Decoupling between ammonia emission and crop production in China due to policy interventions

Running Title: Cropland-NH_3 emission trend in China

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ABSTRACT

Cropland ammonia (NH_3) emission is a critical driver triggering haze pollution. Many agricultural policies were enforced in past four decades to improve nitrogen (N) use efficiency while maintaining crop yield. Inadvertant reductions of NH_3 emissions, which may be induced by such policies, are not well evaluated. Here, we quantify the China's cropland-NH_3 emission change from 1980 to 2050 and its response to policy interventions, using a data-driven model and a survey-based dataset of the fertilization scheme. Cropland-NH_3 emission in China doubled from 1.93 to 4.02 Tg NH_3-N in period 1980-1996, and then decreased to 3.50 Tg NH_3-N in 2017. The prevalence of four agricultural policies may avoid ~3.0 Tg NH_3-N in 2017, mainly located in highly-fertilized areas. Optimization of fertilizer management and food consumption could mitigate three quarters of NH_3 emission in 2050 and lower NH_3 emission intensity (emission divided by crop production) close to the European Union and the United States. Our findings provide an evidence on the decoupling of cropland-NH_3 from crop production in China, and suggest the need to achieve cropland-NH_3 mitigation while sustaining crop yields in other developing economies.

KEYWORDS

ammonia, emission inventory, flux upscaling, decoupling, agricultural management, policy intervention

1. INTRODUCTION

Through its important role in the formation of particulate matter, atmospheric ammonia (NH_3) affects air quality and has implications for human health (Warner et al., 2017). Excess NH_3 in the environment also contributes to soil acidification (Liu et al., 2019), aquatic eutrophication (Elser et al., 2009; Wang et al., 2017; Zhan et al., 2017) and climate change (Hauglustaine et al., 2014). The cropping system, as a source of anthropogenic NH_3 emissions considered second only to animal husbandry, contributes more than one third of atmospheric NH_3 (EDGAR 2017; Paulot et al., 2014; Xu et al., 2019). Cropland NH_3 is considered to consist of emissions due to the application of synthetic fertilizers, manure and crop residue. Reducing these emissions becomes urgent in a situation of increasing food demand due to population growth and a changing diet in future (Fowler et al., 2015). However, NH_3 mitigation from cropping system is challenging as long as agriculture is optimized towards maximum food production.

Actually, high-income countries have long had NH_3 mitigation while sustaining crop yield in their sights (Zhang et al., 2020). For instance, member countries of the European Union (EU) have set a target to reduce NH_3 emissions through the National Emission Ceilings Directive since 2001 (UNECE 1999). In parallel, activities under the UNECE Convention on Long-range Transboundary Air Pollution in the context of the Gothenburg Protocol have set similar targets, including for countries outside of the EU (UNECE 1999). To provide support to the EU member states and parties which have ratified the Gothenburg protocol in attaining these ceilings, an ‘Ammonia Guidance

Document' was developed describing detailed abatement techniques (Bittman 2017), and translated into national plans and legislation in several countries. China promoted abatement options of agricultural NH_3 emissions in the updated Clean Air Action Plan in 2018 (Liu et al., 2019). Although later than the EU, the Chinese government has developed policies that arguably addressed cropland-NH_3 emission mitigation before 2018. For instance, the Agricultural Cost-saving and Efficiency-increasing Program (Wu 2000), and national Soil Testing and Nutrient Recommendation Program (MARA 2015a) were promoted by the government for improving fertilizer use efficiency in 1994 and 2005, respectively. However, cropland-NH_3 reductions associated with these policies are often not well evaluated at regional scale. This further results in an incomplete understanding of the drivers and mechanisms behind changing cropland-NH_3 emissions, and makes future projections and the assessment of further abatement potentials unreliable.

Obstacles of such evaluation lie in the missing methodological approaches to construct linkages between regional cropland-NH_3 emission and agronomic measures or policies. Existing bottom-up models cannot achieve this mainly due to the incomplete model structure and coarse spatial resolution of activity data in connection with agricultural management practices. For example, process-based models e.g. DNDC (Dubache et al., 2019; Li et al., 2019), FAN (Riddick et al., 2016; Vira et al., 2019), DLEM-Bi-NH_3 (Xu et al., 2019) emphasize explicit physicochemical processes of NH_3 transfer across the soil-air interface, but use highly simplified representations of agricultural practices. Data-driven models, which calculate emissions as volatilization rates multiplied by the amount of N-fertilizers applied, could support the analysis of NH_3 trends and patterns in response to historical agricultural management practices beyond alternative climate conditions. However, using temporally consistent activity data on fertilizer schemes may distort the dynamical evolution of cropland-NH_3 emissions (Beusen et al., 2008; Bouwman et al., 2002; Bouwman et al., 1997; Riddick et al., 2016; Vira et al., 2019; Xu et al., 2019).

China has transitioned from an underdeveloped country to the second largest economy globally (Zhou et al., 2020). Driven by demand and policies, the consumption of vegetables, fruits and animal productions is increasing much faster than grain (NBSC 2021). Governmental policies and subsidies are also stimulating the transition of cropping systems from resource dependence (land, fertilizers, water, labor, etc.) to technology-intensive since 1980s (Liu et al., 2016; Jiao et al., 2018). How cropland-NH_3 emissions are responding to technical adoptions and policy interventions over time and space is not well known. To address these knowledge gaps, an updated data-driven model coupled with high-resolution, crop-specific fertilization schemes (rate, form, and placement) was employed to quantify the spatiotemporal pattern of cropland-NH_3 emissions across China for the period 1980-2017. We focused on this period because the most rapid changes took place and the best defined policy interventions in this period and because of data availability. NH_3 emissions from the application of synthetic fertilizers, livestock manure, human excreta, and crop residues returned to croplands were considered. We then identified the driving forces behind changing NH_3 emission patterns by using the Logarithmic Mean Divisia Index method (LMDI, Ang 2015; Guan et al., 2018) and assessed policy-induced NH_3 reductions by translation of the policies into these drivers. Finally, we explored the NH_3 abatement potential for different regions and crops by optimizing the fertilizer management and food consumption in future.

2. MATERIALS AND METHODS

2.1 Data-driven upscaling model

We estimated NH_3 emissions separately for 8 crop types (i.e., rice, maize, wheat, vegetables, fruits, potatoes, legumes, and other upland crops). The NH_3 emissions were calculated as volatilization rate (VR) multiplied by the amount of N-fertilizers applied, whereas environmental conditions and fertilization schemes are considered as correction terms for VRs. This type of function has been applied in previous bottom-up estimates (Huang et al., 2012; Misselbrook et al., 2004; Zhang et al., 2011) as follows:

$$V_{i.k}=VR_{i,k} \times N_{i,k} \times S_{i,k} \tag{1}$$

$$VR_{i,k} = VR_i^0 \times f(pH_{i,k}) \times f(A_{i,k}) \times f(u_{i,k}) \times f(T_{i,k}) \times f(M_{i,k}) \tag{2}$$

where $V_{i,k}$ is NH_3 emission (kg) for crop i in grid k. *VR, N* and *S* represent NH_3 volatilization rate (%), total N application rate (kg N ha^{-1}) and sowing area (ha), respectively. VR^0 is averaged from all available VR data, roughly corresponding to the baseline of VR under reference condition (chamber-based using urea applied through broadcasting with soil/ponded pH of 7 and air temperature of 20°C for upland crops or of 26°C for paddy rice). $f(pH)$, $f(A)$, $f(u)$, $f(T)$, and $f(M)$ represent the correction coefficients that reflect the effects of soil/ponded pH, air temperature and wind speed (as measured 10 m above the surface) during the period of crop growth, the fertilizer type, and the method of fertilizer placement on VR, respectively. To avoid unrealistic values, the estimated $VR_{i,k}$ were capped at 43%, which was consistent with the upper bound of the IPCC Tier 1 default value (Calvo et al., 2019). A detailed introduction and the refinement of the model can be found in Zhan et al. (2021) and supplementary information (Text S1, Figure S1 and Data S1), respectively.

