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Journal of Environmental Management

Meng, Fanlei; Ronda, Reinder; Strokal, Maryna; Kroeze, Carolien; Ma, Lin et al

<https://doi.org/10.1016/j.jenvman.2023.119737>

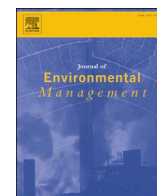
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Research article

Setting goals for agricultural nitrogen emission reduction to ensure safe air and groundwater quality: A case study of Quzhou, the North China Plain

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ARTICLE INFO

Keywords:

Nitrogen emission targets
Ammonia emission
Nitrate leaching
Air quality
Groundwater quality

ABSTRACT

Setting nitrogen (N) emission targets for agricultural systems is crucial to prevent to air and groundwater pollution, yet such targets are rarely defined at the county level. In this study, we employed a forecasting-and-back casting approach to establish human health-based nitrogen targets for air and groundwater quality in Quzhou county, located in the North China Plain. By adopting the World Health Organization (WHO) phase I standard for PM_{2.5} concentration (35 µg m⁻³) and a standard of 11.3 mg NO₃⁻-N L⁻¹ for nitrate in drinking water, we found that ammonia (NH₃) emissions from the entire county must be reduced by at least 3.2 kilotons year⁻¹ in 2050 to meet the WHO's PM_{2.5} phase I standard. Additionally, controlling other pollutants such as sulfur dioxide (SO₂) and nitrogen oxides (NO_x) is necessary, with required reductions ranging from 16% to 64% during 2017–2050. Furthermore, to meet the groundwater quality standard, nitrate nitrogen (NO₃⁻-N) leaching to groundwater should not exceed 0.8 kilotons year⁻¹ by 2050. Achieving this target would require a 50% reduction in NH₃ emissions and a 21% reduction in NO₃⁻-N leaching from agriculture in Quzhou in 2050 compared to their respective levels in 2017 (5.0 and 2.1 kilotons, respectively). Our developed method and the resulting N emission targets can support the development of environmentally-friendly agriculture by facilitating the design of control strategies to minimize agricultural N losses.

1. Introduction

While nitrogen (N) is a key element in ensuring food security, it also has detrimental effects on environmental quality (Penuelas and Sardans, 2022; Misselbrook et al., 2022). In 2018, more than half of the total N input to the global food system was released into the environment, leading to air and water pollution (Gu et al., 2023; Guo et al., 2023; Crippa et al., 2022). This release of N compounds, such as ammonia (NH₃) and nitrate nitrogen (NO₃⁻-N), contributes to pollution, including

the contamination of PM_{2.5} and the accumulation of nitrates in groundwater, posing significant risks to sustainable food production and human health (Min et al., 2022; Erisman et al., 2013; An et al., 2019; Xu et al., 2022; Gu et al., 2021). In 2020, the estimated number of premature deaths associated with PM_{2.5} exceeded 1 million (Cheng et al., 2023). Therefore, it is crucial to establish N emission targets to ensure the air and water quality and protect human health.

The Chinese government has implemented air pollution control policies and climate action measures aimed at reducing fossil fuel

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<https://doi.org/10.1016/j.jenvman.2023.119737>

Received 19 August 2023; Received in revised form 21 November 2023; Accepted 27 November 2023

Available online 7 December 2023

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consumption, leading to significant improvements in air quality. It has been reported that reducing NH_3 emissions is a more cost-effective approach compared to controlling NO_x emissions for mitigating $\text{PM}_{2.5}$ pollution (Gu et al., 2021). While several strategies for reducing NH_3 emissions in crop and livestock production have been developed, including optimizing nitrogen rates, implementing deep fertilizer placement, and using urease inhibitors (Zhang et al., 2021; Kang et al., 2023). However, there is still uncertainty about how to effectively reduce NH_3 emissions and other pollutants (such as SO_2 and NO_x) at a regional scale to meet $\text{PM}_{2.5}$ standards. Moreover, agriculture can significantly impact groundwater quality through N leaching (Wang et al., 2019), but clear targets for controlling NO_3^- -N leaching from agricultural activities in China are currently lacking. Although a method to identify nitrate-vulnerable zones was proposed by Bai et al. (2018), it did not provide precise values for defining these zones. Other studies have attempted to estimate NO_3^- -N leaching targets by multiplying the critical concentration in groundwater (e.g., the WHO limit of $11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$) with the water flux leaching from agricultural soil at a 0.5 by 0.5 -degree grid resolution (Schulte-Uebbing et al., 2022). However, this approach does not specifically address future leaching scenarios. Therefore, it is imperative to establish clear NO_3^- -N leaching targets that align with groundwater quality standards in agricultural production regions. This will aid in effectively managing and mitigating groundwater pollution caused by agricultural activities in the region.

The North China Plain (NCP) is responsible for producing 81% of Chinese wheat and 31% of maize, resulting in substantial fertilizer-related NH_3 losses that account for 43% of the annual Chinese NH_3 emissions (Ju and Zhang, 2017; Wang et al., 2018a). Additionally, in this region, soil nitrate N accumulation in the $0\text{--}4 \text{ m}$ soil layer was 453, 749, 1200, and $2155 \text{ kg N ha}^{-1}$ for wheat, maize, vegetables, and fruit trees, respectively (Zhou et al., 2016). Groundwater pollution is also a concern, as over 34% of monitoring sites in the NCP exceed the drinking water quality standard for nitrate concentrations set by the World Health Organization (WHO) at $11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$ (Ma et al., 2018). Quzhou, located in the NCP, is a representative agricultural county dominated by wheat and maize double-cropping systems, coupled with intensive pig and laying hen production (Zhang et al., 2016; Zhao et al., 2017; Wang et al., 2020). Despite being selected as a demonstration site for Agriculture Green Development, aiming to achieve high resource use

efficiencies and minimal negative environmental impacts in agricultural production (Meng et al., 2022a), Quzhou still faces significant challenges regarding NH_3 emissions and NO_3^- -N leaching from agricultural practices, leading to environmental pollution (Feng et al., 2022; Meng et al., 2022a). Agricultural NH_3 emissions can worsen $\text{PM}_{2.5}$ pollution through chemical reactions with acidifying compounds (SO_2 and NO_2) (Xu et al., 2019). From 2019 to 2021, the annual average $\text{PM}_{2.5}$ concentration in Quzhou was about $56 \mu\text{g m}^{-3}$, which is close to that of the Beijing-Tianjin-Hebei region ($50 \mu\text{g m}^{-3}$) (MEE, 2019, 2020, 2021; Wang et al., 2023). Therefore, Quzhou can serve as a valuable case study to determine the necessary N emission targets for effectively controlling air and groundwater pollution in the NCP.

