pubs.acs.org/JAFC

Challenges for Global Sustainable Nitrogen Management in Agricultural Systems

Lei Liu,* Xiuying Zhang,* Wen Xu, Xuejun Liu, Yi Li, Jing Wei, Meng Gao, Jian Bi, Xuehe Lu, Zhen Wang, and Xiaodi Wu

Cite This: J. Agric. Food Chem. 2020, 68, 3354–3361		🔇 Read O	Read Online	
ACCESS	Metrics & More		Article Recommendations	

ABSTRACT: Nitrogen (N) losses from agricultural production contribute to detrimental impacts on water, soil, air, and human health. However, it is still lacking in evaluating global N budgets in agricultural systems. Hence, we conducted a global analysis on the current status of the N flows in the agricultural systems, explored the possible mitigation measures and challenges, and investigated the existing regulations on controlling N pollution. Globally, agricultural soils received a total of 73 kg of N ha⁻¹ year⁻¹ on average, including N fertilizer plus manure (61%), atmospheric N deposition (10%), and N litters and fixation (29%). The estimated global NH₃ loss to total N inputs was 17%, which led to a loss of \$15 billion year⁻¹. The N use efficiency (NUE) in Eastern China (33%) was much lower than that in the Eastern United States (65%) or Western Europe (61%), leaving much room to enhance the NUE to increase agricultural food production. Meanwhile, higher NH₃ losses from N fertilizers and manure were found in Eastern China (22%) than the Eastern United States (17%) and Western Europe (17%). We highlight the urgency to improve the NUE and decrease NH₃ loss with lower environmental consequences. Our results showed high potentials to mitigate NH₃ volatilization and enhance the NUE by various measures, such as substituting manure N for chemical fertilizer N, applying controlled release fertilizers, and urease inhibitors. These measures should be implemented in combination with the transfer of knowledge to farmers with new technologies and increasing the farm size to enhance the efficiency of agricultural production in the future.

KEYWORDS: nitrogen budgets, NH_3 losses, nitrogen use efficiency, agricultural systems, sustainable agriculture

■ INTRODUCTION

Over the past 50 years, the global nitrogen (N) cycle has undergone tremendous changes,¹ closely related to the extraordinary increase of agricultural synthetic N fertilizer.² Chemical N fertilizer and manure are often used exceeding crop requirements, causing high ammonia (NH₃) volatilization, loss of nitrous oxide, nitrate leaching, and runoff.³ NH₃, as a precursor of fine particulate matter formation, can lead to air quality degradation and then damage human health.⁴ Moreover, excessive atmospheric deposition of ammonium (NH₄⁺) and NH₃ can also degrade sensitive terrestrial and aquatic ecosystems.⁵ This associated NH₃ pollution has become a problem for environmental agencies, scientists, and agricultural policy makers around the world.

Regulation of NH₃ in agricultural systems is needed urgently toward sustainable agricultural production, human health, and environmental benefits.⁶ Before controlling agricultural N loss, understanding N cycle and sources is necessary from both chemical and biological perspectives.⁵ These are linked by many factors, such as climate and field variability, soil characteristics, management practices, cropping systems, and geographical areas.⁷ Any regulation for reducing N loss in the agricultural systems should take into account management practices with consideration of all aspects of the N cycle.⁸ Nitrogen use efficiency (NUE) is an essential indicator in the assessment of the performance of fertilizer application in agricultural systems.⁹ Quantifying the current status of NUE is crucial to enhance N management and decrease detrimental environmental consequences.⁷ Several previous studies have analyzed and quantified the N flows in the fields with N fertilizer application at local scales, the NUE, and the amounts of NH₃ loss.^{7,10,11} However, the field measurements of N flows (such as NH₃ volatilization and the NUE) are still scarce around the globe as a result of the high cost, expertise, and labor.¹⁰

Instead, the process-based models can supply an effective alternative way to study the N budgets in agricultural systems and simulate biogeochemical processes in the soil.^{8,12,13} The denitrification decomposition (DNDC) model can be applied to stimulate N flows in agricultural systems and finely validated by numerous ground-based measurements.^{8,14–17} Previous works have investigated the impacts of agricultural management on N flows at local scales, but very little work quantified comprehensively the management practices and explored mitigation strategies for NH₃ loss on a global perspective. In

Received:	January 14, 2020
Revised:	March 3, 2020
Accepted:	March 4, 2020
Published:	March 4, 2020