2.2 New dataset of fertilization schemes

The data-driven model is forced by multiple gridded input datasets, including a dataset describing the total synthetic-N fertilizer application rate (kg N year^{-1}) developed by Shang et al (2019, see Text S2), and two new datasets associating the fractions of synthetic-N forms and placement to cropland. For N forms, we obtained the crop-specific fraction of three N fertilizers, including ammonium bicarbonate, urea, other N fertilizers at province level from the Statistics of Cost and Income of Chinese Farm Produce for the period 1980 - 2017 (NDRCC 2003; 2020). The placement of synthetic-N fertilizer largely depends on topographic condition, planting density, root depth and crop's economic value (Xi et al., 2013). Consequently, we assumed that all N fertilizers for rice paddies are applied on surface soil as mechanized incorporation is difficult (Zhang et al., 2016); and all N fertilizers for vegetables and fruits are incorporated manually due to their higher economic return and planting density. For field crops such as wheat, maize, potatoes and legumes, machines were typically employed to incorporate basal fertilizers into soil. We therefore assumed that the incorporation proportions of basal N fertilizer could be calculated as a function of the sowing area fertilized by machine divided by total sowing area (data for both from CAAMM 2020) at province level. The criterion and methodology to determine the incorporation proportions are reported in Text S3, Table S1 and Figure S2.

Annual N in livestock manure, human excreta, and crop residues (kg N year^{-1}) returned to croplands were estimated by a Eubolism model at county-scale (Shang et al., 2019). The N amount in organic fertilizers calculated based on county-scale activity data, such as the numbers of livestock by animal, rural population, and yields by crop type from 1980 to 2017 (Shang et al., 2019). In China, farmers usually broadcast the organic fertilizers on soil surface and incorporate them in a short time accompanying with plough or rotary tillage (Beusen et al., 2008; Femke et al., 2019; Xi et al., 2013). Provincial tillage proportion, i.e. sowing areas of tillage (CAAMM 2020) divided by the total (NDRCC 2020), were therefore taken as the incorporation proportion of organic fertilizer following Zhan et al., (2021, details see Text S3 and Figure S3). All the dataset by crop and fertilizer were then disaggregated into grid maps at 1-km spatial resolution within each of the administrative units following the crop-specific Land-Use/cover Dataset produced for China by Liu et al. (2014). This dataset were developed based on Landsat TM\ETM+ images and field investigations at 10-year intervals from the 1980 to 2017.

2.3 Driving forces behind changing NH_3 emissions

To attribute changes in NH_3 emission trends over time to different driving factors, we first applied the Logarithmic Mean Divisia Index (LMDI, Ang 2015; Guan et al., 2018) to evaluate the four main driving factors, i.e. sowing area, cropping structure, N application rate and NH_3-VRs for the period 1980-2017 (Text S4). Next, we analyzed the relative contributions of five secondary driving factors to the trends of cropland's-NH_3 VRs during 1980-2017 using our data-driven model (Text S5 and Table S2). The five factors include air temperature, wind speed, fertilizer forms, incorporation proportion of synthetic-N fertilizer and organic fertilizer.

Fertilization technologies and crop structure in China have experienced substantial transitions during the period from 1980 to 2017. This transition was driven at least by policy interventions. Since the mid-1990s, the Chinese government implemented four policies, i.e. ACE, VTB, EUP and STNR program (Table 1) to develop deep fertilization, adjust cropping structure, optimize fertilizer forms and reduce N application rate, respectively. Here, we translated the effects of these four policies directly on the related driving parameters, and then estimated the potential NH_3 emissions by assuming these policies had not been implemented. The main principle was fixed the four drivers at the level just before the year that policy was implemented, when we estimate the NH_3 emission afterwards. Our data-driven model was employed to calculate the contribution for each policy. Detail descriptions of above scenarios can be found in Table 1, Text S6 and Table S3.

2.4 Future projections

To explore the future NH_3 abatement potential of croplands, we performed four scenario projections in ten-year intervals from 2020 to 2050. In the business-as-usual (BAU) scenario (Table 2), we only consider current (the year 2017) policies and national plans without any further intervention. However, the crop production will increase in line with projected increases of population and gross domestic product (GDP) as projected by Zhang et al (2020). Meanwhile, climate factors, i.e. air temperature and wind speed, changed following a conservative RCP2.6 (stringent mitigation scenario, predicts the global mean temperature increases of up to 2 °C by 2100) future climate change scenario (PICIR 2021). Scenarios OFM and OFC predict the projections based on the same assumptions as BAU, but optimize fertilizer management (OFM) and food consumption (OFC), respectively (Table 2). For scenario OFM, N fertilizer rate was set according to the "N Surplus Benchmarks in China" following Zhang et al. (2019). Meanwhile, the incorporation proportion of synthetic-N fertilizers will achieve 80% for three staple food (i.e. wheat, maize and rice) according to the National Agriculture Mechanization Extension Plan (Zhang et al., 2020). For scenario OFC, the crop production will decrease by optimizing human diet structure following Zhang et al. (2020) and cut 50% of food loss and waste to achieve the Global Sustainable Development Goals (Clark et al., 2020; FAO 2020; Li et al., 2021). To achieve the most ambitious mitigation target, the ALL scenario was propose to combine all the mitigation options identified in OFM and OFC scenarios. Detail descriptions of above scenarios see Table 2, Text S7, and Table S4-S6. It should be noted that for the intermediate year of scenario OFM, OFC and ALL, we assume linear adoption from 2017 until the adoption year (2050), at which point the technologies are entirely adopted

(Clark et al., 2020).

3. RESULTS

3.1 Decoupling of NH_3 emission and crop production

China's cropland-NH_3 emission was 1.93 Tg NH_3-N in 1980, and almost doubled to 3.50 Tg NH_3-N in 2017 (Figure 1). China accounted for about one third of the global cropland-NH_3 emissions, and was equivalent to the triple of the entire cropland-NH_3 emissions of EU and USA combined (Zhan et al., 2021). The emissions were mainly contributed by paddy rice (26-39%), maize (25-38%) and wheat (13-24%), followed by vegetables (1.1-7.9%) and fruits (0.8-4.8%) (Figure 1). However, total cropland-NH_3 emission increase was not linear, instead a rapid increase by 128.7 Gg NH_3-N yr^{-2} from 1980 to 1996 (P<0.05, period P1) and a slight descent of -7.3 Gg NH_3-N yr^{-2} after 1997 (P<0.1, period P2, Figure 1). Spatial analyses further confirmed that the shift from rapid increase to stagnation or slight decrease of cropland-NH_3 emission in P1 and P2, respectively, affected sowing areas that together account for 47.6 % of cropland-NH_3 emission (Figure 2a and 2b). The regions where NH_3 emission decreased are distributed in the North China Plain, the lower Yangtze River Basin and the Sichuan Basin during P2 (Figure 2b).

Our estimate of NH_3 emission from cropland was about one third lower than values derived from previous bottom-up models (EDGAR 2017; Fu et al., 2020; Kang et al., 2016; Ma 2020; Xu et al., 2016; Zhang et al., 2017) (Figure S4). The differences between our estimate and other inventories can be primarily attributed to the updates of crop- and fertilizer-specific fertilization schemes based on sub-national data and the VRs upscaled from globally distributed 499 field observations. Scenario tests showed that the updates of N input data and VRs could explain 66% ~ 100% (for different years) of such discrepancies (Figure S5 and Table S8). The decreased NH_3 emission from cropland at the late stage of P2 (2006-2017) is inconsistent with some earlier estimates (Figure S4), but could explain the observed decreasing trend of atmospheric NH_x depositions (Yu et al., 2019), while NH_3 emissions from livestock and industrial sectors remain stable or increase (EDGAR 2017; Fu et al., 2020; Kang et al., 2016; Ma 2020; Zhang et al., 2017; Meng et al., 2017, Figure S6).

