Main revisions and response to reviewers' comments

Journal: Atmospheric Chemistry and Physics

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Title: Spatial and temporal evolution of future atmospheric reactive nitrogen deposition in China under different climate change mitigation strategies

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We thank very much for the valuable comments and suggestions from the reviewers, which help us improve our manuscript. The comments were carefully considered and revisions have been made in response to suggestions. Following are our point-by-point responses to the comments and corresponding revisions. **Please note that the line numbers mentioned following refer to the clean version of the revised manuscript.**

Reviewer #1 Evaluations:

This study investigates the projected changes in nitrogen deposition in China using the WRF-CMAQ model. It provides valuable insights into the impact of atmospheric nitrogen deposition under different climate pathways and emission scenarios, highlighting the importance of integrating nitrogen deposition considerations into climate strategies. The study follows a sound methodology, including region-specific Chinese national emission targets. However, the manuscript requires significant improvements in presentation and depth of discussion before it can be published.

Overall, the manuscript offers interesting and valuable results, but I have concerns about the clarity of the model description, the methods, and the explanation of some results. These sections were somewhat difficult to follow and would benefit from better organization and clearer explanations. I recommend revising these parts to improve readability and understanding.

Response and revisions:

We show our deepest appreciation and respect to the reviewer for the insightful and constructive suggestions to help us greatly improve the manuscript. These invaluable suggestions offer us a great choice to better evaluate, understand and present the experimental results. The reviewer's positive comments on our work encourage us a lot. Following is our detailed response and revisions according to reviewer's comment.

Specific Comments:

1. L39: Please expand RDN.

Response and revisions:

We thank the reviewer's reminder. We have expanded the "RDN" to "reduced nitrogen (RDN)" **in line 39 in the revised manuscript**.

2. L59-60: Lacks clarity and flow. For example, the transition between the general impact of Nr emissions and the specific situation in China could be smoother. Please consider adding a brief sentence or two to bridge these sections.

Response and revisions:

We warmly thank the reviewer for pointing out the lack of coherence in this section. To bridge the context, the sentence "Influenced by multiple human activities, severe Nr deposition and its subsequent ecological risks in China have attracted growing attentions in recent years (Gu et al., 2012; Liu and Du, 2020)." has been added **in lines 60-62 in the revised manuscript**. We have also added the necessary references **(see below) in lines 845-848 and lines 959-962 in the revised manuscript.**

Reference:

Gu, B., Ge, Y., Ren, Y., Xu, B., Luo, W., Jiang, H, Gu, B., & Chang, J. (2012). Atmospheric reactive nitrogen in China: sources, recent trends, and damage costs. Environmental science & technology, 46(17), 9420-9427. https://doi.org/10.1021/es301446g

Liu, X., & Du, E. (2020). An overview of atmospheric reactive nitrogen in China from a global perspective. Atmospheric Reactive Nitrogen in China: Emission, Deposition and Environmental Impacts, 1-10. https://doi.org/10.1007/978-981-13-8514-8_1

3. L69: The mention of atmospheric chemistry transport models (CTMs) and advanced statistical models is somewhat abrupt. Please explain briefly the importance of modelling techniques in these kinds of studies and how they contribute to understanding Nr deposition trends.

Response and revisions:

We thank the reviewer's constructive suggestion. We have refined the sentence **in lines 72-77 in the revised manuscript** as "Due to the fast change and heterogeneous distribution of emissions and the typically short atmospheric lifetime of most Nr species, there exist challenges in estimating the spatial pattern and long-term trend of Nr deposition across big countries like China, based on observations at individual sites. Atmospheric chemistry transport models (CTMs) or advanced statistical models support analyses of the interannual variations of Nr deposition at multiple spatial scales (Liu et al., 2024; Wen et al., 2024)." The additional references **(see below) have also been added in lines 938-940 and lines 1104-1108 in the revised manuscript.**

Reference:

Liu, L., Wen, Z., Liu, S., Zhang, X., & Liu, X. (2024). Decline in atmospheric nitrogen deposition in China between 2010 and 2020. Nature Geoscience, 17(8), 733-736. https://doi.org/10.1038/s41561-024-01484-4

Wen, Z., Ma, X., Xu, W., Si, R., Liu, L., Ma, M., Zhao, Y., Tang, A., Zhang, Y., Wang, K., Zhang, Y., Shen, J., Zhang, L., Zhao, Y., Zhang, F., Goulding, K., & Liu, X. (2024). Combined short-term and long-term emission controls improve air quality sustainably in China. Nature Communications, 15(1), 5169. https://doi.org/10.1038/s41467-024-49539-9

4. L71: Please integrate these references to strength the narrative. Consider summarizing key findings from the cited studies in a way that builds a unified argument.

Response and revisions:

We thank the reviewer's constructive suggestion. We revisited and summarized these modeling studies. The sentences have been revised as "A series of modeling studies have analyzed the magnitude and spatiotemporal pattern of Nr deposition in China. Yu et al. (2019) applied the Kriging interpolation combined with empirical remote sensing models and estimated that China's annual Nr deposition had increased nearly 60% from 1980s to 2010s. By developing a random forest algorithm, Zhou et al. (2023) quantified the considerable growth of Nr deposition from 2005 to 2012 in eastern China. Gao et al. (2023) revealed the shifting of deposition forms from dominated by wet to more balanced contributions from dry and wet deposition. The national air pollution control actions over the past decade have resulted in a fast decline in emissions of acidic gaseous pollutants (mainly NO_x and SO_2) but relatively stable NH_3 (Zheng et al., 2018). The imbalance in emission reductions for different species has altered the composition of Nr deposition, i.e., a growth in the proportion of RDN (Liu et al., 2020). Zhao et al. (2022) developed the generalized additive model (GAM) and found that the decline in OXN deposition lagged behind NO_x reductions in recent years, attributed partly to the increased precipitation and the strengthening transport of pollution." **in lines 77-92 in the revised manuscript.**

5. L77-81: The weakening response of OXN deposition to NOx emissions and the role of VOCs could be explained in more detail. It might be helpful to briefly describe why the balance of OXN and RDN is crucial and how VOCs affect this balance.

