

## REVIEW ARTICLE OPEN



# Addressing nitrogenous gases from croplands toward low-emission agriculture

Shu-Yuan Pan<sup>1</sup>✉, Kung-Hui He<sup>1</sup>, Kuan-Ting Lin<sup>1</sup>, Chihhao Fan<sup>1</sup> and Chang-Tang Chang<sup>2</sup>

The use of nitrogen fertilizers in agriculture produces significant quantities of nitrogenous gases including ammonia, nitric oxide, and nitrous oxide. Through better farmland management practices, the emission of nitrogenous gases can be reduced while realizing clean water environment and climate-smart agriculture. In this article, we first provided an overview of the international movements on reducing nitrogenous gas emissions from farmlands. Then, we summarized the effect of agricultural management practices on nitrogen use efficiency for various crops, and evaluated their effect on nitrogenous gas emissions. The results indicated the importance of implementing site-specific sustainable management practices to enhance nitrogen use efficiency, and thus mitigate nitrogenous gas emissions. We also addressed the impact of agricultural activities on cropland nitrogen cycles, and highlighted the need to perform systematic trade-off evaluations with a well-defined scope to maximize environmental benefits and maintain ecosystem services. Lastly, we proposed three priority directions by moving toward a low-emission agriculture.

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## INTRODUCTION

In agriculture, sufficient nitrogen provision can ensure the synthesis of numerous non-protein compounds that participate in physiological and metabolic actions of crops, consequently reflecting in the yield and quality of crops<sup>1</sup>. In general, the total nitrogen mass within the top 15 cm of soils are between 0.1% and 0.6% of the soil weight, ranging between 2000 and 12000 kg-N ha<sup>-1</sup>, depending on the types of soil systems<sup>2</sup>. Despite the importance of nitrogen fertilization for growing crops, improper or excessive inputs of nitrogen fertilizers onto farmlands would pose adverse environmental impacts. It is well known that fertilizer applications are the major anthropogenic sources of nitrogenous gas emissions, such as ammonia (NH<sub>3</sub>), nitric oxide (NO), and nitrous oxide (N<sub>2</sub>O). Research has indicated that agricultural activities of fertilization and livestock production are the largest source of NH<sub>3</sub> (accounting for 80–90% of global anthropogenic emissions<sup>3</sup>), the major contributors to tropospheric NO (accounting for 10% of that<sup>4,5</sup>), and the largest anthropogenic source of N<sub>2</sub>O (accounting for 60–70% of that<sup>5</sup>). These nitrogenous gases are critical components in inducing regional- and/or global-scale changes in the atmospheric conditions, such as regional haze formation by NH<sub>3</sub> and NO, and global warming by N<sub>2</sub>O.

NH<sub>3</sub> is a prevailing atmospheric pollutant with a wide variety of adverse impacts. It can neutralize a large portion of acidic species, such as SO<sub>x</sub> and NO<sub>x</sub>, to form ammonium-containing aerosols. These aerosols constitute the major components of fine particulate matter (PM<sub>2.5</sub>), which causes air quality degradation and adverse impacts on human health. According to the estimates by Lelieveld, et al.<sup>6</sup>, the contribution of NH<sub>3</sub> emissions from global agriculture activities to PM<sub>2.5</sub> and associated premature mortality is approximately 20%. In 2014, the global NH<sub>3</sub> emissions from the use of synthetic N fertilizer and manure were 12.3 and 3.8 Tg-N per year, respectively<sup>7</sup>. Also, NH<sub>3</sub> eventually returns to the soils and surface waters through wet or dry deposition<sup>8</sup>, and thus leads to acidification, eutrophication, and biodiversity loss of natural ecosystems. Similarly, NO plays an important role in atmospheric

chemistry as it can catalyze the production of tropospheric ozone and other photochemical oxidants (e.g., nitric acid) in the atmosphere. It was estimated that the global NO emissions from soils were 21 Tg-N per year, with an error at ± 4–10 Tg-N per year<sup>9</sup>. The amounts of NO emissions from soils are generally low; however, prior to being converted into inert nitrogen, significant quantities of N<sub>2</sub>O could be formed under field conditions<sup>2</sup>.

N<sub>2</sub>O is a long-lived greenhouse gas (GHG) with a global warming potential of about 265–298 times greater than CO<sub>2</sub><sup>10</sup>. It can also lead to the depletion of the stratospheric ozone layer. The global N<sub>2</sub>O levels in the atmosphere has increased from 270 ppbv in 1750 to 332 ppbv in 2019<sup>11</sup>. Natural soil is one of the most important natural N<sub>2</sub>O sources (4.9–6.5 Tg-N per year during 2007–2016<sup>12</sup>), followed by emissions from the ocean source (at 2.5–4.3 Tg-N per year during 2007–2016<sup>12</sup>). For the anthropogenic sources, the agriculture sector shares the largest portion of global anthropogenic N<sub>2</sub>O emissions. In agriculture, the N<sub>2</sub>O sources include direct soil emissions from farmland fertilization, manure management, aquaculture, and agri-residue burning<sup>13</sup>. In 2007–2016, the average N<sub>2</sub>O emissions from fertilization on cropland and pasture, manure management, and aquaculture were estimated to range between 2.5 and 5.8 Tg-N per year<sup>12</sup>. It is noted that NH<sub>3</sub> emissions and the subsequent deposition could also contribute to an indirect source of agricultural N<sub>2</sub>O<sup>2</sup>. Other non-agricultural sources of N<sub>2</sub>O emissions include humans, biomass burning, and vehicles<sup>14</sup>.

For the sake of addressing reactive nitrogenous gases from croplands, both emission intensity and emission factor have been widely adopted to evaluate reactive nitrogen emissions from N fertilization. To build emission inventories, in this study, the emission intensity of reactive nitrogenous gases was calculated by subtracting background emissions ( $E_b$ , kg-N ha<sup>-1</sup>) from the total emissions ( $E_t$ , kg-N ha<sup>-1</sup>) per hectare of fertilized farmlands, as shown in Eq. (1). In accordance with the definition suggested by the Intergovernmental Panel on Climate Change (IPCC)<sup>15</sup>, the associated emission factor induced by N fertilization was

<sup>1</sup>Department of Bioenvironmental Systems Engineering, National Taiwan University, Taipei, Taiwan ROC. <sup>2</sup>Department of Environmental Engineering, National Ilan University, Ilan, Taiwan ROC. ✉email: sypan@ntu.edu.tw

**Table 1.** Facts and movements on reactive nitrogen emission reduction from agriculture in representative region or country.

Country/Region	Policy/Regulation	Remarks
European Union	Common Agricultural Policy	<ul style="list-style-type: none"> <li>Reduce anthropogenic air pollutants in Europe.</li> <li>The common agricultural policy is reformed in 2021</li> </ul>
	National Emission Ceilings directive	<ul style="list-style-type: none"> <li>Set the NH<sub>3</sub> ceiling from European countries</li> </ul>
	National Emission Reduction Commitments Directive (2016/2284/EU)	<ul style="list-style-type: none"> <li>Set up the emission reduction commitments for NH<sub>3</sub>, NO<sub>x</sub>, and PM<sub>2.5</sub> in 2020 and beyond.</li> <li>Propose a national advisory code of good agricultural practices</li> </ul>
	European Green Deal Communication	<ul style="list-style-type: none"> <li>Launch strategies and target plans on stepping up 2030 climate ambition.</li> <li>Revise the regulatory framework for the Land Use, Land Use Change and Forestry sector (EU, 2018/841)</li> </ul>
United State	Clean Air Act/National Ambient Air Quality Standard	<ul style="list-style-type: none"> <li>NH<sub>3</sub> is regulated.</li> <li>Reduce nitrogen losses</li> </ul>
	Agricultural Air Quality Conservation Measures	<ul style="list-style-type: none"> <li>Suggest a number of approaches to mitigating NO<sub>x</sub> emissions from agriculture</li> </ul>
Canada	Agri-environmental indicators	<ul style="list-style-type: none"> <li>Include agricultural NH<sub>3</sub>, GHGs, and PM indicators</li> </ul>
China	Air Pollution Action Plan	<ul style="list-style-type: none"> <li>Reduce NH<sub>3</sub> and NO<sub>x</sub> emissions by a number of clean production practices</li> </ul>
	Three-Year Action Plan for Winning the Blue-Sky Defense Battle	<ul style="list-style-type: none"> <li>Decline NO<sub>x</sub> emission by 15% by 2020 compared to 2015 levels</li> </ul>
	No. 1 Central Document and Five-Year Plan	<ul style="list-style-type: none"> <li>Emphasize balanced fertilization and the use of controlled-release fertilizers</li> </ul>
India	Intended Nationally Determined Contributions	<ul style="list-style-type: none"> <li>Propose various mitigation strategies for N<sub>2</sub>O from agricultural practices.</li> <li>Reduce the emission intensity of its GDP by 33–35% by 2030 to 2005 level</li> </ul>
Brazil	National Plan for Low Carbon Emission in Agriculture	<ul style="list-style-type: none"> <li>Implement low carbon agricultural practices</li> </ul>
Russia	The Federal Law of the Russian Federation No. 219-FZ	<ul style="list-style-type: none"> <li>Serve as a basis for deploying the Best Available Techniques</li> </ul>

expressed as a percentage of the emission intensity to the applied nitrogen ( $N_t$ , kg-N ha<sup>-1</sup>), as determined by Eq. (2). In the IPCC Guideline, the default values of emission factors for reactive nitrogen gases are reported with respect to different fertilizers at the global and regional scales<sup>15</sup>. At the national or city scales, extensive studies have been conducted to determine or refine the emission factors, and this review article tends to collect up-to-date inventories from the bottom-up level.

$$\text{Emission Intensity (kg - per ha)} = E_t - E_b \quad (1)$$

$$\text{Emission Factor (\%)} = \frac{E_t - E_b}{N_t} \times 100\% \quad (2)$$

The pathway to a low-emission scheme has been recently pledged by numerous countries and entities. Under this international movement, the emissions of reactive nitrogenous gases from the agriculture sector due to fertilization should be critically mitigated. The trade-offs between crop yields and nitrogenous gas emissions are an essential target for implementing green agricultural practices that aim to reduce the environmental costs while maintaining (or increasing) the associated crop yields. To the best of our knowledge, few attempts have been made to review different agricultural practices on all nitrogenous gas emissions (including NH<sub>3</sub>, NO<sub>x</sub> and N<sub>2</sub>O) from croplands. In this article, we first reviewed the international movements and progresses on this topic and discussed the key components of nitrogen fertilization, including nitrogen use efficiency and sampling techniques. Then, we evaluated the effect of crop types on NH<sub>3</sub>, NO<sub>x</sub>, and N<sub>2</sub>O emissions from the selected studies. For the inventory data of reactive nitrogen emissions, Supplementary Table 1 compiles the background information of the field experiments from the reviewed papers. We also attempted to address the trade-offs among reactive nitrogenous gases from the viewpoint of the nitrogen cycle in croplands. Lastly, we pointed out priority research directions from mitigating nitrogenous gas emissions

from farmlands to realizing a low-emission agriculture. This review should provide insights into the principles and practices of cutting nitrogen-containing gas emissions through more environmentally-friendly approaches.

## THE FACTS AND INTERNATIONAL MOVEMENTS ON MITIGATING NITROGENOUS GAS EMISSIONS

Globally, a number of international conventions, government policies and regulations have been pledged to address the abatement of nitrogenous gas emissions from agriculture. In this section, we provide an overview on the facts and movements of several representative countries in which agriculture occupied a significant portion in its economic structures (or the fertilizer consumption is relatively high in the world), as presented in Table 1).

### European union

In Europe, agriculture activities are under the guidance of the EU Common Agricultural Policy (CAP). Aside from the CAP, remarkable progress on the reduction of anthropogenic air pollutants in Europe has been realized over the past two decades due to the strong incentives initiated by related policies and regulations, such as the National Emission Ceilings directive (NEC, 2016/2284/EC). For instance, the EU Commission has set the NH<sub>3</sub> ceiling for European countries. In 2016, the National Emission Reduction Commitments Directive (2016/2284/EU) further set up the emission reduction commitments for NH<sub>3</sub>, NO<sub>x</sub>, and PM<sub>2.5</sub> in 2020 and beyond, based on the CLRTAP. Under this framework, each EU country needs to propose a national advisory code of good agricultural practices to control these emissions from agriculture<sup>16</sup>.

For NH<sub>3</sub>, it is estimated that 92% of the NH<sub>3</sub> emissions in 2017 came from the agriculture sector<sup>17</sup>. In particular, about 20% of the

NH<sub>3</sub> emissions in the EU were attributed to the use of mineral fertilizers<sup>18</sup>. Therefore, in the Directive, several potential solutions to reduce NH<sub>3</sub> emissions from mineral fertilizers were suggested. One of them is the replacement of urea-based fertilizers with ammonium-nitrate-based fertilizers. Permission granted for the use of urea-based fertilizers should reduce emissions by at least 30% in comparison with the reference method of urea-based fertilizer application<sup>1</sup>. Also, several studies indicated a significant contribution of agricultural emissions to PM<sub>2.5</sub> formation in Europe. For instance, in Germany, it is estimated that ~45% of premature mortality in 2010 is attributed to the PM<sub>2.5</sub> formation originated from agricultural NH<sub>3</sub> emissions<sup>6</sup>.

