

Responses to reviewers (original comments by reviewers are in blue).

Reviewer #1:

Comments on Liu et al., 2024

General summary:

Overall, I think there is clear evidence that a lot of work has been done and much data produced. The writing is clear. The technical analysis seems logical and results reported well, although I cannot speak much to the actual modelling technical components as that is outside my expertise.

My primary critique revolves around the structure and narrative of the paper. Namely, there is so much information presented that it is sometimes difficult to keep track of what the overall main point is that the authors are trying to get across. I think this is exemplified in two ways: first, the title itself about “Exploring...mechanisms”, while accurate to the content, highlights that there is less focus on a definitive point/conclusion than just summarizing and overviewing a lot of results from modelling. Second, there is a massive Results section, but no Discussion. There is some discussion happening within the Results, but the paper would likely improve by having less space dedicated to describing every result from the modelling and more space on what those results mean for things like policy implications, the need to treat interior cities differently from coastal ones in regulations, etc. Overall, I would suggest that the authors take a fresh look at the content of the paper as a whole narrative and re-evaluate if everything in the results needs to be included and described at the detail it currently is at.

That said, I do think that there is a good information here, and a valuable contribution. And I have largely minor critiques on the technical side and scientific content. But for the authors' sake, I think the paper's eventual impact could be greatly improved by focusing the narrative, simplifying/summarizing some of the base results further, and speaking more to the broader implications of this work.

Reply: Thank you for your valuable suggestions for improvement. We agree that the current results section had been overly detailed, which may have detracted from the overall focus of the paper. In response to your feedback, we have implemented the following adjustments:

1. Revised the Title

In line with your suggestions, we had revised the title into: “Enhanced Atmospheric Oxidation and Particle Reductions Driving Changes to Nitrate Formation Mechanisms Across Coastal and Inland Regions of North China.”

2. Streamlined the Results Section

In Section 3.2, we removed some comparisons with previous literature to highlight the unique findings of this study. We streamlined the content by summarizing key trends in a more concise format, with a particular focus on the differences between inland and coastal regions. Additionally, we merged relevant figures, as shown in Figure 11.

3. Enhanced the Discussion

To enhance the discussion, we enriched the results section with additional commentary to emphasize the broader implications and scientific significance of our findings. In Section 3.4 (lines 413–489), we enhanced the discussion of model-isotope comparisons and their implications. Specifically, we proposed concrete policy recommendations, such as intensifying control measures targeting photochemical reactions in inland cities (lines 584–588). We proposed that coastal cities

should focus more on regulations related to aerosol surface area (lines 630–636). Additionally, in Section 3.6, we conducted a comparative analysis of emission reduction experiments between Beijing and Qingdao, providing targeted strategies for improving air quality based on the unique characteristics of each city.

Major points:

I cannot see a supplemental section on the Preprint review page? This made it impossible to examine things referenced in the methods. (Apologies if this was a mistake on my part).

Results: There is a huge amount of information and data both presented and discussed. While I commend the authors for being upfront with their data, it can be a bit overwhelming at times and causes some of the focus to be lost. I would recommend looking back over this section to determine what exactly are the main points and stories you are aiming to get across, and pare down any information and number discussion that distracts away from those points. Perhaps greater summarization of regional trends (e.g., inland vs. coastal) rather than relaying data from multiple cities would help focus the section, too. You do this already some by focusing on Beijing vs. Qingdao, but even further summarization/simplification could help in some spots.

There are a lot of figures, and many of them are similar in theme (e.g., comparing an atmospheric chemical in 2013 and in 2018 and their difference). Perhaps combining many of these into a single, larger figure would be more effective as the reader could cross compare more easily and not hit figure fatigue.

Data availability: This is an unacceptable statement for data availability, as per ACP standards. Data are to be hosted in a publicly accessible location. See further guidance from https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html:

If the data are not publicly accessible at the time of final publication, the data statement should describe where and when they will appear, and provide information on how readers can obtain the data until then. Nevertheless, authors should make such embargoed data available to reviewers during the review process in order to foster reproducibility. The Copernicus review system allows to define such assets as 'access limited to reviewers' and reviewers must then sign that they will use such data only for the purpose of reviewing without making copies, sharing, or reusing.

In rare cases where the data cannot be deposited publicly (e.g., because of commercial constraints), a detailed explanation of why this is the case is required. The data needed to replicate figures in a paper should in any case be publicly available, either in a public database (strongly recommended), or in a supplement to the paper.

Reply: Thank you for your valuable feedback. In response, we have made the following revisions:

1. Supplementary Materials

We have submitted the supplementary materials as required and ensured that all relevant content is clearly labeled. It appears that, perhaps due to a system error, the reviewer may not have been able to access these materials. To resolve this issue, we have added the supplementary materials at the end of the document with all references clearly indicated. We will verify the completeness of these materials in our submission of revised version. We kindly invite you to review them and welcome any further comments

2. Results Section Length

Thank you for highlighting the issue of excessive detail in the results section. We have carefully

revised this section to eliminate repetitive descriptions of secondary information and city-specific data. Specifically, in Section 3.2, we have streamlined the content by summarizing key trends in a more concise format, particularly focusing on the differences between inland and coastal regions. In Section 3.4, we have added detailed comparisons and summaries of model simulations and isotope data for Beijing and Qingdao. Additionally, in Section 3.6, we have placed greater emphasis on the comparative analysis of emission reductions between Beijing and Qingdao, ensuring a clearer and more focused narrative.

3. Number of Charts

Your suggestion regarding the number of charts is greatly appreciated. To streamline the presentation and enhance data comparability, we have merged charts with similar themes. The chemical differences in air composition between 2013 and 2018, including the GR, HONO, N₂O₅, and NO_x concentrations over the North China Plain during winter, have been merged into a single figure (Figure 11).

4. Data Availability Statement

Thank you for your valuable feedback. We recognize that the initial statement may have lacked sufficient detail. We have updated the data availability statement to provide more comprehensive information on how to access the data. All datasets supporting the findings of this study are available through the following channels. Primary datasets and analysis results are available from the corresponding authors upon reasonable request. All plotting data and essential research data have been deposited in a publicly accessible repository on the Baidu Cloud (Data link: <https://pan.baidu.com/s/153rcdB-vTidH-14PPaXu-A>; Access code: egus).

Specific points:

71: Is this coastal or inland Greenland?

Reply: Thank you for your question. The Greenland research station mentioned in the paper is located at (72.6°N, 38.5°W) within Greenland's polar environment, and it is geographically closer to the coastal regions.

73: What is it about the air mass origin that affects the nitrate formation? Or why is this being set apart and discussed here after the review of the coastal vs. inland cities? Isn't air mass origin also the primary reason for those differences? The structure of the paragraph is just confusing me a little bit here.

Reply: Thank you for the question. Regarding the influence of air mass origin on nitrate formation, we have addressed this issue separately in the paper because the source of the air mass is a crucial factor driving the differences between coastal and inland cities, that impacts the underlying mechanisms, specifically the effects of pollutant composition and humidity on nitrate formation pathways.