Figure 1. Total N inputs in cropping systems, including manure amendment, synthetic N fertilizer, atmospheric N deposition, litter (crop residues), and others. (a) Summary of total N inputs in hotspot areas of WEU, EUS, NEI, SCH, and ECH. (b) Each contributor to total N inputs in the hotspot regions. (c) Spatial distribution of total N inputs. The global map of political boundary data was available from https://map.igismap. com/share-map/export-layer/TM_WORLD_BORDERS-0.3/502e4a16930e414107ee22b6198c578f, created by OpenStreetMap contributors and licensed under a CC BY-SA license (https://creativecommons.org/licenses/by-sa/2.0/).

this study, we systematically evaluated N flows in cropping systems, the contribution of $\rm NH_3$ loss to total N inputs, and the NUE in hotspot regions, at the global scale. Previous studies also show management measures are essential for mitigating N loss in the agricultural system, such as improving fertilizer efficiency (applying urease inhibitors, controlled release fertilizers, substitution of manure for chemical N fertilizer, etc.) and implementing scientific farmland management (irrigation immediately, deep placement of N fertilizers, crop residue retention, etc.).^{18–20} Thus, we also demonstrated our insight into current challenges for sustainable N management in global agricultural systems and provide possible pathways to mitigate NH₃ losses.

METHODS

Data Sources. To identify the spatial distribution of global croplands, we used the Global Mosaics of MODIS land-use (MCD12Q1), which is generated by integrated supervised classification algorithms.²¹ Meteorological data included total precipitation, maximum temperature, and minimum temperature at a daily time step from Climate Prediction Center (CPC) global unified data at a resolution of 0.5° grids (https://www.esrl.noaa.gov/psd/). The N fertilizer and manure data were obtained from the Data Center in

NASA's Earth Observing System Data and Information System²² (http://sedac.ciesin.columbia.edu). The crop-specific N fertilizer was gained from Mueller et al.,²³ and the data set of crop-specific N fertilizer can be gained at http://www.earthstat.org/. N deposition data sets were obtained from GEOS-Chem by Geddes et al.²⁴ The soil parameter data of pH, soil organic matter (SOM) fraction, clay fractions, and bulk density were obtained from the Soil Database of China for Land Surface modeling (http://globalchange.bnu.edu.cn). Data on the management, residues, tillage, and cropping systems were adopted by Global Landscapes Initiative (http://www.earthstat.org/), Beach et al.,²⁵ Smith et al.,¹³ and Yu et al.²⁶ The long-term NH₃ volatilization data during 1970–2010 were gained from EDGAR (version 4.3) (https://edgar.jrc.ec.europa.eu/htap.php#), and the trends of NH₃ during 2010–2016 were obtained from satellite data of infrared atmospheric sounding interferometer (IASI) retrievals (http://cds-espri.ipsl.upmc.fr/).

Biogeochemical Modeling of Cropland N Flows by DNDC. DNDC is a N-oriented model, which can be used to simulate crop growth and the biogeochemical processes of decomposition by microbial activities, denitrification, and nitrification in agricultural systems.³ It contains a mass balance methodology for C, N, and water in air, plants, and soil.¹⁴ The impact of N on cropping systems is closely associated with human management practices, soil properties, and climate.¹⁵ We used the DNDC to simulate the N budgets, the NUE, and NH₃ volatilization globally. The baseline scenario was set

pubs.acs.org/JAFC



Figure 2. Ratio of NH₃ volatilization to total N inputs and the NUE in different regions. (a) Spatial distribution of NH₃ volatilization to total N inputs. (b) Annual changes of NH₃ volatilization in hotspot regions during 1970–2010. The global map of political boundaries data was available from https://map.igismap.com/share-map/export-layer/TM_WORLD_BORDERS-0.3/502e4a16930e414107ee22b6198c578f, created by Open-StreetMap contributors and licensed under a CC BY-SA license (https://creativecommons.org/licenses/by-sa/2.0/).

using the 2010 conditions for fertilization (chemical N and manure), crop parameters, soil, irrigation, and land use. NH₃ volatilization was quantified on the basis of soil NH₄⁺, aqueous NH₃, pH, texture, temperature, and wind speed under different agricultural management practices.^{14,27} We used DNDC to assess the impact of management measures on NH₃ volatilization from cropping production systems, which is a well-tested biogeochemical model supporting multipurpose analyses compared to other approaches (http://www.globaldndc. net).^{28,29}

The N budget can be derived by quantifying the soil N inputs and outputs. 30

$$DepN + FerN + ManN + litterN + FixN$$
$$= N_2O + NO + N_2 + NH_3 + cropN + leachN + runoff$$
$$+ dson + dsin$$

Inputs of N include atmospheric deposition (DepN), chemical fertilizer N (FerN), manure N (ManN), litter N from residues of crops (litterN), and biological fixation (FixN). Outputs of N include crop uptake (cropN), NH_3 volatilization, N runoff and leaching (leachN), gaseous N_2O , NO, N_2 , and soil dissolved organic and inorganic N (dson and dsin).