The agriculture sector in the EU also produces considerable amounts of NO<sub>x</sub> and N<sub>2</sub>O emissions. For NO<sub>x</sub>, the emission from agriculture was about 600 kt NO<sub>x</sub>, accounting for 8% of the total NO<sub>x</sub> emission in the EU<sup>17</sup>. For N<sub>2</sub>O, the total emission from agriculture in 2019 was estimated to be 627 kt N<sub>2</sub>O (i.e., 187 Mt CO<sub>2</sub>-eq), sharing about 43.6% of the total GHG emission from agriculture<sup>19</sup>. In 2020, the European Green Deal Communication launched strategies and target plans on stepping up 2030 climate ambition<sup>20</sup>, and revised the regulatory framework for the Land Use, Land Use Change and Forestry sector (EU, 2018/841)<sup>21</sup>. This covers the significant removal of N<sub>2</sub>O resulting from the management of land, forests, and biomass by 2030, and would contribute to the EU's target of emission reduction by 40% compared to 1990.

### The United State and Canada

The agriculture sector is the dominant source of NH<sub>3</sub> emissions in North America<sup>22,23</sup>. In 2018, almost 60% of NH<sub>3</sub> emissions in the US was attributed to agricultural livestock, and the second and the third largest categories were fertilizer application (21%), and agriculture fires and prescribed burning (5%), respectively<sup>22</sup>. Goebes, et al.<sup>24</sup> analyzed county-level monthly data, and reported that the total NH<sub>3</sub> emissions from fertilizer application in the US were estimated to be 590,000–761,000 metric tons, depending upon the emission factors. Another study by Ma, et al.<sup>7</sup> estimated that the NH<sub>3</sub> emission from the use of synthetic N fertilizer in the US was 1.05 Tg-N/year. In the US, the contribution of NH<sub>3</sub> emissions from agriculture to PM<sub>2.5</sub> and associated premature mortality was estimated to be ~29%<sup>6</sup>. In the US, NH<sub>3</sub> is still regulated under the Clean Air Act, and the national ambient air quality standard. Despite that fact, the USEPA recommended a number of ways to reduce nitrogen losses, such as adopting nutrient management techniques, using conservation drainage practices, ensuring year-round ground cover, and implementing conservation tillage.

For the agricultural NO source in the US, a few studies on the emission estimates from the state level have been reported, instead of the country level. For instance, Almaraz, et al.<sup>25</sup> estimated the agricultural NO<sub>x</sub> emissions in California, and found that about 0.16 Mt of NO<sub>x</sub>-N was annually emitted from soil systems, where croplands accounted for ~79% of total emissions. The associated average NO<sub>x</sub> emissions from cropland soils were 19.8 ± 27.3 kg-N per ha per year<sup>25</sup>. For N<sub>2</sub>O, soil management practices (such as fertilization) were the largest source of N<sub>2</sub>O emissions in the US, accounting for 75.4% (about 1156 kt-N<sub>2</sub>O) in 2019<sup>26</sup>. Also, a recent report by the United States Department of Agriculture<sup>27</sup> revealed that the primary GHG sources from agriculture were N<sub>2</sub>O emissions from cropped and grazed soils, which were estimated to be around 264 MMT CO<sub>2</sub>-eq. In October 2012, the USEPA published the Agricultural Air Quality Conservation Measures, suggesting several approaches to mitigating the NO<sub>x</sub> emissions from agriculture, such as equipment modifications. Most of these approaches were related to the direct emissions from the operations of wheels and machinery (e.g., engine combustion).

In Canada, NH<sub>3</sub> emissions have increased by 21% over the period from 1990 to 2018, mainly due to enhanced use of nitrogen fertilizers. NH<sub>3</sub> emissions in Canada are dominated by animal production, which made up 59% of the emissions in 2018, while crop production accounted for 35%. All other combined sources accounted for only 7% of emissions in 2018. Other sources include manufacturing, incineration and waste, and transportation and mobile equipment<sup>18</sup>. Similarly, the N<sub>2</sub>O emission from Canadian agriculture accounts for about half the warming effect of agricultural GHG emissions. During 2007–2016, the soil N<sub>2</sub>O emission in Canada was about 0.2 ± 0.1 Tg-N per year<sup>28</sup>. To measure the performance of the agriculture sector, the Government of Canada has developed a number of agri-environmental indicators, such as the agricultural NH<sub>3</sub> indicator, agricultural GHGs indicator, and PM indicator. Also, numerous management practices have been suggested to reduce the nitrogenous emissions from agriculture, such as adjusting fertilizer rates to coincide with plant needs.

### China

China has recently attached great attention to the development of clean air and green agriculture. In 2013, China implemented the “Air Pollution Action Plan” to reduce the emissions of NH<sub>3</sub> and NO<sub>x</sub> by a number of clean production practices, such as slow-release fertilizer<sup>29</sup>. In 2018, China issued the “Three-Year Action Plan for Winning the Blue-Sky Defense Battle” as the second phase of the 2013 action plan to further improve the air quality<sup>30</sup>. One of the set targets was to decline NO<sub>x</sub> emission by 15% by 2020, compared to 2015 levels. Recently, the promotion of green agriculture has been extensively emphasized in “No. 1 Central Document” and “Five-Year Plan”, with balanced fertilization and controlled-release fertilizers. China is the world's largest fertilizer consumer. In 2018, China imported about 10.6 million tonnes of fertilizer products for agricultural and industrial uses<sup>31</sup>. China is also the world's largest NH<sub>3</sub>-emitting country with annual emissions 3.0 and 2.7 times as much as those in the US and EU, respectively. A previous study indicated that total NH<sub>3</sub> emissions in China over the years of 2005–2008 exceeded the sum of those in the EU and US<sup>32</sup>. This huge amount of NH<sub>3</sub> emission has raised severe degradation of atmosphere quality, such as the formation of secondary PM. For instance, a study reported by Ye, et al.<sup>33</sup> indicated that the total mass of secondary ammonium, nitrate, and sulfate contributes to 25–60% of the total PM<sub>2.5</sub> formation in China. The contribution of NH<sub>3</sub> emissions from agriculture to PM<sub>2.5</sub> and associated premature mortality is estimated to be ~29%<sup>6</sup>.

Several studies in China have developed Chinese NH<sub>3</sub> emission inventories. For instance, Zhang, et al.<sup>34</sup> estimated the NH<sub>3</sub> emissions from agriculture with both top-down (satellite observations) and bottom-up (crop-specific fertilizer application practices with meteorological modulation) approaches. They found that both manure spreading and chemical fertilizer applications accounted for more than 80% of the total NH<sub>3</sub> emissions<sup>34</sup>. In their study, the annual agricultural NH<sub>3</sub> emissions were about 11.7 Tg (using data from the year 2008), where fertilizer application and livestock waste contributed to 43.2% and 45.4% of the total emissions, respectively. While mineral fertilizers are the major sources of NH<sub>3</sub> emission from agricultural soils, considerable uncertainties remain in the national estimates of fertilizer-induced emissions<sup>35</sup>. Xu, et al.<sup>36</sup> conducted a city-level inventory of agricultural fertilizer application based on activity data and regional emission factors. They indicated that the total NH<sub>3</sub> emissions from agricultural fertilizer in China was approximately 8.9–12.3 Tg-NH<sub>3</sub> per year, where livestock manure spreading and synthetic fertilizer use contributed 47.5% and 41.9% to the total emissions, respectively. Another study by Wu, et al.<sup>37</sup> developed both national and agro-region-specific models using high-resolution spatial data. In their study, the annual NH<sub>3</sub> emissions

from cropland were estimated to be 3.64–5.64 Tg  $\text{NH}_3\text{-N}$  ( $p < 0.05$ ; using One-sample  $t$ -test), where the cultivation of paddy rice, maize and wheat accounted for 44%, 20 and 16%, respectively. Despite these available studies, large uncertainties remain in the total  $\text{NH}_3$  emissions due to the significant seasonal variation of emissions which lack detailed activity data and emission factors<sup>38</sup>.

For agricultural  $\text{NO}$  emission, Wang, et al.<sup>39</sup> estimated that the annual  $\text{NO}$  emission from soils was about 657 Gg-N, and approximately 73.7% and 22.0% of the total  $\text{NO}$  emissions in July 1999 originated from arable lands and grasslands, respectively. Another study by Lu, et al.<sup>40</sup> estimated that the annual soil  $\text{NO}_x$  emissions above canopy in 2008–2017 were  $0.77 \pm 0.04$  Tg-N. For comparison, the total anthropogenic  $\text{NO}_x$  emissions, including power plant, industry, transportation, and residential processes, over China in 2010 were estimated to be 27.3 Tg per year (derived from MEIC v1.2)<sup>41</sup>. For the agricultural  $\text{N}_2\text{O}$  source, Gao, et al.<sup>42</sup> estimated the direct  $\text{N}_2\text{O}$  emission from paddy soils in China in 2007 was approximately 35.7 Gg  $\text{N}_2\text{O-N}$  per year, with an annual increase rate of 0.4% since 1980. During 2007–2016, the soil  $\text{N}_2\text{O}$  emission in China was about  $1.4 \pm 0.8$  Tg-N per year<sup>28</sup>.

### India

India is known as a land of agriculture where around two-third of the population relies on agriculture for their livelihood<sup>43</sup>. In fact, the agriculture sector in India contributes to approximately 20% of the nation's Gross Domestic Product in 2020<sup>44</sup>. After the Green Revolution to ensure the food sufficiency of the population, the use of mineral fertilizers intensively increased, which is a significant cause of reactive nitrogenous gas emissions from agricultural croplands. For instance, the  $\text{NH}_3$  emission from the use of synthetic N fertilizer in India was estimated to be 2.37 Tg-N in 2014, ranking as the second highest country worldwide<sup>7</sup>. Agricultural fertilization also made India the third largest emitting country of GHG following China and the US. As per the statistics of 2010, India's agricultural farmlands had a 7% contribution to the global agricultural GHG emissions. The state-wide  $\text{N}_2\text{O}$  emissions of Indian agricultural farmlands ranged between 0.18 and 9.11 kg  $\text{ha}^{-1}$  because of the N-fertilizer applications<sup>45</sup>. As per the International Maize and Wheat Improvement Centre (United Kingdom) report, India could cut down the agricultural GHG emissions by 18% (~94 Mt of  $\text{CO}_2\text{-eq}$  per year) through adopting three measures: efficient fertilizer use, avoidance of tillage, and irrigation management especially in paddy<sup>46</sup>. Under the United Nations Framework Convention on Climate Change, India's Intended Nationally Determined Contributions have proposed various mitigation strategies for  $\text{N}_2\text{O}$  from agricultural practices<sup>47</sup>. India has also announced an ambitious goal of reducing the emission intensity of its Gross Domestic Product by 33–35% by 2030, compared to the 2005 level<sup>48</sup>. This goal should be accomplished by a combination of strategies including transition to sustainable and climate-resilient agricultural systems.

### Brazil

Brazil, as one of the largest consumers of fertilizers around the world, consumed about 4.3 Tg-N of inorganic nitrogen fertilizers in 2018<sup>49</sup>. The N fertilizers in Brazil are mainly applied for sugarcane and corn, where urea shares ~50% of the total N fertilizers<sup>50</sup>. In the case of sugarcane yields, the emission factor of  $\text{NH}_3$  volatilization exhibited a wide range of 1–25% (corresponding to 80 to 100 kg-N  $\text{ha}^{-1}$  fertilization rate) during the warm and wet Brazilian summer<sup>51</sup>. Brazil is also the largest producer of soybeans in the world<sup>52</sup>; therefore, biological fixation represents a large source of reactive nitrogen in the Brazilian nitrogen cycle as leguminous crops (e.g., soybean) can fix inert N into reactive nitrogen through biological symbiosis. Our literature search identified no recent studies on the  $\text{NO}_x$  emission from agricultural fertilization in Brazil. For  $\text{N}_2\text{O}$  emission from farmlands, it is estimated that agriculture

activities contribute over 80% of the anthropogenic  $\text{N}_2\text{O}$  emissions in Brazil<sup>53</sup>. During 2007–2016, the soil  $\text{N}_2\text{O}$  emission in Brazil was about  $1.2 \pm 0.3$  Tg-N per year<sup>28</sup>. To alleviate the environmental impacts, the Brazilian Government announced the “National Plan for Low Carbon Emission in Agriculture” in 2010, especially implementing low carbon agricultural practices.

### Russia

In the early 1990s, nitrogen emissions from agriculture in Russia dropped remarkably due to the significant reduction in husbandry industries after the political-economic transition. For instance, at that time, the available nitrogen in manure (organic fertilizer) was reduced over 85%, and agricultural  $\text{NH}_3$  emission from husbandry was reduced by 60%<sup>54</sup>. Despite a sharp reduction in N-fertilizer uses, about 80% of the nitrogen input to agricultural land still currently comes from mineral fertilizers<sup>55</sup>. A recent study by Bartnicki and Benedictow<sup>56</sup> indicated that the contribution of agriculture to national  $\text{NH}_3$  emissions was more than 90%. Meanwhile, the  $\text{NO}_x$  emissions from agriculture in Russia can be neglected, compared to transportation and combustion (contribution of ~50% and ~45% to total emissions, respectively)<sup>56</sup>. For the  $\text{N}_2\text{O}$  emissions, agriculture has shared a significant portion of the total emissions. The agricultural  $\text{N}_2\text{O}$  emissions in 1994–1999 ranged between 84 and 130 kt- $\text{N}_2\text{O}$  per annum<sup>57</sup>.

