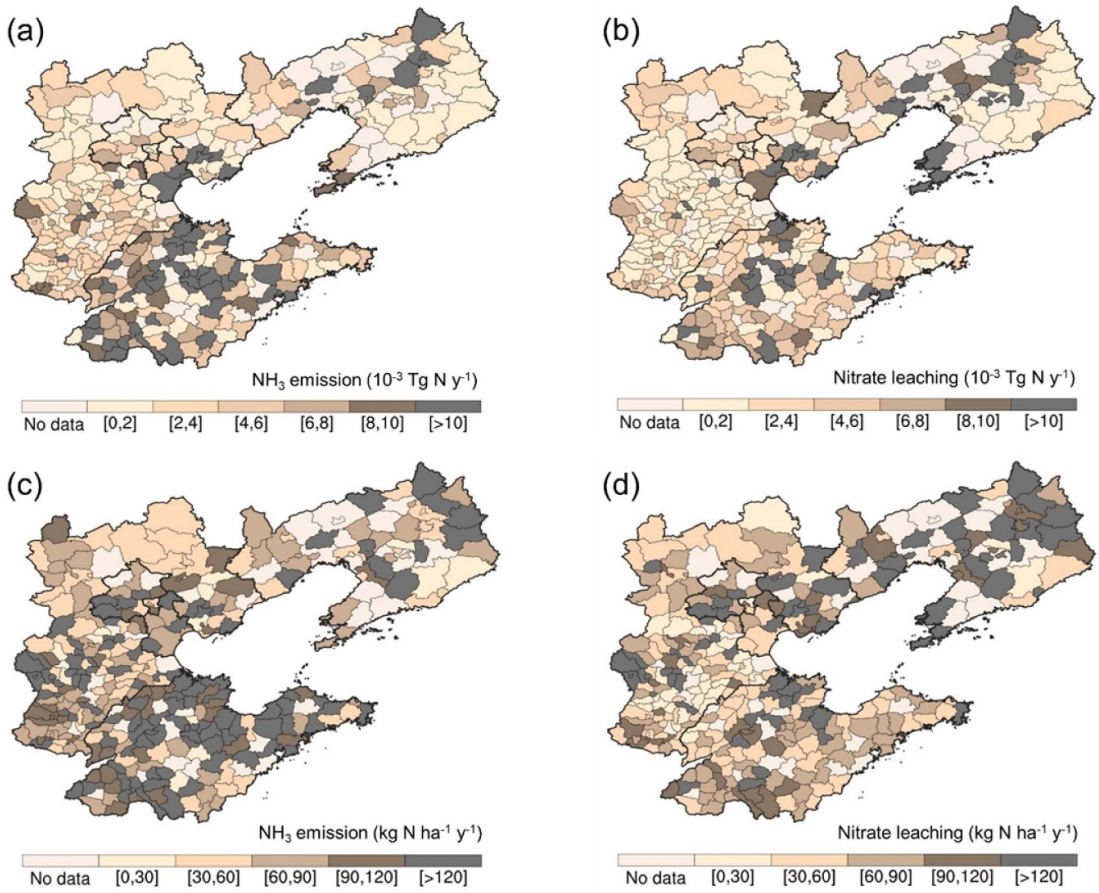

Figure 3. Spatial distribution of (a) NH_3 emissions, (b) nitrate leaching, (c) NH_3 emission intensity and (d) nitrate leaching intensity.

emission is significantly correlated to the application rate of synthetic N fertilizer and manure ($r = 0.93$, $p < 0.01$). Volatilized NH_3 from fertilizers produces a substantial loss of N available for crops, causes economic losses to farmers, and poses detrimental impacts on air quality by the formation of fine particulate matter ($PM_{2.5}$) in the atmosphere (Galloway *et al* 2008, Sanz-Cobena *et al* 2014). Beijing and Liaoning province had relatively higher nitrate leaching intensities of 147 kg N ha^{-1} y^{-1} and 130 kg N ha^{-1} y^{-1}, respectively (figures 3(b) and 3(d)). Croplands in these regions may lead to a high risk to water body contamination (e.g. Liaohe River watershed and coastal areas in south Liaoning province, Chaobaihe River watershed in Hebei province, and Xiaoqinghe River watershed in Shandong province), which should be prioritized for mitigation of nitrate leaching and control of non-point source pollution through both technical and policy approaches. Significant variations in cropland NrSI among counties in the BR were primarily associated with varieties in crops, cropland areas, and the NrSI of different cropping systems analyzed in the following section.

