Author Responses to Referees' Comments on **"Effects of enhancing nitrogen use efficiency in cropland and livestock systems on agricultural ammonia emissions and particulate matter air quality in China"** by Luo et al. (MS No.: egusphere-2025-72)

Our point-by-point responses are provided below. The referees' comments are *italicized*, our new/modified text is highlighted in **bold**. The revised manuscript with tracked changes is also included in the linked file below for the Editor's easy reference:

https://gocuhk-my.sharepoint.com/:w:/g/personal/amostai_cuhk_edu_hk/ETdLdM6V139Kpj5WBZ4xu34BayhK-n8Wwo5I7xnh7kCc3g?e=aScydl

Response to Referee #1

*This article constructed a 1km agricultural NH3 emission inventory for China for the year 2017. Through several agricultural NUE increasing scenarios, they investigated the implications for NH3 emissions and provincial PM2.5 air pollution mitigation. The authors did an in-depth analyses of benefits for various seasons and provinces. The research highlighted the prioritized provinces and crop types for NUE improvements and associated air quality benefits. I recommend its acceptance upon addressing the following comments through minor revision.*

We thank the reviewer for the invaluable comments, which help us improve the manuscript substantially. According to your comments, we have revised accordingly to address the reviewer's concerns.

*1. It's not easy to construct a such high geographical resolution NH3 inventory all based on solid data about activity levels and emission factors, particularly since we do not know below county level the nitrogen fertilizer use situation and manure management information. Although the authors have stated that 'It is assumed that all other crops are distributed uniformly throughout the croplands of each province.' 'The gridded livestock population map at 1 km, including cattle, sheep, goat, pork, and poultry was obtained from Cheng et al. (2023) '. They should disclose more information to what extent current simplification or treatment might affect the NH3 mitigation and PM2.5 mitigation assessment. NH3-contributed PM2.5 may provide a particularly large health impact for populated areas. If the cropland are assumed to be distributed uniformly within one provinces for other major crops, that may lead to large biases in the air pollution impact assessments. It is the same for manure, what is the assumption Cheng et al. 2023 used for allocating livestock population to 1km scale? That assumption would be critical for understanding the validity of livestock NH3 geographical distribution estimated. Also please clarify the EFs, including the geographical resolution and parameterization.*

We thank the reviewer for the very helpful comments. We acknowledge that the absence of certain information could introduce uncertainties into the $NH_3$ emission inventory. As outlined in the introduction, there exist two predominant spatial resolutions for $NH_3$ emission inventories: one at 1 km and the other at 10 km. The coarser resolution relies on a crop and livestock distribution map at a 10 km spatial scale, while the finer resolution utilizes cropland and grassland distribution maps. Previous inventories at 1 km have commonly assumed that $NH_3$ emissions related to crops and livestock are uniformly distributed across croplands and grasslands, respectively (Kang et al., 2016; Huang et al., 2012). Despite this assumption introducing biases into the emission inventory, the accuracy of this inventory has been widely recognized. In our study, to enhance its spatial resolution, we have incorporated high-resolution maps of wheat, maize, rice, cattle, sheep, goats, pork, and poultry.

Wheat, maize, and rice occupy 60% of China's total planting area and contribute to 58% of crop-related $NH_3$ emissions. High-resolution livestock distribution maps were utilized to assess livestock-related $NH_3$ emissions in China. The map combined data from diverse sources, including provincial, municipal, and county statistics, agricultural census data, and intensive farm registration records. Intensive livestock systems, hosting 60% of the total livestock population, are geographically assigned to 1 km grid cells based on the locations of intensive livestock farms. Conversely, extensive livestock farms, primarily comprised of backyard farms involving smallholders, are allocated based on the distribution of rural inhabitants. This dataset excels in accuracy compared to existing livestock distribution maps, particularly in delineating livestock distribution in urban, peri-urban, and rural regions. Additionally, meteorological data (temperature and wind speed) and soil characteristics (soil pH and CEC) are accessible at a 1 km spatial resolution. Our inventory was input into the GCHP model for performance evaluation. In comparison to the MEIC inventory, our model demonstrates superior capabilities in simulating surface $NH_3$ and $PM_{2.5}$ levels.

In summary, ~20% of agricultural $NH_3$ emissions were evenly distributed using simplifying assumptions, which may have led to some uncertainties. However, our inventory has better performance in reproducing the spatial patterns of surface $NH_3$ and $PM_{2.5}$ levels. In our revised manuscript, we have provided more information about livestock map, EFs' resolution and parameterization, as well as the discussion about the uncertainty of our inventory:

Line 129 and 134: "The gridded soil pH and CEC data **(1 km × 1 km)** were obtained from the Harmonized World Soil Database" "where m represents months; $T$ (°C) and $u$ (m s$^{-1}$) are air temperature and wind speed at 2 m height, whereby their gridded values **(1 km × 1 km)** are from Peng et al. (2019) and National Earth System Science Data Center. **The high-resolution climate data were produced by spatially downscaling the 30-min Climatic Research Unit (CRU) time-series dataset with WorldClim climatology using the delta downscaling method.**"

Line 145-146 "**The livestock EFs were first calculated for each livestock type across livestock manure management stages**. Same as Zhang et al. (2018), these emissions EFs are further modulated by the effect of temperature and wind speed following Eq. 4."