The objective of this study is to establish human health-based N targets for NH_3 emissions to air and NO_3^- -N leaching to groundwater from agriculture in Quzhou by 2050. To determine the target for NH_3 emissions, we utilized a forecasting approach based on the human health standard of $\text{PM}_{2.5}$ ($35 \mu\text{g m}^{-3}$). Meanwhile, the target for NO_3^- -N leaching was set to meet the safe drinking groundwater nitrate concentration standard ($11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$) set by WHO, using a back-casting approach. The current N losses from agriculture in Quzhou were also evaluated against these human health targets.

2. Materials and methods

2.1. Study area

Quzhou county, located in the central region of NCP (Fig. 1), is renowned for its intensive agriculture production. It covers an area of 667 km^2 and has a population of 433,000 people. The agricultural sector in Quzhou is primarily consists of a winter wheat and summer maize double cropping system (Zhang et al., 2016). Additionally, the county is characterized by significant laying hen and pig production, aligning with the agricultural structure observed in the larger NCP region (He et al., 2022; Wang et al., 2020). Consequently, Quzhou is facing substantial NH_3 emission, resulting in serious annual mean NH_3 concentrations of approximately $40 \mu\text{g m}^{-3}$ in the air (Feng et al., 2022; Meng et al., 2022b). Moreover, the NO_3^- -N leaching from wheat and maize production was 209 kg N ha^{-1} in Quzhou, exacerbating the risk of groundwater contamination (Niu et al., 2021). Therefore, establishing

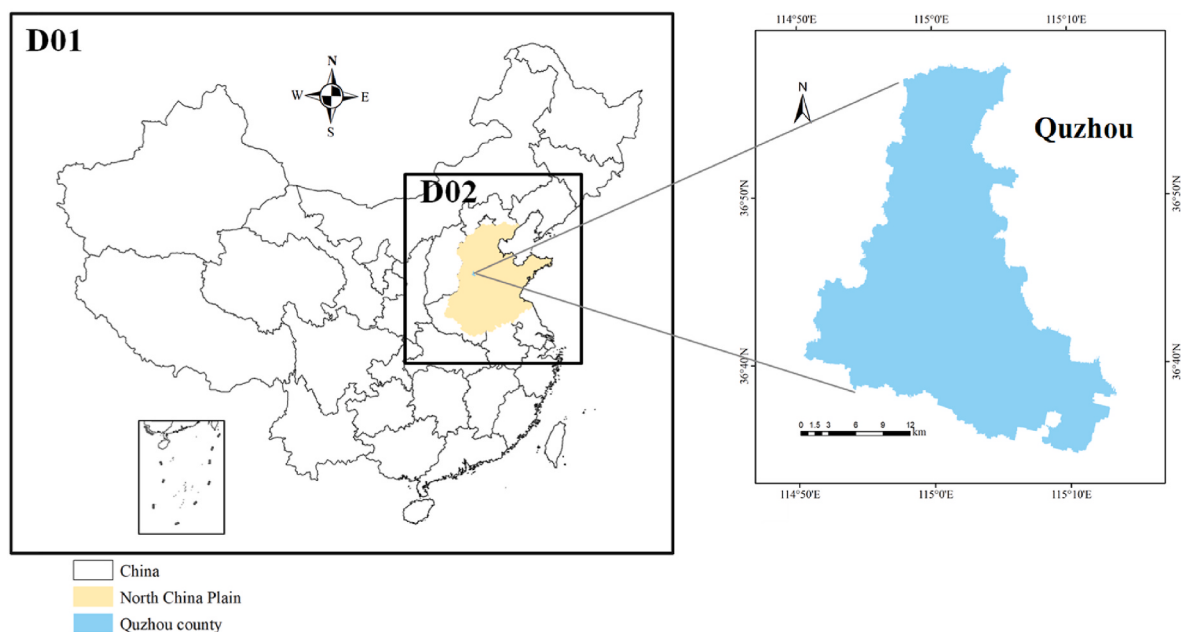


Fig. 1. The study area showing Quzhou, a county located in the North China Plain. (D01 and D02 are two nested air quality modeling domains, which represent the geographical locations of China and North China Plain, respectively).

targets for NH_3 emissions to air and NO_3^- -N leaching to groundwater is crucial for the development of sustainable N management strategies in Quzhou.

2.2. A forecasting-and back-casting approach for nitrogen targets

This study developed human health-based targets for NH_3 emission to air and NO_3^- -N leaching to groundwater in Quzhou (refer to Fig. 2). A forecasting approach was used for the atmospheric part. The NH_3 emission target was derived based on the $\text{PM}_{2.5}$ standard set by the Chinese government ($35 \mu\text{g m}^{-3}$), which aligns with WHO phase I standards. To achieve this, the WRF-CMAQ model was employed to assess the further $\text{PM}_{2.5}$ concentration under different emissions scenarios (see Section 2.2.1). For the groundwater aspect, a back-casting approach was selected. The NO_3^- -N leaching target was established for groundwater quality based on the drinking water quality standard recommended by WHO ($11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$) (see Section 2.2.2).

2.2.1. Forecasting approach for ammonia emission targets

In this paper, a forecasting approach was employed to develop an NH_3 emission target that meets the $\text{PM}_{2.5}$ standard ($35 \mu\text{m}^{-3}$) in Quzhou by 2050 (which is the target year of the study). The forecasting approach is a future scenario approach that considers factors such as technological development, climate change, and policy orientations. It provides valuable insights into the emission levels needed to achieve environmental targets in the future (An et al., 2019; Liang et al., 2020; Yue et al., 2020). The WRF-CMAQ model was utilized to assess $\text{PM}_{2.5}$ concentration, with controlling scenarios for future pollutant emissions serving as input. The meteorological conditions were calculated using WRF version 3.8, and the resulting data was fed to CMAQ version 5.2 to assess the $\text{PM}_{2.5}$ concentrations. The WRF-CMAQ model and its simulation and evaluation were further described in Supplementary Method 1 and Table S1. The development of the NH_3 emission target, derived based on the $\text{PM}_{2.5}$ standards, involved following three steps.