3.3. NrSI of different cropping systems

Altogether 29 types of cropping systems in the BR were analyzed in this study, including 16 mono cropping (figure 4(a)), 10 double cropping (figure 4(b)) and 3 triple cropping systems (figure 4(c)). Large variations in NrSI were found among different cropping systems. For the 16 mono cropping systems, vegetables had the highest NrSI of 355 kg N ha^{-1} y^{-1}, and sugarcane had the lowest NrSI of 41 kg N ha^{-1} y^{-1} with the other cropping systems' NrSI ranged from 58 kg N ha^{-1} y^{-1} to 198 kg N ha^{-1} y^{-1} (figure 4(a)). Among the 10 double cropping systems, winter wheat-vegetable cropping system showed the highest NrSI of 554 kg N ha^{-1} y^{-1}, while oat-rice cropping system presented the lowest NrSI of 52 kg N ha^{-1} y^{-1}, and the rest ranged between 58 kg N ha^{-1} y^{-1} and 213 kg N ha^{-1} y^{-1} (figure 4(b)). For the three triple cropping systems, the NrSI of vegetable-vegetable-vegetable, potato-vegetable-vegetable, and rice-rice-vegetable cropping systems were 1024 kg N ha^{-1} y^{-1}, 484 kg N ha^{-1} y^{-1} and 706 kg N ha^{-1} y^{-1}, respectively (figure 4(c)). Comparable results of the NH_3 volatilization, nitrate leaching, and N_2O emission