## PRINCIPLES OF NITROGEN FERTILIZATION

Nitrogen fertilization is essential for plant growth and development as it controls the vital processes of respiration and photosynthesis. However, its scarcity in soils is one of the common challenges that affect the yield and quality of crops. Begara-Morales<sup>58</sup> addressed the importance of nitrogen fertilization as it not only is essential to nitrate reductase activity for N assimilation, but also can improve phosphorus (P) uptake by crops, especially in P-deficient soils and elevated  $\text{CO}_2$  concentration. Effective use of nitrogen is indispensable for both plant growth and environmental sustainability. In this section, we summarized the types of commonly used N-fertilizers, and then illustrated the nitrogen use efficiency (NUE) and their associated mechanisms in the nitrogen cycles.

### Type of N-fertilizers

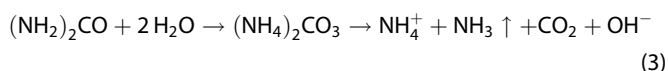
The sources of N-fertilizers include chemical (e.g., urea, urea-ammonium nitrate solution, and ammonium nitrate) and organic fertilizers (e.g., animal manure, compost, and digestate). Chemical N-fertilizers, most notably urea and ammonium nitrate, are synthesized by  $\text{NH}_3$  from the Haber–Bosch process. It is estimated by the Food and Agriculture Organization of the United Nations<sup>59</sup> that the global synthetic N-fertilizer supply is expected to exceed 163 Tg of  $\text{NH}_3\text{-N}$  per year by 2022. Urea ( $\text{CO}(\text{NH}_2)_2$ ) is reported to constitute about 50% of the total N-fertilizers consumption, followed by N-P-K compound (N) at ~14%. As shown in Table 2, urea is an organic amide rich in nitrogen content (up to 46%). After being applied to soil, the urea is hydrolyzed to form ammonium ( $\text{NH}_4^+$ ); therefore, urea is included in the category of mineral fertilizers. Urea-ammonium nitrate (UAN) is a liquid fertilizer produced from 73–78% urea and 93–97% ammonium nitrate solutions, which can be used for a wide range of soils<sup>1</sup>.

The types, doses, timings and methods of fertilizer applications highly relate to the long-term fertility and healthy conditions of soils. Several studies reported the negative impacts of long-term use of chemical fertilizers and/or overfertilization on soils. For instance, Guimarães, et al.<sup>60</sup> observed a reduction of soil pH over years of chemical N-fertilizer use, regardless of the application technique. Also, the soil pH would increase temporarily after the application of urea and/or animal urine. This is attributed to the hydrolysis of urea, which forms ammonium carbonate ( $(\text{NH}_4)_2\text{CO}_3$ )

**Table 2.** Type of fertilizers with their associated basic properties.

Category	Abbre.	Type	Chemical form	N content
Anhydrous ammonia	AA	Liquid	NH <sub>3</sub>	~82%
Urea	-	Dry Solid	CO(NH <sub>2</sub> ) <sub>2</sub>	38–46%
Ammonium bicarbonate	ABC	Dry Solid	(NH <sub>4</sub> )HCO <sub>3</sub>	~17%
Ammonium nitrate	AN	Dry Solid	NH <sub>4</sub> NO <sub>3</sub>	33–34%
Ammonium chloride	AC	Dry Solid	NH <sub>4</sub> Cl	24–25%
Ammonium sulfate	AS	Dry Solid	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	20–21%
Calcium ammonium nitrate	CAN	Dry Solid	5Ca(NO <sub>3</sub> ) <sub>2</sub> ·NH <sub>4</sub> NO <sub>3</sub> ·10H <sub>2</sub> O	20–26%
Calcium nitrate	CN	Dry Solid	Ca(NO <sub>3</sub> ) <sub>2</sub>	11–12%
Urea-ammonium-nitrate	UAN	Liquid	[CO(NH <sub>2</sub> ) <sub>2</sub> ] [NH <sub>4</sub> NO <sub>3</sub> ]	28–32%

that dissociates to produce ammonium, NH<sub>3</sub>, CO<sub>2</sub>, and hydroxide ions, as illustrated in Eq. (3):



According to the above equation, the urea application leads to the NH<sub>3</sub> emission from soils. In fact, different types of fertilizer and their associated application technique have different levels of risks on NH<sub>3</sub> volatilization. This is also highly dependent on the properties and conditions of soils. For instance, research found that the NH<sub>3</sub> emissions from urea and UAN displayed approximately 7 and 4 times higher, respectively, than that from ammonium nitrate<sup>1</sup>. Similarly, Cameron, et al.<sup>2</sup> reported that the NH<sub>3</sub> volatilization losses from urea, ammonium bicarbonate, and ammonium hydroxide fertilizers were higher than that from ammonium sulfate or diammonium phosphate fertilizers.

### Nutrient use efficiency

Farm N indicators are useful to compare farm performance among different farming systems. The loss of fertilizer nitrogen due to NH<sub>3</sub> or other nitrogenous gas (such as NO and N<sub>2</sub>O) emissions can significantly reduce N-fertilizer efficiency. In general, nitrogen use efficiency (NUE) and nitrogen surplus are the most used indicators for evaluating the environmental performance of fertilization, which can be determined by Eqs. (4) and (5), respectively.

$$\text{Nitrogen use efficiency (\%)} = (N_{\text{uptake}}/N_{\text{fer}}) \times 100\% \quad (4)$$

$$\text{Nitrogen surplus (kg - N per ha)} = \sum (N_{\text{inputs}}) - \sum (N_{\text{outputs}}) \quad (5)$$

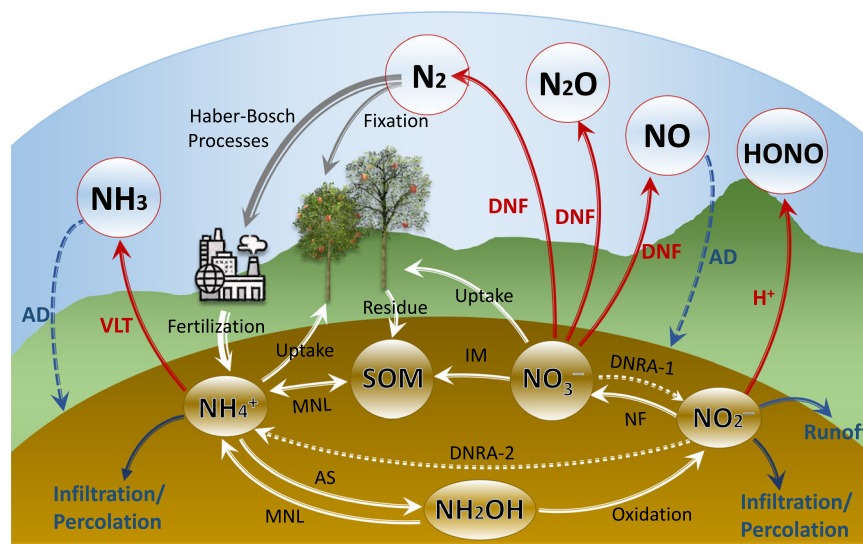
where  $N_{\text{uptake}}$  and  $N_{\text{fer}}$  represent the amount of N uptake by the above-ground crop (kg-N ha<sup>-1</sup>) and the applied N fertilizers (kg-N ha<sup>-1</sup>), respectively.  $N_{\text{inputs}}$  (kg-N ha<sup>-1</sup>) is the N inputs such as fertilization, atmospheric deposition, and irrigation water, and  $N_{\text{outputs}}$  (kg-N ha<sup>-1</sup>) is the N losses such as plant uptake, atmospheric emissions, surface runoff and infiltration. The NUE and N surplus should be cross-referenced from the perspectives of mass balance, with crop analyses such as nitrate content in crops. Several techniques, such as handheld sensors<sup>61</sup> and attenuated total reflectance—Fourier transform infrared spectroscopy<sup>62</sup> have been deployed for this purpose.

Supplementary Table 2 presents the characteristics of different crops with yields, the N uptake, the N surplus, and NUE reported in the literature. The results indicated that the NUE from urea alone in soil-plant systems barely exceeds 50% of the applied nitrogen. For paddy rice using urea alone, the NUE with the N-applied rate of 195 ± 28 kg-N ha<sup>-1</sup> ranged between 14.7% and 38.8% with an average value of 27.8% ± 3.4% ( $n = 8$ ,  $p < 0.05$ ; using One-sample  $t$ -test). For vegetables, the NUE exhibits a wide range between 5.0% and 67.3%, depending upon the types of crops and species.

Quemada, et al.<sup>63</sup> conducted quartile regression analyses on farm-level data ( $n = 1240$ ) from six European countries, and found that the NUE values for half of the arable farms ranged between 45 and 75%. For N surplus, the median value was approximately 68 kg-N ha<sup>-1</sup>. For the crop productivity, three quarters of the arable farms exhibited N outputs over 75 kg-N ha<sup>-1</sup>. They also found that arable lands generally exhibited a lower nitrogen surplus than livestock farms, thereby exhibiting a higher median NUE<sup>63</sup>. For different types of land use, grasslands were found to exhibit higher nitrogen losses than arable lands<sup>64</sup>.

Several studies have proven that the reduction of fertilization intensity could mitigate nitrogenous gas emission while maintaining the crop yields. For instance, Yao, et al.<sup>65</sup> indicated that reducing inputs of chemical fertilizers could increase the NUE and decrease the N surplus and nitrogenous gas emissions without sacrificing the crop yields. In fact, improving the crop's NUE is imperative for reducing the nitrogenous gas emissions from crop growing while obtaining a high yield. A number of green agricultural practices have been developed to increase NUE and crop yield, such as deep fertilization<sup>66</sup>, controlled-release fertilizers<sup>67</sup>, biochar-based fertilizers<sup>68</sup>, and modified-clay composite<sup>69</sup>. It is generally accepted that deep placement of fertilizers is critical for increasing the NUE. For instance, Zhao, et al.<sup>66</sup> found that, in paddy fields, deep fertilization could reduce N loss (20.9–24.8%) directly by the decreases of NH<sub>3</sub> volatilization and denitrification losses, and indirectly by affecting periphytic biofilm development. The development of periphytic biofilms could, despite increasing nitrification-denitrification loss, reduce NH<sub>3</sub> volatilization loss, and thus increase the overall N loss by 3.1–7.1%<sup>66</sup>. In addition, Li, et al.<sup>70</sup> suggested that for mechanical direct-seeded farms, one-time deep placement could effectively improve both the grain yield and NUE, and thus lower GHG emissions.

In terms of advanced or organic fertilizers, Lyu, et al.<sup>67</sup> indicated that the use of controlled-release fertilizers can improve NUE by 30.7–44.0%. Similarly, Puga, et al.<sup>68</sup> found that, compared to the conventional N fertilizers, fertilization (80 kg-N ha<sup>-1</sup>) via side-dressing application of biochar-based N fertilizers can result in a 12% increase in NUE and a 21% increase in corn productivity. Similarly, Mariano, et al.<sup>71</sup> indicated that the use of digestate (or liquid digestate) can replace the use of urea, while maintaining similar or even higher crop production. In addition, the co-application of natural humic substances could assist in increasing NUE while maintaining the crop yields. For instance, Leite, et al.<sup>72</sup> proposed foliar application of urea with humic substances or humic acids to enhance NUE in sugarcane, compared to using urea alone. This practice can induce changes in photosynthesis, intrinsic water use efficiency, and carbon and nitrogen metabolism. Similarly, Shen, et al.<sup>69</sup> developed a bentonite composite material with the interlayer modified by humic acids to enhance NUE. They found that, compared to the unmodified bentonite, the modified one can effectively reduce the nitrogen loss caused by



**Fig. 1 Mechanisms of nitrogen fertilization in soil systems and the associated nitrogen cycles.** VLT Volatilization, DNRA dissimilatory nitrate reduction to ammonia, MNL mineralization, AS assimilation, IM immobilization, NF Nitrification, AD atmospheric deposition.

$\text{NH}_3$  volatilization (by 10.9%) and  $\text{N}_2\text{O}$  emission (by 52.7%) from soil; meanwhile, the leaching loss of  $\text{NH}_4^+\text{-N}$  and  $\text{NO}_3^-\text{-N}$  in soil was much lower. This practice also successfully resulted in a greater yield and nitrogen uptake of wheat.

### Mechanisms and nitrogen cycles

Nitrogen fertilization plays an imperative role in the nitrogen cycle of the soil system, as shown in Fig. 1. Aside from fertilization, both atmospheric deposition<sup>8</sup> and biological fixation of inert nitrogen into reactive nitrogen through leguminous crops<sup>73</sup> (especially rhizobia or legume-associated bacteria) are other major sources of available nitrogen in soils. In the soil system, mineral nitrogen is mainly prone to losses through several pathways, including (i)  $\text{NH}_3$  emissions, (ii) leaching by surface runoff (e.g., removal in drainage water) or subsurface flow (e.g., to groundwater), (iii) denitrification into gaseous forms such as  $\text{N}_2$ ,  $\text{N}_2\text{O}$ , and  $\text{NO}_x$ , and (iv) transformation to nitrous acid (HONO).  $\text{NH}_3$  emissions from agriculture include two major sources: (i) crop foliage emission, and (ii) soil emission due to volatilization (see Eq. (6)). Usually,  $\text{NH}_3$  is deposited much closer to the emission source, and thus may cause eutrophication and acidification of nearby ecosystems.  $\text{NH}_3$  can also readily associate with acid cloud droplets (such as nitrate ( $\text{NO}_3^-$ ) and sulfate ( $\text{SO}_4^{2-}$ )) to form secondary inorganic aerosols, such as ammonium nitrate ( $\text{NH}_4\text{NO}_3$ ) and ammonium sulfites ( $\text{NH}_4\text{HSO}_4$  and  $[\text{NH}_4]_2\text{SO}_4$ ). The aerosols (e.g., containing  $\text{NH}_4^+$  particles) can travel over long distances prior to dry or wet deposition.