Daily NH₃ volatilization is calculated as^{14,28,29}

$$F_{\rm NH_2} = \rm NH_3(l) \times \rm ddf \tag{2}$$

where ddf is the volatilization coefficient, $F_{\rm NH_3}$ is the NH₃ volatilization, and NH₃(l) is the NH₃ concentration in soil water.

Dubache et al.²⁰ found that the ddf was impacted by wind speed, soil temperature, soil depth, vegetation canopy, clay content, soil moisture, and rain-induced canopy wetting. In this study, we revised the DNDC95 code according to Dubache et al.²⁰ using following equations:

$$ddf = f_{wind} f_{temp} f_{depth} f_{canopy} f_{clay} f_{water} f_{rain}$$
(3)
$$\begin{cases} f_{wind} = 0.1 + 1.5 S_{wind} / (1 + S_{wind}) \\ f_{canopy} = 0.4 e^{-0.15lai} + 0.6 \\ f_{depth} = (q - j) / q \\ f_{clay} = 0.4 e^{-0.1clay} + 0.6 \\ f_{temp} = 0.1 + 2.0 T_{soil} / (45 + T_{soil}) \\ f_{water} = 0.45 e^{-10WFPS} + 0.55 \end{cases}$$

$$f_{\rm rain} = 0.65 e^{-1.0 {\rm lai} \times P} + 0.35$$
 (4)

where S_{wind} is the wind speed, lai indicates the leaf area index, q represents the total soil layer number, clay is the soil clay content, T_{soil} is the soil temperature, WFPS is the air-filled porosity, and P is the daily precipitation.

The NUE is a widely used indicator evaluating the efficiency of N inputs (biological N fixation, deposition, manure and fertilizers, and litter N) harvested in biomass, calculated as⁶

(1)



Figure 3. Amendments to control N pollution and normalized NH_3 volatilization during 1970–2016. (a) N-related pollution control regulations. (b) Temporal changes of NH_3 volatilization, normalized to the 1970 level. The trend of NH_3 during 2008–2016 was derived from the trend of IASI NH_3 retrievals.

$$NUE = \frac{cropN}{totalN} \times 100$$
(5)

where cropN indicates N output harvested by crops and totalN represents all N outputs included in eq 1.

Uncertainty Analysis. The driven factors influencing the modeling of N flows are related to the meteorological data, N fertilizer, N manure, N deposition, and soil properties (pH, texture, SOM, bulk density, etc.). We used the minimum and maximum soil properties, temperature, precipitation, and set fluctuations of N fertilizer, N manure, and N deposition within $\pm 20\%$ to assess the possible range of modeling results. All of the results presented in the figures were the mean values, and the annual fluctuations were within $\pm 18\%$ of simulated global cropland N flows. These uncertainties are systematic rather than random and should not affect conclusions of regional comparisons and spatial gradients of N budgets.

RESULTS

Overview of Total N Inputs in Cropping Systems. Globally, cropland soils received a total of 73 kg of N ha⁻¹ year⁻¹ on average in 2010 (Figure 1), including N fertilizer plus manure (61%), atmospheric N deposition (10%), and N litters and fixation (29%). This is consistent with the traditional perspective that a cropland soil is predominately impacted by N fertilizer plus manure, which are common practices in cropping systems around the world.¹ Currently, N manure is an important fertilizer type in cropping systems, 55% of total N fertilizer on average in the global croplands. N litters (from crop residuals) also comprise 29% of N input, highly linked with the activity of the soil microbes mineralizing organic N to $NH_4^{+.16,31}$ With the rapid reactive N emissions, atmospheric N deposition also contributes to 10% of total N inputs. In particular, hotspots of total N inputs were identified globally, including Western Europe (WEU), Eastern United States (EUS), Northeastern India (NEI), Sichuan Basin (SCH), and Eastern China (ECH), as the major areas with intensive agricultural production. The ECH region was the most contaminated N area (221 kg of N ha⁻¹), approximately triple the global average (73), followed by SCH (189), NEI (136), WEU (113), and EUS (91).