Specifically, marine air masses are typically associated with higher humidity, lower NO_x mixing ratios, and longer NO₃ radical lifetimes, which significantly promote reactions via the hetN_2O_5 pathway. In contrast, continental air masses usually exhibit higher NO_x mixing ratios and NH₃ concentrations, which are more favorable for nitrate formation through the OH + NO₂ pathway. To better understand these regional differences, we have compared the nitrate formation mechanisms between coastal and inland cities and further investigated the influence of air mass origin in Section 3.5.3 (lines 608–611). This analysis provides a more comprehensive understanding of the underlying

driving mechanisms behind these regional disparities.

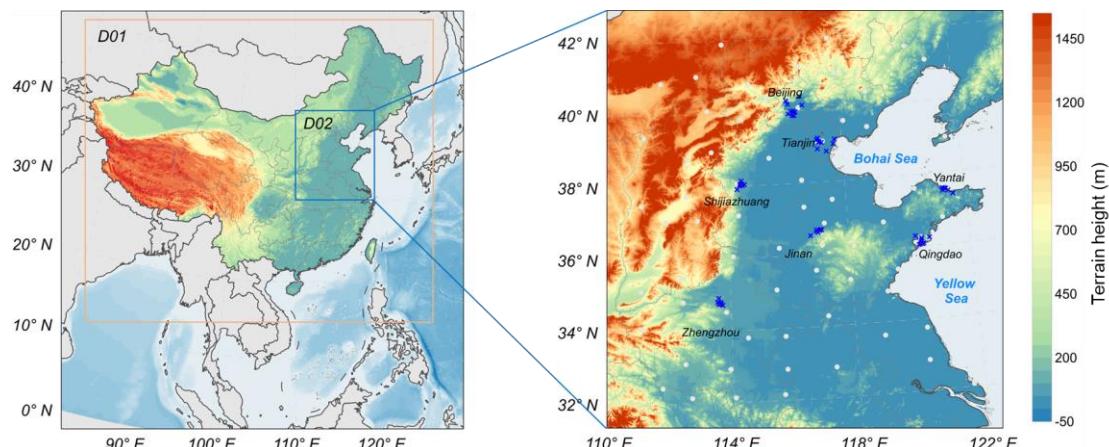
Fig. 2: Data source for terrain heights should be cited

Reply: Thank you for your suggestion. We have supplemented a citation for the terrain elevation data in the manuscript (lines 144–145). The data are sourced from the GEBCO Compilation Group (2024) GEBCO 2024 Grid (doi:10.5285/1c44ce99-0a0d-5f4f-e063-7086abc0ea0f).

165: Was there a specific data network that you were sourcing within that website? For example, that website is just a portal to access many different data networks, such as WMO and GHCN, and if you know the exact data source network, that could be cited here and be more clear.

Reply: We have clarified in the manuscript (line 146) that the terrain elevation data were obtained from the GEBCO website (https://www.gebco.net/data_and_products/gridded_bathymetry_data/) which provides access to the GEBCO 2024 grid dataset.

168: A little more information about these 68 stations would be beneficial, such as are they all within a specific region/geographic bounds? Were there any selection criteria applied to choose the stations?



Reply: These 68 observation stations were all located within the simulated D02 domain, which corresponded to the North China region. The selection of these stations was based on the availability of observational data. Specifically, all stations within this area that had observational data were included.

185: Just to confirm, are all the instrumentation specifics the same that you used here as in this cited paper? You might add a brief line or addition to the end of the sentence currently ending in “denitrifier method” to add the instrumentation used, so that the reader doesn’t have to go look that basic information up in another paper.

Reply: Thank you for your valuable feedback. We now specify that the instrument used was the Gasbench-IRMS (Delta V, Thermo Scientific), which is the same as the one employed in our previous research. This clarification was added at the end of the sentence (lines 210–212) in the revised manuscript.

Revised sentence: The $\delta^{18}\text{O}$ and $\delta^{15}\text{N}$ values of NO_3^- in the TSP samples were determined via the bacterial denitrifier method (Casciotti et al., 2002; Sigman et al., 2001) with the Gasbench-IRMS system (Delta V model, Thermo Scientific).

204: I think that some more information needs to be given here on how you used these indicators to evaluate the simulation effect. You have cited some proposed benchmarks, but it isn't clear to me readily how you will be using this information in your paper. In a very soon following section (3.1) about model evaluation where you present simulated values and some of the benchmarks, I was able to eventually infer how you were doing the evaluation, but it should really be more explicitly clear in the methodology.

Reply: Thank you for your comment. We have included several evaluation metrics and benchmarks in the methodology section (Table 1 and Table 2); however, due to the extensive number of evaluation formulas, we have provided a comprehensive description of the calculation methods and formulas in the supporting information. You can find these details in Table S3 of the supporting information.

220: I'm unclear exactly how the numbers being discussed here from the 68 sites were gathered and compared. Are these pairwise calculations, or overall means, or involving some sort of spatial dimension, etc? Are the comparisons all at hourly resolution, or aggregated to daily or something else? There needs to be more clarity on this, likely in the methodology of the 2.5 section. Also, how did you evaluate parameters that lacked cited benchmarks (perhaps something else that could be included in the methodology?)? For example, some of the Pearson correlation coefficients are somewhat low, for wind especially.

Reply: Thank you for your questions. For the data collected from the 68 observation stations, comparisons were conducted at on an hourly basis. At each station, the observed value was compared with the simulated value from the corresponding model grid cell on a point-by-point, and hour-by-hour basis. These paired comparisons were then aggregated across all 68 stations, and statistical evaluation metrics (e.g., MB, RMSE, and IOA) were generated from the entire dataset to provide an overall assessment of the model's performance.

To make these comparisons, we used the grid cell corresponding to the geographic coordinates of each observation point. This process ensured precise spatial and temporal alignment between the model simulation data and the observational data. Both data processing and comparisons were conducted on an hourly basis, without any daily averaging or other forms of temporal aggregation. Further details on this methodology can be found in Section 2.5 (lines 249–254) of the revised manuscript.

Model evaluation is both a critical and challenging part of model research. The current evaluation framework is based on findings from numerous studies. The relevant literature indicates that the current evaluation metrics are sufficient for analyzing model performance. We aim to further supplement and refine our model evaluation process through these calculations. Regarding the missing cited benchmarks, commonly used methods in the literature, such as those for wind speed and wind direction, were adopted, and we acknowledged that the observed underestimation is a common phenomenon in the model. For instance, the calculation of the Pearson correlation coefficient is intended to assist in assessing model performance. Although some parameters, particularly the correlation coefficient for wind speed, yield lower values, this phenomenon is consistent with the well-documented tendency of the WRF model to overestimate wind speed, which is an issue that is frequently reported in the literature (Tan et al., 2017; Jacobson and Kaufman, 2006). Moreover, evaluating wind direction data is complicated by its cyclic nature (since 360° and 0° represent the same direction), which may lead to some deviations in the correlation and error

metrics.

Tan, J., Zhang, Y., Ma, W., Yu, Q., Wang, Q., Fu, Q., Zhou, B., Chen, J., and Chen, L.: Evaluation and potential improvements of WRF/CMAQ in simulating multi-levels air pollution in megacity Shanghai, China, *Stochastic Environmental Research and Risk Assessment*, 31, 2513-2526, doi:10.1007/s00477-016-1342-3, 2017.