Line 166-174 "The gridded livestock population map at 1 km, including cattle, sheep, goat, pork, and poultry was obtained from Cheng et al. (2023) (Fig. S3). **The livestock map consolidates data from various sources, encompassing provincial, municipal, and county statistics, alongside agricultural census records and intensive farm registration data. Intensive livestock populations, constituting ~60% of the total livestock count, are assigned to 1 km grid cells according to the positions and breeding scales of intensive livestock farms. Meanwhile, extensive livestock populations, primarily comprised of backyard farms involving smallholders, are allocated based on the spatial distribution of rural inhabitants. This dataset offers heightened precision compared to existing livestock distribution maps, particularly in delineating livestock presence across urban, peri-urban, and rural regions.**

**The maps of EFs were created for each grid cell using Eq. 3 and Eq. 4, first by calculating the baseline EFs first and further modulating them by meteorological conditions.**"

Line 630-640: "In addition, the lack of regional-specific EFs poses a challenge to accurately reproduce the spatial pattern of $NH_3$ emissions. **About 20% of agricultural $NH_3$ emissions were evenly distributed using simplifying assumptions, which may have led to uncertainties in gridded allocation. Such uncertainties may induce biases in $NH_3$ and $PM_{2.5}$ mitigation assessment. We utilized the CHANS model to calculate NUE for crop and livestock systems; however, nitrogen budget models like this suffer uncertainties stemming from simplifications of the intricate nitrogen cycle and data deficiencies**

**(Zhang et al., 2021a, b). The estimation uncertainty of nitrogen inputs was noted at ~10%, whereas nitrogen output uncertainty could soar to ~30%, primarily due to challenges in accurately predicting nitrogen levels in individual agricultural products (Zhang et al., 2021b). As for the improved NUE scenarios, while a range of specific actionable strategies can be implemented (Table S9 and S10), it is crucial to acknowledge the challenges associated with executing these measures across different levels, considering the costs and anticipated outcomes. Moreover, our assumption of a simultaneous decrease in all nitrogen losses may not fully account for scenarios where certain measures prioritize $NH_3$ control over other forms of nitrogen loss mitigation.**"

*2. It is relatively easy to use NUE to construct scenarios for crop and livestock management rather than specific technological bundles, however, how realistic are these NUE scenarios? Are technologies available to achieve NUE defined here? Less is known about the potential of improving fruits and vegetables NUE compared to other crops. Furthermore, calculations for crop NUE itself can involve substantial uncertainties and data problems, see Zhang, X., Zou, T., Lassaletta, L. et al. Quantification of global and national nitrogen budgets for crop production. Nat Food 2, 529–540 (2021). https://doi.org/10.1038/s43016-021-00318-5. For livestock, could CHANS model represent flow of TAN across various manure handling stages? Since your emission inventory represent flow of TAN - but I suspect CHANS's representation for manure N would not be as sophisticated. How would the CHANS calculated livestock NUE in China compared to other nitrogen budgets research methods? It still is worthy of conducting a more detailed literature search to understand the uncertainties and give some paraphrases in the Discussion section.*

We appreciate the valuable comments from the reviewer. The NUE target for crop systems aligns with the performance of the top 20% of farmers in China, a localized objective that has demonstrated achievability. Liu et al. (2024) demonstrated in detail the feasibility of such localization goals, highlighting substantial NUE enhancements achievable through optimizing nitrogen fertilizer application rates and adopting advanced technologies. As for livestock, previous studies also showed that certain measures could enhance NUE (Zhang et al., 2020; Bai et al., 2018). We have provided potential strategies for reducing $NH_3$ emissions and enhancing NUE in China, facilitating the development of effective region-specific plans. Notwithstanding the current Chinese primary focus on N management in cereal crops but with less emphasis on fruits and vegetables, it is essential to note that effective measures exist for improving their NUE as well. Strategies such as optimizing fertilizer application rates, urea substitution, utilizing enhanced-efficiency N fertilizers, and integrating irrigation-fertilization management are all useful for enhancing NUE in fruits and vegetables.

We employed the CHANS model to calculate NUE for crop and livestock systems, acknowledging potential uncertainties in its evaluation attributable to the model's simplified processes and parameters. Since NUE is defined as the ratio of nitrogen output to input, uncertainties primarily stem from the estimation of nitrogen inputs and outputs. In the CHANS model, the uncertainties in N inputs and outputs were approximated at ~10% and ~30%, respectively, as a result of the simplified representation of intricate nitrogen cycling processes and data constraints (Zhang et al., 2021). Consistent with our findings, the NUE of cereal crops, deduced from survey data on N inputs, stands at 0.45, with spatial patterns exhibiting similar trends (Zhang, 2021). Overall, the lack of accurate estimates of nitrogen output is the major cause of the bias in nitrogen budget.

In the context of livestock systems, the CHANS model lacks the capability to quantify nitrogen flow across various stages. Following the method of Huang et al. (2012) and Zhang et al. (2018), we estimated the $NH_3$ emissions across different stages. The NUE for ruminants and monogastric animals (such as pork and poultry) is approximately 0.05 and 0.25, respectively, as evaluated by the NUFER model, a widely used localized nitrogen budget model in China, aligning closely with our findings. Regarding spatial distribution, the NUE of livestock system exhibits higher values in the eastern regions and lower values in the western regions, mirroring our own results (Jin et al., 2021).

We have added more information about the performance and uncertainties of CHANS model, as well as available measures for improving NUE. The revisions are as follows:

Line 245-246: "**The available measures to achieve these scenarios with mitigation efficiency in China are shown in Table S9 and S10.**"

**Table S9.** Available mitigation options for croplands in China (derived from meta-analysis of Zhang et al. (2020) and Liu et al. (2021)).

| Aspects | Options | Mitigation efficiency |
|---|---|---|
| Nitrogen application rate | 25% optimal N application rate | 18%–32.4% |
| | 50% optimal N application rate | 25%–48.5% |
| | 75% optimal N application rate | 48.2%–68.3% |
| Application method | Deep placement of fertilizer | 45.1%–79.4% |
| | Irrigation-fertilization integration management | 60.2%–77.4% |
| Cropland management | Recycling straw to croplands | 0%–18.6% |
| | Reducing basal N fertilizer | 26%–62% |
| N fertilizer type | Urea substitution | 8.6%–48.8% |
| | Application of organic fertilizer | 44.7%–63.6% |
| Enhanced-efficiency N fertilizer | Controlled release fertilizer | 46.8%–58.3% |
| | Inhibitor | 21.7%–70.4% |