Response and revisions:

We thank the reviewer's constructive suggestion. The sentences have been refined as "More importantly, the O_3 formation in eastern China has been primarily under the NO_x-saturated condition, and the reduction in NO_x emissions, combined with persistently high volatile organic compounds (VOCs) emissions, has enhanced the O_3 concentration and thereby the capacity of atmospheric oxidation. This has in turn facilitated the conversion of NO_x to nitrate (NO_3^-), and thus weakened the response of OXN deposition to NO_x emission abatement, preventing effective reduction of Nr deposition." **in lines 92-98 in the revised manuscript.**

6. L86-89: Explain how climate change and pollution controls will alter Nr deposition. For instance, describe which meteorological conditions and emission sources are expected to change and how these changes might specifically impact Nr deposition patterns.

Response and revisions:

We thank the reviewer's constructive suggestion. The sentences have been refined as "Future climate change may strengthen the local turbulence and precipitation intensity, which will alter the dry and wet deposition rate, respectively (Toyota et al., 2016; Xia et al., 2024). Meanwhile, the anticipated substantial reduction in Nr emissions through pollution controls will reduce the Nr deposition and change its dominant components." **in lines 103-107 in the revised manuscript.** We have also added the necessary references **(see below) in lines 1077-1080 and 1121-1123 in the revised manuscript.**

Reference:

Toyota, K., Dastoor, A. P., & Ryzhkov, A. (2017). Parameterization of gaseous dry deposition in atmospheric chemistry models: Sensitivity to aerodynamic resistance formulations under statically stable conditions. Atmospheric Environment, 147, 409-422. https://doi.org/10.1016/j.atmosenv.2016.09.055

Xia, W., Wang, Y., Zhang, G. J., & Wang, B. (2024). Light Precipitation rather than Total Precipitation Determines Aerosol Wet Removal. Environmental Science & Technology, in press. https://doi.org/10.1021/acs.est.4c07684

7. L92-94: Please specify the studies.

Response and revisions:

We apologize for the confusing statement regarding previous studies on future Nr deposition. In fact, the specific studies have been introduced later in this paragraph. Here we have reorganized the sentences **in lines 110-120 in the revised manuscript** as "There are only a few studies addressing future Nr deposition in China. They commonly employed coupled climate-chemistry global models to conduct simulations under different predefined greenhouse gas (GHG) emission scenarios. For example, a pioneering study by Galloway et al. (2004) predicted significant growth in Nr deposition in East Asia, exceeding 50 kg N ha^{-1} yr^{-1} by 2050, based on the Intergovernmental Panel on Climate Change IS92a (IPCC92a) emission scenario. The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) presented a multi-model global dataset of Nr deposition from 1850 to 2100 (Lamarque et al. 2013a), with the future emissions obtained from the IPCC Representative Concentration Pathways (RCPs) based on the radiative forcing in 2100 (van Vuuren et al., 2011).".

8. L105-110: Please ensure that the references to Zhang et al. (2019) and Sun et al. (2022) are clearly linked to their respective findings. It might be helpful to briefly summarize each study's focus to avoid ambiguity.

Response and revisions:

We appreciate the reviewer for pointing out this confusing expression. The findings and focus of the two studies have been summarized separately. Specifically, Zhang et al. (2019) focused on OXN deposition and projected future changes under different RCPs, while Sun et al. (2022) examined the future changes in RDN deposition and its increasing proportion in total Nr deposition. Please see **lines 121-128 in the revised manuscript**.

9. L111: Please re-write the sentence for better readability.

Response and revisions:

The sentence has been rewritten as "While previous studies have provided valuable information on the future evolution of Nr deposition in China, they have insufficiently considered the impact of the potentially profound emission reduction resulting from implementation of climate and pollution control policies." **in lines 129-132 in the revised manuscript**.

10. L127-134: Please consider re-writing the sentences for easier understanding and readability.

Response and revisions:

The sentences have been rewritten as "Moreover, the diverse trajectories of emission for various species and regions will change the atmospheric oxidizing capacity and regional transport of pollution, respectively, which will in turn change the response of Nr deposition to the changing precursor emissions. It is essential to evaluate these anticipated changes for a comprehensive understanding of the ecological and environmental impacts of Nr deposition, for a long-term period with continuous air pollution controls and global warming prevention." **in lines 145-151 in the revised manuscript**.

11. L143-145: Please add reference.

Response and revisions:

We thank the reviewer's kind reminder. The necessary additional references **(see below)** have been added **in lines 162-163, 165, 739-742, 991-997, 1004-1009 and 1055-1058 in the revised manuscript**.

Reference:

Alexandrov, G. A., Ginzburg, V. A., Insarov, G. E. & Romanovskaya, A. A. (2021). CMIP6 model projections leave no room for permafrost to persist in Western Siberia under the SSP5-8.5 scenario. Climatic Change, 169(3), 42. https://doi.org/10.1007/s10584-021-03292-w

Meinshausen, M., Nicholls, Z. R. J., Lewis, J., Gidden, M. J., Vogel, E., Freund, M., Beyerle, U., Gessner, C., Nauels, A., Bauer, N., Canadell, J. G., Daniel, J. S., John, A., Krummel, P. B., Luderer, G., Meinshausen, N., Montzka, S. A., Rayner, P. J., Reimann, S., Smith, S. J., van den Berg, M., Velders, G. J. M., Vollmer, M. K., & Wang, R. H. J. (2020). The shared socio-economic pathway (SSP) greenhouse gas concentrations and their extensions to 2500. Geoscientific Model Development, 13(8), 3571-3605.