Current Status of NUE and NH₃ Volatilization. The NUE is an important efficiency index to evaluate the progress toward achieving sustainable agricultural production.^{6,10} The major reason for low NUE is the N loss from the cropland system through NH₃ volatilization, gaseous NO, N₂O, and N₂ emissions, runoff, and leaching.³ Generally, NH₃ volatilization is the main mechanism of many agricultural production systems, as a global problem requiring to be solved.¹¹ Estimated global NH₃ loss percentage was 16.96% in this study, which is close to the estimate (17.60%) by Pan et al.¹¹ and slightly higher than the estimate (10-14%) from the Intergovernmental Panel on Climate Change (IPCC) reports.¹¹ The global average ratio of NH₃ volatilization to total N inputs (16.96%) is equal to a loss of \$15 billion, with the demand for 112 million tons of N [from the Food and Agriculture Organization of the United Nations (FAO)] (~\$350/ton of urea).¹¹ With regard to the hotspot regions of N inputs, the NUE in the ECH was 33% (Figure 2), which was close to the estimate (36-39%) by Gu et al.⁶ China's NUE was much lower than that in the EUS (65%) and WEU (61%), leaving much room to enhance the NUE to increase

pubs.acs.org/JAFC

Perspective



Figure 4. Schematic of the NH_3 -related process. (a) Processes of soil NH_4^+ and NH_3 concentrations in the cropping system. (b) Potential measures for NH_3 mitigation.

agricultural food production in China, with only 7% of the global croplands feeding 20% of the global population. Correspondingly, higher NH₃ losses from N fertilizers and manure were found in the ECH (22%) than the EUS (17%) and WEU (17%). The ECH region has been the largest hotspot of NH₃ volatilization globally, resulting from the highest N inputs to croplands and the low NUE. On the basis of the EDGAR estimates, NH₃ volatilization has increased rapidly during 1970–2010 for ECH (0.4 kg of N ha⁻¹ year⁻¹), EUS (0.04 kg of N ha⁻¹ year⁻¹), and NEI (0.39 kg of N ha⁻¹ year⁻¹), except for WEU with a continuous increase before 1986 (0.10 kg of N ha⁻¹ year⁻¹) but a substantial decrease (-0.04 kg of N ha⁻¹ year⁻¹) since then.

Existing Regulations for Controlling N Pollution. The European Union (EU) Common Agricultural Policy (CAP) was signed in 1958 contributing to the agricultural modernization and experienced several reforms together with a series of water and air quality control regulations (Figure 3a), significantly influencing the NUE and NH_3 loss in the WEU. However, there were very limited regulations for NH_3 in China and the U.S., causing a continuous increase in NH_3 volatilization since 1970 (Figure 3b). With increasing evidence to support the important contribution of NH_3 to fine

particulate matter pollution,^{32,33} in 2019, the United States Environmental Protection Agency (U.S. EPA) has listed NH_3 as a pollutant, supported by the report from the U.S. EPA's Integrated N Panel to the Science Advisory Board. In 2015, China's Ministry of Agriculture formally embarked on the "zero increase action plan (ZIAP)", requiring less than 1% annual increase in N fertilizer use during 2015–2019 and no further increase since 2020.⁶ However, according to the satellite retrievals, NH_3 columns increased continuously after 2015 (Figure 3b), which may be associated with NH_3 loss from organic N manure in livestock production because ZIAP only controlled the N fertilizer amounts.

Measures for Enhancing the NUE and Reducing NH_3 Volatilization. NH_3 volatilization decreases the N available for plants and leads to a series of environmental consequences. Therefore, there is an urgent need to systematically analyze the effect of mitigation strategies toward reducing NH_3 volatilization in crop systems (Figure 4).

First, improving fertilizer efficiency has been widely recognized to enhance the NUE and minimize NH_3 volatilization from cropping systems.^{5,6} Applying urease inhibitors slowing urea hydrolysis⁹ could substantially reduce NH_3 volatilization by 24, 16, and 17% in ECH, the U.S., and

Europe (averaged by 18%), respectively. Controlled release fertilizers extending the availability of N for crop uptake and minimizing urea hydrolysis could substantially decrease NH₃ volatilization by 19, 14, and 15% in ECH, the U.S., and Europe (averaged by 16%), respectively. Substituting 20% chemical N (urea) with equivalent manure can substantially reduce NH₃ volatilization (21%), which could improve the NUE by 15, 25, and 22% in ECH, EUS, and WEU, respectively. Substituting manure for chemical N could enhance immobilized N by microbes and improve the capacity of soil to hold water, microbial activities, and soil structure.⁹ Second, implementing scientific farmland management was another crucial strategy toward improving the NUE and reducing NH₃ volatilization. Irrigation immediately following N fertilizer application by washing N into the deep soil to be adsorbed can enhance the NUE by 14, 14, and 17% in ECH, EUS, and WEU, respectively, and can reduce NH₃ volatilization by 20% on average. Specifically, irrigation immediately contributes to urea hydrolysis, allows NH4⁺ to be adsorbed by plants, and therefore increases the NUE and decreases the conversion of NH4⁺ to NH3 volatilization.^{11,34,35} In addition, deep N fertilizer placement and addition of acidifiers could also decrease NH₃ volatilization significantly by 17 and 11%. Notably, crop residue retention increased NH₃ volatilization as a result of restricted movement of N fertilizer into the deep soil;¹¹ nitrification inhibitors applied for mitigating N₂O emission can unexpectedly increase NH₃ volatilization (~10%).