Nitrogenous oxides emitted from soils are primarily attributed to soil microbial processes, such as nitrification (the conversion of ammonium to nitrate) and denitrification (the conversion of nitrate to nitrogen). Autotrophic nitrification contains two steps, i.e., (i)  $\text{NH}_3$  oxidation, the rate-limiting step in the nitrification process, and (ii)  $\text{NH}_2\text{OH}$  oxidation into nitrite and/or nitrate.  $\text{NH}_3$  oxidation entails the conversion of  $\text{NH}_3$  into  $\text{NH}_2\text{OH}$  by aerobic ammonia-oxidizing bacteria (AOB) and ammonia-oxidizing archaea (AOA). It is noted that AOB is dominant in  $\text{NH}_3$  oxidation in neutral/alkaline or N-rich environment because of their high affinity to  $\text{NH}_3$ <sup>74</sup>. In contrast, AOA play an essential role in acidic or N-limited conditions. With sufficient fertilization, in the case of  $\text{N}_2\text{O}$  emissions, Fu, et al.<sup>75</sup> found that AOB-driven nitrification

should be the major pathway of soil  $\text{N}_2\text{O}$  emissions for both acidic and alkaline soils in paddy fields. The  $\text{N}_2\text{O}$  yields from an AOB-driven pathway in both soils (except in the acidic soil fertilized by ammonium-N) were higher than that from an AOA-driven one. Chen, et al.<sup>76</sup> developed an emission module based on the water and nitrogen management model, and they found that denitrification should be the dominant pathway contributing over 76% of the total  $\text{N}_2\text{O}$  emissions from soils. On the other hand, nitrification and nitrite chemical decomposition accounted for about 52 and 48% of the total  $\text{NO}$  emissions from soils, respectively.

Several recent studies<sup>77</sup> have highlighted the soil HONO emissions due to fertilization. HONO, a precursor of the hydroxyl radical, plays important roles in tropospheric chemistry, human health risk (could damage the respiratory system), and indoor air quality. Wu, et al.<sup>78</sup> noticed that soil reactive nitrogen gas (including HONO) emissions are mainly driven by nitrification and denitrification, which are highly relevant to soil pH, inorganic N content, and microbiological mechanisms. Despite these available studies, a large unknown source of atmospheric HONO (especially during the daytime)<sup>79</sup> and the complex biogeochemical reactions for soil HONO emissions are still not clearly elucidated.

### AMMONIA EMISSION

Agricultural activities are recognized as the major sources of atmospheric  $\text{NH}_3$  in numerous counties, e.g., even contributing up to 96% of national anthropogenic  $\text{NH}_3$  emissions<sup>16</sup>. In this section, we summarized the regulating factors, intensities, and available management practices for  $\text{NH}_3$  emissions from agricultural farmlands.

### Regulating factors for $\text{NH}_3$ emissions

Agricultural  $\text{NH}_3$  emissions have two major sources: (i) crop foliage emission, and (ii) soil emission by volatilization. For the crop foliage emission,  $\text{NH}_3$  is emitted from crop leaves when the internal  $\text{NH}_3$  concentration is relatively higher than that in the surrounding atmosphere. This often occurs during the periods with rapid nitrogen sorption by the roots or senescence inducing N-remobilization from leaves. Sommer, et al.<sup>80</sup> indicated that about 1–4% of shoot nitrogen may be lost through this way. Also,

**Table 3.** Effect of farming systems and management on NH<sub>3</sub> volatilization losses from farm lands.

Category	Factor	Parameter	Factor favors NH <sub>3</sub> volatilization
Agricultural practices	Fertilization	<ul style="list-style-type: none"> <li>• Crop selection and its dose level.</li> <li>• Types of fertilizer (e.g., granulated fertilizers).</li> <li>• Modification to fertilizer</li> </ul>	<ul style="list-style-type: none"> <li>• Timing of applying urea or animal urine<sup>2</sup>.</li> <li>• Urea and animal urine favor volatilization<sup>2</sup></li> </ul>
	Method of application	<ul style="list-style-type: none"> <li>• Broadcasting vs subsurface application.</li> <li>• Deep injection.</li> <li>• Spraying or sprinkling</li> </ul>	<ul style="list-style-type: none"> <li>• Recommended depth for injection is 3–5 cm<sup>2</sup></li> </ul>
	Cultivation system	<ul style="list-style-type: none"> <li>• Clean tillage/No-tillage.</li> <li>• Mulch/Sward.</li> <li>• Fallow rotation.</li> <li>• Type of land-use</li> </ul>	<ul style="list-style-type: none"> <li>• No-tillage farming favors<sup>1</sup>.</li> <li>• Crop residue on the soil surface favors<sup>89</sup></li> </ul>
Soil properties	pH	<ul style="list-style-type: none"> <li>• Fertilizer hydrolysis.</li> <li>• Nitrification by bacteria</li> </ul>	<ul style="list-style-type: none"> <li>• High soil pH favors (especially calcareous soils)<sup>80</sup></li> </ul>
	Fertility	<ul style="list-style-type: none"> <li>• Nitrogen content (or total ammoniacal nitrogen).</li> <li>• Organic matter (soil humus layer, buffering and sorption ability).</li> <li>• Cation exchange capacity (CEC)</li> </ul>	<ul style="list-style-type: none"> <li>• Initial mineral nitrogen content in soil should be considered<sup>84</sup>.</li> <li>• Low clay and organic matter favors<sup>89</sup>.</li> <li>• Low CEC favors<sup>90</sup></li> </ul>
	Moisture	<ul style="list-style-type: none"> <li>• Texture (drainage)</li> </ul>	<ul style="list-style-type: none"> <li>• Initially moist soil followed by drying<sup>89</sup></li> </ul>
	Microbials	<ul style="list-style-type: none"> <li>• The mobility and availability of nitrogen.</li> <li>• Nitrification/Denitrification processes</li> </ul>	<ul style="list-style-type: none"> <li>• Introducing biofertilizers<sup>87</sup> or mixed microorganisms<sup>88</sup> could reduce volatilization</li> </ul>
	Meteorological conditions	Precipitation	<ul style="list-style-type: none"> <li>• Rainfall.</li> <li>• Humidity</li> </ul>
Meteorological conditions	Temperature	<ul style="list-style-type: none"> <li>• Solar radiation.</li> <li>• Seasonal effect</li> <li>• Daily temperature difference</li> </ul>	<ul style="list-style-type: none"> <li>• High temperature (e.g., the peak temperature of the day) favors<sup>2,14</sup></li> </ul>
	Wind speed	<ul style="list-style-type: none"> <li>• Wind speed/Wind erosion</li> </ul>	<ul style="list-style-type: none"> <li>• High wind speed favors<sup>1</sup></li> </ul>

Cameron, et al.<sup>2</sup> found that senescent leaves exhibit a large potential for foliage NH<sub>3</sub> emission.

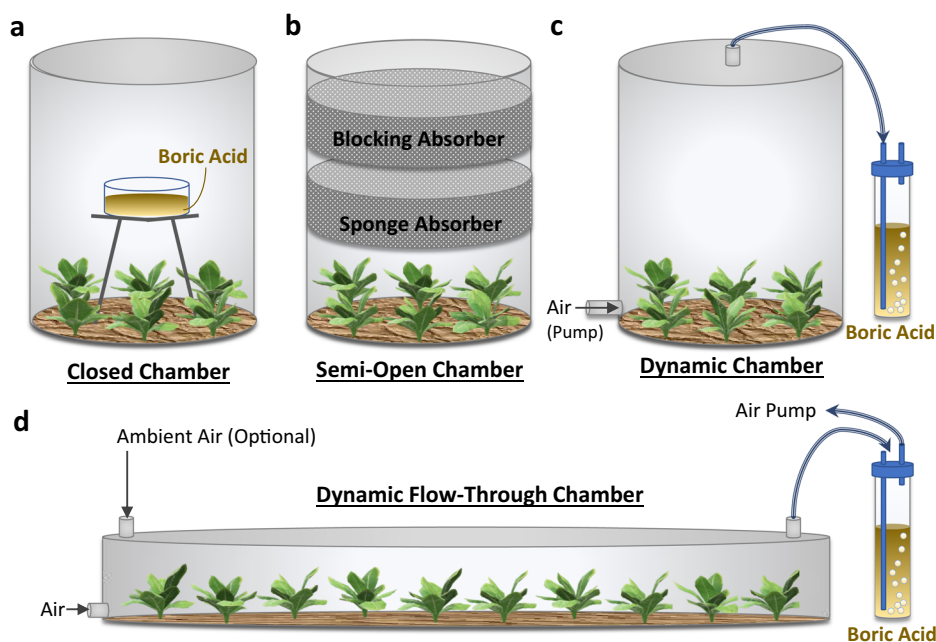
NH<sub>3</sub> soil emission due to volatilization is affected by factors involving agricultural practices, soil physico-chemical properties, and meteorological conditions. In general, NH<sub>3</sub> volatilization occurs due to N-fertilization, application of manure, and volatilization of soil organic matter and plant residues<sup>2</sup>. Sommer, et al.<sup>80</sup> indicated that the proportion of nitrogen loss due to NH<sub>3</sub> volatilization may exceed 50% of the total N fertilizers applied. For the NH<sub>3</sub> soil emission potential, Table 3 presents the factors that influence the effectiveness of fertilization and the NH<sub>3</sub> emission intensity from the perspectives of agricultural management practices, soil physico-chemical properties, and meteorological conditions.

Factors related to agricultural practices include the methods of application, fertilization, and cultivation system. For instance, urea fertilizer application at the surface would increase the soil NH<sub>3</sub> emission potential in the following days. Huang, et al.<sup>81</sup> indicated that deep fertilizer placement would increase crop yields while reducing NH<sub>3</sub> emission intensity. Klimczyk, et al.<sup>1</sup> also found that covering the urea with soil immediately after the application could effectively reduce the emissions by up to 80%. Splitting fertilizer applications (i.e., spreading N fertilizer applications over a time span) would also increase crop yields and reduce NH<sub>3</sub> emission intensity<sup>81</sup>. For liquid fertilizers, Bai, et al.<sup>82</sup> indicated that irrigation water with anhydrous NH<sub>3</sub> would contribute to a higher level of NH<sub>3</sub> emission (e.g., 0.79 ± 0.09 kg-N ha<sup>-1</sup> d<sup>-1</sup>) than broadcasting urea (e.g., -0.06 ± 0.02 kg-N ha<sup>-1</sup> d<sup>-1</sup>). Mencaroni, et al.<sup>83</sup> found that closed-slot injection could reduce NH<sub>3</sub> emissions for both chemical and organic fertilizers. Compared to surface broadcast, for instance, injected application with ammonium nitrate or organic fertilizers could reduce NH<sub>3</sub> emissions in maize by 75 and 96%, respectively, and in winter wheat by 87 and 98%<sup>83</sup>. Similarly, Mariano, et al.<sup>71</sup> observed a higher NH<sub>3</sub> emission rate when

digestate was applied directly to the surface of soils, compared to both urea application and digestate injection.

Factors related to soil physico-chemical properties include the pH, total ammoniacal nitrogen, organic matter, cation exchange capacity, moisture, and microbials. The pH and buffer capacity of the soil and dissolved fertilizer salts are dominant factors controlling NH<sub>3</sub> emission<sup>80</sup>. A naturally high pH (e.g., alkali or calcareous soils) could produce significant amounts of NH<sub>3</sub> emission, especially when urea or animal urine is applied. The NH<sub>3</sub> emission from soils is also related to the concentration of total ammoniacal nitrogen (TAN) in soils. In general, a higher TAN concentration in soils can lead to a higher rate of NH<sub>3</sub> emission. Yang, et al.<sup>84</sup> found that the dynamics of NH<sub>3</sub> emissions in the case of a rice field were mainly affected by the NH<sub>4</sub><sup>+</sup> concentrations of the soil-surface water. Similarly, Shan, et al.<sup>85</sup> indicated that NH<sub>3</sub> volatilization exhibits significantly positive correlation with the increases of both the pH and NH<sub>4</sub><sup>+</sup> concentrations in the top layer of soils. In fact, a number of factors, including types of fertilizers, soil nitrification-denitrification rate, plant uptake rate, and N-immobilization rate, can affect the TAN concentration in soils. For instance, application of NH<sub>3</sub>-based fertilizer, such as urea and animal urine, significantly increases the potential of NH<sub>3</sub> emission<sup>2</sup>. Several studies indicated that replacing a portion of NH<sub>3</sub>-based fertilizers with organic fertilizers could effectively change the soil conditions and thus decrease the NH<sub>3</sub> emission. Dai, et al.<sup>86</sup> found that, after replacing mineral N-fertilizer with organic N-fertilizer, potential nitrification rate increased significantly ( $p < 0.05$ ; using One-sample *t*-test) with the increasing substitution ratio of organic fertilizers in paddy soils. The mobility and availability of nitrogen will also affect the nitrification and denitrification processes, and thus the microbial activity. Several studies have reported that introducing biofertilizers<sup>87</sup> or mixed microorganisms<sup>88</sup> could reduce NH<sub>3</sub> volatilization.