https://doi.org/10.5194/gmd-13-3571-2020

O'Neill, B. C., Carter, T. R., Ebi, K., Harrison, P. A., Kemp-Benedict, E., Kok, K., Kriegler, E., Preston, B. L., Riahi, K., Sillmann, J., van Ruijven, B. J., van Vuuren, D., Carlisle, D., Conde, C., Fuglestvedt, J., Green, C., Hasegawa, T., Leininger, J., Monteith, S., & Pichs-Madruga, R. (2020). Achievements and needs for the climate change scenario framework. Nature Climate Change, 10(12), 1074-1084. https://doi.org/10.1038/s41558-020-00952-0

Su, B., Huang, J., Mondal, S. K., Zhai, J., Wang, Y., Wen, S., Gao, M., Lv, Y., Jiang, S., Jiang, T., & Li, A. (2021). Insight from CMIP6 SSP-RCP scenarios for future drought characteristics in China. Atmospheric Research, 250, 105375. https://doi.org/10.1016/j.atmosres.2020.105375

12. Section 2.1: Please consider rearranging this section slightly to make it in a better flow. I suggest to have subsections for model descriptions, emission data and deposition mechanisms.

Response and revisions:

We thank the reviewer's constructive suggestion. This section has been reorganized according to the newly added subsection titles of "2.1.1 CMAQ model", "2.1.2 Emissions input", "2.1.3 Meteorological driving field" and "2.1.4 Deposition mechanisms". Please see **lines 174-300 in the revised manuscript**.

13. L155-177: The description of the model appears disjoined and seems abrupt at some points. The paragraph jumps from discussing the model to the rationale of selection periods, back to model, and then again to simulation design. Please re-write the paragraph to improve the clarity and flow of the section.

Response and revisions:

We apologize for the disjoined description in this paragraph. We have revised it following the order of model description, model setup, and simulation design. Please see **lines 174-202 in the revised manuscript**.

14. L155-177: The paragraph does not clearly describe what kind of model CMAQ is or how it functions in the context of this study. I recommend including a brief description on important processes included in the model including deposition. Please also add references for the model.

Response and revisions:

We warmly thank the reviewer for pointing out the incomplete presentation of the CMAQ descriptions. More details on model framework and important processes have been added as "As a three-dimensional Eulerian model developed by the United States Environmental Protection Agency (USEPA), CMAQ comprehensively considers the complex atmospheric physical and chemical processes among various air pollutants, primarily including advection, vertical mixing, chemistry of gas and aerosol phase, cloud chemistry, as well as dry and wet deposition (Benish et al., 2022; Fahey et al., 2017)." **in lines 178-183 in the revised manuscript**.

In addition, the necessary references **(see below)** have also been added **in lines 749-752 and 820-824 in the revised manuscript**.

Reference:

Benish, S. E., Bash, J. O., Foley, K. M., Appel, K. W., Hogrefe, C., Gilliam, R., & Pouliot, G. (2022). Long-term regional trends of nitrogen and sulfur deposition in the United States from 2002 to 2017. Atmospheric Chemistry and Physics, 22(19), 12749–12767. https://doi.org/10.5194/acp-22-12749-2022

Fahey, K. M., Carlton, A. G., Pye, H. O. T., Baek, J., Hutzell, W. T., Stanier, C. O., Baker, K. R., Appel, K. W., Jaoui, M., & Offenberg, J. H. (2017). A framework for expanding aqueous chemistry in the Community Multiscale Air Quality (CMAQ) model version 5.1. Geoscientific Model Development, 10(4), 1587-1605. https://doi.org/10.5194/gmd-10-1587-2017

15. L184: Please add reference for DPEC.

Response and revisions:

We thank the reviewer's kind reminder. The necessary additional references of "Cheng et al., 2021a, 2021b" have been added **in line 210 in the revised manuscript**.

16. L206: Please add reference for WRF.

Response and revisions:

We have added the corresponding reference **(see below) in lines 236 and 1051-1054 in the revised manuscript**.

Reference:

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D., Duda, M. G., Huang, X.-Y., Wang, W., & Powers, J. G. (2008). A Description of the Advanced Research WRF Version 3, NCAR technical note, NCAR/TN-475+STR. https://doi.org/10.5065/D68S4MVH

17. L225: Expand CMIP6.

Response and revisions:

We thank the reviewer's reminder. The full name of CMIP6 has been added at its first occurrence in the text. Please see **line 215 in the revised manuscript**.

18. Section 2.2: I understand "Base" is referred to as the simulation of the historical period, but it could be clearer if "Base" is consistently referred to as "the Base case" or "the Base scenario" throughout the text.

Response and revisions:

We appreciate the reviewer's kind suggestion. The term "Base" has been refined as "Base case" in the whole manuscript carefully.

19. L296: While the emission perturbation experiments (Cases 9-12) are described, it would be helpful to explain why a 20% reduction was chosen as the perturbation level. Is this based on realistic policy scenarios, or is it intended as a sensitivity analysis?

Response and revisions:

We thank the reviewer's constructive suggestion. The selection of a perturbation level of 20% reduction was intended as a sensitivity analysis. The value has been widely adopted in numerical modeling projects of atmospheric concentrations and deposition fluxes at both global scales (e.g., the Task Force on Hemispheric Transport of Air Pollution, TF HTAP) and regional scales (e.g., the Air Quality Model Evaluation International Initiative, AQMEII). The choice of 20% was motivated by the consideration that the perturbation would be large enough to produce a sizeable impact (i.e., more than numerical noise), and at the same time an affordable target for an emissions reduction policy (Galmarini et al., 2017).