The concept of decoupling here has been used to describe the relationship between environmental pressure and production growth (Bennetzen et al., 2016). The decreasing emission intensity, which defined as the cropland-NH_3 emission divided by total crop production, could indicate the decoupling of NH_3 emission from crop production. Since 1995, the decelerating and declining NH_3 emissions has sustained an increasing crop production, suggesting a decoupling of NH_3 emissions from crop production at the national level (Figure 1). In 2017, three-fourth of provinces, which supply 96% of total crop yield (in kilocalories), have achieved the decoupling of NH_3 emissions with crop production. These provinces showed a clear northwestward trends (Figure 2c). Eastern coastal provinces (e.g., Zhejiang, Fujiang and Guangdong) decoupled NH_3 emission from crop production before 1995; while the major crop-production provinces in east and central China decoupled in mid-1990s (Figure 2c). Provinces of coupled NH_3 emissions and crop production are mainly located in two regions. The first one comprises some rich municipalities in eastern coastal parts, such as Beijing, Tianjin, and Shanghai, where sowing areas were diminished due to their economic development. The second one covers most parts of the less-developed provinces in western China, which account for only 4.0% of national sowing areas (Figure 2c).

3.2 Drivers of China's cropland-NH_3 emission trends

Changes in N application rates were the dominant driver of the NH_3 emission trends for the past four decades (Figure 3a). This factor alone led to the increasing NH_3 emission by 34% at the end of Period P1 (1980-1996), then its contribution decreased from 83% in 2003 to 60% in 2017 (Figure 3a). To feed the growing population, China's government introduced the Household Responsibility System to stimulate farmers' enthusiasm to farm since 1980 (Jiao et al., 2018). Economic benefits of crop yield growth incentivized synthetic fertilizer applications, that is, N application rate increased from 121 kg N ha^{-1} in 1980 to 219 kg ha^{-1} in 2007 (Figure 4e). However, N application rate started to decline continuously at an average of 0.82 kg ha^{-1} yr^{-2} after 2007 (Figure 4e). This notable decline appears to be mainly associated with the intervention of STNR Program, which launched in 2005 to match the supply of nutrients with demand during field application. By the year 2013, the implementation area of the STNR program was increased six-fold (Figure 4e). Due to the timing of introduction of STNR, there appears to be an association between the decrease N application rate and NH_3 reductions in time, which suggests that the measures of STNR have played a role. The NH_3 reduction which promoted by STNR probably reached 1.8 Tg NH_3-N in 2017 based on our scenario estimates (Table S3), especially for North China Plain and Sichuan Basin (Figure 4a and 5d).

As the second most important driver, NH_3-VR increased cropland-NH_3 emission by 14% by the end of period P1 (1980-1996), but decreased largely after 1994 (Figure 3a). After 2010, the NH_3-VR even exerted as a negligible factor (5%, Figure 3a). By further decomposing the effect of NH_3-VR into climate and fertilizer scheme drivers, we find that climate change and the increasing shares of ABC and urea contributed largely (38% and 73%) to promote NH_3-VR in P1 (Figure 3b). And the pronounced decreases of NH_3-VRs were almost entirely related to the increasing proportion of deep fertilization by machine and diminished ratio of ammonium bicarbonate after 1994 (Figure 3b). Such technology innovations seem to be supported by the ACE program and EUP guideline (Table 1) started in mid-1990s. To increase fertilizer efficiency, Chinese government implemented the ACE Program to promote deep fertilization in 1994. For field crops (i.e. wheat, maize, potatoes and legumes), almost one third of sowing area was deep-fertilized using machines in 2017 (Figure 4b). At the same time, most medium- and small- size manufacturers in China had upgraded their production devices towards high concentration nitrogen fertilizer (i.e. urea, with 46% N content) to replace ammonium bicarbonate (only 17% N content but 1.47 - 2.29 fold VR compared to urea, Figure S1). The consumption of urea has increased 1.5 times between 1996 and 2017, while the ammonium bicarbonate decreased by almost 69% in the same period (Figure 4d). These two policy interventions triggered innovations on fertilization method and fertilizer types. According to our estimates, the subsequent reduction of

NH_3 emissions may have amounted to 0.23 (ACE) and 0.95 (EUP) Tg NH_3-N in 2017, especially for agricultural intensive regions (Figure 4e, 5a and 5c).

Another 23% increase in NH_3 emissions was driven by arable land expansion, but was partially offset by crop mix adjustment (Figure 3a). For example, in order to meet increased consumption of cash crops, Chinese government launched the VTB Program (Table 1) in 1988. Driven by this program, the sowing areas of vegetables and fruits increased by 185% and 79% during 1990 to 2003, respectively. Meanwhile, the areas sown with wheat and paddy rice declined by 29% and 20% at the same period (Figure 4c). This structural transition in cropping patterns that occurred in P1 resulted in decreasing NH_3 emissions. The reason is that vegetables and fruits have lower VRs (about 78%) than that of staple crops due to their widespread deep placement (Figure S7). This transition probably resulted in NH_3 emission reductions of 0.12-0.27 Tg NH_3-N yr^{-1} by around 2000, but did not play a critical role after the mid-2000s due to the government's guideline to prevent the further decrease on sowing area of cereal crops (Figure 4c). Additionally, the effect of shift in crop mix compensated for each other across different regions (Figure 5b). For example, the increase in cash crop cultivation drove emission down in south China but up in North China Plain due to the area expansion of maize (Figure 5b).

Throughout the time period considered, policies appear to accelerate technical improvement and NH_3 emission reductions in cropland. Since 1995, policy interventions seemed play key roles to promote the decoupling of NH_3 emission from crop production for the provinces in east and central China (Figure 2c). Without these policies, cropland-NH_3 emissions in China would remain coupled with crop production by the end of 2020s (Figure S8). The most effective technologies to achieve the decoupling of NH_3 emission from crop production were N application rate reduction and a wider application of urea, supported by the national STNR and EUP program, respectively (Figure 4a).

3.3 Targeted mitigation opportunities by 2050

Despite the fact that China has decoupled its NH_3 emissions from crop production at the national level, its emissions intensity in 2017 (1.37 g NH_3-N kcal^{-} yr^{-1}) was still 3 times more than the EU and the USA in 2000 (Zhan et al., 2021). We therefore explored the NH_3 mitigation potential for the next 30 years (2020-2050) by implementing strategies including optimization of fertilizer management and demand-side measures for diets.

China's crop demand is projected to increase by 140% by 2050 considering both economic development and population growth. This would require an additional sowing area of 35.4 Mha, with the total NH_3 emissions achieving 4.9 Tg NH_3-N by 2050 if maintaining the 2017 management practice under increasing temperature conditions (BAU, Figure 6a). Under BAU, cropland emissions of NH_3 in 2030 (4.15 Tg NH_3-N) would exceed the peak level in 1996 (4.02 Tg NH_3-N) and steadily increase until 2050 (Figure 6a). NH_3 abatement through optimizing diet composition and cutting food losses and waste (OFC) could reduce NH_3 emission by 18.4% in 2050 compared with BAU (Figure 6a). When conducting optimal fertilizer management (OFM), N fertilizer consumption would reduce by 50.5%, inducing a subsequent NH_3 reduction of 67.4% compared with BAU in 2050 (Figure 6a). To achieve the most ambitious mitigation target, the ALL scenario combined all the mitigation options identified in OFW and OFC. The estimated NH_3 emissions of the ALL scenario are 1.28 Tg NH_3-N in 2050 (73.6% reduction relative to BAU, Figure 6a). Under scenario ALL, China would show a quite low cropland-NH_3 emission intensity (0.43 g NH_3-N $kcal^{-1}$ yr^{-1}) in 2050, which is closer to that of the USA (0.42 g NH_3-N $kcal^{-1}$ yr^{-1}) and the EU (0.39 g NH_3-N $kcal^{-1}$ yr^{-1}).

Spatially explicit information of NH_3 mitigation potential could help us to identify specific crops and hotspot areas, which may be attractive 'mitigation targets'. We ranked gridded mitigation potentials from largest to smallest, and then added the value to the sum of its predecessors, resulting in cumulative mitigation potential up to a given point of sowing area. Figure 6b and 6c shows the uneven distribution of NH_3 mitigation potentials across Chinese croplands. A half of the NH_3 emission reduction could be achieved on 24% of sowing area for maize, 30% for wheat, 19% for rice, and 26% for all crops together (Figure 6b). Total mitigation potentials were concentrated in Huaihe (Yellow River) Basin, which contributed about half of the total. This result implies the importance of this region on crop production and highlights the benefit of focusing on a small area that could deliver large NH_3 mitigation.