Firstly, we evaluated $\text{PM}_{2.5}$ concentrations simulated by WRF-CMAQ using observed $\text{PM}_{2.5}$ collected in the NCP. The current (2017) emissions of air pollutants were used as input for WRF-CMAQ in this evaluation.

The current status_2017 emissions, denoted as C0, represent anthropogenic emissions for 2017 and were obtained from the Multi-resolution Emission Inventory (<http://meicmodel.org>). These emissions were then prepared for input into the WRF-CMAQ model (see Table 1 and Supplementary Method 2).

Secondly, we assessed future $\text{PM}_{2.5}$ concentrations using the WRF-CMAQ model, with future emissions scenarios as input. The B0 and S0 represent emissions scenarios for the year 2050 under high economic growth and ambitious carbon neutral goals, respectively. The B0 scenario reflects anthropogenic emissions under SSP5-RCP8.5 scenario, which assumes high economic growth and an intensification of environmental degradation, with China gradually catching up to developed countries in terms of end-of-pipe control technologies (Tong et al., 2020). On the other hand, the S0 scenario reflects anthropogenic emissions under an ambitious carbon neutral goal scenario with reduced NH_3 emission. This scenario assumes that China has implemented ambitious climate policies to achieve net-zero CO_2 emissions and has fully deployed the best available end-of-pipe control technologies across

Table 1

Scenarios for future emissions as used to identify the ammonia emission targets to meet the $\text{PM}_{2.5}$ standard in Quzhou.

Scenario	Objective	Descriptions
C0	Current Status_2017	Evaluation of WRF-CMAQ
B0	High economic growth_2050	To identify the NH_3 emission targets under future emission scenarios
S0	Carbon Neutral Goals + NH_3 mitigation_2050	Base simulation under the 2017 emission condition 2050 emissions under the SSP5-RCP8.5 pathway (high economic growth) while assuming that end-of-pipe control technologies are fully applied. 2050 emissions in a scenario based on ambitious carbon neutral goals, and 50% NH_3 emission reduction between 2017 and 2050

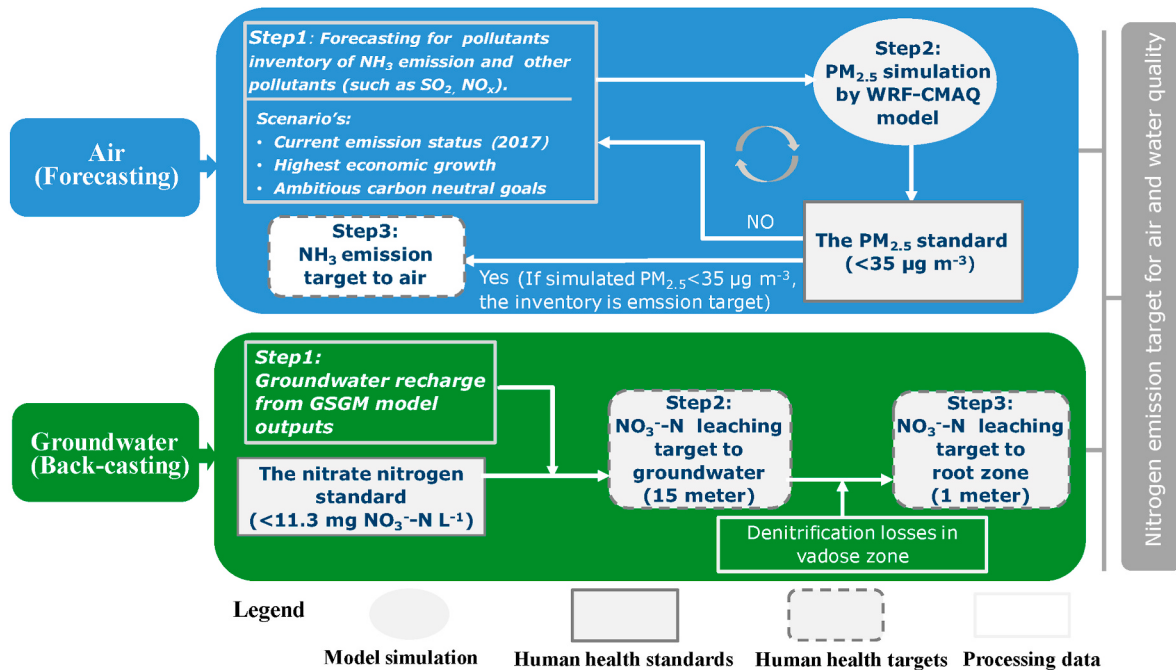


Fig. 2. The forecasting-and back-casting approach to quantify targets for ammonia (NH_3) emission to air and nitrate nitrogen (NO_3^- -N) leaching to groundwater. WRF-CMAQ stands for the Weather Research and Forecasting model and Community Multiscale Air Quality model, while GSGM refers to the Global-scale Surface Water-Groundwater Model. Detailed descriptions of the approach can be found in Sections 2.2.1 and 2.2.2.

all sectors to improve air quality (Cheng et al., 2021). In our study, we further improved the carbon neutral goals scenario by incorporating a 50% reduction in NH_3 emissions from 2017 to 2050. This reduction is based on the largest current technical reduction potential reported in the literature (Meng et al., 2022b; Zhang et al., 2020; Liu et al., 2019). The emission inventories for the B0 and S0 scenarios were obtained from the outputs of the Dynamic Projection model for emissions in China (DPEC) (<http://meicmodel.org>), and were prepared for input into the WRF-CMAQ (see Table 1 and S2 and Supplementary Method 2).

Thirdly, we determined the NH_3 emission target based on simulated $\text{PM}_{2.5}$ concentrations from the WRF-CMAQ model, with a focus on meeting the $\text{PM}_{2.5}$ standard of $35 \mu\text{g m}^{-3}$. Initially, we analyzed whether the simulations under scenarios B0 and S0 meet the standard. If the simulated $\text{PM}_{2.5}$ concentration still exceeds $35 \mu\text{g m}^{-3}$, we further analyze the potential for intensified emission reduction based on the contribution of emissions to $\text{PM}_{2.5}$, as indicated by the simulation results. Once the simulated $\text{PM}_{2.5}$ concentration falls below the $\text{PM}_{2.5}$ standard for the first time during this assessment loop (Fig. 2), the corresponding NH_3 emission level is identified as the NH_3 emission reduction target.