Soil organic matter in humus possesses the multi-functions of stabilizing the soil structure and minimizing the risk of soil



**Fig. 2** Several examples of chambers for sampling and measuring  $\text{NH}_3$  volatilization. **a** Closed static chamber; **b** semi-open chamber; **c** dynamic chamber-capture system; **d** dynamic flow-through chamber.

erosion<sup>1</sup>. It is found that soils with a high organic matter content exhibit a greater ammonium sorption capacity, thereby reducing nitrogen losses by  $\text{NH}_3$  volatilization<sup>89</sup>. Similarly, a higher cation exchange capacity (CEC) could retain ammonium ions on the surface of soil clays and organic matters through electrostatic attraction, thereby reducing the concentration of available ammonium in soils. A higher CEC could also enhance the buffering capacity and thus help the soil against pH change. In practice, information of basic soil properties can be used to map the soil  $\text{NH}_3$  emission potential, regardless the fertilization practices. For instance, Duan and Xiao<sup>90</sup> proposed the classification of  $\text{NH}_3$  emission potentials by identifying the pH and CEC threshold levels as follows:

- Very low ( $\text{pH} < 7$ ;  $\text{CEC} \geq 20 \text{ cmol kg}^{-1}$ ):  $\text{NH}_3$  volatilization  $< 0.10 \text{ cmol kg}^{-1}$ .
- Low ( $\text{pH} < 7$ ;  $\text{CEC} < 20 \text{ cmol kg}^{-1}$ ):  $\text{NH}_3$  volatilization ranges from  $0.10\text{--}0.20 \text{ cmol kg}^{-1}$ .
- Medium ( $7 \leq \text{pH} < 8$ ;  $\text{CEC} > 10 \text{ cmol kg}^{-1}$ ):  $\text{NH}_3$  volatilization ranges from  $0.20\text{--}1.00 \text{ cmol kg}^{-1}$ .
- High ( $7 \leq \text{pH} < 8$ ;  $\text{CEC} < 10 \text{ cmol kg}^{-1}$ ):  $\text{NH}_3$  volatilization ranges from  $0.60\text{--}1.00 \text{ cmol kg}^{-1}$ .
- Very high ( $\text{pH} \geq 8$ ;  $\text{CEC} < 10 \text{ cmol kg}^{-1}$ ):  $\text{NH}_3$  volatilization  $> 1.00 \text{ cmol kg}^{-1}$ .

Several studies applied these criteria to identify the regions with a high  $\text{NH}_3$  emission potential. For instance, Mencaroni, et al.<sup>83</sup> mapped the  $\text{NH}_3$  emission potential of the Veneto region in northeast Italy. It should be noted that the criteria of the presented  $\text{NH}_3$  emission potential is independent of the agricultural practices and fertilizations.

Several meteorological factors, such as temperature, wind speed and precipitation, will interact with  $\text{NH}_3$  emission potentials. For instance, a higher temperature will increase the rate of  $\text{NH}_3$  transfer from the soil into the atmosphere;  $\text{NH}_3$  emissions from the nitrogen fertilizer application were found to generally peak at the time of highest daily temperature<sup>2</sup> or during the summer<sup>14</sup>. Similarly, Yang, et al.<sup>84</sup> found that solar radiation was the dominant factor, especially during the rice panicle formation stage, for intra-day  $\text{NH}_3$  emissions. In addition, significant intensities of irrigation or rainfall right after urea-based

applications could reduce the  $\text{NH}_3$  emissions. The introduced water (either rainfall or irrigation) can hydrolyze the urea into ammonium and transport them below the surface of the top soils, thereby keeping the  $\text{NH}_3$  concentration at surface low.

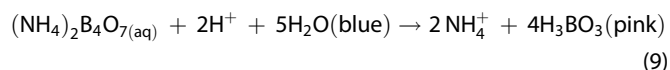
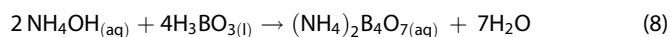
### Sampling of $\text{NH}_3$ emissions from farmlands

Sampling techniques determine the precision and accuracy of observations for the reactive nitrogen emissions from farmlands. The design of the  $\text{NH}_3$  sampling system is particularly important as ammonia is highly soluble and different designs affect the sensitivity and representativeness of emission intensity. In fact, it is still difficult to accurately measure the  $\text{NH}_3$  emission from agricultural sources. To date, a few  $\text{NH}_3$  sampling techniques have been developed, such as the dynamic chamber-capture system<sup>91,92</sup>, passive sampler<sup>93,94</sup>, static chamber with absorbents<sup>95</sup>, and absorptive sponges<sup>96</sup>. Figure 2 shows several examples of chambers, such as closed static chamber, semi-open chamber, and dynamic chamber-capture system for sampling and measuring  $\text{NH}_3$  volatilization. In the case of using an air pump to actively introduce the gas in the chamber to the boric acid solution, one should note that the amount of air inputs should be designated to be approximately the volume of the chamber. Several designs include a pressure gauge on the top of the chamber cap to ensure that the pressure within the chamber maintains a positive pressure throughout the sampling procedure. Otherwise, the gases within the voids and/or pores of soils would flow out to interfere with the determination of the real emission intensity. In addition, several studies have indicated the underestimation of  $\text{NH}_3$  concentrations by passive samplers, and thus their effective sampling rate should be corrected by both theoretical and practical approaches (e.g., mass transfer correction factor)<sup>94</sup>.

In general, the  $\text{NH}_3$  sampling system introduces and dissolves  $\text{NH}_3$  into the boric acid ( $\text{H}_3\text{BO}_3$ ) solution with sufficient contact times, as described by Eqs. (7–8).  $\text{NH}_3$  captured in boric acid is then measured via titration with the  $\text{H}_2\text{SO}_4$  solution using an indicator of bromocresol green and methyl red, as described by Eq. (9). These procedures are well known as the Kjeldahl titration method, which is used to quantify the nitrogen content in food and soils. Therefore, the sensitivity of the Kjeldahl titration on the



NH<sub>3</sub> concentration in the ambient air should be critically evaluated as the original Kjeldahl method is used for a relatively high concentration of NH<sub>3</sub><sup>91</sup>.



### Emission intensity of different crops

Supplementary Table 3 compiles the NH<sub>3</sub> emission intensity and emission factor for different crops in the literature. Across the collected literature, the NH<sub>3</sub> emission intensities from all fertilized treatments exhibits a wide range between 0.5 and 172 kg-N ha<sup>-1</sup>, corresponding to the emission factors of 0.3–34.0%, depending upon the types of crops, species and fertilizers. The average NH<sub>3</sub> emission factor is 12.5 ± 1.5% ( $n = 29$ ;  $p < 0.05$ ; using One-sample  $t$ -test). The available data of NH<sub>3</sub> emission factor reported in the literature are quite different. For instance, Ma, et al.<sup>7</sup> reported the worldwide average NH<sub>3</sub> emission factor of 12.6% and 14.1% ( $n = 324$ ) for synthetic fertilizer and manure, respectively. Another evaluation by Mikkelsen<sup>89</sup> suggested a state-wide average NH<sub>3</sub> emission factor of 2.4% for all types of N fertilizer applications.

For rice in paddy fields, the average NH<sub>3</sub> emission intensity with the N-applied rate of 243 ± 20 kg-N ha<sup>-1</sup> is 32.0 ± 4.5 kg-N ha<sup>-1</sup> ( $n = 9$ ,  $p < 0.05$ ; using One-sample  $t$ -test), regardless the types of fertilizers. This emission intensity corresponds to a NH<sub>3</sub> emission factor of 14.4 ± 3.0%. Rice generally had greater increases in NH<sub>3</sub> emission intensities and emission factors in response to inorganic N addition (with increasing proportions of basal N, as well as soil organic carbon and total nitrogen) than other crops<sup>81</sup>. Huang, et al.<sup>81</sup> found that fertilizer-induced NH<sub>3</sub> emission intensities and emission factors for rice paddies were significantly higher than those for upland crops. Several studies have revealed that replacing a portion of urea with organic fertilizers could reduce the NH<sub>3</sub> emissions in paddy fields. For instance, Li, et al.<sup>97</sup> applied a mixture of biogas slurry and hydrothermal carbonization aqueous phase to replace urea. In their study, they found that the NH<sub>3</sub> volatilization from rice plant soils can be reduced by up to 65.5%.

For vegetables and fruits, the average NH<sub>3</sub> emission intensity with the N-applied rate of 237 ± 28 kg-N ha<sup>-1</sup> is 36.0 ± 7.8 kg-N ha<sup>-1</sup> ( $n = 29$ ,  $p < 0.05$ ; using One-sample  $t$ -test), corresponding to an emission factor of 12.3 ± 1.6%. It is also found that, among all the studied fruits and vegetables, banana exhibits the highest NH<sub>3</sub> emission intensity ranging between 100 and 172 kg-N ha<sup>-1</sup>, probably due to its high N-applied ratio of around 500 kg-N ha<sup>-1</sup>. Cabbages and fruits (such as pineapple and peach) also were found to have relatively high NH<sub>3</sub> emissions. Similarly, application of slow release fertilizers could effectively reduce the NH<sub>3</sub> emissions; for instance, the NH<sub>3</sub> emissions for peach with a fertilization rate of 436.4 kg-N ha<sup>-1</sup> could be reduced significantly from 77.2 to 36.9 kg-N ha<sup>-1</sup> if the urea-based composite fertilizers were replaced with bag-controlled release fertilizers<sup>95</sup>.

For the effect of different types of fertilizers on NH<sub>3</sub> volatilization, the collected information in this study (Supplementary Table 3) was further categorized into three groups: (i) urea, (ii) organic fertilizers, and (iii) urea with urease inhibitors or slow release fertilizers. As shown in Supplementary Fig. 1, the results indicated the NH<sub>3</sub> emission factor for the urea group was 13.7 ± 2.2% ( $n = 15$ ,  $p < 0.05$ ; using One-sample  $t$ -test), while the factors for organic fertilizers, and urea with nitrification inhibitors or slow release fertilizers were similar (at about 12.5%).

### Practical methods of reducing ammonia emissions

The levels of NH<sub>3</sub> emissions from farmlands are highly dependent on fertilization practices, soil properties (soil-water chemistry), and meteorological conditions. From the chemistry viewpoint, several practical methods have been developed to reduce NH<sub>3</sub> emissions from agricultural farmlands, such as

- Applying improved fertilization techniques: subsurface applications, deep injection (e.g., anhydrous NH<sub>3</sub>), urea fertilizer application before the onset of rain, and irrigation after the urea fertilization<sup>2</sup>.
- Using slow release fertilizers (or controlled-release fertilizers)<sup>98</sup>, or N fertilizers with a urease inhibitor coating<sup>2</sup>.
- Introducing biofertilizers (such as *Bacillus subtilis*<sup>87</sup>), modified composite materials in soil systems<sup>69</sup>, or mixed microorganisms<sup>88</sup>.

The method and timing of fertilizer application affect NH<sub>3</sub> volatilization remarkably, especially for urea-based fertilizers. Several improved fertilization practices have been recommended to reduce the NH<sub>3</sub> emission, such as subsurface applications (including deep injection of liquid fertilizers) before the onset of rain or introducing irrigation water right afterwards. In addition to improved fertilization techniques, another reliable approach is to use slow release fertilizers (or controlled-release fertilizers) and urease/nitrification inhibitors. This can significantly reduce the intensity of NH<sub>3</sub> emissions<sup>81</sup>.

Slow release fertilizers employ organic polymer materials (such as thermoplastics and resins) or acidifying minerals (such as sulfur) as the coating or encapsulation of urea granules. The coating layers can serve as a physical barrier, and thus gradually release the nutrients. Shan, et al.<sup>85</sup> conducted a 3-year field trial for cabbage cultivation using different types of slow release fertilizers. In their study, compared to conventional urea fertilizer, the NH<sub>3</sub> volatilization using sulfur-coated urea, biological Carbon Power® urea, and bulk-blend controlled-release fertilizer were significantly reduced by 60.7–68.8%, 71.9–79.0%, and 77.7–83.1%, respectively.

In urea applications without incorporation by machinery, rainfall, or irrigation, a significant quantity of nitrogen loss by NH<sub>3</sub> volatilization occurs through enzymatic hydrolysis of urea by urease. This can be effectively reduced by coating urea with a stabilizer, including (i) a urease inhibitor, such as N-(*n*-butyl) thiophosphoric triamide (NBPT)<sup>99</sup> and phosphorodiamidate (PPDA)<sup>100</sup>; (ii) a nitrification inhibitor, such as dicyandiamide (DCD)<sup>71</sup>, 3,4-dimethylpyrazole phosphate (DMPP)<sup>101</sup>, and nitrapyrin<sup>102</sup>. For instance, Yang, et al.<sup>99</sup> used NBPT as a urease inhibitor, and found that NH<sub>3</sub> volatilization from paddy fields was reduced by 61.1–63.6%. Urease inhibitors can effectively reduce the rate of urea hydrolyzation by deactivating the urease enzyme. In a meta-analysis performed by Silva, et al.<sup>103</sup>, NH<sub>3</sub> emissions accounted for about 30% of surface-applied urea-N in tropical and temperate soils, and this ratio can be further reduced to 14.8% in the case of NBPT-treated urea. In addition, application of biofertilizers in replacing conventional N fertilizers has gained great attention to reduce NH<sub>3</sub> emission from crop lands. For instance, Sun, et al.<sup>87</sup> applied *Bacillus subtilis* biofertilizer for the leafy vegetables, and they found that biofertilizers could effectively reduce NH<sub>3</sub> volatilization by 71%, compared with conventional fertilization.