Jacobson, M. Z. and Kaufman, Y. J.: Wind reduction by aerosol particles, *Geophysical Research Letters*, 33, doi:10.1029/2006GL027838, 2006.

Section 3.4: The model has output for “Others” but your isotopic method doesn’t. However, I don’t see any discussion of this in this section, but I feel it needs addressed in some form. If 5-8% of reactions are “others” in the model, but you don’t distinguish those in the isotopic method, does that mean that you assume you are attributing those “others” reactions to either OH+NO₂ or hetN₂O₅? Is that baked into the uncertainties in any way, or handled specifically?

Reply:

Thank you for your valuable comments. We apologize for the lack of detailed information on our methods, which may have caused confusion.

The CMAQ model took a comprehensive approach by including the contributions of 5-8% of “other” pathways, which was its main advantage. Conversely in our isotopic calculations, we focused only on the ·OH+NO₂ and hetN₂O₅ pathways because the $\delta^{18}\text{O}$ end-member values for multiple reaction pathways were quite similar. Hence, those 5–8% of “other” pathways were attributed to the ·OH+NO₂ and hetN₂O₅ pathways in our isotope calculations, potentially resulting in an overestimation of these two pathways compared to the CMAQ model. Notably, that the uncertainty in the pathway contribution calculations based on stable isotopes was approximately 10–30%. Nevertheless, as a receptor model, the isotopic method, based on observational data, provided robust evidence that supported the general patterns revealed by CMAQ (Figure R1). Additionally, we supplemented isotope calculation uncertainties in the manuscript and added a detailed comparison and discussion of both isotopic and CMAQ model results in the relevant section. A detailed explanation follows.

In our previous research, we compiled theoretical calculations of the thermodynamic fractionation of $\delta^{18}\text{O}$ for the commonly considered atmospheric NO₃[−] formation pathways (Table R1), and P7, P8, and P9 were considered as the “other” reactions (Luo et al., 2020). However, due to the end-member values of $\delta^{18}\text{O}$ being too close to those of hetN₂O₅, such as in the calculations for the winter of 2018 in Qingdao, the end-member $\delta^{18}\text{O}$ value for the hetN₂O₅ pathway was $102.6 \pm 4.2 \text{ ‰}$. Conversely, the end-member values for P7, P8, and P9 were $105.8 \pm 4.8 \text{ ‰}$, $126.4 \pm 4.8 \text{ ‰}$, and $135.0 \pm 4.8 \text{ ‰}$, respectively. This consideration was immature in actual pathway calculations; in more extensive research (Table R2), researchers primarily consider ·OH+NO₂ and hetN₂O₅ the two main formation pathways when calculating atmospheric NO₃[−] formation.

In our isotope analysis, we employed the Stable Isotope Mixing Model in R (SIAR) to determine the relative contributions of different formation pathways to atmospheric NO₃[−]. The main sources of uncertainty stem from the standard deviations (SD) in each pathway’s end-member calculations and the posterior probability distributions generated by the model. As mentioned before, focusing on the most dominant pathways, rather than multiple pathways (Zhang et al., 2021; Luo et al., 2022), enhances the model’s reliability and reduces overall uncertainty. The posterior probability for each

pathway's contribution is obtained using the Hilborn sampling-importance-resampling method. For detailed information about the model framework and computational methods, please refer to Moore and Semmens (2008). We have added an explanation of model uncertainties in the Methods section (lines 237–239) and in Section 3.4 (lines (413–430) of the manuscript.

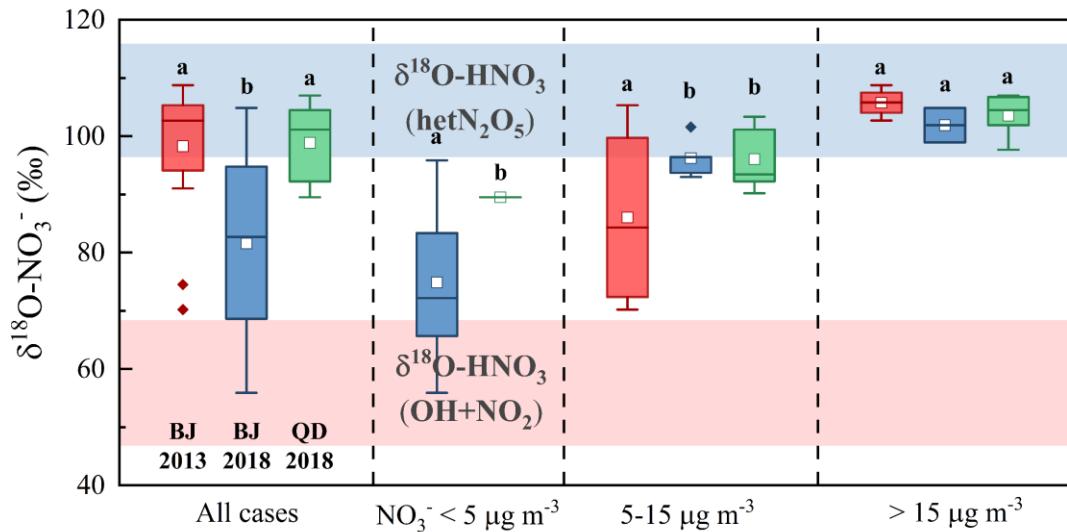


Figure R1. Boxplot of $\delta^{18}\text{O}$ of atmospheric NO_3^- collected in Beijing in the winters of 2013 and 2018 and in Qingdao in the winter of 2018 under different nitrate conditions. The shadows of red and blue indicate the ranges of $\delta^{18}\text{O}-\text{NO}_3^-$ generated via the daytime and nocturnal pathways, respectively. Categories that share common letters do not differ in significance, which is set to 0.05.

Table R1. Calculated $\delta^{18}\text{O}$ values of NOy for each nitrate production pathway (Luo et al., 2020).

Pathway	Expression
R1	$\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$
R2	$\text{NO} + \text{RO}_2/\text{HO}_2 \rightarrow \text{NO}_2 + \text{O}_2$
R3	$\text{NO}_2 + \text{O}_3 \rightarrow \text{NO}_3$
R4	$\text{NO}_2 + \text{NO}_3 \rightarrow \text{N}_2\text{O}_5$
R5	$\text{NO}_2 + \text{OH} \rightarrow \text{HNO}_3$
R6	$\text{N}_2\text{O}_5 + \text{H}_2\text{O} \rightarrow \text{HNO}_3$
R7	$\text{NO}_3 + \text{HC/DMS} \rightarrow \text{HNO}_3$
R8	$\text{N}_2\text{O}_5 + \text{Cl}^- \rightarrow \text{pNO}_3^-$
R9	$\text{ClNO}_3 + \text{H}_2\text{O} \rightarrow \text{HNO}_3$
R10	$\text{NO}_2 + \text{H}_2\text{O} \rightarrow \text{HNO}_3$