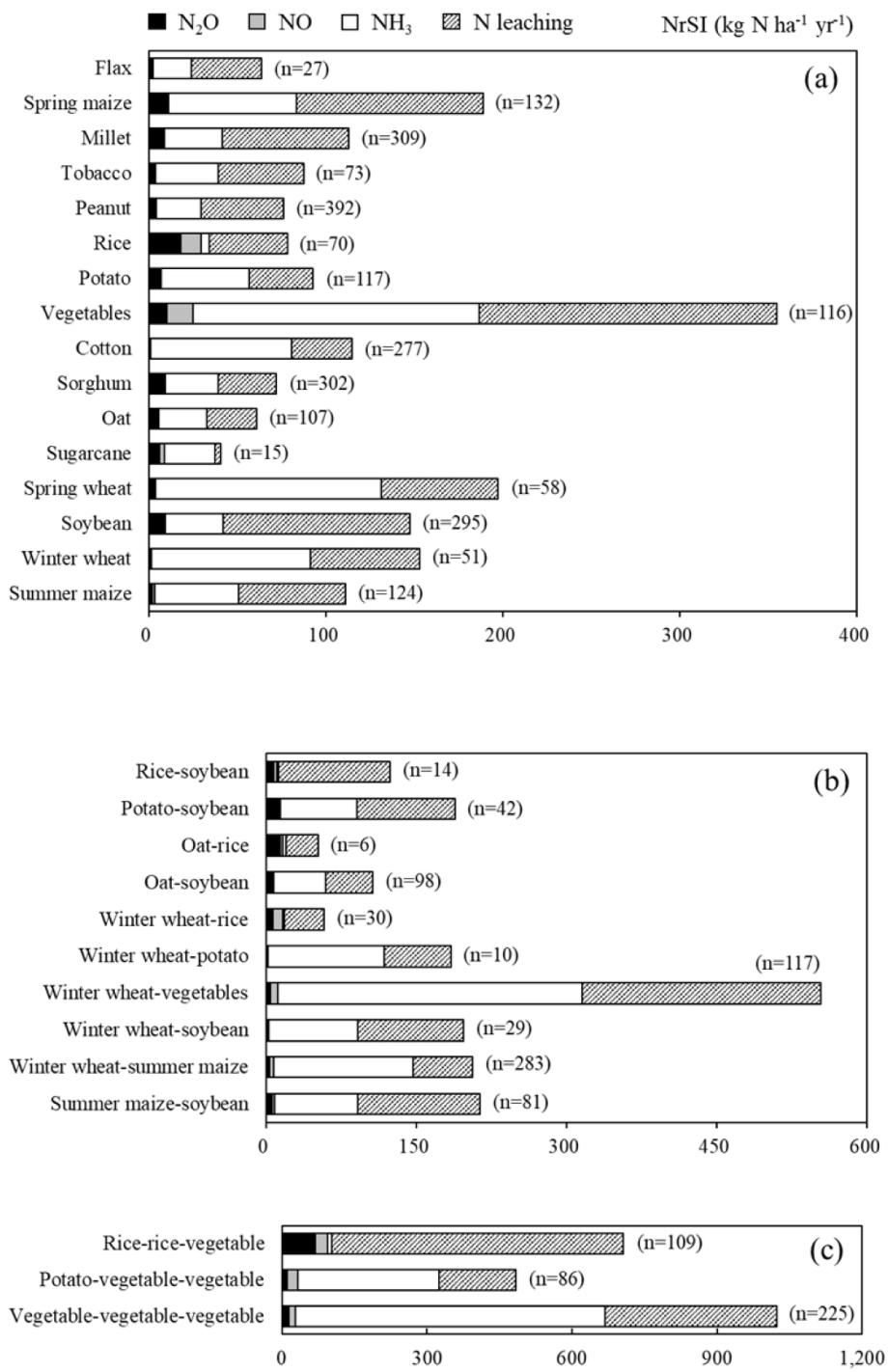

Figure 4. NrSI for 16 mono-cropping systems (upper panel), 10 double cropping systems (middle panel) and 3 triple cropping systems (lower panel). Note: n represents the number of counties included in the calculation for each cropping system.

were found between our study and other published observations for major cropping systems in the BR except the N_2O emissions from spring maize cropping system (table 2). An exponential relationship between fertilizer N rate and N_2O emission, especially in cereal cropping systems was reported by Shcherbak *et al* (2014), which indicates the higher fertilizer N application rate may lead to a larger N_2O emission from spring maize cropping system in the BR. Since there are significant differences across sites or years in Nr losses from a particular cropping system (table 2), this integration approach is able to provide an overall picture of Nr losses induced by various cropping systems simultaneously on regional level.

For all cropping systems, NH_3 volatilization and nitrate leaching dominated the Nr losses during crop production. Their contribution to the total on-farm Nr losses ranged from 62% to 99%. Except the over-use of fertilizers, the high level of NH_3 volatilization is

Table 2. Comparison of N inputs into and Nr losses from major cropping systems in the BR between our study and other published observations.

Year	Region	Cropping system	N input (kg N ha^{-1} y^{-1})	Nr loss intensity (kg N ha^{-1} y^{-1})			Studies
				NH_3 volatilization	Nitrate leaching	N_2O emission	
2008	The BR	Winter wheat-summer maize	607	139.1	59.2	3.5	This study
		Vegetable-vegetable-vegetable	1404	639.6	356.9	13.8	
		Vegetables	536	161.3	168.4	10.3	
		Spring maize	354	72.3	105.4	10.7	
2000s	Northern China Plain	Winter wheat-summer maize	545	120	136	16[a]	Zhao *et al* 2009
2003–2004	Hebei	Winter wheat-summer maize	368–968		11–145		Li *et al* 2007
2008–2009	Shandong	Winter wheat-summer maize	600			4.0	Cui *et al* 2012
2010–2013	Beijing	Winter wheat-summer maize	560		21.4–60.2		Huang *et al* 2017
2012	Shandong	Summer maize	100–200	6.5–51.8	16.3–77.2		Jia *et al* 2014
2012	Beijing	Summer maize	216			2.02	Yan *et al* 2014b
2012–2013	Beijing	Summer maize	300	10–37			Li *et al* 2017a
2012–2013	Hebei	Summer maize	270	55–108			
2009	Liaoning	Spring maize	270			2.56	Li *et al* 2012
2010	Liaoning	Spring maize	270			2.71	
2010	Liaoning	Spring maize	265			1.11	Yang *et al* 2014
2011–2012	Liaoning	Spring maize	229		123.7		Xu *et al* 2014
2005	Shandong	Vegetable	493–1316		152–347		Song *et al* 2009
2004	Shandong	Tomato	870			8.5	He *et al* 2009
2005	Shandong	Tomato	630			9.1	
2009	Beijing	Romanesco broccoli	820		301.3[b]		Guo *et al* 2012a
2009	Shandong	Onion	466			2.92	Yao *et al* 2017
2010	Shandong	Onion	487			2.76	
2010–2011	Shandong	Tomato	895		287		Fan *et al* 2014
2010	Hebei	Tomato	410	14.8		7.7	Guo *et al* 2012c
2012	Beijing	Cabbage-celery	703.5			5.51	Yan *et al* 2014b
2012	Beijing	Cucumber	703.5			3.7	
2017	Hebei	Tomato/cucumber-cabbage	390–780	65.1–125.2		6.0–20.5	Zheng *et al* 2018

[a]This value represents the total N loss through denitrification

[b]This value is the N losses through all pathways

also caused by the wide use of urea in China (accounting for over 50% of the total N fertilizer consumption) and the surface broadcasting of NH_3-based fertilizers without soil covering (Li *et al* 2015). Among all the cropping systems, rice-based cropping systems, such as rice mono system and oat-rice rotations, showed relatively higher proportion of Nr losses via N_2O emission due to alternate anaerobic and aerobic cycling that could considerably enhance N_2O emission compared to constant aerobic and anaerobic conditions (Smith and Patrick 1983) as frequent dryness of the rice paddy after the first mid-season aeration is a widely adopted practice for rice cultivation in China (Cai *et al* 1997).