4. DISCUSSION & CONCLUSIONS

Our study provides evidence in the decoupling of NH_3 emission from crop production since 1995 at the national level. Four critical policies (Table 1) since mid-

1990s contributed to a decoupling and probably cut nearly half of the cropland-NH_3 emission in 2017. Of all, national STNR Program and EUP guide appear to be the most effective policies. Still, increasing population, GDP and climate warming indicate a 140% increase in crop NH_3 emissions in 2050 when compared with 2017. Our result reveals both the achievements in alleviating cropland-NH_3 emission in past few decades and future challenges in re-increasing NH_3 emission of China.

Fertilizer-induced increase in NH_3 emissions are universal worldwide after the invention of the Haber-Bosch process. To mitigate the negative effects, some directive, policy and mitigating options were implemented in high-income countries at the beginning of 21st century (Bittman 2017; UNECE 1999). Though the lack of the comprehensive assessment of these policies on NH_3 mitigation, we can see a declining cropland-NH_3 emission trend (at −0.6 Gg N year^{-1}) in Europe and a stagnation in cropland-NH_3 emissions from North America since the 1980s (Xu et al., 2019). As the largest emitter of cropland-NH_3 emissions in the world (Zhan et al., 2021), China has also implemented action plans to improve N use efficiency and reduce environmental pollution since 1990s (Jiao et al., 2018). Our results provide evidence that cropland-NH_3 emissions have been increasingly mitigated in China while not compromising crop production.

Challenges of NH_3 abatement are universal across the rapidly developing countries of the world. Developing countries which fall into two groups need to pay more attention to NH_3 mitigation while improving crop yield. The first category includes Pakistan and India (Shahzad et al., 2019), which sustain the crop yields largely by relying on high N application rate (Zhan et al., 2021). The second category mainly includes countries in sub-Saharan Africa, where agricultural production needs to improve urgently to keep pace with the rapid population growth (Hong et al., 2021). All the situations portend an intensive application of N-fertilizer to the cropland in these countries, a situation similar to that of China. China's experience could provide a guide and a paradigm shift for above-mentioned countries, on managing N cycles under the balance of agricultural development and controlling NH_3 pollution. However, not all the measures can be applied well for other regions, some techniques are restricted in applicability by their effectiveness or practical limitation. These limitations may be of very different nature, caused by local climate, soil conditions (pH, slope), farm size, financial and technical issues. Therefore, implementation of NH_3 abatement measures should follow their applicability and be adjusted to local conditions (Zhang et al., 2020).

Even if our results show that the cropland-NH_3 emission can be effectively managed by related policies across China (Figure 4), further work needs to be done to determine the reliability of our estimates. In this study, we translated the effects of four policies on the related key driving parameters directly. Physical and socio-economic barriers, farmers' adaptive behavior from policy enactment to implementation need to be considered through specific approaches, such as econometric models (Huang et al., 2016; Wang et al., 2015) and socioeconomic studies (Scrieciu 2011). Therefore, our estimates may provide the most optimistic NH_3 reductions of these policies. Another limitation is that our model does not take irrigation practices into account (Sommer et al., 2004) , which may lead to the overestimation of NH_3 VRs and emissions. Besides, we assumed the consistent fertilizer placement for rice, vegetables, fruits and other crops according to the universal practice in China. This may distort the spatiotemporal trend of NH_3-VRs for above crops. For example, few farmers also deployed manual deep fertilization or side-deep fertilizer machinery in paddy fields, which largely reduced the NH_3-VRs of rice when compared with broadcasting application.

Future growth in population and incomes is likely to further boost food demand and hinder previous efforts to suppress the increasing cropland-NH_3 emissions (Figure 6a). The Chinese government has strictly limited the input of synthetic fertilizer as well as setting ambitious goals to improve crop NUE (Liu et al., 2016). China also launched the "Strategy of taking potato as the fourth staple food" in 2015 (MARA 2015b). This policy showed a large potential to reduce NH_3 emissions because potatoes, which generally grow in cold regions, exhibit lower VRs (8.8%) than rice (19.1%), maize (20.7%) and wheat (11.5%) (Figure S7). However, barriers exist to promote further technologies to mitigate crop-NH_3 emission in China. First, adjustment of fertilizer types (e.g. replacing urea by nitrate N-fertilizer) and deep placement often result in pollution swapping between environmental media. For example, fertilizer incorporation can reduce NH_3 emissions, but may lead to increased nitrate leaching, especially in wet climates (Zhan et al., 2021). Second, given that poor smallholder farmers still dominate China's agricultural production, the transition to large-scale and mechanized fertilization in China is restricted by inherent social barriers and weak technical foundation, which takes time and effort to overcome (Zhang et al., 2020).

Future reductions in consumption of NH_3-intensive fertilizers, machines and services need to be further supported by research, policies and financial incentives for all the major NH_3 emitters of the world. Promoting balanced diets and reducing food waste to mitigate NH_3 emissions may be critical for the developed countries and rapid growing economies. Adopting regionally specific-approaches is another efficient pathway to achieve NH_3 mitigation particularly across the emission hotspots. Our spatially explicit cropland-NH_3 emission data could be used to support and guide the development of such interventions, which may include inter-provincial cooperation, national or international food trade (Shan et al., 2021). The ambitious goal should be designed in segments, and cost-benefit analysis could be helped to provide guidance for emerging policy priorities in reducing NH_3 pollution (Zhang et al., 2020). Meanwhile, China plays an important role in the South-South co-operation via South-South trade and the Belt and Road Initiative, especially in the technology extension of crop planting and machine application (Shan et al., 2021). The experience and status quo of NH_3 emissions and policy induced abatement in China may have implications for other developing economies to achieve cropland's NH_3 mitigation while sustaining crop yields.

SUPPORTING INFORMATION

Extended explanation of cropland-NH_3 VR model, datasets, scenario simulation, comparison with previous estimates, and associated supplementary Tables and Figures are all available free of charge at http://pubs.acs.org.

ACKNOWLEDGMENT

This study was supported by the National Key Research and Development Program of China (2018YFC0213304), the National Natural Science Foundation of China (41907087; 71961137011), Central Public-interest Scientific Institution Basal Research Fund (BSRF201905), and S&T Program of Hebei (21326403D). The work of Stefan Reis was supported by the UK Natural Environment Research Council (NERC) National Capability award NE/R000131/1 (Sustainable Use of Natural Resources to Improve Human Health and Support Economic Development, SUNRISE).

AUTHORSHIP CONTRIBUTIONS STATEMENT

Wulahati Adalibieke: Methodology, Formal analysis, Visualization. Xiaoying Zhan: Investigation, Results Interpretation, Writing original draft. Xiaoqing Cui: Resources, Data curation. Stefan Reis: Writing - review & editing. Wilfried Winiwarter: Writing - re-view & editing. Feng Zhou: Conceptualization, Writing - review & editing, Funding acquisition, Project administration.

CONFLICTS OF INTEREST

The authors declare no conflicts of interest.

REFERENCES

1. Ang, B. W. (2015). LMDI decomposition approach: A guide for implementation. *Energy Policy* 86: 233-238.
2. Bai, Z. H., Ma, W. Q., Ma, L., Velthof, G. L., Wei, Z. B., Havlik, P., . . . Zhang, F. S. (2018). China's livestock transition: Driving forces, impacts, and consequences. *Science Advances* 4: 11.