Table R2. Comparison of the contributions of major atmospheric NO_3^- formation pathways in China

Region	City	Lon (°E)	Lat (°N)	Method	Year	Season	$\cdot\text{OH}+\text{NO}_2$	hetN_2O_5 (%)	$\text{NO}_3\cdot+\text{HC}$	Reference
NCP	Zibo	118.0547	36.8258	$\Delta^{17}\text{O}$	2022-2023	Winter	17.7	32.6	49.7	Feng et al., 2023
	Zhoukou	114.6464	33.6033				53.4	31.1	15.5	
	Beijing	117.1231	39.0601	$\Delta^{17}\text{O}$	2021	Winter	18.0	29.1	52.9	Yan et al., 2023
						Spring	26.9	35.2	37.9	
	Beijing	116.3660	39.9746	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	17.4	61.9	20.7	Luo et al., 2023
	Tianjin	117.4177	39.5557	$\Delta^{17}\text{O}$		Winter	29.7	34.8	35.5	
				2022	Spring	51.0	31.9	17.0	Zhang et al., 2024	
					Summer	41.8	33.6	24.6		
					Autumn	13.2	34.7	52.1		
	Shijiazhuang	114.4942	38.0980	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	6.3	66.0	27.7	Luo et al., 2023
	Tianjin	117.3350	38.9944	$\delta^{18}\text{O}$		Winter	31.4	68.6		
				2019-2020	Spring	57.4	42.6		Zhang et al., 2022	
					Summer	84.1	15.9			
	Jiaozuo	113.2606	35.1865	$\delta^{18}\text{O}$	2020	Winter	30.0	70.0		Li et al., 2022a
						Summer	61.0	39.0		
	Beijing	116.3711	39.9744	$\Delta^{17}\text{O}$	2016 2017	Winter	42.6	25.4	31.7	Fan et al., 2022
	Summer	46.2	23.0	31.4						
	Beijing	116.4167	40.0333				67.4	32.6		
	Tianjin	117.1500	39.0833	$\delta^{18}\text{O}$	2017-2018	Winter	58.1	41.9		Zhang et al., 2021a
	Shijiazhuang	114.6377	38.0128				50.5	49.5		

Jinan	117.0500	36.6667				50.6	49.4		
Jiaozuo	113.2667	35.1833				50.7	49.3		
Beijing	117.7000	40.0667	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	45.3	46.5	8.2	Zhang et al., 2021b
					Autumn	51.8	48.2		
Shijiazhuang	114.6377	38.0128	$\delta^{18}\text{O}$	2017-2018	Winter	22.5	77.5		Luo et al., 2021
					Spring	51.5	48.5		
					Summer	71.1	28.9		
Beijing	116.3400	39.9300	$\delta^{18}\text{O}$	2013-2014	Spring	18.8	81.2		
					Summer	41.2	58.8		Zong et al., 2020
					Autumn	17.9	82.1		
					Winter	17.3	82.7		
Beijing	116.3667	39.9667	$\delta^{18}\text{O}$	2018	Winter	52.0	48.0		Fan et al., 2020
Beijing	116.4167	40.0333	$\delta^{18}\text{O}$	2017-2018	Winter	48.0	52.0		Zhang et al., 2020a
Beijing	116.3800	39.9800	$\delta^{18}\text{O}$	2013	Spring	56.5	27.2	16.3	Luo et al., 2020a
Jinan	117.0500	36.6667	$\delta^{18}\text{O}$	2018	Summer	39.1	60.9		Zhang et al., 2020b
Beijing	116.7000	40.0667	$\Delta^{17}\text{O}$	2015	Winter	29.5	35.1	35.6	Song et al., 2020
Beijing	116.4167	40.0333	$\delta^{18}\text{O}$	2017-2018	Winter	47.0	53.0		Zhang et al., 2020c
Beijing	116.7000	40.0667	$\Delta^{17}\text{O}$	2014	Spring	27.0	35.0	38.0	
					Summer	40.9	36.1	32.9	
					Autumn	25.0	35.0	40.0	Wang et al., 2019
					Winter	31.0	34.0	34.0	
Beijing	116.6800	40.4100	$\Delta^{17}\text{O}$	2014-2015	Winter	23.5		76.5	He et al., 2018
Dongying	118.9833	37.7500	$\delta^{18}\text{O}$	2013	Summer	47.4	52.6		Zong et al., 2018
Central	Wuhan	114.3000	30.5300	$\delta^{18}\text{O}$	2018-2019	Spring	35.0	65.0	
					Summer	69.7	30.3		Deng et al., 2024
					Autumn	34.5	65.5		

YRD	Shiyan	111.4000	33.2000	$\delta^{18}\text{O}$	2021	Winter	7.8	92.2		
						Spring	64.0	19.0	17.0	
						Summer	84.0	8.0	8.0	
						Autumn	64.0	19.0	18.0	
						Winter	25.0	38.0	37.0	
	Wuhan	104.3600	30.5200	$\delta^{18}\text{O}$	2013-2014	Spring	17.9	82.1		
						Summer	60.4	39.6		
						Autumn	28.5	71.5	Zong et al., 2020	
						Winter	11.2	88.8		
	Lanzhou	103.8551	36.0305	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	22.2	57.6	20.4	Luo et al., 2023
	Shanghai	121.4279	31.2595	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	18.3	47.9	33.8	Luo et al., 2023
	Shanghai	121.5000	31.3000	$\delta^{18}\text{O}$	2019	Summer	70.5	29.5		
	Shanghai	121.5100	31.3400	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2018	Winter	16.7	28.3	55.0	Zhu et al., 2021
	Shanghai	121.5700	31.2600	$\Delta^{17}\text{O}$	2016	Summer	56.1	24.8	19.1	
	Shanghai	121.5000	31.2900	$\delta^{18}\text{O}$	2013-2014	Winter	63.8	36.2		
						Spring	62.7	37.3		He et al., 2020
						Summer	32.6	67.4		
						Autumn	60.0	40.0		Zong et al., 2020
	Nanjing	118.7000	32.2000	$\Delta^{17}\text{O}$	2018	Winter	27.4	34.0	47.1	
						Summer	18.9	34.0		
						Autumn	24.1	34.8	41.1	
						Spring	18.3	35.0	46.7	Fan et al., 2023
						Summer	40.5	35.2	24.3	