**Table S10.** Available mitigation options for livestock in China (derived from meta-analysis of Zhang et al. (2020) and Liu et al. (2021)).

| Stages | Options | Mitigation efficiency |
|---|---|---|
| Feeding | Low crude protein feeding | 10%–46% |
| | Dietary additives | 33%–45% |
| | Phase feeding | ~10% |
| Housing | Floor adaption | 10%–50% |
| | Bedding materials | 20%–50% |
| | Air scrubbing techniques or bio-filter | 70%–95% |
| | Frequent manure removal | 25%–30% |
| | Rapid manure drying | 70%–90% |
| Storage | Solid-liquid separation | 20%–30% |
| | Improvement in storage facility | 26%–62% |
| | Manure surface covers | 40%–60% |
| | Acidification by additives | 18%–70% |
| | Composting | ~55% |
| | Cooling | 20%–30% |
| Spreading | Band spreading | 38%–75% |
| | Incorporation | 45%–65% |
| | Injection (slurry only) | 80%–90% |
| Grazing | Adjusting the grazing time | ~10% |

Line 461-463: "In China, a west-to-east trend of increasing NUE of livestock systems is observed, as shown in Figure S6b**. Similar NUE values and spatial patterns have been reported in another localized nitrogen budget model (NUFER model) (Bai et al., 2018; Jin et al., 2021). Based on this model, the NUE of ruminants and monogastric animals are ~0.05 and ~0.25, respectively.**"

Line 630-640: "**About 20% of agricultural $NH_3$ emissions were evenly distributed using simplifying assumptions, which may have led to uncertainties in gridded allocation. Such uncertainties may induce biases in $NH_3$ and $PM_{2.5}$ mitigation assessment. We utilized the CHANS model to calculate NUE for crop and livestock systems; however, nitrogen budget models like this suffer uncertainties stemming from simplifications of the intricate nitrogen cycle and data deficiencies (Zhang et al., 2021a, b). The estimation uncertainty of nitrogen inputs was noted at ~10%, whereas nitrogen output uncertainty could soar to ~30%, primarily due to challenges in accurately predicting nitrogen levels in individual agricultural products (Zhang et al., 2021b). As for the improved NUE scenarios, while a range of specific actionable strategies can be implemented (Table S9 and S10), it is crucial to acknowledge the challenges associated with executing these measures across different levels, considering the costs and anticipated outcomes. Moreover, our assumption of a simultaneous decrease in all nitrogen losses may not fully account for scenarios where certain measures prioritize $NH_3$ control over other forms of nitrogen loss mitigation.**"

Bai, Z., Ma, W., Ma, L., Velthof, G. L., Wei, Z., Havlík, P., Oenema, O., Lee, M. R. F., and Zhang, F.: China's livestock transition: Driving forces, impacts, and consequences, Sci. Adv., 4, https://doi.org/10.1126/sciadv.aar8534, 2018.
Jin, X., Zhang, N., Zhao, Z., Bai, Z., and Ma, L.: Nitrogen budgets of contrasting crop-livestock systems in China, Environ. Pollut., 288, 117633, https://doi.org/10.1016/j.envpol.2021.117633, 2021.
Liu, X., Sha, Z., Song, Y., Dong, H., Pan, Y., Gao, Z., Li, Y., Ma, L., Dong, W., Hu, C., Wang, W., Wang, Y., Geng, H., Zheng, Y., and Gu, M.: China's Atmospheric Ammonia Emission Characteristics, Mitigation Options and Policy Recommendations, Res. Environ. Sci., 34, 149–157, 2021.
Liu, Y., Zhuang, M., Liang, X., Lam, S. K., Chen, D., Malik, A., Li, M., Lenzen, M., Zhang, L., Zhang, R., Zhang, L., and Hao, Y.: Localized nitrogen management strategies can halve fertilizer use in Chinese staple crop production, Nat. Food, 5, 825–835, https://doi.org/10.1038/s43016-024-01057-z, 2024.
Zhang, Q.: Nitrogen, phosphorus and potassium nutrient balance and optimization approaches of major crops in China, China Agricultural University, 2021.
Zhang, X., Gu, B., van Grinsven, H., Lam, S. K., Liang, X., Bai, M., and Chen, D.: Societal benefits of halving agricultural ammonia emissions in China far exceed the abatement costs, Nat. Commun., 11, 4357, https://doi.org/10.1038/s41467-020-18196-z, 2020.

*3. The GCHP simulation is done for one year for China in quite high resolution. I wonder what is the computing resources and time taken for completing the baseline simulation?*

We perform monthly simulations, and it takes about 11 hours for one-month simulation using 60 cores.

*4. Atmospheric background emissions, which affect contribution of NH3 to PM2.5, have changed a lot between 2017 and the present. Could the authors comment on the implications for the effectiveness of these NUE-increasing scenarios?*

Since the initiation of China's Air Clean Action Strategy in 2013, there has been a significant reduction in $SO_2$ and $NO_x$ emissions, particularly in the case of $SO_2$, leading to a notable alleviation of $PM_{2.5}$ pollution. However, effective control measures for $NH_3$ emissions are lacking, potentially offsetting the air quality improvements achieved through the reduction of $SO_2$ and $NO_x$. There is growing evidence of the importance of $NH_3$ for $PM_{2.5}$ control in China. Fu et al. (2017) indicated that the rise in $NH_3$ concentrations has undermined the benefits of reducing SNA concentrations (especially for nitrate) via emissions control of $SO_2$ and $NO_x$. Comparing air pollution in China before and after the COVID-19 lockdown, Xu et al. (2022) observed that while there was a sharp reduction in $SO_2$ and $NO_x$ emissions during the lockdown, the concurrent increase in $NH_3$ concentrations may have contributed to the persistent high levels of $PM_{2.5}$ pollution.