3. Bennetzen, E. H., Smith, P., Porter, J. R. (2016). Decoupling of greenhouse gas emissions from global agricultural production: 1970-2050. *Global Change Biology* 22: 763-781.
4. Beusen, A. H. W., Bouwman, A. F., Heuberger, P. S. C., Van Drecht, G., Van Der Hoek, K. W. (2008). Bottom-up uncertainty estimates of global ammonia emissions from global agricultural production systems. *Atmospheric Environment* 42: 6067-6077.
5. Bittman, S., Dedina, M., Howard C.M., Oenema, O., Sutton, M.A. (2017). Options for ammonia mitigation (Edinburgh, UK: Centre for Ecology and Hydrology).
6. Bouwman, A. F., Boumans, L. J. M., Batjes, N. H. (2002). Estimation of global NH_3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Global Biogeochemical Cycles* 16: 16.
7. Bouwman, A. F., Lee, D. S., Asman, W. A. H., Dentener, F. J., VanderHoek, K. W., Olivier, J. G. J. (1997). A global high-resolution emission inventory for ammonia. *Global Biogeochemical Cycles* 11: 561-587.
8. CAAMM (2020). China Association of Agricultural Machinery Manufacturers (CAAMM). China Agricultural Machinery Industry Yearbook. https://data.cnki.net/yearbook/Single/N2020080630
9. Calvo, B., E.; , Tanabe, K., Kranjc, A., Baasansuren, J., Fukuda, M., Ngarize, S., . . . Federici, S. (2019). 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories. In. Switzerland.
10. Clark, M. A., Domingo, N. G. G., Colgan, K., Thakrar, S. K., Tilman, D., Lynch, J., . . . Hill, J. D. (2020). Global food system emissions could preclude achieving the 1.5 degrees and 2 degrees climate change targets. *Science* 370: 705-708.
11. Dubache, G., Li, S. Q., Zheng, X. H., Zhang, W., Deng, J. (2019). Modeling ammonia volatilization following urea application to winter cereal fields in the United Kingdom by a revised biogeochemical model. *Science of the Total Environment* 660: 1403-1418.
12. EDGAR (2017). Emissions Database for Global Atmospheric Research (EDGAR) V4.3.2. https://edgar.jrc.ec.europa.eu/overview.php?v=432_AP.

13. Elser, J. J., Andersen, T., Baron, J. S., Bergstrom, A. K., Jansson, M., Kyle, M., . . . Hessen, D. O. (2009). Shifts in Lake N:P Stoichiometry and Nutrient Limitation Driven by Atmospheric Nitrogen Deposition. *Science* 326: 835-837.
14. FAO (2020). Food and Agricultural Organization of the United Nations (FAO). FAOSTAT. http://www.fao.org/faostat/en/#data/QC
15. Femke, L., Jetse, J. S., Christoph, M. (2019). Options to model the effects of tillage on N_2O emissions at the global scale. *Ecological Modelling* 392: 212–225.
16. Fowler, D., Steadman, C. E., Stevenson, D., Coyle, M., Rees, R. M., Skiba, U. M., . . . Galloway, J. N. (2015). Effects of global change during the 21st century on the nitrogen cycle. *Atmospheric Chemistry and Physics* 15: 13849-13893.
17. Fu, H., Luo, Z. B., Hu, S. Y. (2020). A temporal -spatial analysis and future trends of ammonia emissions in China. *Science of the Total Environment* 731: 138897.
18. Guan, D. B., Meng, J., Reiner, D. M., Zhang, N., Shan, Y. L., Mi, Z. F., . . . Davis, S. J. (2018). Structural decline in China's CO_2 emissions through transitions in industry and energy systems. *Nature Geoscience* 11: 551-555.
19. Hauglustaine, D. A., Balkanski, Y., Schulz, M. (2014). A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate. *Atmospheric Chemistry and Physics* 14: 11031-11063.
20. Hong, C. P., Burney, J. A., Pongratz, J., Nabel, J., Mueller, N. D., Jackson, R. B., Davis, S. J. (2021). Global and regional drivers of land-use emissions in 1961-2017. *Nature* 589: 554-561.
21. Huang, J. K., Ding, J. P. (2016). Institutional innovation and policy support to facilitate small-scale farming transformation in China. *Agricultural Economics* 47: 227–237.
22. Huang, X., Song, Y., Li, M. M., Li, J. F., Huo, Q., Cai, X. H., . . . Zhang, H. S. (2012). A high-resolution ammonia emission inventory in China. *Global Biogeochemical Cycles* 26: 1-14.
23. Jiao, X. Q., Mongol, N., Zhang, F. S. (2018). The transformation of agriculture in China: Looking back and looking forward. *Journal of Integrative Agriculture* 17: 755-764.

24. Kang, Y. N., Liu, M. X., Song, Y., Huang, X., Yao, H., Cai, X. H., . . . Zhu, T. (2016). High-resolution ammonia emissions inventories in China from 1980 to 2012. *Atmospheric Chemistry and Physics* 16: 2043-2058.
25. Li, S. Q., Zheng, X. H., Zhang, W., Han, S. H., Deng, J., Wang, K., . . . Liu, C. Y. (2019). Modeling ammonia volatilization following the application of synthetic fertilizers to cultivated uplands with calcareous soils using an improved DNDC biogeochemistry model. *Science of the Total Environment* 660: 931-946.
26. Li, X., Liu, X. J., Lu, S. J., Cheng, G. Y., Hu, Y. C., Liu, J. G., . . . Liu, G. (2021). China's food loss and waste embodies increasing environmental impacts 2: 519–528
27. Li, Z. J. (2009). Development course and experience on fertilizer industry in past six decades. *Economic Analysis of China* 10: 26-30.
28. Liu, J. K., W.; Zhang, Z.; Xu, X.; Qin, Y.; Ning, J.; Zhou, W.; Zhang, S.;, Li, R. Y., C.; et al. (2014). Spatiotemporal Characteristics, Patterns and Causes of Land Use Changes in China since the Late 1980s. *Acta Geogr. Sin* 69: 3–14.
29. Liu, M. X., Huang, X., Song, Y., Tang, J., Cao, J. J., Zhang, X. Y., . . . Zhu, T. (2019). Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain. *Proceedings of the National Academy of Sciences of the United States of America* 116: 7760-7765.
30. Liu, X. J., Vitousek, P., Chang, Y. H., Zhang, W. F., Matson, P., Zhang, F. S. (2016). Evidence for a historic change occurring in China. *Environmental Science & Technology* 50: 505-506.
31. Ma, S. Y. (2020). High-resolution assessment of ammonia emissions in China: Inventories, driving forces and mitigation. *Atmospheric Environment* 229: 117458.
32. MARA (2015a). Ministry of Agriculture and Rural Affairs (MARA). Implementation plan for soil testing and formulation fertilization for 10 million farms. 2015, http://www.moa.gov.cn/nybgb/2005/dliuq/201806/t20180618_6152460.htm.
33. MARA (2015b). Ministry of Agriculture and Rural Affairs (MARA). Strategy to promote potato as the fourth staple food in China.

http://cpc.people.com.cn/n/2015/0107/c83083-26340225.html.

34. Meng, W. J., Zhong, Q. R., Yun, X., Zhu, X., Huang, T. B., Shen, H. Z., . . . Tao, S. (2017). Improvement of a global high-resolution ammonia emission inventory for combustion and industrial sources with new data from the residential and transportation sectors. *Environmental Science & Technology* 51: 2821-2829.

35. Misselbrook, T. H., Sutton, M. A., Scholefield, D. (2004). A simple process-based model for estimating ammonia emissions from agricultural land after fertilizer applications. *Soil Use and Management* 20: 365-372.

36. NBSC (2021). National Bureau of Statistics of China (NBSC). http://data.stats.gov.cn/easyquery.htm?cn=C01.

37. NDRCC (2003). National Development and Reform Commission of China (NDRCC). Statistics of cost and income of Chinese farm produce since the establishment of the nation from 1950-1997.

38. NDRCC (2020). National Development and Reform Commission of China (NDRCC). Statistics of cost and income of Chinese farm produce. https://data.stats.gov.cn/easyquery.htm?cn=C01

39. Paulot, F., Jacob, D. J., Pinder, R. W., Bash, J. O., Travis, K., Henze, D. K. (2014). Ammonia emissions in the United States, European Union, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH_3). *Journal of Geophysical Research-Atmospheres* 119: 4343-4364.

40. PICIR (2021). Potsdam-Institute for Climate Impact Research (PICIR). Inter-Sectoral Impact Model Intercomparison Project. https://esg.pik-potsdam.de/search/isimip/.

41. Riddick, S., Ward, D., Hess, P., Mahowald, N., Massad, R., Holland, E. (2016). Estimate of changes in agricultural terrestrial nitrogen pathways and ammonia emissions from 1850 to present in the Community Earth System Model. *Biogeosciences* 13: 3397-3426.