	Xiamen	118.0900	24.4360	$\delta^{18}\text{O}$	2019-2021	Cold Warm	20.2 30.3	38.2 31.5	21.6 18.3	Li et al., 2022b	
	Nanchang	115.8085	28.6832	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	17.2	43.6	39.2	Luo et al., 2023	
	Nanchang	115.9333	28.6833	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	30.4	36.8	32.8	Zhang et al., 2021c	
Southeast	Nanchang	115.8085	28.6832	$\delta^{18}\text{O}$	2017-2018	Autumn	18.0	38.6	43.4		
						Winter	7.5	33.8	58.7	Luo et al., 2020b	
						Spring	33.2	34.5	32.3		
						Summer	58.7	28.1	13.2		
	Nanchang	115.9000	28.7000	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017	Autumn	37.1	60.3	2.6	Xiao et al., 2020	
	Ganzhou	114.7600	25.6600	$\delta^{18}\text{O}$	2019	Winter Summer	41.6 73.5	29.5 14.1	28.9 12.4	Cheng et al., 2022	
	Chengdu	104.0527	30.5598	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	23.5	40.8	35.7	Luo et al., 2023	
	Chengdu	104.3800	30.6400	$\delta^{18}\text{O}$	2013-2014	Spring	46.5	53.5			
						Summer	61.8	38.2		Zong et al., 2020	
						Autumn	29.1	70.9			
						Winter	11.3	88.7			
Southwest	Kunming	102.7000	25.0667	$\delta^{18}\text{O}$	2017-2018	Autumn	85.0	15.0			
						Winter	74.4	25.6		Guo et al., 2021	
	Nanning	108.2833	22.8333			Autumn	87.9	12.1			
						Winter	68.4	31.6			
	Guiyang	106.7167	26.5667	$\delta^{18}\text{O}$	2013-2014 2016	Winter	38.3	61.7		Li et al., 2021a	
						Summer	80.2	19.8			
	Nanning	108.3192	22.7990	$\delta^{15}\text{N}$ and $\delta^{18}\text{O}$	2017-2018	Winter	47.5	27.8	24.7	Luo et al., 2023	
						Spring	70.4	22.8	6.8		
						Summer	85.6	10.9	3.5		
PRD	Guangzhou	113.3689	23.1938	$\delta^{18}\text{O}$	2015-2018	Autumn	86.1	10.6	3.3	Xi et al., 2023	

						Winter	72.0	21.0	7.1
Guangzhou	113.3397	23.1075	$\Delta^{17}\text{O}$	2018	Autumn	61.0	12.0	27.0	Wang et al., 2023
Guangzhou	113.3600	23.1500	$\delta^{18}\text{O}$	2013-2014	Spring	48.0	52.0		
					Summer	66.9	33.1		Zong et al., 2020
					Autumn	33.9	66.1		
					Winter	9.7	90.3		
Harbin	126.6333	45.7469	$\Delta^{17}\text{O}$	2022-2023	Winter	55.3	28.2	16.5	Feng et al., 2023
Shenyang	123.4300	41.7700			Spring	65.8	25.6	8.6	
					Summer	95.3	3.0	1.7	
					Autumn	83.0	12.1	4.8	
			$\Delta^{17}\text{O}$	2015-2018	Winter	57.7	30.9	11.4	Li et al., 2022c
Fushun	124.9400	41.8500			Spring	40.0	34.5	25.5	
					Summer	85.7	9.6	4.7	
					Autumn	46.0	35.9	18.1	
					Winter	37.6	33.2	29.2	
Northeast	Harbin	126.7400	45.7300			69.4	30.6		
	Changchun	125.4000	43.8500	$\delta^{18}\text{O}$	2017-2018	50.2	49.8		Zhao et al., 2021
	Harbin	126.5300	45.8400		Winter	50.1	49.9		
	Yushu	126.5300	44.8600			37.3	62.7		
	Harbin	120.6817	45.7539	$\delta^{18}\text{O}$	2017-2018	Spring	46.4	53.6	
					Summer	68.7	31.2		Sun et al., 2020
					Autumn	56.1	43.9		
					Winter	44.8	55.2		
	Changchun	125.4000	44.0000	$\delta^{18}\text{O}$	2017-2018	Summer	79.7	20.3	
					Autumn	56.1	43.9		Zhao et al., 2020
					Winter	55.9	44.1		

							Spring	58.0	42.0	
Island	Beihuangcheng Island	120.9167	36.4000	$\delta^{18}\text{O}$	2015		Autumn	35.0	65.0	
							Winter	24.0	76.0	Zong et al., 2017
							Spring	47.0	53.0	
							Summer	68.0	32.0	
							Dongsha Island	66.8	10.2	Yang et al., 2023
	Pengjiayu Island	116.7167	20.7000	$\delta^{18}\text{O}$	2013	Spring	62.1	23.4	14.5	
		122.1333	26.0500							
Marine	Bohai Sea and Yellow Sea		32 - 34 34 - 36 36 - 38 38 - 40	$\Delta^{17}\text{O}$	2018	Spring	60.0	24.0	16.0	
							49.0	31.0	19.0	Zhao et al., 2024
							26.0	35.0	39.0	
							14.0	34.0	52.0	
							2014	Summer	65.9	34.1
NCP	Bohai Sea		38 - 40	$\delta^{18}\text{O}$	2017	Summer	43.9	56.1		
							2016	Winter	49.3	50.7
							2017	Summer	41.1	58.9
							2018	Winter	73.6	Zong et al., 2022
							2019	Summer	26.4	
					2018	Summer	73.6	26.4		
							2019	Winter	26.4	Zong et al., 2022
							2019	Summer	61.0	
							2019	Winter	32.0	
							2019	Summer	61.7	39.0
NCP	Beijing	116.4125	40.2352		2013		2019	Winter	32.0	Liu et al., under review
							2013	Summer	67.8	
							2018	Winter	77.5	
							2018	Summer	19.7	
							2013	Winter	15.1	
NCP	Tianjin	117.4812	39.0929	CMAQ	2013	Winter	2013	Summer	73.8	Liu et al., under review
							2018	Winter	14.2	
NCP	Shijiazhuang	114.5562	37.8247		2013		2013	Summer	73.8	Liu et al., under review
							2018	Winter	14.2	

				2018	83.6	8.3	
Jinan	116.9187	36.8712		2013	63.7	27.7	
				2018	72.9	19.9	
				2013	77.6	10.6	
Zhengzhou	113.8719	34.7968		2018	79.2	12.7	
				2013	51.5	41.5	
				2018	56.5	37.0	
Qingdao	120.5000	36.1533		2013	48.2	45.7	
				2018	50.4	43.5	
				2021	Winter	51.0	45.0
NCP	115.4817	36.6111	WRF-Chem	2017	Winter	48.0	30.3
NCP	114.8000	39.5000	WRF-Chem	2021	Winter	69.3	28.4
Shanghai	120.8662	28.0818		Spring	81.8	15.3	
				Summer	82.9	12.2	
				Autumn	86.9	11.1	
				Winter	59.2	36.1	
Nanjing	118.6491	28.7744		Spring	73.1	5.4	
				Summer	74.7	17.9	
				Autumn	69.7	25.4	Sun et al., 2022
YRD			CMAQ	2017	Winter	66.9	27.1
				Spring	78.5	16.5	
				Summer	81.7	10.4	
				Autumn	72.5	21.8	
Hefei	117.3881	28.5389		Winter	68.9	26.8	
				Spring	74.9	20.9	
				Summer	78.7	14.3	
Changzhou	119.6087	28.4776		Winter	68.9	26.8	
				Spring	74.9	20.9	
				Summer	78.7	14.3	