Additional explanations have been added as "The 20% emissions reduction was regarded as a reasonable perturbation to achieve a significant change (Galmarini et al., 2017)." **in lines 328-330 in the revised manuscript**. We have been also added the necessary reference **(see below) in lines 831-836 in the revised manuscript.**

Reference

Galmarini, S., Koffi, B., Solazzo, E., Keating, T., Hogrefe, C., Schulz, M., Benedictow, A., Griesfeller, J. J., Janssens-Maenhout, G., Carmichael, G., Fu, J., & Dentener, F. (2017). Coordination and harmonization of the multi-scale, multi-model activities HTAP2, AQMEII3, and MICS-Asia3: simulations, emission inventories, boundary conditions, and model output formats. Atmospheric Chemistry and Physics, 17(2), 1543-1555. https://doi.org/10.5194/acp-17-1543-2017

20. L310-312: Please cite the previous study.

Response and revisions:

We thank the reviewer's valuable suggestion. The previous study mentioned here is Ma et al. (2023). The sentences have been revised as "We selected 28 sites for dry deposition fluxes and 53 sites for wet deposition fluxes, for which at least two-year continuous measurement data were available, to evaluate model performance. Details of monitoring stations can be found in our previous study (Ma et al., 2023)." **in lines 342-345 in the revised manuscript**.

21. L349: NME for the DDEP OXN in Table 2 is 34.76%. Which one is correct?

Response and revisions:

We thank the reviewer for pointing out the inconsistent description of statistical indicators related to Nr deposition. The value in Table 2 is correct. The NME for the OXN_DDEP has been changed to 34.76% **in line 383 in the revised manuscript**.

22. Table 2: Please be consistent on acronyms throughout the text. Table header has OXN DDEP and caption and description uses DDEP OXN.

Response and revisions:

We warmly thank the reviewer for pointing out the inconsistent description of acronyms. The acronyms related to nitrogen deposition for different components and forms throughout the manuscript have been uniformly provided as OXN_DDEP, RDN_DDEP, OXN_WDEP, and RDN_WDEP. Such revisions include **lines 382-383, 396-397, 463, 467 and 472 in the revised manuscript**.

23. L364-365: Based on the results, it is quite strong speculation that the underestimation of precipitation could indeed explain the lower wet deposition values. Have you done some kind of sensitivity analysis by adjusting the precipitation inputs in the model to better understand their dependence?

Response and revisions:

We thank the reviewer's important and insightful comment. Unfortunately, we did not conduct specific sensitivity analysis by adjusting the precipitation inputs. The relationship between wet deposition and precipitation is affected by the frequency, duration, and intensity of the rainfall as well as the ambient concentration. Several studies have reported that this relationship is typically nonlinear on hourly and daily time scales, but it becomes closer to linear on an annual accumulation basis (Sahu et al., 2010; Zhang et al., 2019).

Therefore, we have expanded the sentence as "Part of the reason may be underestimation of precipitation (Figure 1d), given the closely linear relationship between wet deposition and precipitation on an annual accumulation basis (Sahu et al., 2010; Zhang et al., 2019)." **in lines 398-401 in the revised manuscript.** The additional reference **(see below)** has also been added **in lines 1029-1032 in the revised manuscript**.

Reference:

Sahu, S. K., Gelfand, A. E., & Holland, D. M. (2010). Fusing point and areal level space–time data with application to wet deposition. Journal of the Royal Statistical Society Series C: Applied Statistics, 59(1), 77-103. https://doi.org/10.1111/j.1467-9876.2009.00685.x

24. L387-410: Please consider rearranging the paragraph so that the explanation of the projected reduction in NOx emissions precedes the discussion of its impact on OXN and RDN deposition. Also, I recommend including Figure S2 in the main text to provide better guidance for readers.

Response and revisions:

We thank the reviewer's constructive suggestion. The order of this paragraph has been rearranged to first analyze the projected changes in emissions before discussing the resulting deposition changes. Please see **lines 424-455 in the revised manuscript**. Additionally, we have moved Figure S2 in the original supplement to the main text **as Figure 2 in the revised manuscript**.

25. L486-493: The assumption that OXN deposition is smaller than that of NOx changes in WC are mainly due to reduced atmospheric transport to EC seems reasonable. Please add explain this more, also taking into account the atmospheric chemistry.

Response and revisions:

We apologize for the insufficient explanation and discussion in this section. To better support this assumption, we further calculated the ratio of changes in OXN outflow to changes in NO_x emissions ($\Delta T/\Delta E$) by combining the sensitivity simulation cases with fixed WC emissions as 2010s, and such information has been added in Figure 6 in the revised manuscript (Figure 5 in the original submission; **see below**). Under the baseline emission scenario, $\Delta T/\Delta E$ is greater than 1, indicating that "efficacy" of eastward transport of OXN would be enhanced, especially for WC. This resulted in a growing OXN deposition that would greatly lag behind the growth of emissions. The meteorological conditions of high wind speeds and low humidity in WC would hinder the conversion of aerosol NO_3^-, resulting in a high proportion of NO_2 in total OXN. Gaseous NO_2 usually has stronger long-distance transport capability, thus contributing to the high transport efficacy of OXN. Under the two emission reduction scenarios, the efficacy of eastward transport of OXN would decrease ($\Delta T/\Delta E < 1$), resulting in a larger decline in deposition compared to that in emissions.

The additional explanations and clarifications have been summarized **in lines 546-557 and 1232-1234 in the revised manuscript**.