It is noteworthy that the effectiveness of $PM_{2.5}$ control through $NH_3$ reduction diminishes as $SO_2$ and $NO_x$ levels decrease further (Liu et al., 2021b). In $NH_3$-rich environments, such as agricultural regions, there is a scarcity of adequate acidic gases to neutralize the $NH_3$, thereby constraining the formation of $NH_4^+$ within $PM_{2.5}$. Despite the diminished air quality improvements resulting from $NH_3$ mitigation, the significance of $NH_3$ control remains paramount. From a cost perspective, the expense of $NH_3$ abatement is only ~10% of that associated with $NO_x$ abatement (Gu et al., 2021). As China intensifies its efforts to reduce $NO_x$ emissions, the abatement costs are anticipated to escalate. Furthermore, enhancing NUE not only curbs $NH_3$ emissions, but also lower $N_2O$ emissions, water nitrogen leaching, nitrogen deposition, and lowers fertilizer expenses. Furthermore, reductions in $NH_3$ emissions lead to decreased atmospheric nitrogen deposition. Therefore, it is essential to improve the NUE of agricultural systems.

We have now added more information in the concluding section:

Line 614-624: "**Since the initiation of China's Air Clean Action Strategy in 2013, there have been significant reductions in $SO_2$ and $NO_x$ emissions, notably alleviating $PM_{2.5}$ pollution. However, effective control measures for $NH_3$ emissions are lacking, potentially limiting further air quality improvements. It is noteworthy that the effectiveness of $PM_{2.5}$ control through $NH_3$ reductions will likely diminish as $SO_2$ and $NO_x$ levels decrease further (Liu et al., 2021b), because in $NH_3$-rich environments, there may be a scarcity of adequate acidic gases to neutralize $NH_3$, thereby constraining ammonium formation. Nevertheless, the significance of $NH_3$ control via improving NUE remains. From a cost perspective, the expense of $NH_3$ abatement is only ~10% of that associated with $NO_x$ abatement (Gu et al., 2021). As China intensifies its efforts to reduce $NO_x$ emissions, the abatement costs are anticipated to rise. Furthermore, enhancing NUE not only curbs $NH_3$ emissions, but also lowers $N_2O$ emissions, nitrogen leaching to water, nitrogen deposition, and fertilizer expenses, thus offering climatic, ecological and socioeconomic co-benefits.**"

Fu, X., Wang, S., Xing, J., Zhang, X., Wang, T., and Hao, J.: Increasing Ammonia Concentrations Reduce the Effectiveness of Particle Pollution Control Achieved via SO 2 and NO X Emissions Reduction in East China, Environ. Sci. Technol. Lett., 4, 221–227, https://doi.org/10.1021/acs.estlett.7b00143, 2017.
Gu, B., Zhang, L., Dingenen, R. Van, Vieno, M., Grinsven, H. J. Van, Zhang, X., Zhang, S., Chen, Y., Wang, S., Ren, C., Rao, S., Holland, M., Winiwarter, W., Chen, D., Xu, J., and

Sutton, M. A.: Abating ammonia is more cost-effective than nitrogen oxides for mitigating PM2.5 air pollution, Science (80-. )., 374, 758–762, https://doi.org/10.1126/science.abf8623, 2021.
Liu, Z., Zhou, M., Chen, Y., Chen, D., Pan, Y., Song, T., Ji, D., Chen, Q., and Zhang, L.: The nonlinear response of fine particulate matter pollution to ammonia emission reductions in North China, Environ. Res. Lett., 16, https://doi.org/10.1088/1748-9326/abdf86, 2021b.
Xu, W., Zhao, Y., Wen, Z., Chang, Y., Pan, Y., Sun, Y., Ma, X., Sha, Z., Li, Z., Kang, J., Liu, L., Tang, A., Wang, K., Zhang, Y., Guo, Y., Zhang, L., Sheng, L., Zhang, X., Gu, B., Song, Y., Van Damme, M., Clarisse, L., Coheur, P.-F., Collett, J. L., Goulding, K., Zhang, F., He, K., and Liu, X.: Increasing importance of ammonia emission abatement in PM2.5 pollution control, Sci. Bull., 67, 1745–1749, https://doi.org/10.1016/j.scib.2022.07.021, 2022.

Response to Referee #2

*This study develops a high-resolution NH3 emission inventory for Chinese agriculture and integrates it with nitrogen flow and air quality models to assess mitigation potentials. Results show that cropland NUE improvements and organic fertilizer use offer greater NH3 reduction than livestock measures, with distinct regional effectiveness. Specifically, organic fertilizers are found to be most effective in grain-producing regions, while NUE enhancement benefits southern coastal areas. The analysis particularly highlights severe over-fertilization in vegetable/fruit production as a critical mitigation target. These findings provide scientific support for China's emerging agricultural NH3 control policies while revealing data gaps for future refinement, demonstrating how optimized nitrogen management can simultaneously address air pollution and sustainable development goals.*

*The study is well organized and conducted. Below are some moderate comments for further clarification of the manuscript.*

> We appreciate the reviewer for the invaluable comments and suggestions, which help us improve the manuscript substantially. According to your comments, we have replied and revised accordingly to address the reviewer's concerns.