### NITROGEN OXIDE EMISSIONS

Soil nitrogen oxides are produced by microbial reactions through nitrification and denitrification primarily due to the nitrogen fertilization. In this section, we summarize the emission intensity and factor for nitrogen oxides, including nitric oxide (NO) and nitrous oxide (N<sub>2</sub>O), and illustrate available management practices for NO and N<sub>2</sub>O emissions from agricultural farmlands.

## Regulating factors for nitrogen oxide emissions

Soil microbial processes of nitrification and denitrification are dependent on various factors, such as agricultural management practices (such as fertilizer type and rate), soil physico-chemical properties (such as fertilizer type and rate), and meteorological conditions (such as temperature and moisture). With respect to agricultural practices, soil NO and N<sub>2</sub>O emissions generally increase with the increase of N-applied rate. In fact, the mechanisms and their correlations are quite complex; Yao, et al.<sup>65</sup> observed that these trace emissions may exhibit a non-linear threshold response to the N-applied rate of fertilizers. Similarly, You, et al.<sup>104</sup> conducted a global-scale meta-analysis to explore the effect of N addition on the N functional genes and N fertilizer-induced N<sub>2</sub>O emissions in croplands. They found that the functional genes that encode enzymes involved in nitrification (AOA and AOB) and in the transformation of N<sub>2</sub>O to N<sub>2</sub> (i.e., *nosZ*) were the major mechanisms for N<sub>2</sub>O emissions<sup>104</sup>. FAO<sup>105</sup> also indicated that subsurface application or injection of nitrogen fertilizers generally resulted in higher N<sub>2</sub>O emissions (but lower NO formations), compared to broadcasting synthetic fertilizers and manure.

For the soil properties, several studies indicated that the contents of soil inorganic nitrogen, oxygen, and water should be the dominant factors driving NO and N<sub>2</sub>O emissions<sup>101,106</sup>. The concentrations of both readily bioavailable organic carbon and inorganic nitrogen in soil are also positively correlated to the denitrification rate. For instance, Cameron, et al.<sup>2</sup> introduced organic carbon to the soil systems, and found that the microbial growth (especially soil denitrifiers) and respiration were stimulated and enhanced, respectively. Maaz, et al.<sup>107</sup> also suggested that N<sub>2</sub>O emissions would increase by ~5% with a unit increase in soil organic carbon (%) for a given N-fertilization rate. For the water content, when soils with water contents are below the field capacity, N<sub>2</sub>O would be produced predominantly through nitrification. However, when the soil water content is above the field capacity, N<sub>2</sub>O is generated predominantly through denitrification. For the NO production, it would exceed the N<sub>2</sub>O production especially when soil water contents are below field capacity. For the soil pH, acidic soils with pH < 5.0 would exhibit slower denitrification rates than neutral pH soils, resulting in higher N<sub>2</sub>O emission intensities.

For the meteorological conditions, several studies have indicated that the emissions of N<sub>2</sub>O and NO are significantly correlated with soil temperature<sup>108</sup>. For instance, Pang, et al.<sup>109</sup> found that soil temperature should be the most significant factor in controlling NO emission, followed by fertilization intensity and gravimetric soil water content, according to the results of multiple linear regression analysis.

## Sampling of nitrogen oxide emissions from farmlands

NO<sub>x</sub> and N<sub>2</sub>O emissions from a farmland can be collected by chambers presented in Fig. 2, mostly by a sealed static chamber method<sup>110</sup>. In particular, N<sub>2</sub>O sampling should reference the chamber-based trace gas flux measurements protocols suggested by the USDA<sup>111</sup>, and/or to the new procedure implemented in an R package<sup>112</sup>. The gas sample is usually collected with a plastic syringe, or stored in a gas sampling bag prior to analysis. The concentrations of NO<sub>x</sub> and N<sub>2</sub>O are analyzed via a gas chromatograph, usually equipped with an electron capture detector.

## Emission inventory

Substantial NO emissions can be incurred by N fertilizer application during the growing periods. As presented in Supplementary Table 4, the NO emission intensity from all fertilized treatments exhibits a wide range between between

0.06 and 39.8 kg-N ha<sup>-1</sup> across the collected literature. Regardless the types of fertilizers, the average NO emission intensity is found to be 4.04 ± 1.30 kg-N ha<sup>-1</sup> ( $n = 37$ ,  $p < 0.05$ ; using One-sample  $t$ -test), corresponding to the emission factor of 0.32 ± 0.10%. For the paddy rice, the average NO emission intensity and factor with the applied-N ratio of 203 ± 23 kg-N ha<sup>-1</sup> are 0.16 ± 0.04 kg-N ha<sup>-1</sup> and 0.06 ± 0.01% ( $n = 5$ ,  $p < 0.05$ ; using One-sample  $t$ -test), respectively. Unlike the NH<sub>3</sub> emissions, the NO emissions of rice from paddy fields were relatively lower compared to other vegetables and fruits. For vegetables and fruits, the average NO emission intensity was found to be 4.64 ± 1.48 kg-N ha<sup>-1</sup> ( $n = 32$ ,  $p < 0.05$ ; using One-sample  $t$ -test), depending on the types and species of crops. This corresponded to the average NO emission factor of 0.40 ± 0.13%, which was six-fold higher than that of paddy rice. Crops, such as garlic, cabbage, radish, tomato, and cucumber, could emit intensive NO from soils during their growth.

NO emissions generally increase with the increase of chemical N fertilizer application rates. Macdonald, et al.<sup>113</sup> found that the concentration of available mineral N appeared to be an important driver of NO<sub>x</sub> emission in the case of sugarcane fields. As aforementioned, several soil properties play an important role in soil NO emission. Das, et al.<sup>92</sup> indicated a significantly positive correlation ( $p < 0.01$ ; using One-sample  $t$ -test) between NO flux and soil pH, i.e., the NO flux tend to peak near a soil pH of 7. Also, the NO flux during the day time were much higher than the night time counterparts, indicating that NO emissions should be maximized during the day time when the temperatures of both soils and ambiance remain high<sup>92</sup>. For soil water content, Lan, et al.<sup>114</sup> found that, under aerobic conditions of 60% water holding capacity, the ammonium-N pool via nitrification should be the dominant source of NO in paddy soils. However, another study<sup>113</sup> showed that soil water-filled pore space may not be the key driver of NO<sub>x</sub> emissions. Furthermore, Geng, et al.<sup>115</sup> indicated that the fallow period in the vegetable system should be an important period for NO emissions. Similar results were reported by Zhang, et al.<sup>108</sup> that NO fluxes were pronounced during the fallow periods prior to the next cropping seasons.

For the effect of different types of fertilizers on NO emission factors, the collected information in this study (Supplementary Table 4) was further categorized into four groups: (i) urea, (ii) organic fertilizer, (iii) slow-release fertilizer, and (iv) composite fertilizer. As shown in Supplementary Fig. 2, the results indicated the NO emission factors for the organic fertilizer group was 2.39 ± 1.54% ( $n = 7$ ,  $p < 0.05$ ; using One-sample  $t$ -test), followed by the composite fertilizer group (2.09 ± 0.38%,  $n = 6$ ,  $p < 0.05$ ; using One-sample  $t$ -test) and the urea group (0.50 ± 0.26%,  $n = 9$ ,  $p < 0.05$ ; using One-sample  $t$ -test). The NO emission factor for the slow-release fertilizer group was the lowest, i.e., 0.26 ± 0.07% ( $n = 5$ ,  $p < 0.05$ ; using One-sample  $t$ -test).

Supplementary Table 5 compiles the N<sub>2</sub>O emission intensity and emission factor for different crops in the literature. Among the collected literature, the N<sub>2</sub>O emission intensities were in the range between 0.02 and 36.2 kg-N ha<sup>-1</sup>, depending on the the types of crops and fertilizers. The average N<sub>2</sub>O emission intensity is found to be 3.82 ± 0.70 kg-N ha<sup>-1</sup> ( $n = 72$ ,  $p < 0.05$ ; using One-sample  $t$ -test), corresponding to the emission factor of 1.15 ± 0.22%, regardless the types of fertilizers. For the paddy rice, the average N<sub>2</sub>O emission intensity and factor were found to be 1.75 ± 0.56 kg-N ha<sup>-1</sup> and 0.81 ± 0.22% ( $n = 11$ ,  $p < 0.05$ ; using One-sample  $t$ -test), respectively. As one of the powerful GHG, the IPCC has suggested a default value for the N<sub>2</sub>O emission factor of below 1% (i.e., the amount of N<sub>2</sub>O-N emission to the amount of the applied N-fertilizers)<sup>10</sup>. It is noted that the average N<sub>2</sub>O emission factor for rice meets the default value suggested by IPCC. Also, similar to the NO emission, the N<sub>2</sub>O emission of rice from paddy fields is relatively low compared to other vegetables and fruits. Maaz, et al.<sup>107</sup> also reported the same finding that the N<sub>2</sub>O emission from rice paddy was lower by ~70% than that from maize fields.

For vegetables and fruits, the average  $\text{N}_2\text{O}$  emission intensity and factor were found to be  $4.20 \pm 0.82 \text{ kg-N ha}^{-1}$  ( $n = 61$ ,  $p < 0.05$ ; using One-sample  $t$ -test) and  $1.21 \pm 0.26\%$  ( $n = 52$ ,  $p < 0.05$ ; using One-sample  $t$ -test), respectively. Similar results were observed in the study reported by Yang, et al.<sup>116</sup> that, according to a meta-analysis, the average  $\text{N}_2\text{O}$  emission factor of all studied vegetables was about 1.41% ( $n = 223$ ; CI: 1.19–1.64%), where stem vegetables exhibited the lowest emission factor (0.71%; CI: 0.47–0.98%). In their study, the  $\text{N}_2\text{O}$  emission factors of vegetables were also found to be significantly different among vegetable species, which should be critically considered for the global or regional estimation<sup>116</sup>. As presented in Supplementary Table 5, the  $\text{N}_2\text{O}$  emission intensities for corn, banana, and sugarcane were relatively high among all studied vegetables and fruits. The average  $\text{N}_2\text{O}$  emission intensities for corn, banana, and sugarcane were  $16.7 \pm 7.2$  ( $n = 5$ ),  $5.07 \pm 2.21$  ( $n = 5$ ), and  $4.81 \pm 0.90$  ( $n = 12$ )  $\text{kg-N ha}^{-1}$ , respectively. For leguminous crops including soybean and faba bean, the  $\text{N}_2\text{O}$  emission intensity was found to be in the range between 0.19 and  $10.4 \text{ kg-N ha}^{-1}$ .

In fact, soil  $\text{N}_2\text{O}$  emissions are largely attributed to biochemical reactions of nitrification and denitrification, which principally increase with the intensity of N application. Pinheiro, et al.<sup>117</sup> observed an increase in  $\text{N}_2\text{O}$  emission after N-fertilizer application followed by an increase in the soil nitrate concentration. This suggested that nitrification should be the major pathway involved in soil  $\text{N}_2\text{O}$  emission. Different types of fertilizers would also significantly affect the  $\text{N}_2\text{O}$  emission. To evaluate the effect of different types of fertilizers on  $\text{N}_2\text{O}$  emission factors, the collected information in this study (Supplementary Table 5) was further categorized into eight groups, as shown in Supplementary Fig. 3. The average  $\text{N}_2\text{O}$  emission factors for the groups with urea applications were generally higher (1.92–2.38%) than other groups, such as organic (i.e.,  $0.38 \pm 0.16\%$ ) or composite (i.e.,  $1.55 \pm 0.12\%$ ) fertilizers. Also, the average  $\text{N}_2\text{O}$  emission factors for the groups with slow-release fertilizers were relatively low among all groups, ranging from  $0.31 \pm 0.13\%$  ( $n = 5$ ,  $p < 0.05$ ; using One-sample  $t$ -test) for slow-release fertilizers to  $0.43 \pm 0.10\%$  ( $n = 6$ ,  $p < 0.05$ ; using One-sample  $t$ -test) for manure with slow-release fertilizers. Similarly, in a study with sugarcane in Brazil, Degaspari, et al.<sup>118</sup> reported  $\text{N}_2\text{O}$  emissions for urea and a nitrate-based fertilizer (CAN). They found that the  $\text{N}_2\text{O}$  intensities for the unfertilized control, urea, and CAN were 11.4, 19.9, and 16.3  $\text{mg N}_2\text{O-N per kg-stalk}$ , respectively. Zeng and Li<sup>119</sup> also noted that urea substitution treatments will reduce  $\text{N}_2\text{O}$  emission by 26–58% while increasing the yield of paddy rice by 15–23%, compared to urea-only fertilization. Similarly, Ikezawa, et al.<sup>120</sup> evaluated the effect of fertilizers on  $\text{N}_2\text{O}$  emissions for deep placement, and they found that the cumulative  $\text{N}_2\text{O}$  emissions for urea and lime nitrogen were 3.1 and 1.8  $\text{kg-N ha}^{-1}$ , respectively. These indicate that the choice of N-fertilizers would significantly affect the magnitude of  $\text{N}_2\text{O}$  emissions, regardless the types of soils or crops.