42. Scrieciu, S. S. (2011). Socioeconomic and environmental impacts on agriculture in the new Europe: post-communist transition and accession to the European Union

(Routledge: United Nations Environment Programme, Paris, France.).

43. Shahzad, A. N., Qureshi, M. K., Wakeel, A., Misselbrook, T. (2019). Crop production in Pakistan and low nitrogen use efficiencies. *Nature Sustainability* 2: 1106-1114.

44. Shan, Y., Fang, S., Cai, B., Zhou, Y., Li, D., Feng, K., Hubacek, K. (2021). Chinese cities exhibit varying degrees of decoupling of economic growth and CO_2 emissions between 2005 and 2015. *One Earth* 4: 124-134.

45. Shang, Z. Y., Zhou, F., Smith, P., Saikawa, E., Ciais, P., Chang, J. F., . . . Sabbatini, S. (2019). Weakened growth of cropland-N_2O emissions in China associated with nationwide policy interventions. *Global Change Biology* 25: 3706-3719.

46. Sommer, S. G., Schjoerring, J. K., Denmead, O. T. (2004) Ammonia emission from mineral fertilizers and fertilized crops. in Sparks, D. L. (ed.), *Advances in Agronomy* (Elsevier Academic Press Inc: San Diego).

47. UNECE (1999). United Nations Economic Commission for Europe (UNECE). Protocol to Abate Acidification, Eutrophication and Ground-level Ozone. https://unece.org/environment-policyair/protocol-abate-acidification-eutrophication-and-ground-level-ozone

48. Vira, J., Hess, P., Melkonian, J., Wieder, W. R. (2019). An improved mechanistic model for ammonia volatilization in earth system models: flow of agricultural nitrogen, version 2 (FAN v2). *Geoscientific Model Development Discussions*: 1-49.

49. Wang, J. X., Yang, Y., Huang, J. K., Chen, K. (2015). Information provision, policy support, and farmers' adaptive responses against drought: An empirical study in the North China Plain. *Ecological Modelling* 318: 275-282.

50. Wang, R., Goll, D., Balkanski, Y., Hauglustaine, D., Boucher, O., Ciais, P., . . . Tao, S. (2017). Global forest carbon uptake due to nitrogen and phosphorus deposition from 1850 to 2100. *Global Change Biology* 23: 4854-4872.

51. Warner, J. X., Dickerson, R. R., Wei, Z., Strow, L. L., Wang, Y., Liang, Q. (2017). Increased atmospheric ammonia over the world's major agricultural areas detected from space. *Geophysical Research Letters* 44: 2875-2884.

52. Wu, C. Y. (2000). Technology of Agricultural Cost-saving and Efficiency-

increasing Program (in Chinese). *Agricultural Machinery Extension* 3: 28.

53. Xi, Z. B., Huang, P. Z., Duan, J. X. (2013). Modern chemical fertilizer (expanded edition) (China Agriculture Press: Beijing).

54. Xu, P., Liao, Y. J., Lin, Y. H., Zhao, C. X., Yan, C. H., Cao, M. N., . . . Luan, S. J. (2016). High-resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008. *Atmospheric Chemistry and Physics* 16: 1207-1218.

55. Xu, R. T., Tian, H. Q., Pan, S. F., Prior, S. A., Feng, Y. C., Batchelor, W. D., . . . Yang, J. (2019). Global ammonia emissions from synthetic nitrogen fertilizer applications in agricultural systems: Empirical and process-based estimates and uncertainty. *Global Change Biology* 25: 314-326.

56. Yu, G. R., Jia, Y. L., He, N. P., Zhu, J. X., Chen, Z., Wang, Q. F., . . . Goulding, K. (2019). Stabilization of atmospheric nitrogen deposition in China over the past decade. *Nature Geoscience* 12: 424-429.

57. Zhan, X. Y., Adalibieke, W., Cui, X. Q., Winiwarter, W., Reis, S., Zhang, L., . . . Zhou, F. (2021). Improved estimates of ammonia emissions from global croplands. *Environ. Sci. Technol* 55: 1329-1338.

58. Zhan, X. Y., Zhou, F., Liu, X. J., Paerl, H. W., Shen, J. L., Wang, R., . . . Tang, X. Y. (2017). Evidence for the importance of atmospheric nitrogen deposition to eutrophic Lake Dianchi, China. *Environmental Science & Technology* 51: 6699-6708.

59. Zhang, C., Ju, X. T., Powlson, D., Oenema, O., Pete, S. (2019). Nitrogen Surplus Benchmarks for Controlling N Pollution in the Main Cropping Systems of China. *Environmental Science & Technology* 53: 6678−6687.

60. Zhang, W. F., Yi, J. J., Zhang, F. S. (2016). China fertilizer development research report (In Chinese) (China Agricultural University Press: Beijing).

61. Zhang, X. M., Gu, B. J., van Grinsven, H., Lam, S. K., Liang, X., Bai, M., Chen, D. L. (2020). Societal benefits of halving agricultural ammonia emissions in China far exceed the abatement costs. *Nature Communications* 11: 4357.

62. Zhang, X. M., Wu, Y. Y., Liu, X. J., Reis, S., Jin, J. X., Dragosits, U., . . . Gu, B. J.

(2017). Ammonia emissions may be substantially underestimated in China. *Environmental Science & Technology* 51: 12089-12096.

63. Zhang, Y. S., Luan, S. J., Chen, L. L., Shao, M. (2011). Estimating the volatilization of ammonia from synthetic nitrogenous fertilizers used in China. *Journal of Environmental Management* 92: 480-493.
64. Zhou, F., Bo, Y., Ciais, P., Dumas, P., Tang, Q. H., Wang, X. H., . . . Wada, Y. (2020). Deceleration of China's human water use and its key drivers. *Proceedings of the National Academy of Sciences of the United States of America* 117: 7702-7711.

TABLE 1 Policies on fertilization and crop structure issued by the Chinese Government since mid-1990s

Policy name	Acronym	Starting year	Related parameter driving NH_3 emissions	Description
Agricultural Cost-saving and Efficiency-increasing Program	ACE	1994	Incorporation proportion of synthetic-N fertilizer	Implement deep fertilization machine to increase fertilizer use efficiency and save agricultural cost for field crops (Wu 2000)
Vegetable Basket Program	VTB	Phase I: 1988 Phase II: 1995	Crop structure	Encourage the growth of cash crops, especially vegetables and fruits, around cities to meet increased consumption requirements (Bai et al., 2018)
Encouragement of urea production guideline	EUP	1996	Fertilizer form	Encourage medium- and small- size manufacturers upgraded production devices towards high concentration N fertilizer (i.e. urea, with 46% N content) to replace ammonium bicarbonate (17% N content) (Li 2009)
National Soil Testing and Nutrient Recommendation Program	STNR	2005	N application rate	Optimize nutrient management through soil testing (MARA 2015a)

TABLE 2 Cropland-NH_3 mitigation pathways in future

Scenario	Acronym	Main consequence	Key indicators in 2050		
			Sowing area (10^8 ha)	N fertilizer rate (kg N ha^{-1})	N fertilizer input (Tg N yr^{-1})
Business as usual	BAU	Increased sowing area and N fertilizer input; Increased NH_3 loss in cropland	2.0	213.5	42.7
Optimized fertilizer management	OFM	Reduced use of chemical fertilizer; Reduced NH_3 loss in cropland; Improved N use efficiency	2.0	105.5	21.1
Optimized food consumption	OFC	Reduced food loss and waste; Reduced net land requirement and N fertilizer input for crop production	1.8	203.9	36.7
Combined all the mitigation measures	ALL	Combined consequence of scenarios OFM and OFC	1.8	100	18