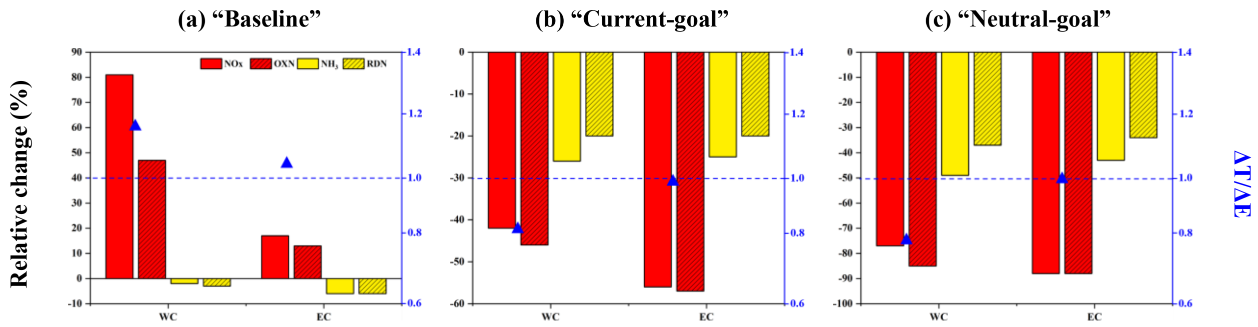

Figure 6 in the revised manuscript: Relative changes in Nr emissions and deposition as well as the ratio of changes in OXN outflow to changes in NO_x emissions (ΔT/ΔE) in WC and EC from 2010s to 2060s under different emission scenarios.

26. L510-515: I recommend to replace absolute change with relative change.

Response and revisions:

We thank the reviewer's kind suggestion. The sentences have been revised as "Under the "Baseline" scenario, the outflow fluxes from EC in 2060s would increase by **19%** compared to the case with the emissions maintained at the 2010s level" and "In contrast, the outflow fluxes under the scenarios of "Current-goal" and "Neutral-goal" would respectively decline by **49%** and **89%** attributable to the emission abatement in EC" **in lines 564-568 in the revised manuscript**.

27. Sect. 3.4: Given that the interactions between VOCs, NOx, and O_3 formation can vary greatly depending on regional atmospheric conditions, could you please explain how does the model account for these non-linear and region-specific interactions?

Response and revisions:

We sincerely thank the reviewer's question. The CMAQ model describes the non-linear interactions between NO_x, VOCs, and O_3 through an embedded module of photochemical mechanism (e.g., CB05 and SAPRC). These complex mechanisms include hundreds of chemical reaction equations, with reaction rates depending on reactant concentrations, temperature, and solar radiation conditions. For example, different ratios of NO_x and VOCs concentrations can significantly affect the O_3 formation rate. For photolysis reactions that are crucial for O_3 formation (e.g., NO_2 photolysis), the model dynamically calculates grid-level reaction rate constants at each time step based on atmospheric conditions such as solar radiation intensity, thereby further introducing non-linear effects.

Regional emission and meteorological conditions, as provided by the emission inventories and meteorological driving fields, affect the local photochemical reaction pathways and formation rates. This reflects the region-specific O_3-VOCs-NO_x interactions. For instance, O_3 formation is generally VOCs-limited in urban areas with high NO_x emissions, while in suburban areas with low NO_x, it is more likely to be NO_x-limited.

We have added a sentence as "The model incorporates the temporal and spatial variations of chemical mechanisms, emissions and meteorology, thus effectively accounting for nonlinearity and regional transport (Liu et al., 2010).". Please see **lines 183-185 in the revised manuscript**.

28. Bonus question: Do you think your results would be consistent in other models with the same horizontal resolution? What key factors should be taken into account when applying these findings to broader global or regional air quality assessments?

Response and revisions:

We sincerely thank the reviewer's insightful question. Under similar horizontal resolution, meteorological driving fields, and emission processing methods, other atmospheric chemistry models that incorporate photochemical and heterogeneous chemical processes (e.g., WRF-Chem, CAMx, GEOS-Chem, etc.) could also yield similar trends in the response of Nr deposition to emission reductions. Using the WRF-Chem model, Liu et al. (2022) also found a similar weak response of OXN deposition to NO_x emissions in China during the 2010s based on sensitivity simulations. However, it should be noted that there are differences in the parameterization of specific chemical processes among these models, which can affect the sensitivity of pollutants to emission changes (Yu et al., 2010). For example, different mechanisms produce varying rate constants for the reaction "$NO_2 + OH \rightarrow HNO_3$" (CB4: 1.15E-11 cm^3 $molecule^{-1}$ s^{-1}, CB05: 1.05E-11 cm^3 $molecule^{-1}$ s^{-1}, SAPRC-99: 8.98E-12 cm^3 $molecule^{-1}$ s^{-1}).

Our findings emphasize the non-linear relationship between pollutant deposition and the precursor emission reductions, which is influenced by the combined effects of climate change, regional transport, and gas-aerosol phase chemistry. When conducting broader air quality assessments, these factors should be considered comprehensively.

Reference:

Liu, M., Shang, F., Lu, X., Huang, X., Song, Y., Liu, B., Zhang, Q., Liu X., Cao, J., Xu, T., Wang T., Xu, Z., Xu, W., Liao W., Kang L., Cai, X., Zhang, H, Dai, Y., & Liu, X. (2022). Unexpected response of nitrogen deposition to nitrogen oxide controls and implications for land carbon sink. Nature Communications, 13(1), 3126. https://doi.org/10.1038/s41467-022-30854-y

Yu, S., Mathur, R., Sarwar, G., Kang, D., Tong, D., Pouliot, G., & Pleim, J. (2010). Eta-CMAQ air quality forecasts for O_3 and related species using three different photochemical mechanisms (CB4, CB05, SAPRC-99): comparisons with measurements during the 2004 ICARTT study. Atmospheric Chemistry and Physics, 10(6), 3001-3025. https://doi.org/10.5194/acp-10-3001-2010

Reviewer #2 Evaluations:

The authors present a modeling study of wet and dry deposition of oxidized and reduced nitrogen species in China under several emissions and climate scenarios. For the most part, these simulations are clearly described, as are the findings. I recommend publication subject to these minor revisions.