*1) The agricultural crop-related $NH_3$ emissions in this study are significantly higher than those in other emission inventories. Could this be attributed not only to the use of localized emission factors for China but also to other potential reasons? Was the total nitrogen application amount constrained in the calculations? Additionally, the assumption that crops other than the major ones are uniformly distributed across provincial croplands—what is the basis for this method, and how does it impact the subsequent NH3 mitigation potential analysis?*

> We appreciate the feedback. Nitrogen usage has been constrained for each province. China's total nitrogen fertilizer consumption was 29.62 Tg N in 2017, with our analysis indicating that ~17% of this amount volatilizes as $NH_3$. The $NH_3$ losses from fertilizers were estimated to be around 17–18% of the synthetic nitrogen inputs based on two widely used nitrogen mass flow models in China, namely, the CHANS and NUFER models (Ma et al., 2010; Gu et al., 2015). Additionally, we have presented a comparison of emission inventories in Table 1, demonstrating that our estimates do not significantly exceed those of other studies. The discrepancy between our estimates and Kang et al. (2016) can be primarily attributable to the utilization of localized emission factors. Kang's study employed European emission factors, which are notably lower than Chinese emission factors due to advanced management and technology. In addition to bottom-up estimation, satellite-based top-down estimates suggested that the current emission inventory (e.g., MEIC inventory) underestimates emissions (Jin et al., 2023; Luo et al., 2022). Furthermore, based on the simulation outcomes of surface $NH_3$ concentrations, our inventory demonstrates enhanced accuracy. Consequently, we are confident with our fertilizer-related $NH_3$ emissions.
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> Previous inventories at 1 km have commonly assumed that $NH_3$ emissions related to crops and livestock are uniformly distributed across croplands and grasslands, respectively (Kang et al., 2016; Huang et al., 2012). This assumption rests on the premise that, in the absence of precise data on the distribution of other crops, the average fertilizer intensity used across all crops remains relatively consistent on croplands. Despite this assumption introducing biases into the emission inventory, the accuracy of this method has been widely recognized. In our study, to enhance the spatial resolution inventory, we have incorporated high-resolution maps of wheat, maize, rice, cattle, sheep, goats, pork, and poultry. About 20% of agricultural $NH_3$ emissions were evenly distributed using simplifying assumptions, which may have led to some uncertainties. These uncertainties primarily pertain to the spatial distribution of $NH_3$ emissions and have minimal impact on total $NH_3$ emission estimates. The impact of this simplifying assumption on assessing the mitigation potential of $NH_3$ emissions is limited

compared to the air quality improvements resulting from such mitigation efforts. Nonetheless, our inventory excels in replicating the spatial patterns of surface $NH_3$ and $PM_{2.5}$ levels. In our revised manuscript, we have included a discussion addressing the uncertainty associated with our simplified assumption. The revisions are as follows:

Line 630-640: "**About 20% of agricultural $NH_3$ emissions were evenly distributed using simplifying assumptions, which may have led to uncertainties in gridded allocation. Such uncertainties may induce biases in $NH_3$ and $PM_{2.5}$ mitigation assessment. We utilized the CHANS model to calculate NUE for crop and livestock systems; however, nitrogen budget models like this suffer uncertainties stemming from simplifications of the intricate nitrogen cycle and data deficiencies (Zhang et al., 2021a, b). The estimation uncertainty of nitrogen inputs was noted at ~10%, whereas nitrogen output uncertainty could soar to ~30%, primarily due to challenges in accurately predicting nitrogen levels in individual agricultural products (Zhang et al., 2021b). As for the improved NUE scenarios, while a range of specific actionable strategies can be implemented (Table S9 and S10), it is crucial to acknowledge the challenges associated with executing these measures across different levels, considering the costs and anticipated outcomes. Moreover, our assumption of a simultaneous decrease in all nitrogen losses may not fully account for scenarios where certain measures prioritize $NH_3$ control over other forms of nitrogen loss mitigation.**"


Gu, B., Ju, X., Chang, J., Ge, Y., and Vitousek, P. M.: Integrated reactive nitrogen budgets and future trends in China, Proc. Natl. Acad. Sci., 112, 8792–8797, https://doi.org/10.1073/pnas.1510211112, 2015.

Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., and Zhang, H.: A high-resolution ammonia emission inventory in China, Global Biogeochem. Cycles, 26, 1–14, https://doi.org/10.1029/2011GB004161, 2012.

Jin, J., Fang, L., Li, B., Liao, H., Wang, Y., Han, W., Li, K., Pang, M., Wu, X., and Xiang Lin, H.: 4DEnVar-based inversion system for ammonia emission estimation in China through assimilating IASI ammonia retrievals, Environ. Res. Lett., 18, 034005, https://doi.org/10.1088/1748-9326/acb835, 2023.

Kang, Y., Liu, M., Song, Y., Huang, X., Yao, H., Cai, X., Zhang, H., Kang, L., Liu, X., Yan, X., He, H., Zhang, Q., Shao, M., and Zhu, T.: High-resolution ammonia emissions inventories in China from 1980 to 2012, Atmos. Chem. Phys., 16, 2043–2058, https://doi.org/10.5194/acp-16-2043-2016, 2016.

Luo, Z., Zhang, Y., Chen, W., Van Damme, M., Coheur, P.-F., and Clarisse, L.: Estimating global ammonia (NH3) emissions based on IASI observations from 2008 to 2018, Atmos. Chem. Phys., 22, 10375–10388, https://doi.org/10.5194/acp-22-10375-2022, 2022.

Ma, L., Ma, W. Q., Velthof, G. L., Wang, F. H., Qin, W., Zhang, F. S., and Oenema, O.: Modeling Nutrient Flows in the Food Chain of China, J. Environ. Qual., 39, 1279–1289, https://doi.org/10.2134/jeq2009.0403, 2010.


*2) In the NUE-C and OUR mitigation scenarios, the proposed measures may also alter the corresponding emission factors (EFs). Did the study only consider reductions in nitrogen input (activity data)? Could you further explain how the mitigation measures were integrated with the emission inventory? (Page 9)*

We thank the reviewer's comments. It is true that various measures to increase the NUE of crops, such as deep placement of fertilizer and urea substitution, prove effective in reducing EFs. By lowering EFs, a smaller portion of nitrogen input is lost to the environment while a larger proportion is utilized by the crop, resulting in a decreased nitrogen input. Thus, viewed holistically, any efforts to enhance NUE can ultimately be interpreted as a reduction in system nitrogen inputs, consequently lowering $NH_3$ emissions while ensuring nitrogen outputs. We also acknowledge that $NH_3$ reduction potential evaluation may be subject to some uncertainty, e.g., we assumed a simultaneous reduction of all nitrogen losses, such as $NH_3$ emissions and water nitrogen leakage, under the improved NUE scenario, however, some measures might emphasize $NH_3$ control over other forms of nitrogen loss mitigation.