DISCUSSION

Research on agricultural NH₃ volatilization and the NUE at the local scales has been undertaken separately in recent years^{5,6,11} but lacking a global perspective on the current status of total N budgets, the possible mitigation measures and challenges, and the existing regulations on controlling N pollution. Building on these issues, we present the status of the N budgets in the cropping systems and highlight the urgency to improve the NUE and decrease NH₂ loss to reduce the environmental consequences. Agricultural N fertilizer and manure have a substantial impact on environmental quality, given the low NUE and high NH₃ loss globally. Currently, the NUE ranged from 31 to 65% in the selected hotspots with intensive N inputs. In particular, China's NUE (31%) is much lower than that of the U.S. (65%) and Europe (61%), while NH₃ volatilization in China is triple that in the U.S. and Europe. Moreover, a continuous increase in NH₃ volatilization since 1970 occurred in China and the U.S. as a result of the lack of effective regulations. The increase rate of NH₃ volatilization in China is 10 times greater than that in the U.S. since 1970. Our results show that a great potential exists to mitigate NH₃ volatilization and improve the NUE by various strategies, for instance, substituting manure for chemical N and applying controlled release fertilizers and urease inhibitors. However, these identified measures, such as substituting manure for chemical N, certainly need additional expenditure from farmers, and therefore, a fertilizer subsidy policy should also be carried out on agricultural systems to match these measures.

The total N input in ECH's croplands is more double than that in the EUS and WEU. The reason that Chinese farmers apply so much more N chemicals than the other regions may be closely linked with the small farm size (~ 0.1 ha on average for each parcel of cropland) in China.³⁶ It is difficult to persuade farmers to limit their input of chemical fertilizers

because many people still believe higher crop yields can be achieved through more chemical fertilizers.¹⁰ Moreover, in such a small scale, it is difficult to transfer knowledge of modern management practice (such as 4R nutrient stewardship) to farmers and apply technological innovations (such as increasing feed conversion ratio in the livestock subsystem), with the high financial cost.¹⁰ In the future, transfer of knowledge to farmers with new technologies should be conducted toward better agricultural managements, together with increasing the farm size to enhance the efficiency of agricultural production.

In contrast, the WEU is the only region having a continuous decrease in NH₃ volatilization since the late 1990s, with numerous mature regulations on N pollutions. N flows in Europe are regulated by various policy measures, such as input control (agricultural application restriction), emission control (manure application and storage), N concentration restriction in air and water, N exposure restriction, and critical N load.³⁷ China and the U.S. need to learn from the WEU regarding the NH3 emission control framework and measures toward achieving sustainable agricultural production. Previously, the effects of NH₃ on air quality and human health were debated heavily for the limit targets to be achieved in China and the U.S. Fortunately, the U.S. EPA has listed NH₃ as a pollutant in 2019 as a result of its critical contribution to fine particular matter,³⁸ under the background that the acidic gases (SO₂ and SO₂) have decreased in a recent decade.^{39,40} Also, China has started ZIAP for controlling the amounts of N fertilizer in 2015. Now, it is time to manage agricultural N to reduce the N-related environmental consequences globally, toward a more sustainable outcome. Non-chemical or organic farming established on the basis of biological principles may have the prospect for addressing the N loss pollution of chemical farming.⁴¹ It depends upon various biological processes to maintain soil fertility, meet crop nutrition, and establish a biological system to prevent weeds and pests.⁴¹⁻⁴³

AUTHOR INFORMATION

Corresponding Authors

- Lei Liu College of Earth and Environmental Sciences, Lanzhou University, Lanzhou, Gansu 730000, People's Republic of China; Ocicid.org/0000-0003-2889-8900; Email: liuleigeo@lzu.edu.cn
- Xiuying Zhang International Institute for Earth System Science, Nanjing University, Nanjing, Jiangsu 210023, People's Republic of China; Email: zhangxy@nju.edu.cn

Authors

- Wen Xu College of Resources and Environmental Sciences, National Academy of Agriculture Green Development, China Agricultural University, Beijing 100193, People's Republic of China
- Xuejun Liu College of Resources and Environmental Sciences, National Academy of Agriculture Green Development, China Agricultural University, Beijing 100193, People's Republic of China; ⊚ orcid.org/0000-0002-8367-5833
- Yi Li SailBri Cooper, Incorporated, Beaverton, Oregon 97008, United States
- Jing Wei State Key Laboratory of Remote Sensing Science, College of Global Change and Earth System Science, Beijing Normal University, Beijing 100875, People's Republic of China; Department of Atmospheric and Oceanic Science, Earth System Science Interdisciplinary Center, University of Maryland, College