Response and revisions:

We warmly thank the reviewer and show our deepest respect to the reviewer for the constructive and insightful suggestions. We are encouraged by the positive comments from the reviewer on our work. Following is our detailed response and revisions according to reviewer's comment.

Comments:

1. I do not understand the point of the Western China vs Eastern China analysis. The authors conclude that WC emissions contribute to deposition in EC, while emissions in EC contribute to pollutant outflow to the western Pacific. Is any of this surprising or informative?

Response and revisions:

We apologize for the insufficient explanation and discussion in this section. The motivation for the analysis of Western China (WC) versus Eastern China (EC) is to explore the potential changes in regional transport intensity induced by different emission control scenarios. To better understand the transport intensity, we further calculated the ratio of changes in OXN outflow to changes in NO_x emissions ($\Delta T/\Delta E$) in WC and EC, respectively, as shown in Figure 6 in the revised manuscript (improved based on Figure 5 in the original submission; **see below**).

Under the baseline emission scenario, $\Delta T/\Delta E$ is greater than 1 for both WC and EC, indicating that the "efficacy" of eastward transport of OXN would be enhanced. This resulted in a growing OXN deposition that would lag behind the growth of emissions. The transport efficacy of OXN from WC would be greater than that from EC, due partly to the higher wind speeds and lower humidity in WC. The specific meteorological conditions hinder the conversion of aerosol NO_3^-, resulting in a high proportion of NO_2 in total OXN. Gaseous NO_2 usually has stronger long-distance transport capability, thus contributing to the high transport efficacy of OXN. Under the two emission reduction scenarios, the transport efficacy of OXN from WC to EC would decline, with ΔT/ΔE around 0.8, resulting in a larger reduction in deposition compared to that in emissions. In contrast, the ΔT/ΔE in EC is closer to 1, which indicates more similar changes in NO_x emissions and OXN deposition.

Overall, the disparity in transport efficacy between WC and EC reflects the different relationships between the changing deposition and emissions, and highlights the important role of inter-regional transport in the evolution of source-sink relationship of air pollutants.

We have been added the additional explanations and clarifications **in lines 546-557, 569-574 and 1232-1234 in the revised manuscript.**

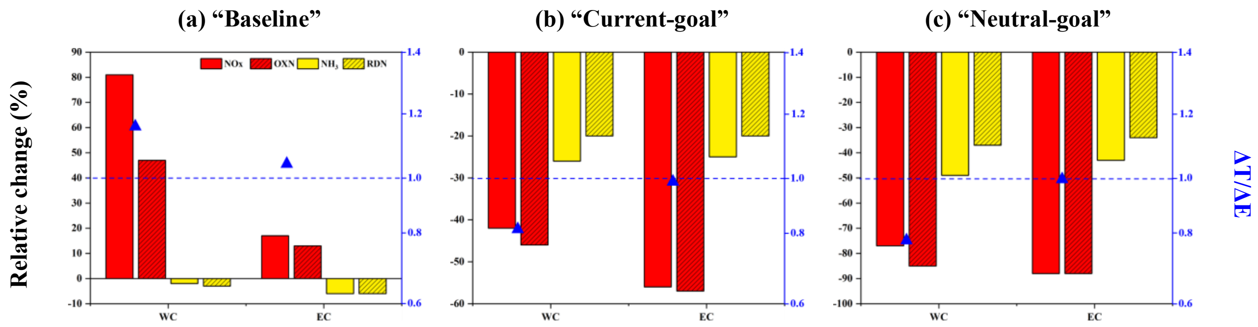

Figure 6 in the revised manuscript: Relative changes in Nr emissions and deposition as well as the ratio of changes in OXN outflow to changes in NO_x emissions (ΔT/ΔE) in WC and EC from 2010s to 2060s under different emission scenarios.

2. It would be useful to add a few sentences in the discussion comparing the results of the present study to prior studies. Obviously the projected deposition totals are far lower than those in Galloway et al. The authors may wish to comment on why their projections were so high, even in comparison to the relatively pessimistic SSP5-8.5 scenario. Are your results in line with ACCMIP and Lamarque et al. (2013b)?

Response and revisions:

We thank the reviewer’s constructive suggestion. Overall, our predictions of future total Nr deposition were generally lower than those from previous global-scale studies, particularly the results of Galloway et al. (2004). They found that Nr deposition flux in most of East Asia would exceed 50 kg N ha^{-1} yr^{-1} by 2050 under the old emission scenario of IPCC92a. The results from ACCMIP datasets that relied on RCPs framework are more comparable to our study. Specifically, Lamarque et al. (2013b) reported that the region-averaged Nr deposition in East Asia would reach 6.9-10.2 kg N ha^{-1} yr^{-1} by 2100, which is roughly in line with our results (9.0-15.4 kg N ha^{-1} yr^{-1}) in the 2060s.

Such difference arises from the different assumptions on the changing air pollutant emissions in China across studies. By considering China's near-term strict clean air actions and the anticipated long-term emissions controls, the air pollutant emission levels in DPEC (used in this study) are lower than those in any existing CMIP emission scenarios and the earlier IPCC92a scenario. This would result in predictions with lower air pollutant concentrations and deposition. For example, Cheng et al. (2021a) compared $PM_{2.5}$ concentrations in China driven by the CMIP6 and DPEC emission scenarios. As shown in Figure R1, the declining rate of simulated $PM_{2.5}$ for CMIP6 cases were much smaller than DPEC-driven cases between 2015 and 2019, and the latter were close to in-situ observations.

The discussions about the comparison of total Nr deposition between our results and the previous studies have been summarized **in lines 657-669 in the revised manuscript**.