Given the absence of detailed information regarding implementation rates and effectiveness of individual $NH_3$ abatement measures across various crop and livestock systems, in this study we devised $NH_3$ abatement scenarios based on NUE. We calculated $NH_3$ emission reductions utilizing mitigation efficiency, as defined by the following equation:

$$Mitigation\ efficiency = \frac{NUE_t - NUE_b}{NUE_t}$$
$$NH_{3,t} = NH_{3,b}(1 - Mitigation\ efficiency)$$

where $t$ and $b$ represent the scenario of target and baseline. $NUE_t$ and $NUE_b$ refer to the NUE of target and baseline scenarios. $NH_{3,t}$ and $NH_{3,b}$ refer to the $NH_3$ emissions of target and baseline scenarios.

We have added the mitigation measures available for improving NUE, and discussion about the uncertainty of our scenarios, as detailed in our response to the question above and cited revised text in Line 608-615 of the manuscript, and also:

Line 245-246: "**The available measures to achieve these scenarios with mitigation efficiency in China are shown in Table S9 and S10.**"

**Table S9.** Available mitigation options for croplands in China (derived from meta-analysis of Zhang et al. (2020) and Liu et al. (2021)).

| Aspects | Options | Mitigation efficiency |
|---|---|---|
| Nitrogen application rate | 25% optimal N application rate | 18%–32.4% |
| | 50% optimal N application rate | 25%–48.5% |
| | 75% optimal N application rate | 48.2%–68.3% |
| Application method | Deep placement of fertilizer | 45.1%–79.4% |
| | Irrigation-fertilization integration management | 60.2%–77.4% |
| Cropland management | Recycling straw to croplands | 0%–18.6% |
| | Reducing basal N fertilizer | 26%–62% |
| N fertilizer type | Urea substitution | 8.6%–48.8% |
| | Application of organic fertilizer | 44.7%–63.6% |
| Enhanced-efficiency N fertilizer | Controlled release fertilizer | 46.8%–58.3% |
| | Inhibitor | 21.7%–70.4% |

**Table S10.** Available mitigation options for livestock in China (derived from meta-analysis of Zhang et al. (2020) and Liu et al. (2021)).

| Stages | Options | Mitigation efficiency |
|---|---|---|
| Feeding | Low crude protein feeding | 10%–46% |
| | Dietary additives | 33%–45% |
| | Phase feeding | ~10% |
| Housing | Floor adaption | 10%–50% |
| | Bedding materials | 20%–50% |

| | | |
|---|---|---|
| | Air scrubbing techniques or bio-filter | 70%–95% |
| | Frequent manure removal | 25%–30% |
| | Rapid manure drying | 70%–90% |
| | Solid-liquid separation | 20%–30% |
| Storage | Improvement in storage facility | 26%–62% |
| | Manure surface covers | 40%–60% |
| | Acidification by additives | 18%–70% |
| | Composting | ~55% |
| | Cooling | 20%–30% |
| | Band spreading | 38%–75% |
| Spreading | Incorporation | 45%–65% |
| | Injection (slurry only) | 80%–90% |
| Grazing | Adjusting the grazing time | ~10% |

Liu, X., Sha, Z., Song, Y., Dong, H., Pan, Y., Gao, Z., Li, Y., Ma, L., Dong, W., Hu, C., Wang, W., Wang, Y., Geng, H., Zheng, Y., and Gu, M.: China's Atmospheric Ammonia Emission Characteristics, Mitigation Options and Policy Recommendations, Res. Environ. Sci., 34, 149–157, 2021.
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*3) It is generally believed that poultry contributes significantly to livestock-related agricultural NH3 emissions, yet in this study, poultry accounts for a relatively small proportion (17.6%). What might explain this discrepancy? (Page 10)*

We appreciate the reviewer's feedback. We have compared the poultry-related $NH_3$ emissions with several studies. Our results align closely with the estimates presented by Li et al. (2021) and Yang et al. (2023). The method of calculating livestock-related $NH_3$ emissions in this study is similar to Li et al. (2021). Nonetheless, as highlighted by the reviewer, certain studies reported the substantial $NH_3$ emissions from poultry. In terms of $NH_3$ EF, the average EF of poultry in our study is consistent with most existing studies. Notably, Xu et al. (2016) reported higher emissions while having a smaller EF.

| Reference | The share of livestock related $NH_3$ emissions | The $NH_3$ EF of poultry (g $NH_3$ head$^{-1}$ day$^{-1}$) |
|---|---|---|
| This study | 17.6% | 0.54 |
| Li et al. (2021) | 21.2% | Similar method and data but without detail information |
| Yang et al. (2023) | 12.7% | 0.55 |
| Xu et al. (2016) | 26.2% | 0.14 |
| Xu et al. (2015) | 34% | 0.43 |
| Gao et al. (2013) | 26.6% | 0.54 |

To identify potential reasons for these discrepancies, we have examined the livestock-related $NH_3$ calculations in each study. The equation of poultry-related $NH_3$ emissions of our method is as follows:

$$NH_3 = Livestock\ population \times raising\ days \times EF$$

where livestock population is the annual slaughter number. The raising days for poultry producing meat are 50–75 days. EF is the daily EF. However, in the study of Xu et al. (2016) and Gao et al. (2013), there was a lack of focus on the shorter duration of poultry rearing, and it was unclear whether the poultry population was stock number or slaughter number. They estimated $NH_3$ from annual EF values multiplied by activity data without further clarification. Hence, we posit that the disparities may stem from uncertainties of raising days and livestock numbers. This study adopted the livestock-related $NH_3$ emission estimation method proposed by Kang et al. (2016) and Huang et al. (2012), a widely utilized approach in China known for its accuracy. Our findings on the magnitude of livestock-related $NH_3$ emissions align closely with those of Zhang et al. (2018) and Li et al. (2021), who employed the same method.