						Autumn	77.6	18.3	
						Winter	59.7	35.5	
						Spring	70.5	23.3	
						Summer	76.4	10.7	
						Autumn	73.8	21.3	
YRD	Hangzhou	119.7671	27.1448						
YRD	Shanghai	120.9890 121.5330	31.0970 31.2280	F0AM	2019	Winter	42.9 36.8	55.5 62.1	Zang et al., 2022
NCP		114.8000	39.5000			Summer	60.4	39.6	
YRD		121.0000	31.1000	WRF-Chem	2016	Winter	91.9	8.1	Li et al., 2021b
NCP		114.8000	39.5000	WRF-Chem	2016	Summer	75.3	24.7	
Taiwan		120.6753	21.1892	WRF-CMAQ PA	2017	Winter	86.2	13.8	
PRD		113.3600	23.1500	WRF-CMAQ PA	2015	Spring	53.4	46.6	Liu et al., 2020
Central	Xi'an	108.9552	34.2919	WRF-Chem	2017	Winter	> 45.0	30.0	Chuang et al., 2022
NCP	Beijing	116.6800	40.4100	GEOS-Chem	2014-2015	Winter	47.0	34.0	Qu et al., 2021
NCP		114.8000	39.5000	WRF-CMAQ PA	2017	Winter	34.4	44.9	Chan et al., 2021
NCP	Beijing	116.6800	40.4100	Box model	2016-2017	Winter	43.0	44.0	Fu et al., 2020
YRD	Nanjing	116.6800	40.4100	Box model	2015	Winter	68.8	31.2	Chen et al., 2020
NCP	Beijing	118.6491	28.7744	CMAQ	2017	Winter	80		Sun et al., 2018
NCP	Beijing	116.6800	40.4100		2017	Winter	42		Qiu et al., 2019
NCP		114.8000	39.5000	WRF-Chem	2015	Winter	30.1		Liu et al., 2019
PRD		113.3600	23.1500	WRF-Chem	2013	Winter	57.4		Li et al., 2016

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Technical points:

45 : Do you mean “adsorbed” here rather than “absorbed”? The use of “onto” makes it seem like you might be referring to adsorption rather than absorption.

Reply: Thank you for the suggestion. We have replaced “absorbed” with “adsorbed” in line 47.

65: This paragraph is excessively long and should be broken up by paragraph breaks to aid readability.

Reply: Thank you for the suggestion. We have restructured the paragraph to improve the readability by splitting it into two separate sections, which can now be found in lines 59–80 and lines 105–124 of the revised manuscript.

133: I think the use of a colon (:) here is more appropriate than “i.e.”

Reply: Thank you for your suggestion. We have revised the text by replacing “i.e.” with a colon (:) to improve clarity, as recommended. This change has been made in line 176.

182: Perhaps refer to it as the “bacterial denitrifier method” just to be explicitly clear.

Reply: You are correct. This method employs the *Pseudomonas aureofaciens* (ATCC13985) strain to completely reduce NO_3^- to N_2O gas for isotope analysis. We have revised the text accordingly, and the change can be found in lines 206–209.

Revised sentence: The $\delta^{18}\text{O}$ and $\delta^{15}\text{N}$ values of NO_3^- in the TSP samples were determined via the bacterial denitrifier method (Casciotti et al., 2002; Sigman et al., 2001) with the Gasbench-IRMS system (Delta V model, Thermo Scientific).

Figure 3: The star symbol is used four times in total, but I’m guess you are only referring to the two times it is used for the R value? Perhaps just state in the caption that the R value is significant at a $p < 0.05$ level.

Reply: Thank you for your suggestion. We have revised the figure caption to clarify the use of the star symbol. The figure now states that the * indicates R values significant at the $p < 0.05$ level. This change can be found in the caption of Figure 3 in the revised manuscript.

Figure 4: Maybe consider putting a larger label on the vertical left side for $\text{PM}_{2.5}$, NO_3^- , NH_4^+ , and SO_4^{2-} to make it more clear what each row of data is representing.

Reply: Thank you for your suggestion. We have updated Figure 4 by adding larger labels on the left side to clearly indicate $\text{PM}_{2.5}$, NO_3^- , NH_4^+ , and SO_4^{2-} for each row of data. This revision can be found in Figure 4 of the revised manuscript.

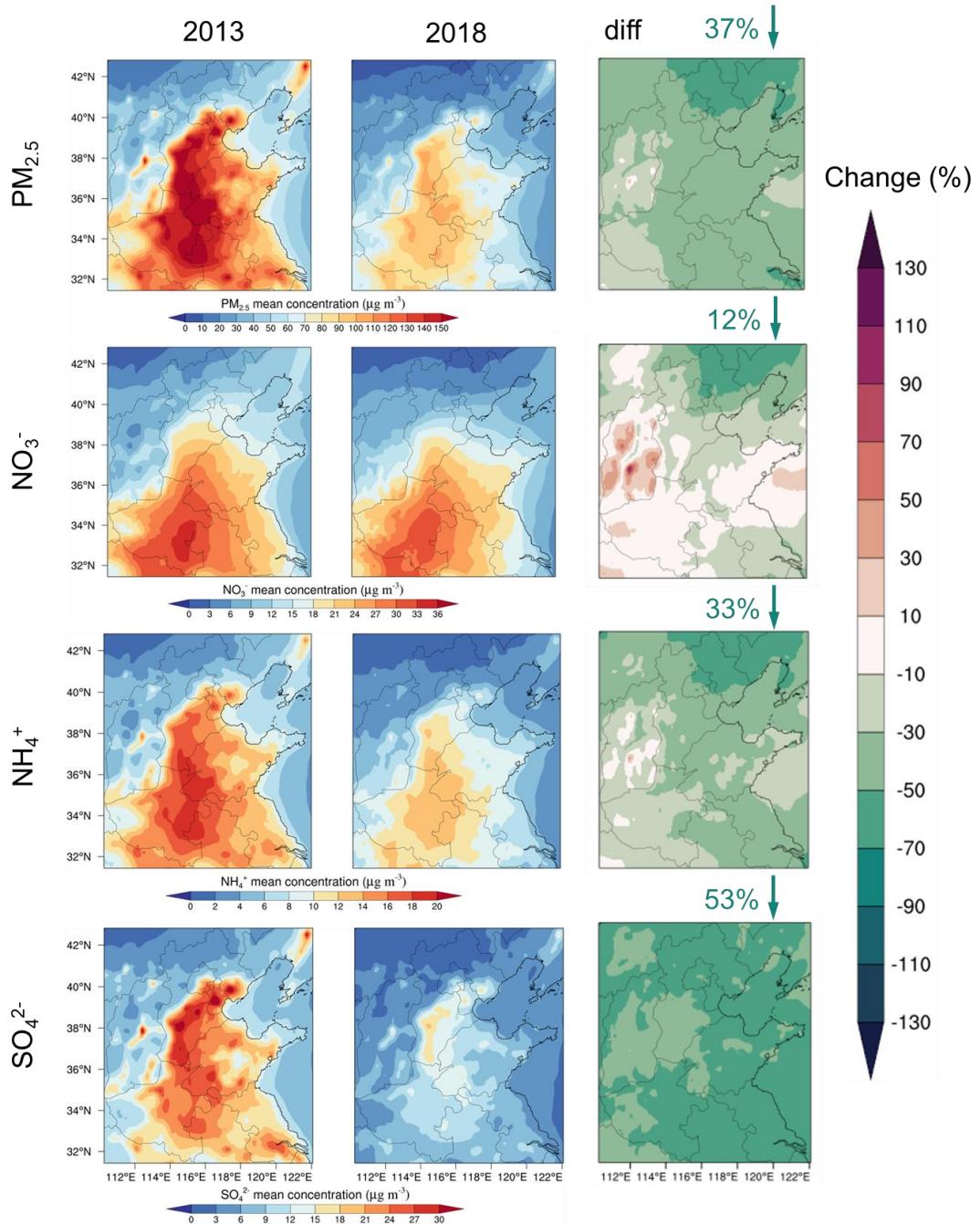


Figure 4 Spatial distribution and changes in $\text{PM}_{2.5}$ and its components in the NCP during the winters of 2013 and 2018. The up arrows indicate increases, and the down arrows indicate decreases.