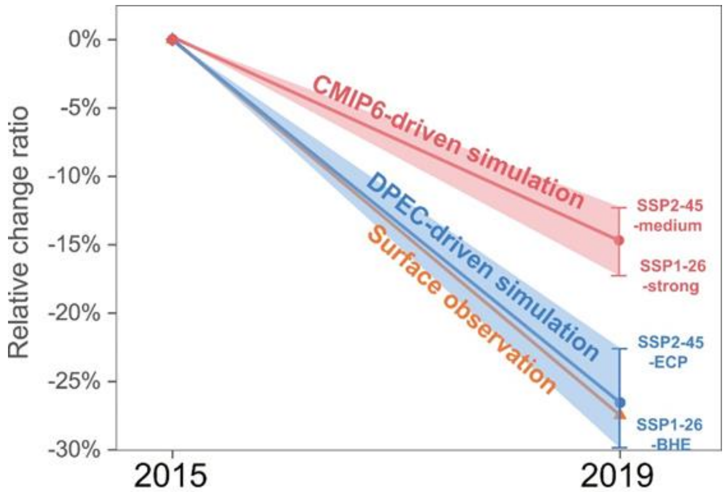

Figure R1 Relative change ratios of surface in-situ observed and simulated $PM_{2.5}$ concentrations during 2015-2019 driven by DPEC (blue) and CMIP6 (red) emissions under the strong and medium mitigation pathways.

Reference:

Cheng, J., Tong, D., Liu, Y., Yu, S., Yan, L., Zheng, B., Geng, G., He, K., & Zhang, Q. (2021a). Comparison of current and future PM2.5 air quality in China under CMIP6 and DPEC emission scenarios. Geophysical Research Letters, 48(11), e2021GL093197.

https://doi.org/10.1029/2021GL093197

3. The authors imply that simulating 5 years is sufficient to capture interannual variability in annual deposition. From my own work I suspect this is true, but the authors could easily demonstrate this with an additional figure in the supplement. How much Quantifying variability is there in annual total deposition from year to year, either in observations or in the model, particularly when emissions are held constant from year to year?

Response and revisions:

We thank the reviewer's constructive suggestion. At the site scale, the temporal correlation coefficient (R) between the simulated and observed annual Nr deposition for different components and forms were 0.63-0.82, as shown in Table 2 in the text. This indicates that our 5-year simulations effectively capture the interannual variability.

Thus, we further presented the annual total Nr deposition for 2010-2014 (Base case) and for 2060-2064 (blue) under the SSP2-4.5 (Case 1) and SSP5-8.5 (Case 2), as shown in a newly added Figure S3 in the revised supplement. The standard deviation (SD, kg N ha^{-1} yr^{-1}) was calculated to quantify the interannual variability. With the joint influence of emissions and meteorology, the SD for 2010-2014 was 0.78 kg N ha^{-1} yr^{-1}. In the future simulations, the emissions in Case 1 and Case 2 were held constant from year to year. The interannual variability in Nr deposition during 2060-2064 would result solely from meteorological fluctuations, with the SD estimated at 0.27 and 0.45 kg N ha^{-1} yr^{-1}, respectively.

Description of the 5-year interannual variability has been added in **lines 410-411 and 448-455 in the revised manuscript**.

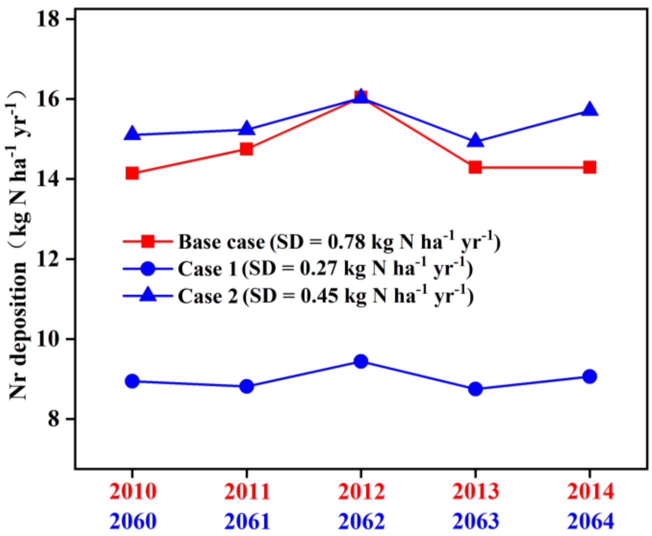

Figure S3 in the revised supplement: Annual Nr deposition flux (kg N ha^{-1} yr^{-1}), five-year standard deviation (SD, kg N ha^{-1} yr^{-1}) for 2010-2014 (red) and for 2060-2064 (blue) under the SSP2-4.5 (Case 1) and SSP5-8.5 (Case 2).

Technical corrections by line number:

4. L63 "hotpots" should be "hot spots" or "hotspots"

Response and revisions:

The "hotpots" has been revised as "hotspots" **in line 65 in the revised manuscript**.

5. L70 fluxes has increased --> fluxes have increased.

Response and revisions:

The sentence has been refined as "Yu et al. (2019) applied the Kriging interpolation combined with empirical remote sensing models and estimated that China’s annual Nr deposition had increased nearly 60% from 1980s to 2010s" **in lines 79-81 in the revised manuscript**.

6. L76 e.g. should be i.e.

Response and revisions:

The "e.g." has been revised as "i.e." **in line 88 in the revised manuscript**.

7. L81-82 resulted in only less than 80% of OXN deposition --> resulted in less than 80%

abatement of (or "reduction in") OXN deposition.

Response and revisions:

The "resulted in only less than 80% of OXN deposition" has been revised as "resulted in less than 80% abatement of OXN deposition" **in lines 98-99 in the revised manuscript**.

8. L84 are --> is.

Response and revisions:

The "are" has been revised as "is" **in lines 101 in the revised manuscript**.