2.2.2. Back-casting approach for NO_3^- -N leaching targets

In this study, a back-casting approach was developed to identify a human health-based target for NO_3^- -N leaching. The back-casting approach proposes an environmental target for emission reduction in the future based on the turning point of environmental pollution changes (Li et al., 2019). The target was estimated by multiplying the drinking water quality standard of nitrate concentration ($11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$ from WHO) by the water value for groundwater recharge in Quzhou in 2050. Groundwater recharge is the process by which the volume or flow of water reaches down to the water table, forming an addition to the groundwater reservoir (de Vries and Simmers, 2002). The estimation of NO_3^- -N leaching targets based on the back-casting approach involved three steps.

The first step involved deriving the groundwater recharge in Quzhou in 2050 (Fig. S1). The groundwater recharge was derived using outputs of the Global-scale surface water-groundwater model (GSGM) developed by de Graaf et al. (2019). The GSGM simulated groundwater recharge as the flux between groundwater and soil moisture, considering recharge and capillary rise. The simulations were conducted under the Representative Concentration Pathway 8.5 from the Coupled Model Intercomparison Project (CMIP5). Detailed information about the model description and its simulation of the GSGM can be found in de Graaf et al. (2019). In our study, we averaged simulated groundwater recharge over a 30-year period (2035–2065) to represent groundwater recharge in Quzhou in 2050. The period 2035–2065 was chosen by considering 2050 as the midpoint and expanding the timeframe by 15 years on either side. In addition, to convert the groundwater recharge from the raster scale to the county scale, we first extracted the groundwater recharge grid data based on the administrative area of Quzhou county, and then multiplied it by the area corresponding to the grid based on the administrative area.

In the second step, the environmental target for NO_3^- -N leaching to shallow groundwater at a depth of 15 m was calculated (Chen, 2019; MWR, 2017). The target for NO_3^- -N leaching to groundwater was derived by multiplying environmental standard for groundwater ($11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$) by the groundwater recharge in Quzhou from the first step, using equation (1).

$$L_g = Q_C \times L_P \times 10^{-6} \quad (1)$$

where L_g represents the target for nitrate leaching to groundwater (ton year^{-1}); Q_C represents the groundwater recharge in Quzhou ($\text{m}^3 \text{ year}^{-1}$); L_P is the standard for nitrate concentration for groundwater ($11.3 \text{ mg NO}_3^- \text{ N L}^{-1}$ recommended by the WHO) in Quzhou (mg N L^{-1}). 10^{-6} is the unit conversion for ton year^{-1} .

In the third step, our study estimated the human health-based target for NO_3^- -N leaching to the root zone, which has a soil thickness of approximately 1 m. This estimation considered the N losses from NO_3^- -N transport processes via denitrification in the vadose zone, which has a soil thickness of about 14 m. The amount of NO_3^- -N leaching to the root zone is therefore estimated based on the proportions of NO_3^- -N leaching to groundwater, using equation (2).

$$L_r = L_g \div (1 - N_d) \quad (2)$$

where, L_r represents the target for nitrate leaching to the root zone (ton year^{-1}); L_g represents the target for nitrate leaching to groundwater (ton year^{-1}); N_d represents the ratio of N losses through denitrification in the vadose zone, which is assumed to be 50%. This estimation is based on field measurements conducted in regions near our study site in the NCP (Niu et al., 2021; Wang et al., 2019).

2.3. Gaps between current agricultural emissions and their future targets

In this section, we compared Quzhou's current N losses from agriculture with the identified future human health-based targets. The NH_3 emissions to air and NO_3^- -N leaching to groundwater from agriculture in Quzhou in 2017 were obtained from Meng et al. (2022a), using the NUTrient flows in Food chains, Environment and Resources use (NUFER) model (Meng et al., 2022a). The human health-based target for NH_3 emissions to air was derived from Section 2.2.1, while the target for NO_3^- -N leaching to the root zone is based on Section 2.2.2. The gap between the current values and the targets for NH_3 emissions and NO_3^- -N leaching was calculated using equation (3)–(6):

$$\text{NH}_{3\text{gap}} = \text{NH}_{3\text{current}} - \text{NH}_{3\text{target_agr}} \quad (3)$$

$$\text{NH}_{3\text{current}} = \frac{17}{14} \times \text{NH}_{3\text{NUFER}} \quad (4)$$

$$\text{NH}_{3\text{target_agr}} = \text{Ratio}_{\text{agr}} \times \text{NH}_{3\text{target_county}} \quad (5)$$

where $\text{NH}_{3\text{gap}}$ represents the gap between the current NH_3 emission and future NH_3 emission target from agriculture ($\text{kilotons year}^{-1}$); $\text{NH}_{3\text{current}}$ is the current NH_3 emission from agriculture in Quzhou in 2017 ($\text{kilotons year}^{-1}$); $\frac{17}{14}$ is the conversion coefficient for NH_3 -N emissions to NH_3 emissions; $\text{NH}_{3\text{NUFER}}$ represents current NH_3 emission from agriculture calculated by the NUFER model in Quzhou in 2017 ($\text{kilotons year}^{-1}$); $\text{NH}_{3\text{target_agr}}$ is the future NH_3 emission target from agriculture in Quzhou in 2050 ($\text{kilotons year}^{-1}$); $\text{Ratio}_{\text{agr}}$ is the proportion of agricultural NH_3 emissions to total NH_3 emissions in Quzhou. The agricultural NH_3 emission accounted for 96.3% of the total NH_3 emission (refer to Fig. S2); $\text{NH}_{3\text{target_county}}$ is future NH_3 emission target from the entire Quzhou county ($\text{kilotons year}^{-1}$);

$$\text{NO}_3^- \text{ N leaching}_{\text{gap}} = \text{NO}_3^- \text{ N leaching}_{\text{current}} - \text{NO}_3^- \text{ N leaching}_{\text{target}} \quad (6)$$

where $\text{NO}_3^- \text{ N leaching}_{\text{gap}}$ is the gap between current NO_3^- -N leaching and future NO_3^- -N leaching target from agriculture ($\text{kilotons year}^{-1}$);

$\text{NO}_3^- \text{ N leaching}_{\text{current}}$ is the current NO_3^- -N leaching from agriculture in Quzhou in 2017, which was calculated by the NUFER model ($\text{kilotons year}^{-1}$); $\text{NO}_3^- \text{ N leaching}_{\text{target}}$ is the future NO_3^- -N leaching target from agriculture in Quzhou in 2050 ($\text{kilotons year}^{-1}$).