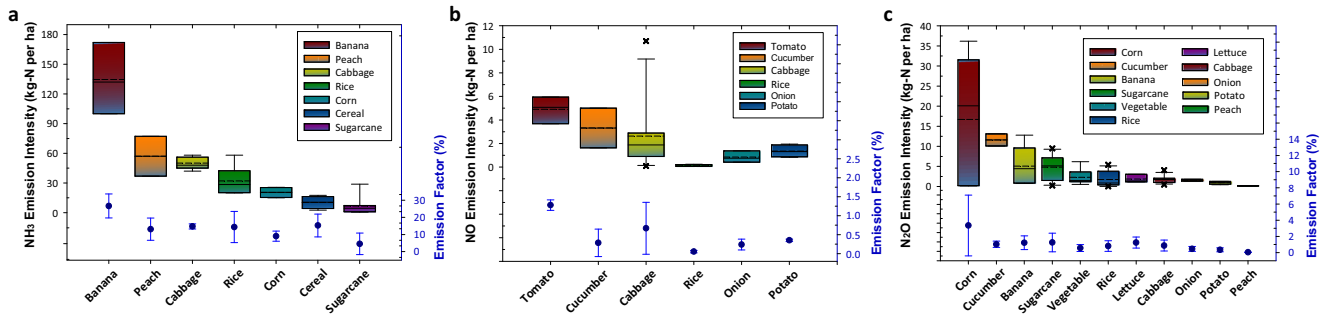
In the case of co-applications of inorganic fertilizer and organic manure, Yang, et al.<sup>121</sup> applied  $^{15}\text{N}$ -labeled ammonium sulfate as the inorganic N source, and found that the  $(\text{NH}_4)_2\text{SO}_4$ -derived  $\text{N}_2\text{O}$  emissions accounted for about 0.01–1.18% of the total  $\text{N}_2\text{O}$  flux. Zhang, et al.<sup>122</sup> performed 3-year field experiments with ten consecutive vegetable crops, and noticed that organic fertilizer application could increase ecosystem respiration by 13.9% without significant effects on  $\text{N}_2\text{O}$  emission, compared to conventional chemical fertilization. Similar results were observed that the replacement of inorganic N fertilizer with manure increased the yield and N agronomic efficiency of overall vegetables; whereas, this did not significantly affect the scaled  $\text{N}_2\text{O}$  emissions<sup>116</sup>. However, different observations were still found in the literature. For instance, Maaz, et al.<sup>107</sup> indicated that, in the Asia-Africa regions, the co-applications of organic manure with mineral fertilizers would result in an increase in  $\text{N}_2\text{O}$  emission by 7.5%. In addition, combined application of K fertilizer with  $\text{NH}_3$  fertilizer

would increase the abundance of *norB*-type denitrifiers (especially the genera *Streptomyces* and *HypHomicrobium*), thereby promoting the biochemical transformation of nitrite to  $\text{N}_2\text{O}$  and resulting in a significant increase in  $\text{N}_2\text{O}$  emissions<sup>123</sup>. In contrast, they found that the combined application of K with  $\text{NH}_3$ -based fertilizer would increase  $\text{N}_2\text{O}$  emissions by 22.7%; in contrast, the combined application of K with nitrate-based fertilizer would reduce the average  $\text{N}_2\text{O}$  emissions by 28.3% (compared with no K-fertilizer addition).

### Practical methods of reducing $\text{NO}_x$ and $\text{N}_2\text{O}$ emissions

Both balanced fertilization and improved NUE are always the most effective strategies to reduce nitrogenous oxide emissions from farmlands. Aside from the above front-end approaches, the NO emission can be controlled by a number of back-end practices, such as (i) adjustments of soil moisture, (ii) the application depth of N fertilizer, (iii) the use of organic fertilizers, and (iv) the use of controlled-release fertilizers. The increase in the application depth of fertilizers could effectively reduce the NO emission because of potential NO sorption by soils<sup>124</sup>. For urea, deep placement (e.g., 0.12 m deep in the case of Andisols<sup>125</sup>) would be highly effective in reducing NO emissions; however, relatively less effective on  $\text{N}_2\text{O}$  emissions. For the organic farming system, a number of studies have proven that organic fertilizers could greatly reduce nitrogenous oxide emissions from various crops, such as managed vegetable systems<sup>115</sup>. Organic fertilizers could result in a low NO emission intensity as the denitrification could be enhanced by the increase of soil organic carbon and pH<sup>115</sup>. Cheng, et al.<sup>124</sup> also noticed that banded controlled-release urea can significantly reduce the NO emission by 78.8–82.6%, in comparison with the conventional urea. However, the effect of organic farming on  $\text{N}_2\text{O}$  emission reduction is still a topic of discussion. For instance, a recent study<sup>126</sup> indicated that the use of livestock manures could reduce both NO (by 46.5–59.8%) and  $\text{N}_2\text{O}$  (by 41.4–49.6%) emissions in comparison to urea fertilizer. Abbasi, et al.<sup>127</sup> also found that the use of organic manure in corn growing seasons would produce less  $\text{N}_2\text{O}$  emissions, compared to inorganic ammonium nitrate; however, it resulted in a higher  $\text{N}_2\text{O}$  emission in unfertilized soybean seasons.

For reducing the  $\text{N}_2\text{O}$  emission, several practical methods have been developed and deployed, such as (i) keeping soils in aerobic conditions by optimum irrigation-drainage management, and avoidance of soil compaction by animals or traffic<sup>2</sup>, (ii) using slow release fertilizers<sup>128</sup>, urease inhibitor<sup>129</sup>, or nitrification inhibitor<sup>130</sup>, (iii) incorporating (bio-)organic fertilizers<sup>131</sup> and biochars<sup>132</sup> in soil-plant systems, and (iv) sowing legume crops in the fallow period between crop cycles<sup>133</sup>. In particular, the green practice of using inhibitors has been greatly advocated by numerous studies. Subbarao and Searchinger<sup>134</sup> proposed the concept of maintaining the status of fertilizers in soil systems as a “more ammonium solution” by applying biological nitrification inhibitors. Biological nitrification inhibitors typically work at least 10 cm underground in the rhizosphere; therefore, the  $\text{NH}_3$  emission from soils, on the other hand, would not increase<sup>135</sup>. Wang, et al.<sup>130</sup> has critically reviewed the effect of biological nitrification inhibitors on the  $\text{N}_2\text{O}$  emission. Nitrification inhibitors can be transported through the roots to the active sites for nitrification in the soils to increase NUE and yield, thereby reducing  $\text{N}_2\text{O}$  emissions. For instance, the use of the urease and/or nitrification inhibitors can significantly reduce  $\text{N}_2\text{O}$  emissions, e.g., by up to 65.4% in the case of NBPT and DCD<sup>129</sup>. Maaz, et al.<sup>107</sup> also reported a wide range of  $\text{N}_2\text{O}$  emission reduction by 8–100% when introducing nitrification inhibitors or combined with urease inhibitors. Cheng, et al.<sup>124</sup> also noticed that banded controlled-release urea can significantly reduce the  $\text{N}_2\text{O}$  emission by 31.6–40.5%, in comparison with the conventional urea.



**Fig. 3** Effect of different crops on emission intensity and factor for nitrogenous gases. **a**  $\text{NH}_3$  emission for different crops, and the detailed data are compiled in Supplementary Table 3; **b**  $\text{NO}$  emission for different crops, and the detailed data are compiled in Supplementary Table 4; **c**  $\text{N}_2\text{O}$  emission for different crops, and the detailed data are compiled in Supplementary Table 5. All error bars are determined at the 0.05 confidence level (Student's *t*-test).

The  $\text{N}_2\text{O}$  emissions from farmlands can be affected by the tillage system; however, its effect is still a topic of discussion. Koga<sup>13</sup> found that reduced tillage with green manure application could effectively reduce  $\text{N}_2\text{O}$  emission from rotation crop lands. Langeroodi, et al.<sup>136</sup> also suggested that a no-tillage system would result in a lower cumulative  $\text{N}_2\text{O}$  emissions flux compared to conventional tillage for a wheat-soybean rotation, especially when fertilizer was applied. Similar observations by Fiorini, et al.<sup>137</sup> found that  $\text{N}_2\text{O}$  emissions in a no-tillage system were 40–55% lower than that in conventional tillage. However, a recent study by Badagliacca, et al.<sup>138</sup> indicated that, in the case of faba beans, higher  $\text{N}_2\text{O}$  emissions were observed in a no-tillage system ( $0.259 \text{ g-N m}^{-2}$ ) than that in conventional tillage ( $0.171 \text{ g-N m}^{-2}$ )<sup>13</sup>. Similarly, Gong, et al.<sup>139</sup> observed higher  $\text{N}_2\text{O}$  emissions under a no-tillage system that those under moldboard plowing in the organic soybean field. On the other hand, no-tillage would also result in a higher annual SOC sequestration compared to moldboard plowing, thereby fully compensating the global warming potential caused by an increased  $\text{N}_2\text{O}$  emission<sup>139</sup>.

For the use of biomaterials, Wang, et al.<sup>140</sup> proved that biochar amendment in paddy soil could reduce the soil nitrate concentration by promoting  $\text{NH}_3$  oxidation and total nitrogen uptake, thereby reducing soil  $\text{N}_2\text{O}$  flux. With the biochar amendment, the  $\text{N}_2\text{O}$  denitrification was decreased due to the decreased bulk density of soils<sup>140</sup>. Similar results were observed that the cumulative  $\text{N}_2\text{O}$  emissions could be effectively reduced by 52.2% and 97.8% with the biochar additive doses of 1 and 3  $\text{kg m}^{-2}$ , respectively<sup>141</sup>. Yi, et al.<sup>142</sup> also indicated that, for reducing  $\text{N}_2\text{O}$  emissions caused by using urea, the biochar amendment should be superior to DCD, particularly in the late growth stage of leafy vegetables. Another study reported by Pokharel and Chang<sup>102</sup> indicated that biochar can significantly interact with nitrification inhibitor, and thus affect the  $\text{N}_2\text{O}$  emission intensity, e.g., the efficacy of nitrification inhibitor being reduced when co-applied with biochar largely depending on water-filled pore space.

Furthermore, the use of crop residues in soil is a common practice in agriculture. This is believed to be beneficial to soil carbon sequestration while improving physico-chemical properties of the soil and crop yield<sup>143,144</sup>. However, crop residues are known to be one of the major sources of  $\text{N}_2\text{O}$  emissions in agricultural ecosystems. Crop residues are produced from agriculture in large amounts around the world. Globally, the amount of solid residues from cereal crops, food crops, and legumes is estimated to be over 9.6 billion metric tons per year<sup>145</sup>. Akiyama, et al.<sup>146</sup> suggested that crop residues with a low C/N ratio (e.g., less than 35 such as vegetables) could enhance bacterial and fungal denitrification, and thus lead to a high  $\text{N}_2\text{O}$  emission. In their study, large  $\text{N}_2\text{O}$  peaks were observed after crop residues were placed on the surface of the soil for more than one week, especially in summer. In fact, the crop residues can be

utilized via numerous approaches, and converted into bio-energy<sup>145</sup> or bioresources<sup>147</sup>. Another recent study by Rothardt, et al.<sup>110</sup> provided similar findings that replacement of crop residues with a higher C/N ratio straw could mitigate  $\text{N}_2\text{O}$  emission by up to 45%. However, Lal<sup>145</sup> also raised a concern that even a partial removal (30–40%) of crop residue from land could exacerbate soil erosion, deplete soil organic carbon, and accentuate GHG emissions from soils. Therefore, one of the priority research directions should be focused on practices that mitigate  $\text{N}_2\text{O}$  emissions by deploying crop residues with a high C/N with different harvest practices.

#### TRADE-OFFS AMONG NITROGENOUS GASES FROM CROPLANDS

Reactive nitrogen emissions from soils are dependent largely on climatic and soil physico-chemical properties, as well as the nature of the crops and management differences. Several studies have revealed wide variances on reactive nitrogen emissions across countries and regions<sup>7,28,107</sup>. Since the pathways and mechanisms for  $\text{NH}_3$ ,  $\text{NO}_x$ , and  $\text{N}_2\text{O}$  emissions are different, attempts at determining trade-offs among these formation pathways have been conducted by several studies, e.g., with the use of nitrification inhibitors or other green practices<sup>148</sup>. Theoretically, reduced  $\text{NH}_3$  volatilization could lead to greater  $\text{NO}_x/\text{N}_2\text{O}$  emissions and/or other downstream losses, if the NUE of crops and N-uptake by microbes remain constant without adjusting the N-fertilization rate. A holistic evaluation from the nitrogen cycle point of view to determine the trade-offs (or synergies), however, is still limited. For some sustainable managements or green practices, reactive nitrogen emissions could be potentially mitigated, while other eco-environmental and economic benefits, such as soil carbon sink, water quality restoration, improved public health, and increased food production, could be simultaneously realized. However, their overall environmental benefits and trade-offs with economic viability are still a topic of discussion among scientists and policy makers due to the high spatial and temporal variability<sup>149</sup>. This highlights the need to perform holistic and systematic evaluations with a well-defined scope to maximize the overall environmental benefits and maintain ecosystem services. Moreover, global climate change would pose difficulties on identifying the trade-off points; for instance, Ma, et al.<sup>7</sup> indicated that increased temperature due to climate change could significantly stimulate fertilizer-induced  $\text{NH}_3$  emissions from managed ecosystems.

In this article, we briefly reviewed the effect of different crops on emission intensity and factor for nitrogenous gases, including  $\text{NH}_3$ ,  $\text{NO}_x$ , and  $\text{N}_2\text{O}$  (as shown in Fig. 3). Significant differences in nitrogenous gas emissions among crop species were observed. The mean intensities of  $\text{NH}_3$ ,  $\text{NO}_x$ , and  $\text{N}_2\text{O}$  emissions from all

fertilized treatments were found to be 0.5–172, 0.06–39.8 (some outliers were not presented in Fig. 3), and 0.02–36.2 kg-N ha<sup>-1</sup>, respectively. The associated emission factors of NH<sub>3</sub>, NO<sub>x</sub>, and N<sub>2</sub>O were 0.3–34.0%, 0.02–11.3%, and 0.02–10.1%, respectively. For comparison, in the case of sugarcane fields, Macdonald, et al.<sup>113</sup> indicated that the average emission rate of N<sub>2</sub>O was greater than that of NO<sub>x</sub> and NH<sub>3</sub> from the fertilized soil. Aside from the crop species, the differences in emission intensities and factors were highly dependent on agricultural management practices, such as the types and methods of fertilization, among regions. This reveals the importance of implementing site-specific sustainable management practices to enhance the NUE of crops and thus mitigate the nitrogenous gas emissions. In fact, the real conditions in agricultural management differ greatly.