Park, Maryland 20742, United States; o orcid.org/0000-0002-8803-7056

- **Meng Gao** Department of Geography, Hong Kong Baptist University, Kowloon Tong, Hong Kong Special Administrative Region of the People's Republic of China
- Jian Bi College of Earth and Environmental Sciences, Lanzhou University, Lanzhou, Gansu 730000, People's Republic of China
- **Xuehe Lu** International Institute for Earth System Science, Nanjing University, Nanjing, Jiangsu 210023, People's Republic of China
- **Zhen Wang** International Institute for Earth System Science, Nanjing University, Nanjing, Jiangsu 210023, People's Republic of China
- **Xiaodi Wu** International Institute for Earth System Science, Nanjing University, Nanjing, Jiangsu 210023, People's Republic of China

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.jafc.0c00273

Funding

This study is supported by the National Natural Science Foundation of China (41471343, 41425007, and 41101315) as well as the Chinese National Programs on Heavy Air Pollution Mechanisms and Enhanced Prevention Measures (Project DQGG0208).

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors acknowledge Dr. Jia Deng at the University of New Hampshire and Dr. Feng Zhang at Lanzhou University for the help using the DNDC model. The authors also thank Jeffrey Geddes at Boston University for providing the global N deposition datasets.

REFERENCES

(1) Galloway, J. N.; Townsend, A. R.; Erisman, J. W.; Bekunda, M.; Cai, Z.; Freney, J. R.; Martinelli, L. A.; Seitzinger, S. P.; Sutton, M. A. Transformation of the nitrogen cycle: Recent trends, questions, and potential solutions. *Science* **2008**, 320 (5878), 889–892.

(2) Galloway, J. N.; Dentener, F. J.; Capone, D. G.; Boyer, E. W.; Howarth, R. W.; Seitzinger, S. P.; Asner, G. P.; Cleveland, C. C.; Green, P. A.; Holland, E. A.; Karl, D. M.; Michaels, A. F.; Porter, J. H.; Townsend, A. R.; Vöosmarty, C. J. Nitrogen cycles: Past, present, and future. *Biogeochemistry* **2004**, 70 (2), 153–226.

(3) Li, C. S. Modeling trace gas emissions from agricultural ecosystems. *Nutr. Cycling Agroecosyst.* **2000**, 58 (1–3), 259–276.

(4) Liu, X.; Zhang, Y.; Han, W.; Tang, A.; Shen, J.; Cui, Z.; Vitousek, P.; Erisman, J. W.; Goulding, K.; Christie, P.; Fangmeier, A.; Zhang, F. Enhanced nitrogen deposition over China. *Nature* **2013**, *494* (7438), 459–462.

(5) Gu, B.; Ju, X.; Chang, J.; Ge, Y.; Vitousek, P. M. Integrated reactive nitrogen budgets and future trends in China. *Proc. Natl. Acad. Sci. U. S. A.* **2015**, *112* (28), 8792–8797.

(6) Gu, B.; Ju, X.; Chang, S. X.; Ge, Y.; Chang, J. Nitrogen use efficiencies in Chinese agricultural systems and implications for food security and environmental protection. *Reg. Environ. Change* **2017**, *17*, 1217–1227.

(7) Li, C.; Narayanan, V.; Harriss, R. C. Model estimates of nitrous oxide emissions from agricultural lands in the United States. *Global Biogeochem. Cy.* **1996**, *10* (2), 297–306.

(8) Li, C.; Salas, W.; Zhang, R.; Krauter, C.; Rotz, A.; Mitloehner, F. Manure-DNDC: A biogeochemical process model for quantifying

greenhouse gas and ammonia emissions from livestock manure systems. *Nutr. Cycling Agroecosyst.* **2012**, *93* (2), 163–200.

(9) Xia, L.; Lam, S. K.; Chen, D.; Wang, J.; Tang, Q.; Yan, X. Can knowledge-based N management produce more staple grain with lower greenhouse gas emission and reactive nitrogen pollution? A meta-analysis. *Global Change Biol.* **2017**, *23* (5), 1917–1925.

(10) Ju, X.-T.; Xing, G.-X.; Chen, X.-P.; Zhang, S.-L.; Zhang, L.-J.; Liu, X.-J.; Cui, Z.-L.; Yin, B.; Christie, P.; Zhu, Z.-L.; Zhang, F.-S. Reducing environmental risk by improving N management in intensive Chinese agricultural systems. *Proc. Natl. Acad. Sci. U. S. A.* **2009**, *106* (9), 3041–3046.

(11) Pan, B.; Lam, S. K.; Mosier, A.; Luo, Y.; Chen, D. Ammonia volatilization from synthetic fertilizers and its mitigation strategies: A global synthesis. *Agric., Ecosyst. Environ.* **2016**, *232*, 283–289.