3. Results and discussion

3.1. Reducing ammonia emissions to air to meet $\text{PM}_{2.5}$ standard

Two future emission scenarios were developed to determine the NH_3 emission target in Quzhou and assess the required reduction in NH_3 emissions while controlling other emissions such as SO_2 and NO_x (Fig. 3). Under the high economic growth scenario (B0), $\text{PM}_{2.5}$

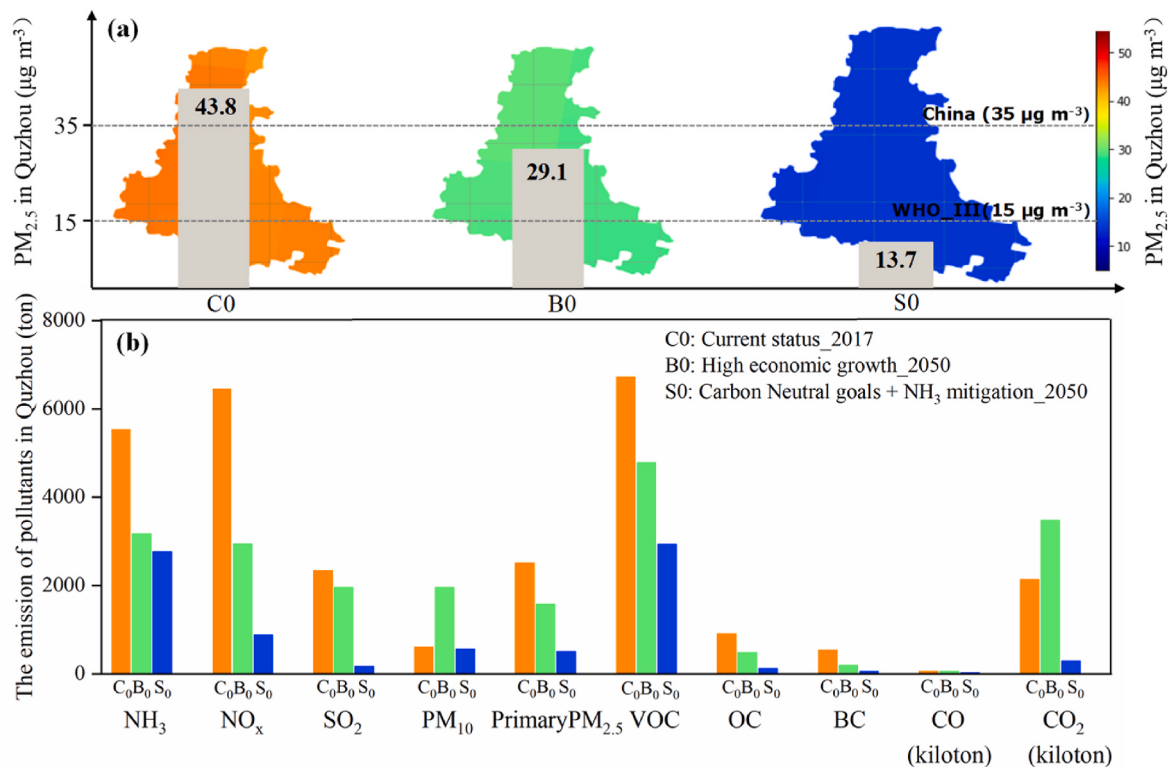


Fig. 3. The simulated $PM_{2.5}$ concentrations (a, $\mu g m^{-3}$) and emissions of pollutants to air (b, kilotons year $^{-1}$) in Quzhou in 2017 and 2050. In Subpanel 3a, the left vertical axis represents the $PM_{2.5}$ concentration of the column chart, while the right vertical axis represents the $PM_{2.5}$ concentration displayed on the map. The C0 scenario reflects the year 2017, representing the baseline conditions. The B0 scenario represents anthropogenic emissions projected for 2050 under the Shared Socioeconomic Pathway (SSP) 5 and Representative Concentration Pathway (RCP) 8.5, which represents the high economic growth. The S0 scenario represents anthropogenic emissions projected for 2050 under ambitious carbon neutral goals. The emission of pollutants considered in this study includes ammonia (NH_3), sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), PM_{10} , $PM_{2.5}$, carbon dioxide (CO_2), volatile organic compounds (VOC), element carbon (EC), and organic carbon (OC). Further details on the approach can be found in Section 2.2.1.

concentrations can meet the $PM_{2.5}$ standard ($35 \mu g m^{-3}$) when the total NH_3 emissions in Quzhou are reduced by at least 3.2 kilotons year $^{-1}$ in 2050. This reduction relies on synergistic control of NH_3 emissions along with other pollutants, such as SO_2 and NO_x . Our study reveals varying emission reduction rates (16–64%) for different pollutants in Quzhou (Fig. 3b). With widespread implementation of end-of-pipe exhaust treatment technologies, reductions of 64%, 47%, 54%, and 16% are projected for BC, OC, NO_x and SO_2 respectively, from 2017 to 2050. This offers the potential to achieve the $PM_{2.5}$ standard. However, the emissions of certain pollutants, such as PM_{10} , primary $PM_{2.5}$ and CO, are expected to increase by 223%, 63%, and 5% respectively during the same period. This is mainly due to the continued dominance of fossil fuels, particularly in the transportation and power sectors, under the SSP5-RCP8.5 scenario. Consequently, there is significant potential to further reduce $PM_{2.5}$ concentrations through synergistic control of NH_3 and other pollutant emissions.

In the ambitious carbon neutral goals scenarios (S0), it is projected that $PM_{2.5}$ concentration can reach $13.7 \mu g m^{-3}$ when NH_3 emissions are limited to less than 2.8 kilotons year $^{-1}$ and other pollutants such as SO_2 and NO_x are reduced by 6–93% during the period of 2017–2050. Specifically, under scenario S0, SO_2 and NO_x emissions are projected to decrease by 86% and 93% respectively during this period. This significant reduction can be attributed to the improved new energy structure, primarily driven by the carbon-neutral policy implementation (Cheng et al., 2021). These results suggest that it is possible for the annual mean $PM_{2.5}$ concentration to meet the WHO phase III guidance ($15 \mu g m^{-3}$) in Quzhou by 2050. However, the $PM_{2.5}$ concentration still falls short of the WHO health standard ($5 \mu g m^{-3}$). Our findings indicate that achieving the WHO health standard would be a highly ambitious goal,

requiring stringent limits on emissions of SO_2 , NO_x , NH_3 , VOC, and primary $PM_{2.5}$ in Quzhou. Additionally, it would necessitate significant efforts to address transboundary air pollution from the NCP, other regions in China, and countries outside of China (Jiang et al., 2023).