Fig 5: Period missing at end of caption. This diverging color scheme is also a bit confusing as used here, because it is the same color scheme used in Fig 4 to show representative change (pos = red, neg = green), but here it is a unidirectional scale. I'd recommend a different color scheme to avoid confusion or unintentional misleading.

Reply: We are sorry for the confusion. We have revised Figure 5 by updating the color scheme to avoid confusion with Figure 4 and to better align it with its unidirectional scale. Additionally, we have added a period at the end of the title. This revision can be found in Figure 5 of the revised manuscript.

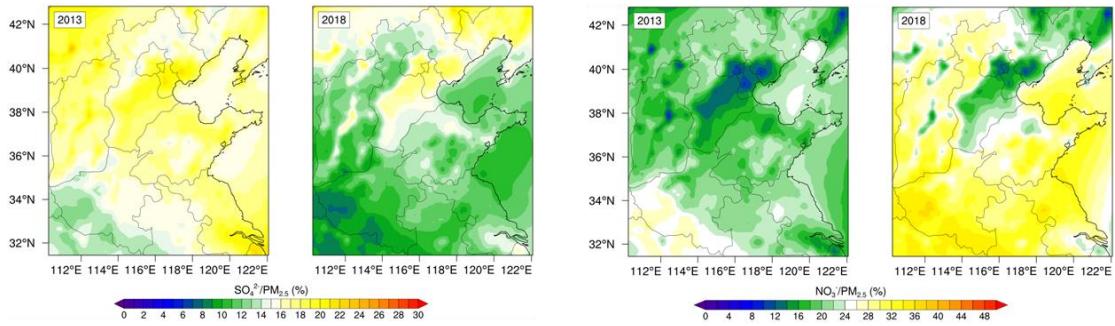


Figure 5 Spatial distributions of the nitrate and sulfate proportions in PM_{2.5} in the NCP region during the winters of 2013 and 2018.

Figure 6: Humans are pretty bad at estimating angular areas. You might consider alternatives such as treemaps or waffle charts. Not required from me, but just put here for consideration. This is also a pretty simple figure, and since you have so many figures, you might consider merging it with another or whether it is necessary.

Reply: Thank you for your valuable suggestion. We believe that alternative visualizations, such as treemaps or waffle charts, can indeed improve the clarity and readability of the figure. After further review, we find that Figure 6 contributes relatively little to the overall manuscript; therefore, we have removed it.

Fig 9: The color choices could be changed to improve the visual story. For example, the OH +NO₂ on both sides would ideally both be blue, or shades of blue. And hetN₂O₅ both be orange or shades of orange. That would make it more clear that we should be directly relating them.

Reply: Thank you for your insightful suggestion. We have revised the figure according to suggestion. This revision can be found in Figure 9 of the revised manuscript.

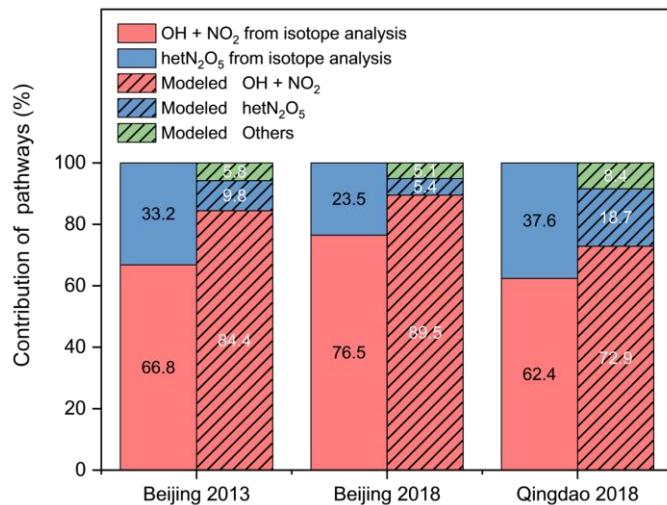
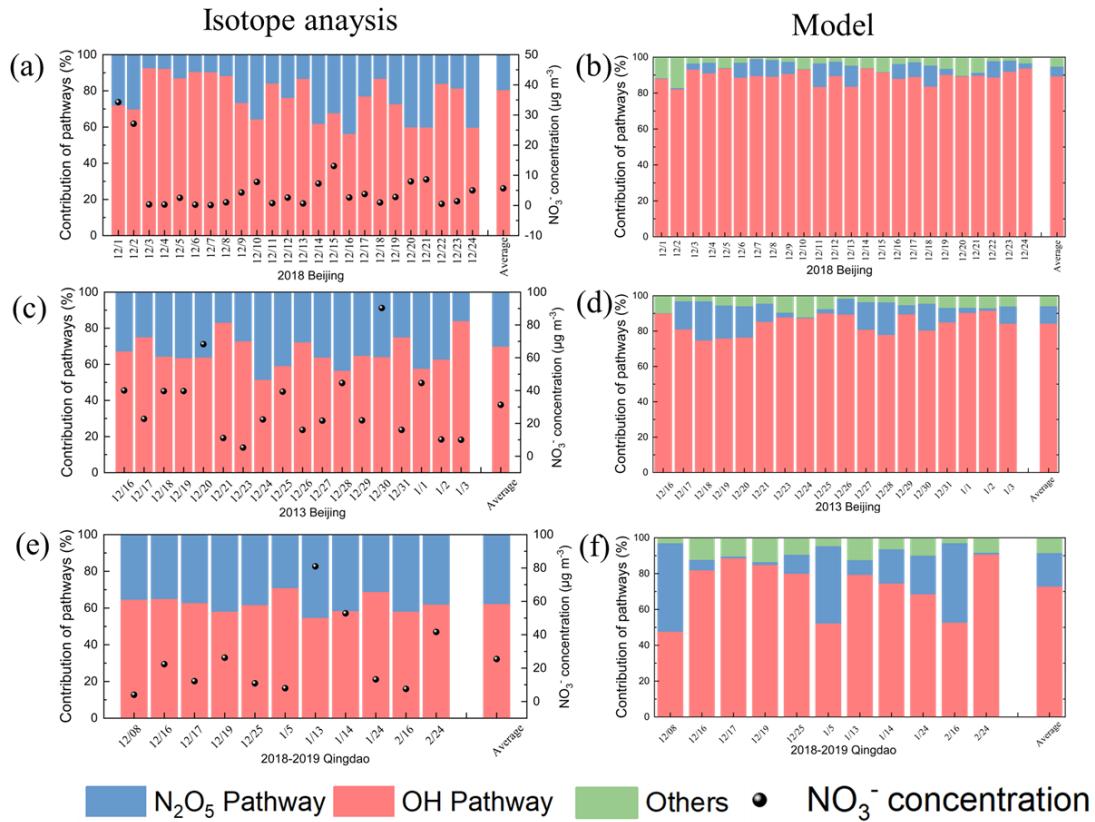


Figure 9 Comparison of the contributions of the atmospheric NO₃⁻ formation pathways based on the dual-isotope results and model simulations for Beijing in 2013 and 2018 and for Qingdao in 2018.