(12) Congreves, K. A.; Grant, B. B.; Dutta, B.; Smith, W. N.; Chantigny, M. H.; Rochette, P.; Desjardins, R. L. Predicting ammonia volatilization after field application of swine slurry: DNDC model development. *Agric., Ecosyst. Environ.* **2016**, *219*, 179–189.

(13) Goglio, P.; Grant, B. B.; Smith, W. N.; Desjardins, R. L.; Worth, D. E.; Zentner, R.; Malhi, S. S. Impact of management strategies on the global warming potential at the cropping system level. *Sci. Total Environ.* **2014**, *490*, 921–933.

(14) Li, C. S. Modeling trace gas emissions from agricultural ecosystems. In *Methane Emissions from Major Rice Ecosystems in Asia;* Wassmann, R., Lantin, R. S., Neue, H.-U., Eds.; Springer: Dordrecht, Netherlands, 2000; pp 259–276, DOI: 10.1007/978-94-010-0898-3 20.

(15) Giltrap, D. L.; Li, C.; Saggar, S. DNDC: A process-based model of greenhouse gas fluxes from agricultural soils. *Agric., Ecosyst. Environ.* **2010**, *136* (3–4), 292–300.

(16) Li, H.; Wang, L.; Li, J.; Gao, M.; Zhang, J.; Zhang, J.; Qiu, J.; Deng, J.; Li, C.; Frolking, S. The development of China-DNDC and review of its applications for sustaining Chinese agriculture. *Ecol. Modell.* **2017**, *348*, 1–13.

(17) Dutta, B.; Congreves, K. A.; Smith, W. N.; Grant, B. B.; Rochette, P.; Chantigny, M. H.; Desjardins, R. L. Improving DNDC model to estimate ammonia loss from urea fertilizer application in temperate agroecosystems. *Nutr. Cycling Agroecosyst.* **2016**, *106*, 275– 292.

(18) Canfield, D. E.; Glazer, A. N.; Falkowski, P. G. The Evolution and Future of Earth's Nitrogen Cycle. *Science* **2010**, 330 (6001), 192–196.

(19) Bussink, D.; Oenema, O. Ammonia volatilization from dairy farming systems in temperate areas: A review. *Nutr. Cycling Agroecosyst.* **1998**, *51* (1), 19–33.

(20) Dubache, G.; Li, S.; Zheng, X.; Zhang, W.; Deng, J. Modeling ammonia volatilization following urea application to winter cereal fields in the United Kingdom by a revised biogeochemical model. *Sci. Total Environ.* **2019**, *660*, 1403–1418.

(21) Friedl, M. A.; Sulla-Menashe, D.; Tan, B.; Schneider, A.; Ramankutty, N.; Sibley, A.; Huang, X. MODIS Collection 5 global land cover: Algorithm refinements and characterization of new datasets. *Remote Sens. Environ.* **2010**, *114* (1), 168–182.

(22) Potter, P.; Ramankutty, N.; Bennett, E. M.; Donner, S. D. Characterizing the Spatial Patterns of Global Fertilizer Application and Manure Production. *Earth Interact.* **2010**, *14* (2), 1–22.

(23) Mueller, N. D.; Gerber, J. S.; Johnston, M.; Ray, D. K.; Ramankutty, N.; Foley, J. A. Closing yield gaps through nutrient and water management. *Nature* **2012**, *490* (7419), 254–257.

(24) Geddes, J. A.; Martin, R. V. Global deposition of total reactive nitrogen oxides from 1996 to 2014 constrained with satellite observations of NO₂ columns. *Atmos. Chem. Phys.* **2017**, 17 (16), 10071–10091.

(25) Beach, R. H.; Creason, J.; Ohrel, S. B.; Ragnauth, S.; Ogle, S.; Li, C.; Ingraham, P.; Salas, W. Global mitigation potential and costs of reducing agricultural non-CO2 greenhouse gas emissions through 2030. *Journal of Integrative Environmental Sciences* **2015**, *12* (sup1), 87–105.

Journal of Agricultural and Food Chemistry

(26) Yu, C.; Huang, X.; Chen, H.; Godfray, H. C. J.; Wright, J. S.; Hall, J. W.; Gong, P.; Ni, S.; Qiao, S.; Huang, G.; Xiao, Y.; Zhang, J.; Feng, Z.; Ju, X.; Ciais, P.; Stenseth, N. C.; Hessen, D. O.; Sun, Z.; Yu, L.; Cai, W.; Fu, H.; Huang, X.; Zhang, C.; Liu, H.; Taylor, J. Managing nitrogen to restore water quality in China. *Nature* **2019**, *567* (7749), 516–520.