Line 353-355: "**We acknowledge the disparities in estimating livestock waste-related $NH_3$ emissions. For example, our poultry $NH_3$ estimates differ from those reported by Xu et al. (2015) and Gao et al. (2013), likely reflecting uncertainties in raising days and livestock numbers.**"

Gao, Z., Ma, W., Zhu, G., and Roelcke, M.: Estimating farm-gate ammonia emissions from major animal production systems in China, Atmos. Environ., 79, 20–28, https://doi.org/10.1016/j.atmosenv.2013.06.025, 2013.
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Zhang, L., Chen, Y., Zhao, Y., Henze, D. K., Zhu, L., Song, Y., Paulot, F., Liu, X., Pan, Y., Lin, Y., and Huang, B.: Agricultural ammonia emissions in China: Reconciling bottom-up and top-down estimates, Atmos. Chem. Phys., 18, 339–355, https://doi.org/10.5194/acp-18-339-2018, 2018.

*4) Does the HHH region include Hebei or Hubei? Spatially, Hebei (Beijing-Tianjin-Hebei region) has long been considered a hotspot for NH3 emissions. Why does this inventory instead show Jiangsu Province as having higher emissions? (Page 13)*

The HHH region encompasses Beijing, Tianjin, Hebei, Henan, and Shandong provinces. As illustrated in Figures 3b and 3c, the Beijing-Tianjin-Hebei area emerges as a significant hotspot for $NH_3$ emissions, with Henan, Shandong, and Jiangsu also exhibiting substantial agricultural $NH_3$ losses. Figure 3a depicts the $NH_3$ emission intensity rather than the total magnitude; when considering the total emissions, Hebei's agricultural $NH_3$ emissions surpass Jiangsu's by 617 Gg compared to 520 Gg (Table S12). Similar spatial patterns have been observed in previous studies by Zhang et al. (2018) and Li et al. (2021). In terms of agricultural activity data, Jiangsu province utilized 1.84 Tg of synthetic N fertilizer in 2017, slightly higher than the 1.77 Tg consumed by Hebei province. However, Hebei boasts a larger livestock population compared to Jiangsu province.

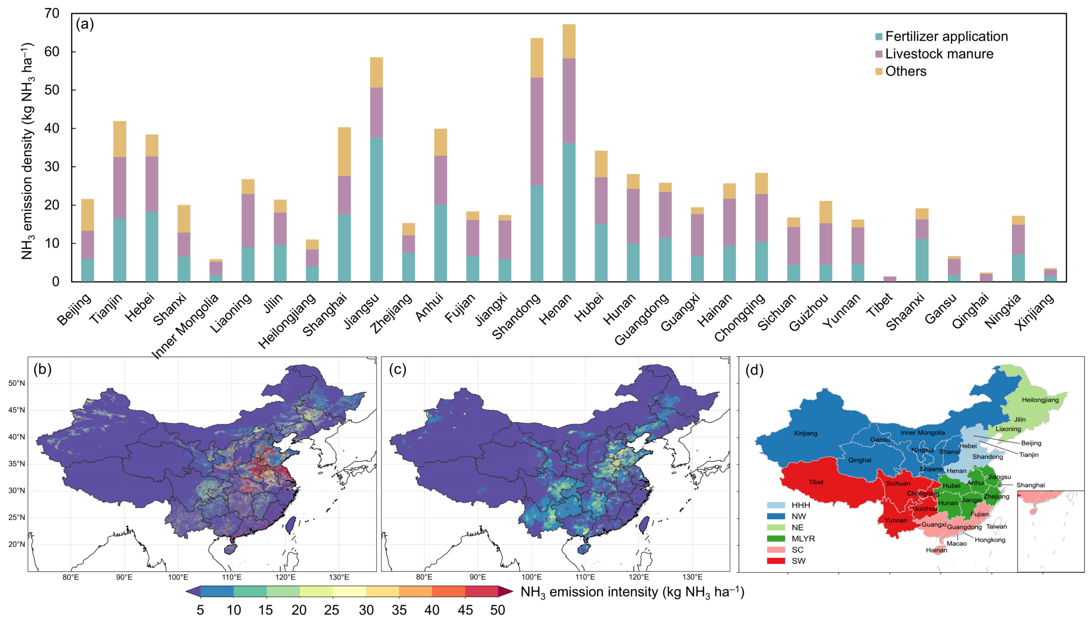


**Figure 3.** Provincial total $NH_3$ emissions (**a**), and the spatial distribution of $NH_3$ emissions from fertilizer application (**b**), livestock waste (**c**); Agricultural sub-regions and provinces distribution in China **(d)**. Agricultural sub-regions include Huang-Huai-Hai region (HHH), Middle and Lower Yangtze River region (MLYR), Northwest region (NW), Northeast region (NE), Southwest region (SW), and Southern China region (SC).

**Table S12.** Total agricultural $NH_3$ emissions and reduction potential of provinces (Gg)

| Province | Agricultural $NH_3$ | NUE-C | OUR | NUE-L | COMB |
|---|---|---|---|---|---|
| Beijing | 21.89 | 7.75 | 2.73 | 0.99 | 9.26 |
| Tianjin | 38.74 | 7.65 | 7.41 | 2.62 | 14.74 |
| Hebei | 617.37 | 117.81 | 128.83 | 38.76 | 237.72 |
| Shanxi | 201.58 | 45.42 | 30.17 | 19.83 | 82.21 |
| Inner Mongolia | 621.88 | 79.45 | 62.14 | 70.96 | 183.71 |
| Liaoning | 339.19 | 42.66 | 44.36 | 36.60 | 108.40 |
| Jilin | 338.24 | 14.03 | 75.28 | 29.35 | 112.77 |
| Heilongjiang | 400.03 | 17.35 | 55.56 | 35.42 | 104.67 |
| Shanghai | 19.34 | 6.80 | 3.86 | 0.90 | 9.11 |
| Jiangsu | 520.00 | 165.94 | 162.56 | 27.48 | 285.31 |
| Zhejiang | 123.54 | 50.04 | 30.49 | 8.65 | 69.36 |