9. L97-99 It is a bit confusing to cite Galloway et al. (2004) after saying studies primarily used RCPs, since Galloway et al. preceded the RCPs. Also, from skimming the text of Galloway et al. it appears a variety of model types was used, not just coupled climate-chemistry models. One approach would be to leave the "A pioneering study by Galloway et al..." as-is, but move it up earlier in the paragraph.

Response and revisions:

We appreciate the reviewer for pointing out this confusing expression. The statement about Galloway et al. (2004) should indeed appear before the studies that primarily used RCPs. Here we have reorganized the sentences as "For example, a pioneering study by Galloway et al. (2004) predicted significant growth in Nr deposition in East Asia, exceeding 50 kg N ha^{-1} yr^{-1} by 2050, based on the Intergovernmental Panel on Climate Change IS92a (IPCC92a) emission scenario. The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) presented a multi-model global dataset of Nr deposition from 1850 to 2100 (Lamarque et al. 2013a), with the future emissions obtained from the IPCC Representative Concentration Pathways (RCPs) based on the radiative forcing in 2100 (van Vuuren et al., 2011)." **in lines 112-120 in the revised manuscript**.

10. L100 datasets --> dataset.

Response and revisions:

The "datasets" has been revised as "dataset" **in line 117 in the revised manuscript**.

11. L111 "Even" should be "Even though", "Though", or "While".

Response and revisions:

The "Even" has been revised as "While". Additionally, for better readability, the sentence has been rewritten as "While previous studies have provided valuable information on the future evolution of Nr deposition in China, they have insufficiently considered the impact of the potentially profound emission reduction resulting from implementation of climate and pollution control policies." **in lines 129-132 in the revised manuscript**.

12. L127 "progresses" is not the correct word. Perhaps "trajectories".

Response and revisions:

We are grateful to the reviewer for pointing out the improper word choice. The sentence has been revised as "Moreover, the diverse trajectories of emission for various species and regions will change the atmospheric oxidizing capacity and regional transport of pollution, respectively, which will in turn change the response of Nr deposition to the changing precursor emissions." **in line 145-148 in the revised manuscript**.

13. L187-188 "corrects the erroneous emission trends" is rather harsh: isn't it the case that the CMIP scenarios were projections based on then-current legislation? Also, which phase of CMIP are you referring to?

Response and revisions:

We thank the reviewer for pointing out this imprecise expression. The sentence has been revised as "It thus better depicts the emission trends of China compared to the results in the sixth Coupled Model Intercomparison Project (CMIP6) scenarios" **in lines 213-215 in the revised manuscript.**

14. L219 commonly practical --> common practice.

Response and revisions:

The "commonly practical" has been revised as "common practice" **in line 247 in the revised manuscript**.

15. L230 Since you are using WRF, a dynamical model, this is "dynamical" rather than "statistical" downscaling.

Response and revisions:

We appreciate the reviewer for pointing out the error and the “statistical” has been revised as “dynamical” **in line 258 in the revised manuscript**.

16. L291 - The text here says Case 7 is based on Case 3, but Table 1 indicates it is based on Case 2. Which meteorological and emissions data were used for Case 7?

Response and revisions:

We thank the reviewer for pointing out the inconsistency regarding the simulation cases. The description in Table 1 is accurate. The meteorological and emissions data used for Case 7 are based on Case2, i.e., following the SSP5-8.5 climate pathway and “Baseline” emission scenario. The incorrect description has been corrected in **in line 321 in the revised manuscript**.

17. Figure 1: In my view, it is inappropriate to compute NMB and NME statistics for temperatures in degrees Celsius. It is not meaningful to say that there is a 10% error in temperature, since if you measured temperature in kelvins, the bias and error statistics would be different (better). Instead one should compute MB and ME statistics, in degrees.

Response and revisions:

We warmly thank the reviewer for the important insights regarding the calculations of meteorological verification. The statistics of NMB and NME involve ratios in their calculations, while temperature errors are typically expressed in absolute values. We have replaced them with mean bias (MB) and mean error (ME). The equations **(see below)** have been added in the supplement. The results show that the T2 was generally underestimated with the MB and ME calculated at -0.94 ℃ and 1.54 ℃, respectively. Moreover, the verifications of other meteorological parameters have also been recalculated as MB and ME for consistency. The relevant descriptions in Section 2.3, Section 3.1 and Figure 1 have been revised **in lines 345-347, 366-378 and 1269 in the revised manuscript**.

$$MB = \frac{1}{n}\sum_{i=1}^{n}(S_i - O_i) \quad \text{(S1)}$$

$$ME = \frac{1}{n}\sum_{i=1}^{n}|S_i - O_i| \quad \text{(S2)}$$

18. L362 - "Compared to dry deposition, wet deposition of OXN and RDN was simulated to be far lower than the observations." I do not understand this sentence. Are you referring to absolute quantities? It seems not, since later (line 385-385) you say the ratio of wet dep to total dep is 0.54. But if you mean that the wet deposition bias is much lower, that does not make sense either. The NMB for OXN_WDEP is greater (more negative), while the bias for RDN_WDEP is lower (less negative).

Response and revisions:

We apologize for the incorrect statement regarding the comparison of wet deposition between simulations and observations. To clarify, we have revised the sentence as "The wet deposition of OXN and RDN (OXN_WDEP and RDN_WDEP) were also underestimated compared to observations" **in lines 396-397 in the revised manuscript**.

19. L449 - The text refers to a spatial correlation of 0.89 (between Fig 3b and 3c?), but the value shown in Fig 3b is 0.97.

Response and revisions:

We thank the reviewer for pointing out this inconsistency. The true spatial correlation value is 0.97 in the figure, and we have corrected this error **in line 493 in the revised manuscript.**

20. L496, 509 - 100 hPa should probably be 50 hPa here, since that was the model top stated on line 171.

Response and revisions:

We thank the reviewer for pointing out this expression error. The "100 hpa" has been revised as "50 hpa" **in lines 539 and 563 in the revised manuscript.**