3.2. Targets for nitrogen leaching to groundwater

The human health-based target for NO_3^- -N leaching to groundwater at a depth of 15 m is estimated at 0.8 kilotons year $^{-1}$ for 2050 (Fig. 4). The target was calculated by multiplying the standard of nitrate concentrations in groundwater ($11.3 mg NO_3^- N L^{-1}$ from WHO) by the estimated groundwater recharge from the GSGM model outputs. Our analysis indicates that groundwater recharge is projected to be approximately $7.5 \times 10^7 m^3$ in Quzhou in 2050 (Fig. S1). Furthermore, we calculated the target for NO_3^- -N leaching to the root zone, which corresponds to a soil thickness of about 1 m. Our results show that the target for NO_3^- -N leaching to the root zone in Quzhou in 2050 is 1.7 kilotons year $^{-1}$ (Fig. 4). This target effectively supports agricultural policies aimed at controlling NO_3^- -N leaching.

3.3. Comparison of current nitrogen emissions with their future targets

In 2017, NH_3 emissions from agriculture in Quzhou totaled 5.0 kilotons, while NO_3^- -N leaching amounted to 2.1 kilotons, both exceeding the identified N emission target (Fig. S3, Meng et al., 2022a). To meet N emission targets for air quality ($PM_{2.5}$) and groundwater quality, it is necessary to reduce NH_3 emissions and NO_3^- -N leaching from agriculture by at least 50% and 21%, respectively, between 2017 and 2050. According to Meng et al. (2022a), NH_3 emissions from agriculture in

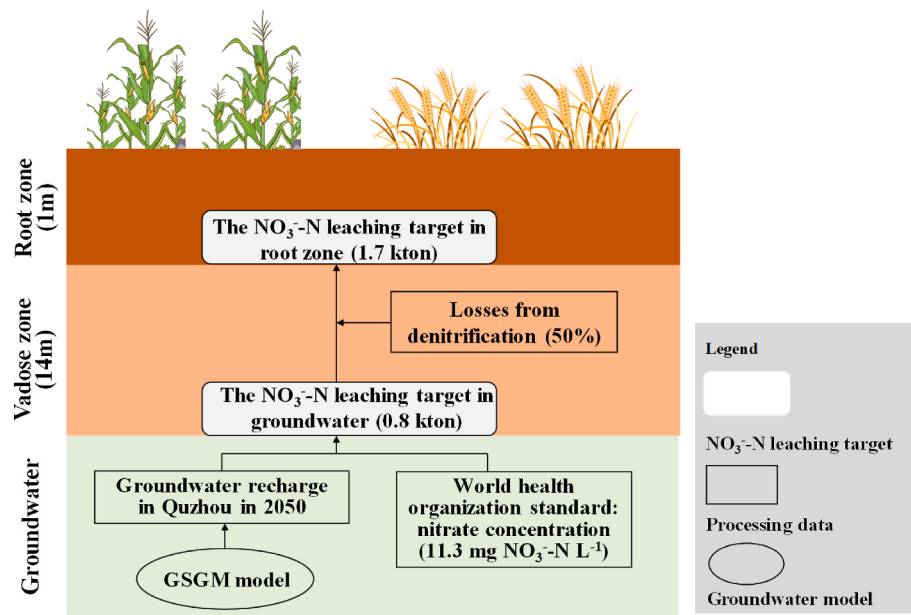


Fig. 4. Nitrate ($\text{NO}_3\text{-N}$) leaching to groundwater and its environmental target in Quzhou in 2050. GSGM denotes the Global-scale surface water-groundwater model. $\text{NO}_3\text{-N}$ is nitrate nitrogen. For details on the approach see Section 2.2.2.

Quzhou were primarily derived from wheat, maize, vegetables, pigs, and laying hens. In 2017, wheat, maize, and vegetables accounted for 77% of NH_3 emissions from crop production, while pigs and laying hens were responsible for 63% of NH_3 emissions from livestock production. Similarly, $\text{NO}_3\text{-N}$ emissions from agriculture were primarily associated with wheat, maize, and vegetable production. These crops were responsible for 81% of the $\text{NO}_3\text{-N}$ leaching into groundwater during crop production in 2017. This highlights the importance of prioritizing improvements in nutrient management for crops and animals that contribute the most to N losses. Additionally, it is anticipated that future food production and consumption, including fertilizer application, intensive animal production, animal feed, and human diets, will be influenced by socioeconomic development and global changes (Wang et al., 2019; Ren et al., 2023). Therefore, it is crucial to establish additional targets for reducing N losses in agriculture to better understand potential future scenarios.

3.4. Model evaluation

To support our conclusion regarding the NH_3 emission target in Quzhou, we evaluated and discussed the results from the WRF-CMAQ model in three ways.

Firstly, we conducted a comparison between simulated meteorological data and locally observed data, as well as between observed and simulated $\text{PM}_{2.5}$ concentrations (Figs. S4–S8). The results showed that the model accurately predicted meteorological parameters, with correlation coefficients (r) of 0.8, 0.8, and 0.5 for temperature, relative humidity, and wind speed, respectively (Figs. S4–S7). However, there was a tendency for the simulated wind speed to be overestimated in Quzhou, consistent with previous studies (Lu et al., 2019). This discrepancy may be attributed to limitations in the representation of underlying surface parameters that influence wind speed in the WRF model configuration. Additionally, our study compared observed and simulated daily $\text{PM}_{2.5}$ in January and October 2017 (Figs. S8c and d), respectively. The model accurately captured the temporal variations of $\text{PM}_{2.5}$ in Quzhou, with a normalized mean bias (NMB) value of 40%, normalized mean error (NME) of 43.5%, and r value of 0.6 (Figs. S8c and d). Furthermore, we compared the distributions of simulated daily mean $\text{PM}_{2.5}$ concentrations in January, April, July, and October 2017 in Quzhou and Beijing (Zhang et al., 2021) (Figs. S8a and b). The simulated $\text{PM}_{2.5}$ concentrations aligned with the observed trend distribution, with r values of 0.76

and 0.87, respectively. Importantly, all NMB and NME values fell within the criteria ($\text{NMB} \leq \pm 60\%$, $\text{NME} < 75\%$) set by the U.S. Environmental Agency (Wang et al., 2022).