| | | | | | |
|---|---|---|---|---|---|
| Anhui | 460.64 | 87.88 | 111.15 | 29.83 | 194.22 |
| Fujian | 200.35 | 60.07 | 33.55 | 29.80 | 98.94 |
| Jiangxi | 267.73 | 21.02 | 32.00 | 34.09 | 79.31 |
| Shandong | 837.44 | 71.59 | 136.83 | 89.47 | 270.97 |
| Henan | 973.60 | 142.53 | 259.58 | 79.29 | 417.51 |
| Hubei | 507.64 | 122.35 | 116.37 | 45.79 | 232.86 |
| Hunan | 513.96 | 82.68 | 72.59 | 63.61 | 188.16 |
| Guangdong | 422.05 | 131.94 | 85.02 | 60.04 | 221.37 |
| Guangxi | 418.00 | 60.18 | 61.90 | 70.09 | 168.85 |
| Hainan | 76.88 | 24.35 | 14.25 | 11.95 | 39.94 |
| Chongqing | 188.82 | 46.43 | 25.05 | 23.23 | 78.96 |
| Sichuan | 694.36 | 77.33 | 62.53 | 107.32 | 221.49 |
| Guizhou | 268.66 | 50.91 | 8.17 | 43.86 | 96.89 |
| Yunnan | 553.91 | 119.55 | 62.25 | 81.63 | 221.76 |
| Tibet | 165.81 | 2.28 | 0.32 | 38.39 | 40.69 |
| Shaanxi | 335.44 | 162.91 | 87.55 | 20.55 | 208.70 |
| Gansu | 271.62 | 46.60 | 3.15 | 43.43 | 92.03 |
| Qinghai | 149.81 | 3.92 | 0.00 | 33.68 | 37.69 |
| Ningxia | 98.96 | 34.66 | 12.48 | 7.53 | 45.21 |
| Xinjiang | 530.04 | 145.57 | 71.00 | 59.96 | 229.46 |

Li, B., Chen, L., Shen, W., Jin, J., Wang, T., Wang, P., Yang, Y., and Liao, H.: Improved gridded ammonia emission inventory in China, Atmos. Chem. Phys., 21, 15883–15900, https://doi.org/10.5194/acp-21-15883-2021, 2021.

Zhang, L., Chen, Y., Zhao, Y., Henze, D. K., Zhu, L., Song, Y., Paulot, F., Liu, X., Pan, Y., Lin, Y., and Huang, B.: Agricultural ammonia emissions in China: Reconciling bottom-up and top-down estimates, Atmos. Chem. Phys., 18, 339–355, https://doi.org/10.5194/acp-18-339-2018, 2018.

*5) Although the emission inventory in this study achieves a 1 km resolution, the meteorological reanalysis data likely do not match this resolution. Could you provide more details on the data processing methods?*

We apologize for the unclear data description. Meteorological data (temperature and wind speed) and soil characteristics (soil pH and CEC) in this study are all at a 1 km spatial resolution. We have provided the spatial resolution in the data description.

Line 129 and 134 "The gridded soil pH and CEC data **(1 km × 1 km)** were obtained from the Harmonized World Soil Database" "where m represents months; $T$ (°C) and $u$ (m s$^{-1}$) are air temperature and wind speed at 2 m height, whereby their gridded values **(1 km × 1 km)** are from Peng et al. (2019) and National Earth System Science Data Center. **The high-resolution climate data were produced by spatially downscaling the 30-min Climatic Research Unit (CRU) time-series dataset with WorldClim climatology using the delta downscaling method.**"

*6) When comparing model results with observations, could seasonal comparisons be included for a more comprehensive evaluation?*

Thanks for your helpful suggestions. We have added the comparison between simulation and observations. Our inventory demonstrated superior accuracy in modeling surface $NH_3$ concentrations compared to MEIC in all seasons, particularly during summer. Regarding $PM_{2.5}$, our inventory's simulations exhibited stronger spatial correlation with observations than MEIC simulations, although they were slightly overestimated.

Line 391-395: "Additionally, we examined the seasonality of $NH_3$ concentrations for sub-regions in Table S6 **and conducted seasonal comparison between simulations and observations in Table S7**. The temporal correlation between IASI-derived $NH_3$ and $NH_3$ modeled by our inventory is better than that for MEIC. **Our inventory demonstrates superior accuracy in modeling surface $NH_3$ concentrations compared to MEIC in all seasons, particularly during summer. Regarding $PM_{2.5}$, simulations with our inventory exhibit a stronger spatial correlation with observations than with MEIC, although concentrations are slightly overestimated.**"

**Table S7.** The simulation performance of our inventory and MEIC inventory regarding surface $NH_3$ and $PM_{2.5}$ levels.

| Season | This study | | | | MEIC | | | |
|---|---|---|---|---|---|---|---|---|
| | $NH_3$ | | $PM_{2.5}$ | | $NH_3$ | | $PM_{2.5}$ | |
| | R | RMSE | R | RMSE | R | RMSE | R | RMSE |
| Spring (March, April, May) | 0.72 | 3.6 | 0.5 | 27.9 | 0.72 | 4.1 | 0.49 | 26.8 |
| Summer (June, July, August) | 0.76 | 6.2 | 0.68 | 36.1 | 0.67 | 7 | 0.65 | 34.95 |
| Autumn (September, October, November) | 0.65 | 3.1 | 0.62 | 31.4 | 0.61 | 2.9 | 0.61 | 29.8 |
| Winter (December, January, February) | 0.6 | 2.5 | 0.6 | 30.4 | 0.54 | 2.8 | 0.59 | 31.6 |