(27) Giltrap, D.; Saggar, S.; Rodriguez, J.; Bishop, P. Modelling NH_3 volatilisation within a urine patch using NZ-DNDC. *Nutr. Cycling Agroecosyst.* **2017**, *108* (3), 267–277.

(28) Deng, J.; Zhu, B.; Zhou, Z.; Zheng, X.; Li, C.; Wang, T.; Tang, J. Modeling nitrogen loadings from agricultural soils in southwest China with modified DNDC. *J. Geophys. Res.* **2011**, *116* (G2), G02020.

(29) Deng, J.; Li, C.; Wang, Y. Modeling ammonia emissions from dairy production systems in the United States. *Atmos. Environ.* **2015**, *114*, 8–18.

(30) Ladha, J. K.; Tirol-Padre, A.; Reddy, C. K.; Cassman, K. G.; Verma, S.; Powlson, D. S.; van Kessel, C.; de B. Richter, D.; Chakraborty, D.; Pathak, H. Global nitrogen budgets in cereals: A 50year assessment for maize, rice, and wheat production systems. *Sci. Rep.* **2016**, *6* (1), 19355.

(31) Congreves, K. A.; Grant, B. B.; Dutta, B.; Smith, W. N.; Chantigny, M. H.; Rochette, P.; Desjardins, R. L. Predicting ammonia volatilization after field application of swine slurry: DNDC model development. *Agric., Ecosyst. Environ.* **2016**, *219*, 179–189.

(32) Wu, Y.; Gu, B.; Erisman, J. W.; Reis, S.; Fang, Y.; Lu, X.; Zhang, X. PM_{2.5} pollution is substantially affected by ammonia emissions in China. *Environ. Pollut.* **2016**, *218*, 86–94.

(33) Xu, Z.; Liu, M.; Zhang, M.; Song, Y.; Wang, S.; Zhang, L.; Xu, T.; Wang, T.; Yan, C.; Zhou, T.; Sun, Y.; Pan, Y.; Hu, M.; Zheng, M.; Zhu, T. High efficiency of livestock ammonia emission controls in alleviating particulate nitrate during a severe winter haze episode in northern China. *Atmos. Chem. Phys.* **2019**, *19* (8), 5605–5613.

(34) Xu, J.; Peng, S.; Yang, S.; Wang, W. Ammonia volatilization losses from a rice paddy with different irrigation and nitrogen managements. *Agr. Water Manage*. **2012**, *104*, 184–192.

(35) Bouwman, A. F.; Boumans, L. J. M.; Batjes, N. H. Estimation of global NH₃ volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Global Biogeochem. Cycles* **2002**, *16* (2), 8-1–8-14.

(36) Wu, Y.; Xi, X.; Tang, X.; Luo, D.; Gu, B.; Lam, S. K.; Vitousek, P. M.; Chen, D. Policy distortions, farm size, and the overuse of agricultural chemicals in China. *Proc. Natl. Acad. Sci. U. S. A.* 2018, 115 (27), 7010–7015.

(37) The European Nitrogen Assessment: Sources, Effects and Policy Perspectives; Sutton, M. A., Howard, C. M., Erisman, J. W., Billen, G., Bleeker, A., Grennfelt, P., van Grinsven, H., Grizzetti, B., Eds.; Cambridge University Press: Cambridge, U.K., 2011; DOI: 10.1017/CBO9780511976988.

(38) Wei, J.; Li, Z.; Guo, J.; Sun, L.; Huang, W.; Xue, W.; Fan, T.; Cribb, M. Satellite-Derived 1-km-Resolution PM_1 Concentrations from 2014 to 2018 across China. *Environ. Sci. Technol.* **2019**, 53 (22), 13265–13274.

(39) Geddes, J. A.; Martin, R. V.; Boys, B. L.; van Donkelaar, A. Long-term trends worldwide in ambient NO_2 concentrations inferred from satellite observations. *Environ. Health Perspect.* **2016**, 124 (3), 281.

(40) Wei, J.; Huang, W.; Li, Z.; Xue, W.; Peng, Y.; Sun, L.; Cribb, M. Estimating 1-km-resolution PM_{2.5} concentrations across China using the space-time random forest approach. *Remote Sens. Environ.* **2019**, 231, 111221.

(41) Bond, W.; Grundy, A. C. Non-chemical weed management in organic farming systems. *Weed Res.* **2001**, *41* (5), 383–405.

(42) Albrecht, H. Development of arable weed seedbanks during the 6 years after the change from conventional to organic farming. *Weed Res.* **2005**, 45 (5), 339–350.

(43) Darmency, H. Does genetic variability in weeds respond to non-chemical selection pressure in arable fields? *Weed Res.* **2019**, *59* (4), 260–264.