Secondly, we validated the representativeness of the typical seasonal months (January, April, July, and October) by comparing the proportion of pollutant emissions and the $\text{PM}_{2.5}$ concentrations during these four months in Quzhou from 2015 to 2020 (Figs. S9 and S10). Interestingly, the emissions of pollutants during these four months accounted for one-third of Quzhou's total annual pollutant emissions in 2017 (Fig. S9). This suggests that the emission inventory adequately captures the overall annual conditions. Furthermore, we observed seasonal variations in $\text{PM}_{2.5}$ levels, and the average $\text{PM}_{2.5}$ concentration during the four-month period was comparable to the annual average (Fig. S10).

Thirdly, we compared our NH_3 emission target with previous studies (Liu et al., 2021b). Our study is the first to estimate how to meet $\text{PM}_{2.5}$ thresholds through NH_3 emission reduction. Previous studies focused on predicting the effect of NH_3 emissions reductions on $\text{PM}_{2.5}$ concentrations in specific months, such as January and July (Liu et al., 2021b; Ye et al., 2022). They found that the efficiency of $\text{PM}_{2.5}$ reduction is highly dependent on the magnitude of NH_3 emission reduction. Typically, they observed that a significant reduction of 40–60% in NH_3 emissions leads to a high control efficiency for $\text{PM}_{2.5}$ (Liu et al., 2021b). These findings are consistent with our conclusion of meeting environmental targets through a 50% reduction in NH_3 emissions from agriculture.

3.5. Uncertainties of our approach to define targets

This study employed forecasting and back-casting techniques to establish N emission targets from agriculture for air and groundwater quality, with the aim of protecting human health at the county level.

The $\text{NO}_3\text{-N}$ leaching target approach has been successfully implemented in various regions globally (Schulte-Uebbing et al., 2022), including European (de Vries et al., 2021) and China (Ying et al., 2020). However, estimating the target is subject to uncertainties arising from denitrification losses in the vadose zone (Li et al., 2022). Monitoring denitrification in the vadose zone is challenging due to limitations in soil depth and monitoring methods (Yuan et al., 2019). To address this uncertainty, a range of denitrification losses (20–50%) in the vadose zone was considered for the $\text{NO}_3\text{-N}$ leaching target in different regions of the NCP (Niu et al., 2021; Wang et al., 2018b, 2019; Fig. S11). In this study,

we improved the approach by incorporating long-time series groundwater recharge data and local monitoring of denitrification losses in the vadose zone specific to Quzhou (Niu et al., 2021). This enhances the reliability of implementing the NO_3^- -N target approach in Quzhou. Future research could focus on more frequent monitoring of N denitrification losses in the vadose zone, which could contribute to further improving the accuracy of environmental target estimates. To support our conclusion regarding the NO_3^- -N leaching target in Quzhou, we compared the simulated groundwater recharge and observed groundwater recharge from a published paper in Quzhou in 2017 (See Table S3). According to the GSGM model results, the average annual groundwater recharge in Quzhou county in 2017 was estimated to be 158 mm yr^{-1} , which is consistent with the current published results. Additionally, the GSGM model has been evaluated and accepted in the literature (de Graaf et al., 2019). We believe the simulated groundwater recharge give trust in our study.

The forecasting approach utilized in this study aids in the development of an NH_3 emission target that aligns with the $\text{PM}_{2.5}$ standard ($35 \mu\text{g m}^{-3}$) for human health. This approach considers factors such as technological development, climate change, and policy orientations, thereby providing insights into the emissions levels required to achieve environmental targets in the future (An et al., 2019; Liang et al., 2020; Yue et al., 2020). It is important to note that our NH_3 emissions target is not a critical value that directly determines whether the $\text{PM}_{2.5}$ standard is met. One limitation is that the NH_3 emission target is influenced by the extent to which emissions of other pollutants, such as SO_2 and NO_x , are reduced. With technological advancements and the adoption of new energy structures, the emission of pollutants could be further reduced. To address the uncertainty surrounding pollutant emissions, we constructed future emission inventories based on scenarios that encompass the highest future economic increment and ambitious carbon neutral goals (Cheng et al., 2021; Tong et al., 2020). These scenarios incorporate the latest technologies and existing knowledge of China's environmental policies, thereby reflecting potential future development pathways. However, this study does not consider natural emissions induced by climate change or the changes in future meteorological conditions. Nevertheless, some studies show that substantial emission reductions could greatly improve air quality, surpassing the influence of future meteorology changes, particularly in the context of carbon neutrality targets (Zhang et al., 2019; Liu et al., 2021a). Additionally, the meteorological condition in 2017 were assessed without specific climate events (Cheng et al., 2021; Ding et al., 2021). Despite these considerations, the forecasting approach remain valuable in establishing the target for NH_3 emission to air from agricultural activities.

3.6. Implication and outlook

This study focuses on Quzhou as a case study to quantify N emission targets from agricultural activities and their impacts on air and groundwater quality at the county level. The findings of this study are valuable in guiding the development of agri-environmental policies and regional N management to safeguard human health.

Firstly, this study supports Quzhou in developing effective environmental policies by setting targets for reducing reactive N losses. By reducing NH_3 emissions and NO_3^- -N leaching, improvements can be achieved in air and groundwater quality, which are recognized objectives in the NCP (Bai et al., 2018; Liu et al., 2021b). Our study provides an approach and information to determine the necessary reductions in NH_3 emissions and NO_3^- -N leaching to attain these goals. Future studies can further investigate specific reduction targets for different animal and crop types, enhancing the specificity and effectiveness of environmental measures.

Secondly, this study provides new insight into regional control of pollutant emissions based on $\text{PM}_{2.5}$ standard. While most studies focus on analyzing the response of $\text{PM}_{2.5}$ concentration to the reduction of specific pollutants (Zhao et al., 2021; Meng et al., 2022b; Xu et al.,

2022), our approach explores the potential to meet regional $\text{PM}_{2.5}$ standard by reducing NH_3 emissions and other pollutants such as SO_2 and NO_x . The findings suggest that under high socioeconomic growth scenarios (B0), 96% of counties in the NCP can meet the $\text{PM}_{2.5}$ standard ($35 \mu\text{g m}^{-3}$) by controlling NH_3 emissions and other pollutants (Fig. 5). Furthermore, under ambitious carbon neutral goals scenarios (S0), 96% of counties in the NCP can meet even more stringent $\text{PM}_{2.5}$ standards, such as WHO-III standard ($15 \mu\text{g m}^{-3}$). Setting targets for NH_3 emissions to the air will contribute to improving regional air quality in the future.