The cropping systems involved with vegetable production had a relatively higher NrSI, primarily due to the continuous over-fertilization and shallow root system (Chen *et al* 2004, Ju *et al* 2004, Xiong *et al* 2006). For example, the average synthetic N fertilizer inputs for vegetable mono-cropping in 116 counties and triple-vegetable cropping in 225 counties were 402 kg N ha^{-1} y^{-1} and 1241 kg N ha^{-1} y^{-1} in 2008. In contrast, the average synthetic N fertilizer input was only 100 kg N ha^{-1} y^{-1} for sugarcane, 90 kg N ha^{-1} y^{-1} for oat and 70 kg N ha^{-1} y^{-1} for flax mono-cropping systems. Considerable N was accumulated in the soil after the continuous over-fertilization, which could easily result in nitrate leaching, especially for vegetables with a shallow root system (FAO 2006). The NrSI of various cropping systems could help identify systems with particularly high Nr losses, thus providing opportunities for future sustainable agricultural production by adjusting and optimizing the temporal and spatial layout for different cropping systems.

3.4. Sources of uncertainty

Uncertainties in our analysis mainly originate from the quality of input data (Li *et al* 2017a). Although the county-level datasets were highly reliable, the assumption of homogeneous soil properties and climate conditions will inevitably introduce uncertainties to some extent. In addition, the simplification of parameters, such as the ratios of crop residue returning and animal excreta utilized as manure, and the limited data on farming practices obtained through field surveys, might also result in potential uncertainties. Since the study area is flat overall, we did not consider surface runoff in the regional modelling, leading to an underestimation of Nr losses. However, treating the soil as a series of discrete horizontal layers and assuming some soil properties uniform within each layer/across all layers (0–50 cm soil profile), the DNDC model may overestimate nitrate leaching (2014). Future improvement of the DNDC model with fine input data and more field observations for model validation would benefit the application of this integration method for Nr hotspots assessment to other regions.

3.5. Significance and future application of the integrated approach

Integrating the DNDC model and the NrSI framework is a novel step forward for identifying Nr hotspots. Since the NrSI framework has been proposed to quantify the environmental burdens of Nr losses (Liang *et al* 2018), its application is mainly limited by the development of VNFs for agricultural products. VNFs play an important role to accurately assess the Nr losses associated with crop production. Currently, however, few countries have developed their own VNFs (Shibata *et al* 2017). The integration of the DNDC model resolves this problem as it can track soil N dynamics, N gas fluxes and nitrate leaching across crops and their rotation regimes. Meanwhile, with the consideration of local soil properties, meteorological conditions and conventional farming practices (Li 2009), it can more accurately estimate Nr losses and differentiate their multiple pathways from local to regional scale. This integrated N assessment method, therefore, provides a more comprehensive estimation of N balance, Nr losses and the NrSI associated with crop production on multiple levels (e.g. county, provincial, and regional), across various cropping systems and via different Nr loss pathways (i.e. NH_3 volatilization, nitrate leaching, N_2O and NO emissions). The DNDC model has been applied and expanded by researchers worldwide in a range of countries and cropping systems (Giltrap *et al* 2010, Li *et al* 2017a). The comprehensive database of 62 crops and 12 soil types embedded in the DNDC model (Li *et al* 2017a) enables the application of this integrated N assessment method at a wide range of regions and cropping systems in the future.

4. Conclusion

This study develops a new approach by integrating the DNDC model and the NrSI framework to identify hotspots of Nr losses induced by various cropping systems and through different pathways on regional scale. We illustrate the approach with a case study of the BR, China. The analysis provides a comprehensive estimation of N balance, Nr losses and the NrSI associated with crop production on multiple levels (i.e. county, provincial and regional), across various cropping systems (i.e. mono, double and triple cropping) and via different Nr loss pathways (i.e. NH_3 volatilization, nitrate leaching, N_2O and NO emissions) in the BR. The application of this integrated N assessment method could be used to identify areas and/or cropping systems with particularly high Nr losses and NrSI to provide basic information for setting Nr mitigation priorities on a wide range of regions and cropping systems.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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