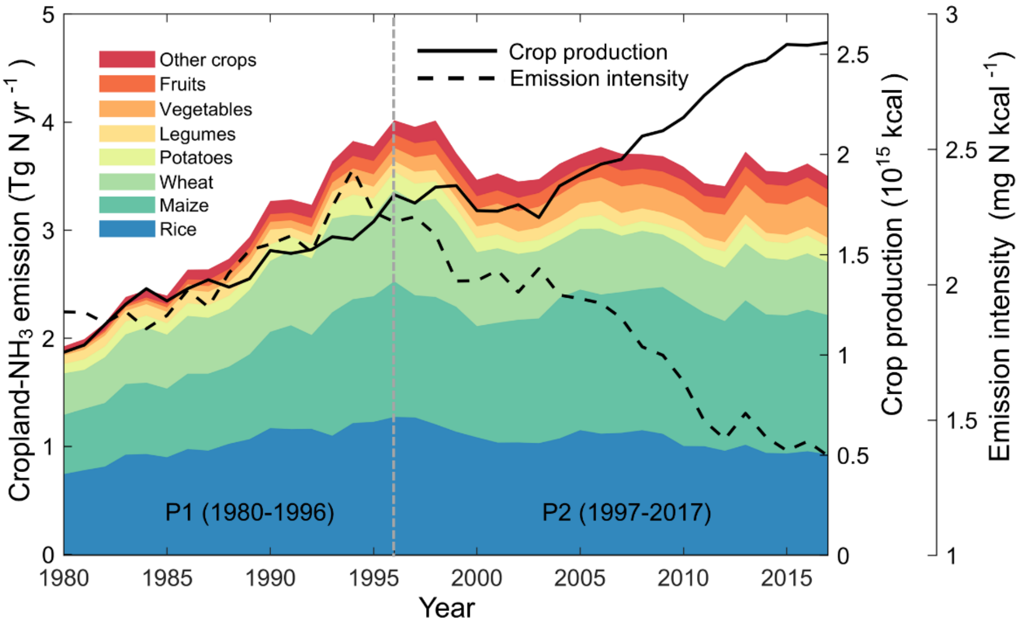


FIGURE 1 The interannual variabilities of cropland-NH_3 emissions, crop production and NH_3 emission intensity in China. The national mean emission intensity was defined as the cropland-NH_3 emission divided by total crop production (in kilocalories, Table S7) at national scale.

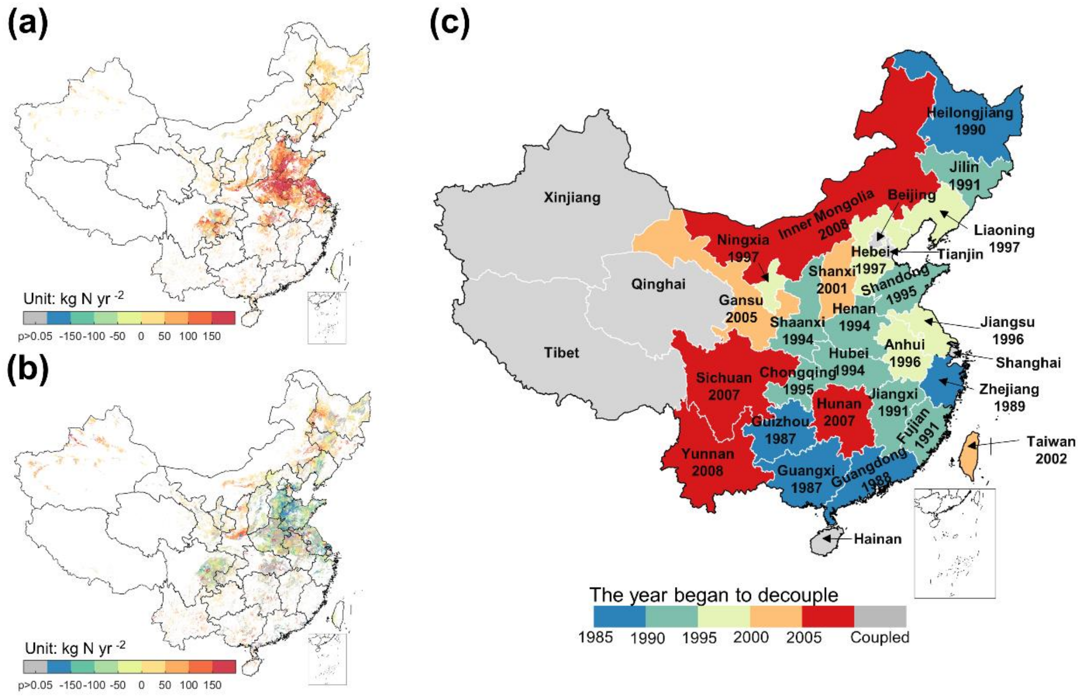


FIGURE 2 Spatial pattern of China's cropland-NH_3 emission trends and the breakpoint at province scale. Panels **a** and **b** represent the spatial pattern of cropland-NH_3 emission trends in P1 (1980-1996) and P2 (1997-2017) respectively. Panel **c** represents the year began to decouple its NH_3 emission from crop production, that is, the year which emission intensity turned to significant decrease (P<0.05) at province scale. Piecewise linear regression was applied to detect the provincial breakpoint following Zhou et al. (2020, see Text S8).

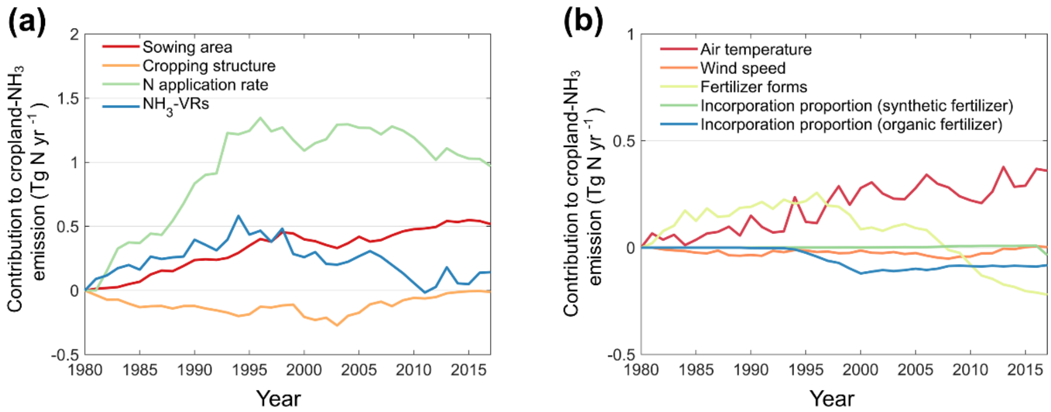


FIGURE 3 Contributions of driving factors to China's cropland-NH_3 emission and NH_3-VRs. Panels **a** represents four main driving factors' contributions to cropland-NH_3 emission. Panels **b** represents five secondary driving factors' contributions to NH_3-VRs.