4. Conclusions

We have developed an innovative approach to quantify N emission targets based on human health considerations. This approach was applied to Quzhou county in the NCP as a representative case study, with objective of determining N emission targets for NH_3 emission and NO_3^- -N leaching from agriculture activities in order to meet air ($\text{PM}_{2.5}$) and groundwater quality standards.

Our findings reveal that to meet WHO's phase I target for $\text{PM}_{2.5}$ in Quzhou, a minimum reduction of 3.2 kilotons year⁻¹ in NH_3 emissions by 2050 is required, assuming a scenario of high economic growth. Achieving the more stringent WHO's phase III target would necessitate the implementation of an ambitious environmental policy, with NH_3 emissions at the county-scale needing to be reduced to less than 2.8 kilotons year⁻¹, along with reductions of 6–93% in other pollutants during the period of 2017–2050. Additionally, to meet the standard for groundwater quality, the target for NO_3^- -N leaching into groundwater at a depth of 15 m was estimated to be 0.8 kilotons year⁻¹ by 2050 in Quzhou. Therefore, in order to meet the health-related targets in Quzhou, NH_3 emissions and NO_3^- -N leaching from agriculture need to be reduced by at least 50% and 21% respectively, compared to 2017 levels.

Our approach not only supports the establishment of health-related N emission reduction targets for agriculture but also assists in formulating effective strategies to promote agricultural green development. Future research could focus on identifying specific reduction targets for different animal and crop types as well as feasible options to achieve these targets.

CRedit authorship contribution statement

Fanlei Meng: Data curation, Formal analysis, Investigation, Methodology, Resources, Software, Validation, Visualization, Writing - original draft, Writing - review & editing. **Reinder Ronda:** Methodology, Software, Supervision, Visualization, Writing - original draft, Writing - review & editing. **Maryna Stokol:** Conceptualization, Supervision, Writing - original draft, Writing - review & editing. **Carolien Kroeze:** Conceptualization, Supervision, Writing - original draft, Writing - review & editing. **Lin Ma:** Conceptualization, Supervision, Writing - original draft, Writing - review & editing. **Maarten Krol:** Methodology, Writing - original draft, Writing - review & editing. **Inge de Graaf:** Methodology, Resources, Writing - original draft, Writing - review & editing. **Yuanhong Zhao:** Methodology, Resources, Writing - original draft, Writing - review & editing. **Yutong Wang:** Methodology, Resources, Writing - original draft, Writing - review & editing. **Xiaohui Du:** Methodology, Resources, Writing - original draft, Writing - review & editing. **Xuejun Liu:** Methodology, Writing - original draft, Writing - review & editing. **Wen Xu:** Conceptualization, Funding acquisition, Methodology, Resources, Software, Supervision, Writing - original draft, Writing - review & editing. **Fusuo Zhang:** Conceptualization, Methodology, Resources, Supervision, Writing - original draft, Writing - review & editing, Funding acquisition. **Mengru Wang:** Conceptualization, Methodology, Resources, Supervision, Writing - original draft, Writing - review & editing.

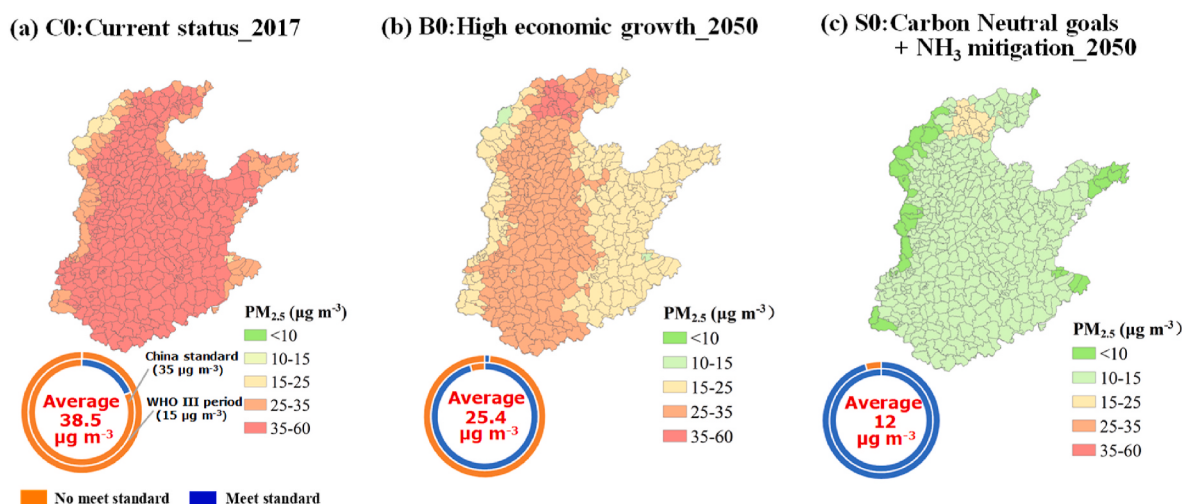


Fig. 5. The simulated PM_{2.5} concentration (μg m⁻³) and the percentage of counties meeting PM_{2.5} standard (%) in the North China Plain. The inner circle shows the Chinese standard (35 μg m⁻³), while the outer circle shows the WHO-III standard of 15 μg m⁻³. (a) Scenario C0 reflect the year 2017, proving a baseline for comparison; (b) Scenario B0 represents anthropogenic emissions projected for 2050 under Shared Socioeconomic Pathway (SSP) 5 and Representative Concentration Pathway (RCP) 8.5, indicating high economic growth. (c) Scenario S0 represents anthropogenic emissions projected for 2050 under an ambitious carbon neutral goals scenario.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

This work was sponsored by the National Natural Science Foundation of China (42175137), the National Key Research and Development Program of China (2021YFD1700900), the China Scholarship Council (No.201913043) and Hainan University, and the High-level Team Project of China Agricultural University. We would like to acknowledge Mingchen Ma from the Ocean University of China and Yibo Zhang from Zhejiang University help for WRF-CMAQ configuration and its improvement.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2023.119737>.

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