To synergistically realize the mitigation of NH<sub>3</sub>, NO<sub>x</sub>, and N<sub>2</sub>O from farmlands, here we summarize three major management practices: (i) balanced fertilization with appropriate application methods, (ii) fertilizer modifications and inhibitors, and (iii) better farmland management. First, the guidelines of balanced N-fertilization should follow a site-specific approach with appropriate fertilizer management, with respect to the 4R (right source, right timing, and right placement at a right rate) principles. The site-specific approach depends on the crop and soil properties to implement the 4R principles. The possible strategies include deep injection of organic fertilizer 3–5 cm below the soil surface, splitting fertilizer applications, and application of urea before the onset of rain. Similar results were observed that using a 15-cm deep injection of liquid digestate can replace the synthetic fertilizer with low NH<sub>3</sub> emission<sup>150</sup>. However, it is a complex task to determine appropriate levels of nitrogen fertilization and irrigation for croplands. The first step to optimizing fertilization and irrigation is to measure the initial mineral nitrogen content and nitrogen budget in the soil systems. It is noted that the farmland's nitrogen balance should be <30 kg-N ha<sup>-1</sup>, which is the level for a safe environment<sup>63</sup>.

Second, in addition to balanced fertilization, several approaches to fertilizer modifications and inhibitors have been developed to reduce nitrogen losses from fertilizers, including fertilizer coatings, urease/nitrification inhibitors, or the addition of calcium salts. In fact, controlling nitrification in the soil systems is critical to increase the status in crop NUE and reduce nitrogenous gas emissions. Several studies have suggested that the biological nitrification inhibitors should be widely applied to reduce the nitrogenous gas emission from farmlands. In some cases, the biological nitrification inhibitors can be used in corporation with slow release fertilizers or urea inhibitors, especially in the case of urea-based fertilizers. This practice ensures the development of suitable nitrogen synergists to effectively improve the NUE while reducing environmental pollutions.

Third, farmland management practices might introduce significant quantities of nitrogenous gas emissions. For instance, the use of crop residues into soil is a common agricultural practice to enhance soil organic carbon while improving soil physical properties. However, crop residues with a low C/N ratio also produce high N<sub>2</sub>O emissions. Therefore, priority research directions should include (i) development of alternative practices that mitigate N<sub>2</sub>O emissions from deploying low C/N crop residues, and (ii) utilization of crop residues in biorefinery industries to produce bio-based chemicals. Similarly, Yao, et al.<sup>106</sup> reported that novel water-saving practices for ground cover rice production systems with integrated nutrient management is a green farming practice for maximizing environmental benefits (e.g., NO and N<sub>2</sub>O emissions) and yields. In other words, the issue of water use and nutrient fertilization should be simultaneously addressed to achieve a total solution to sustainable farming systems. Furthermore, sound fallow management strategies, such as sowing soybean in the fallow period, is highly essential for reducing NO<sub>x</sub> and N<sub>2</sub>O emissions. For instance, De Antoni Migliorati, et al.<sup>133</sup>

found that the combined use of soybean fallows with a nitrification inhibitor (e.g., DMPP) was the most effective practice to synergistically reduce nitrogen losses while maintaining (or even increasing) crop yields.

Lastly, this study highlighted the need to further evaluate potential trade-offs among N loss pathway, as well as carbon-nitrogen management in cropping systems. In addition to reactive nitrogen emission, carbon-bearing gases (such as CO<sub>2</sub> and CH<sub>4</sub>) from farmlands are of great concern in the face of global climate heating<sup>11</sup>. This complex relationship among all trace gas emissions attributed to soil nutrient cycles could be approached by biogeochemical simulation models, such as DayCent<sup>133</sup>. As we discussed in Section "Practical methods of reducing NO<sub>x</sub> and N<sub>2</sub>O emissions", the use of crop residues could benefit soil carbon sequestration, soil quality improvement, and crop yield; on the flip side, crop residues are the major sources of soil N<sub>2</sub>O emissions. Unfortunately, our literature search identified no studies on addressing the carbon-nitrogen nexus for available green practices (such as applications of green manure and biochars), which should be one of the priority research directions in the future.

## PERSPECTIVES AND PROSPECTS TOWARD LOW-EMISSION AGRICULTURE

Farmland management practices should comprise a broader vision, such as sustainable nutrient management, linkage with climate actions, and optimization of carbon-nitrogen nexus. To address nitrogenous gas emissions from farmlands, we propose three priority directions moving toward a low-emission agriculture, including (i) managing nitrogenous gas emissions by closing nutrient cycles, (ii) reducing front-end emissions by cleaner and alternative fertilizer productions, and (iii) addressing the carbon-nitrogen nexus by a more holistic consideration.

### Managing nitrogenous gas emissions by closing nutrient cycles

Although nitrogen in the mineral form (e.g., ammonia-N) is useful for plant nutrition, if improperly deployed, it can potentially cause severe environmental concerns, such as nitrates (NO<sub>3</sub><sup>-</sup>) leaching and nitrogenous gas (NH<sub>3</sub>, NO, N<sub>2</sub>O and HONO) emissions to the atmosphere. In other words, the best farmland management practice should comprise a broader vision embracing clean water environment and climate-smart agriculture from the perspective of the nutrient cycle (see discussion in Section "Mechanisms and nitrogen cycles"). Linkages with water protection (e.g., nitrate leaching) and climate policies require attention to avoid negative side effects from measures of nitrogenous gas emission abatement, thereby realizing synergies and profits. Several studies found that deep fertilizer placement and injection can effectively reduce the NH<sub>3</sub> emission from farmlands. However, Mencaroni, et al.<sup>83</sup> also noticed that a certain increase in nitrate leaching from farmlands was observed, mostly in the case of winter wheat. In the cases of ammonium-nitrate injection and organic fertilizers, the nitrate leaching from farmlands was enhanced by 24 and 89%, respectively. Therefore, a holistic approach to evaluating the effect of alternative agricultural practices on the whole nitrogen cycle (or even nutrient cycle) and their associated nitrogen loss pathways (e.g., through nitrate leaching) should be conducted.

Few attempts have been made to apply simulation models for evaluating N losses to drain flow and reactive nitrogen gas emissions, such as the Root Zone Water Quality Model<sup>151</sup>. From the technical aspect, one of the recent promising practices is the reuse of return water from agricultural drainage systems. Langholtz, et al.<sup>152</sup> indicated that the return water reuse could enhance the nutrient recycle and reduce the intensity of nutrient loss to runoff (a major non-point source pollution from

agriculture). In fact, agricultural return water, compared to conventional irrigation water, usually contains higher levels of salinity, thereby requiring careful management on the reuse targets of crops and soils<sup>153</sup>. However, our literature search identified no studies have been conducted yet on assessing the effect of return water reuse on watershed quality improvement, climate change mitigation and public health protection, which should be one of the future priority research directions.

### Reducing front-end emissions by cleaner and alternative fertilizer productions

For the back-end field emissions, we have highlighted several management practices to reduce the nitrogenous gas emissions (see discussion in Section “Trade-offs among nitrogenous gases from croplands”), such as appropriate fertilization for the respective crop and soil type, and proper timing of fertilizations with uptake demand. Using nitrification inhibitors is another feasible practice to decrease NH<sub>3</sub> and N<sub>2</sub>O emissions. Also, less use of synthetic fertilizers would benefit air conditions, water quality, and the climate. In fact, regarding the front-end emissions, the production of synthetic fertilizers requires huge amounts of fossil fuels, such as natural gas, and the subsequent use of synthetic fertilizers in farmlands would contribute to significant quantities of nitrogenous gas emissions.

From the life-cycle perspective, the front-end (indirect, considered as the scope 3 in inventory) emissions of air pollutants for fertilization include the upstream energy-consuming production for NH<sub>3</sub> synthesis and P/K fertilizers<sup>154</sup>. Chemical N-fertilizers are synthesized by NH<sub>3</sub> from the Haber–Bosch process (see Fig. 1). It is noteworthy that, despite approaching the thermodynamic limits, the Haber–Bosch process is energy intensive with an energy consumption of ~12.1 kWh per kg NH<sub>3</sub>-N<sup>155</sup>, and responsible for ~2% of global energy consumption<sup>156</sup>. The associated GHG emission intensity from N-fertilizer manufacture is estimated to be ~2.89 kg-CO<sub>2</sub> per kg NH<sub>3</sub><sup>157</sup>, corresponding to 1.44% of global CO<sub>2</sub> emission<sup>156</sup>. The front-end emissions (i.e., due to Haber-Bosch processes) could potentially be reduced via a number of cleaner practices, such as electrocatalytic NH<sub>3</sub> synthesis<sup>158,159</sup>, photocatalytic synthesis<sup>160</sup>, biomass-based chemical looping<sup>161</sup>, and green hydrogen-based fertilizer production. It is noted that these cleaner practices are highly related to the new processes incorporated with the concept of circular bioeconomy systems, which should be one of the priority research directions in the near future.

### Addressing carbon-nitrogen nexus by a more holistic consideration

As we mentioned in Section “Trade-offs among nitrogenous gases from croplands”, the carbon-nitrogen nexus in a farmland should be synergistically addressed for optimization, especially in the face of urgent climate actions. Without a transformational breakthrough in current crop production systems, it is difficult to reach a real zero emission farmland. The implementation of site-specific sustainable management practices depending on the crop and soil properties can effectively reduce and even prevent the nitrogenous gas emissions to a certain low level, thereby realizing a low-emission agriculture. Recently, to meet the long-term goals of the Paris Agreement, the concept of “net zero agriculture” has set up deeper agricultural emission cuts for GHGs, including N<sub>2</sub>O and CO<sub>2</sub>. Agricultural farmlands play essential roles in achieving the goal of net-zero emissions as they can provide numerous ecosystem service functions, such as carbon sinks, biomass resources, and nutrient cycle. Several countries, such as the United Kingdom<sup>162</sup>, have ambitiously announced goals, pathways and action plans toward a net-zero agriculture. A long-term strategic approach should be developed for (i) the role of national land-use plans and agriculture sector, (ii) along with a

combination of changes in farm management, and (iii) the interaction of agriculture with other sectors.

Aside from these strategic plans, negative emission practices should be developed and deployed to facilitate the progress of net-zero emission agriculture. Available negative carbon emission technologies include soil sequestration<sup>163</sup>, biochar<sup>143</sup>, bioenergy with carbon capture and storage<sup>164</sup>, and air capture<sup>165</sup>. For instance, bioenergy is a “carbon-neutral” energy as the emitted CO<sub>2</sub> during bioenergy use could be captured afterward by plants (or energy crops) through photosynthesis to form biomass. The formed biomass can be further processed and converted into bioenergy, thereby realizing a carbon neutral. If the emitted CO<sub>2</sub> during bioenergy use is captured and stored by other means, the overall CO<sub>2</sub> generation becomes negative. Another important case is soil carbon sink, where CO<sub>2</sub> is removed from the atmosphere and stored in the soil carbon pool. This process is primarily mediated by crops and plants through photosynthesis, with carbon stored in the form of soil organic carbon. Therefore, soil carbon sink could help moderate the greenhouse effect by reducing atmospheric CO<sub>2</sub> enrichment, and thus realize net-zero emission agriculture. Furthermore, we also noticed few advanced technologies based on photocatalysts to remove multiple air pollutants and non-CO<sub>2</sub> GHGs (such as N<sub>2</sub>O). For instance, de\_Richter, et al.<sup>166</sup> critically reviewed large-scale atmospheric solar photocatalysis processes, and indicated the importance of future focuses on GHGs photocatalytic removal from sources such as agricultural greenhouses associated with sewage sludge treatments or manure applications.

To look toward to the future of research on this topic, the carbon-nitrogen nexus in agriculture will be the most challenging issue in the coming decade. For instance, the synergetic effect of available green practices (such as return water reuse, and deploying crop residues, green manure, and biochars) on the simultaneous mitigation of carbon and reactive nitrogen emissions should be investigated. The scope of synergies should broadly embrace overall eco-environmental benefits, such as water quality improvement, climate change mitigation, and public health protection.

### DATA AVAILABILITY

Derived data in this study are available from the corresponding author upon reasonable request.

### CODE AVAILABILITY

The source codes for the analysis of this study are available upon request from the corresponding author.

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## AUTHOR CONTRIBUTIONS

S.Y.P.: Conceptualization, Methodology, Supervision, Funding acquisition, Writing-Review and Editing, Project administration, Resources. All authors contributed to revising and finalizing the manuscript. K.H.H.: Formal analysis, Investigation, Data Curation. K.T.L.: Investigation, Data Curation. C.F.: Investigation, Data Curation. C.T.C.: Methodology, Investigation. All authors contributed to revising and finalizing the manuscript.

### COMPETING INTERESTS

The authors declare no competing interests.

### ADDITIONAL INFORMATION

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**Correspondence** and requests for materials should be addressed to Shu-Yuan Pan.

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