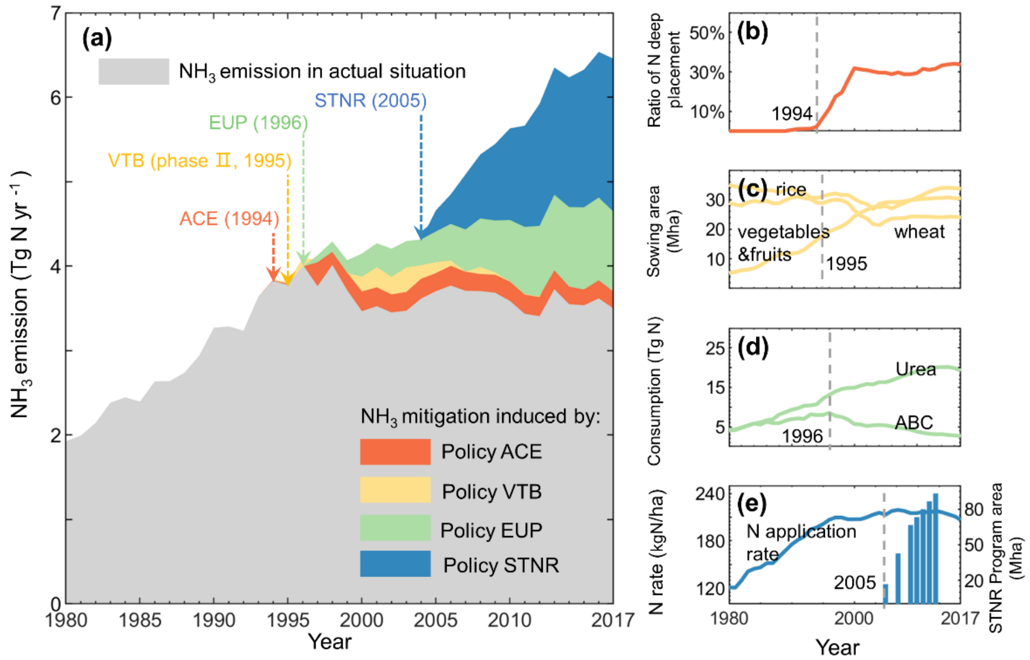
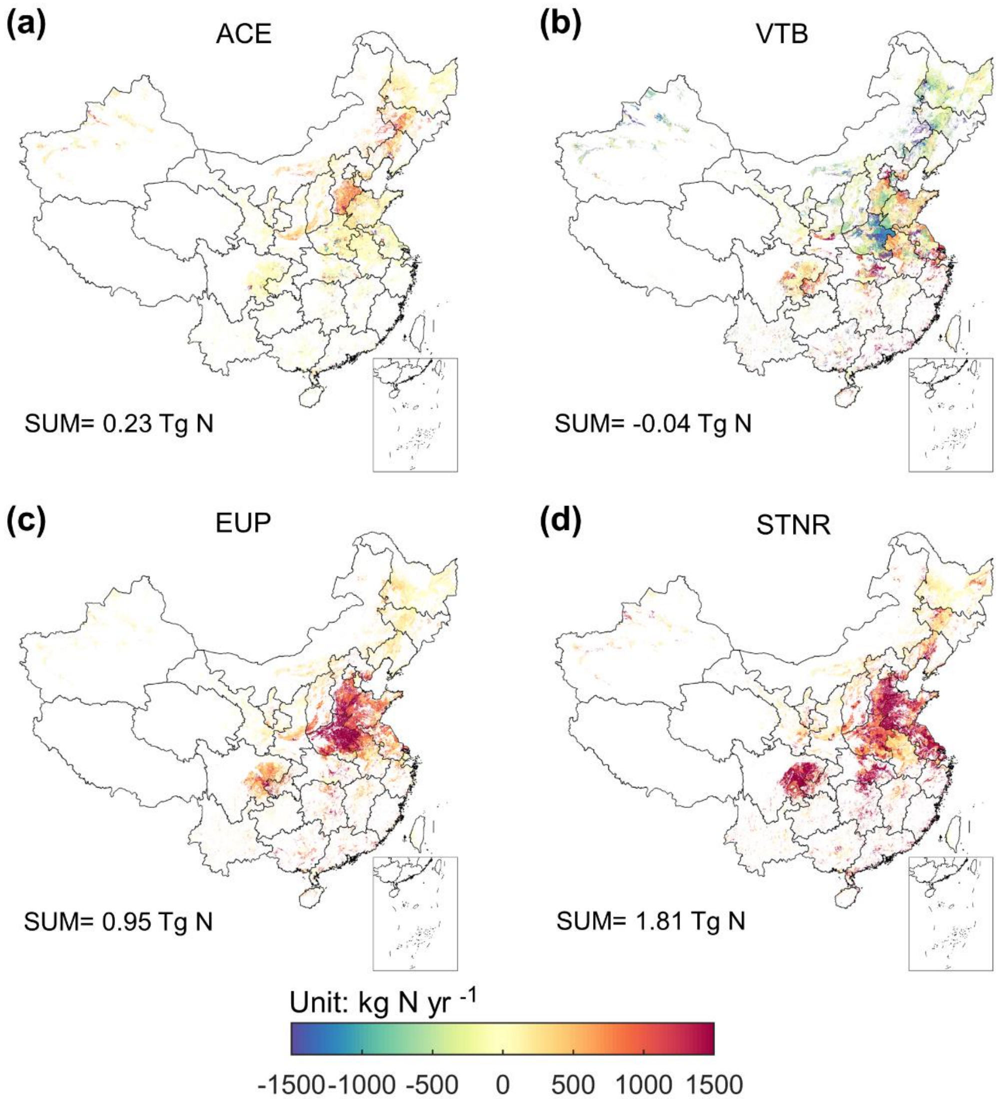


FIGURE 4 Changes of N application rate, forms, placement, crop structure and their potential effects on cropland-NH3 emission from 1980 to 2017. (a) ACE, VTB, EUP and STNR Program represent Agricultural Cost-saving and Efficiency-increasing Program, Vegetable Basket Program (Phase II), Encouragement of urea production guideline, National Soil Testing and Nutrient Recommendation Program, respectively. Detailed descriptions of above four policies can be found in Table 1. **(b)** Share of basal fertilizer incorporated by machine for four field crops, i.e. wheat, maize, potatoes and legumes. **(c)** Sowing areas of rice, wheat, and vegetables & fruits in China. **(d)** Consumption of two forms of alkaline fertilizer, i.e. urea and ammonium bicarbonate (ABC). **(e)** N application rate (line), and implemention area of the STNR program at national scale (column). After 2013, implemention area of the STNR program is not publicly available.



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207 **FIGURE 5 Cropland-NH_3 mitigation induced by policies implement in 2017.** Detail

208 descriptions of four policies can be found in Table 1. Values denote the probable NH_3

209 reductions induced by each policy at national scale.

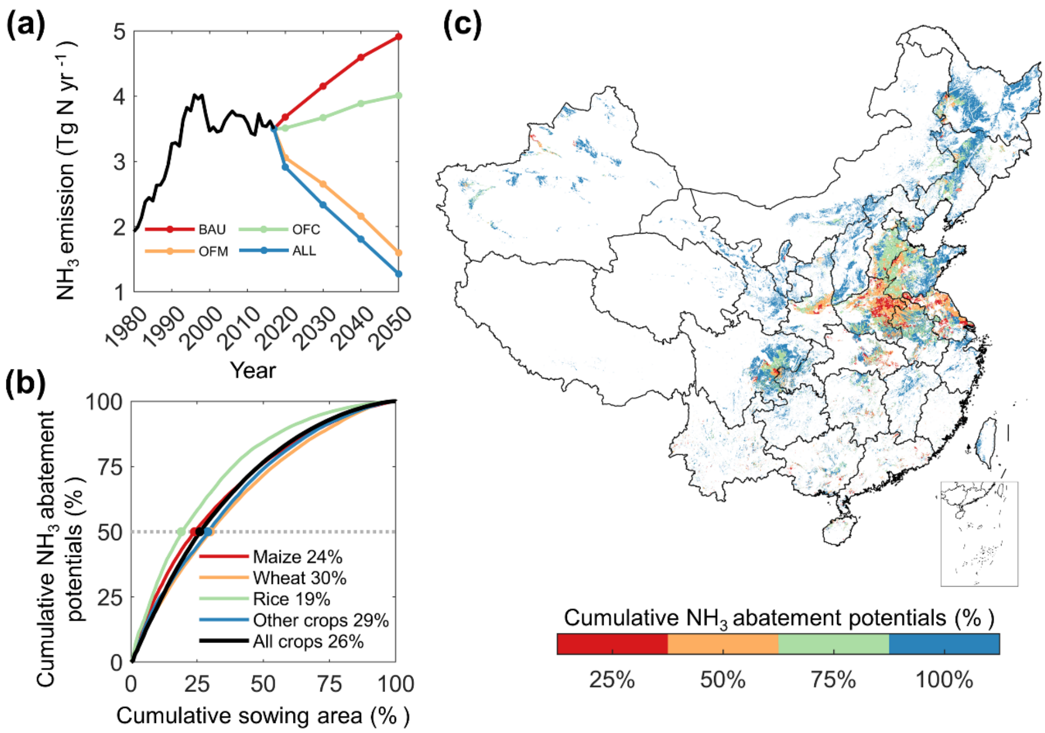

FIGURE 6 Mitigation potentials of China's cropland-NH_3. (a) Future NH_3 emissions under four scenarios; (b) China's cropland-NH_3 mitigation potentials by crop under scenario ALL; (c) Spatial pattern of China's cumulative NH_3 abatement potentials under scenario ALL.