Fig 10: The legend for the dot looks like it is just connected to OH Pathway, and it should be labelled as NO_3^- concentration or $[\text{NO}_3^-]$ not just NO_3^- . Missing a period at end of caption. Subfigures should probably either be all in one column OR the 2018/19 Qingdao be under the 2018 Beijing chart.

Reply: Thank you for your valuable feedback. We have updated the legend to clearly label the dots as "NO₃⁻ concentration" and added a period at the end of the caption. Additionally, we have adjusted the layout by placing the 2018/19 Qingdao chart directly below the 2018 Beijing chart, ensuring a more logical and visually consistent arrangement. This revision can be found in Figure 8 of the revised manuscript.



432: There is a comma splice in this sentence.

Reply: We have revised the sentence to correct this issue. In addition, we have checked the manuscript carefully to ensure proper sentence structure and improve readability in lines 518–520 of the revised version. We appreciate your careful review and valuable feedback.

Revised sentence: This hypothesis could be verified from an emission perspective. The total ammonia emissions in China increased from 9.64 to 9.75 Tg from 2013–2015 and then gradually decreased to 9.12 Tg by 2018 (Liao et al., 2022).

Fig 13: The scaling seems poor or wrong in the difference map. The HONO concentrations only cover <2 ppb but the scaling on the difference is ± 50 .

Reply: We apologize for the confusion. The differential scale represents the percentage change in HONO concentration in 2018 relative to 2013. For clarity, we have adjusted the scale to $\pm 100\%$ to more accurately depict the range of HONO concentration variations. This revision has been implemented in Figure 11 of the revised manuscript.

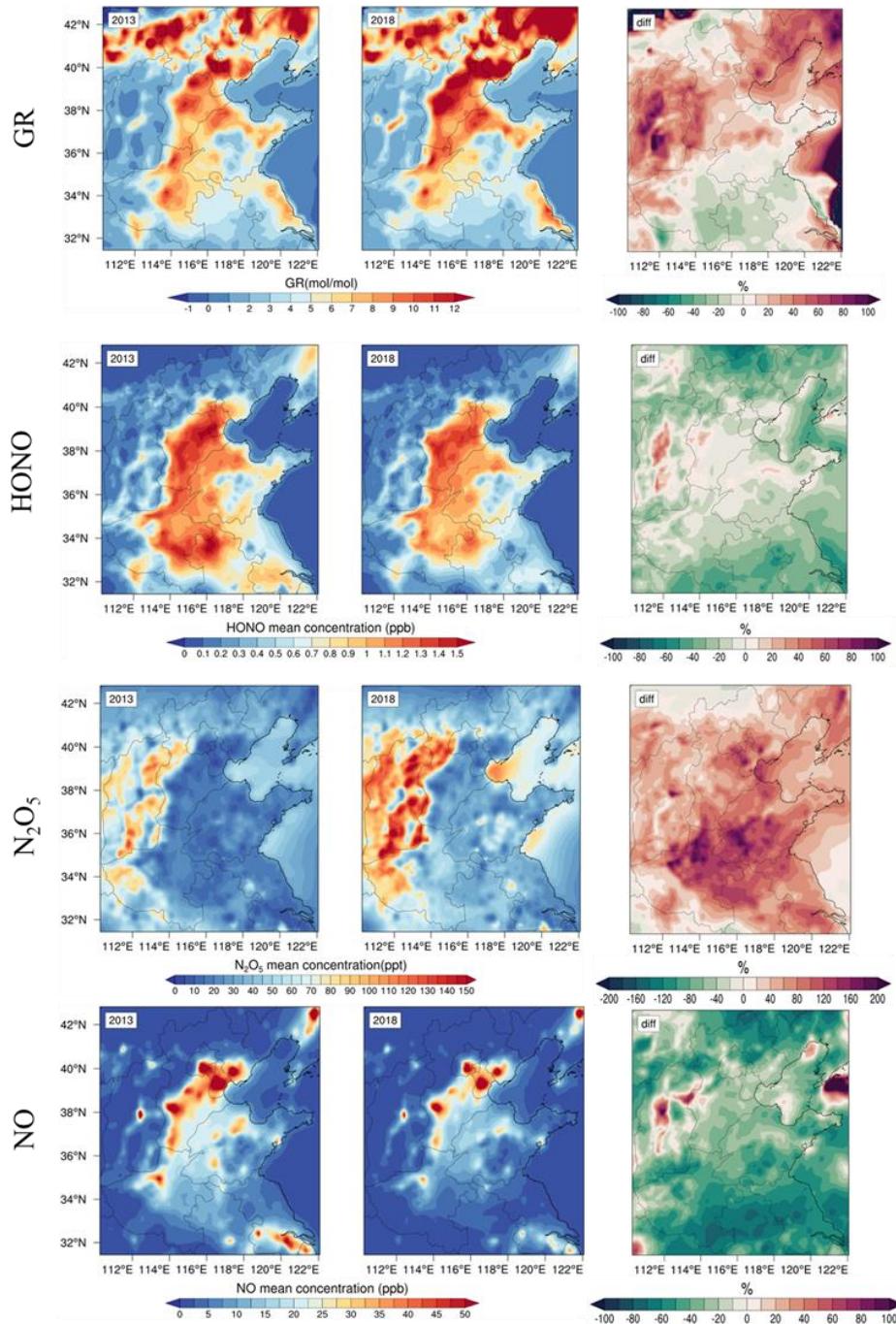


Figure 11 Spatial distributions and interannual variations in the GR, HONO, N_2O_5 , and NO_x concentrations over the North China Plain during the winters of 2013 and 2018. The percentage changes (diff) represent the relative differences between 2018 and 2013.

Supporting Information

Enhanced Atmospheric Oxidation and Particle Reductions Driving Changes to Nitrate Formation Mechanisms across Coastal and Inland Regions of North China

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Text S1

Our isotope blank measurements followed the same procedure as the sample isotope analysis. Specifically, in the sample measurements, after purging with high-purity nitrogen, 20 nmol of nitrogen was added to the headspace vial containing the *Pseudomonas aureofaciens* (ATCC13985) strain. For the blank measurements, no sample was added, and after 24 hours, 10 M NaOH was directly injected to quench the reaction before the analysis. The peak area in the chromatogram represents the absolute amount of N₂O reduced by the strain, and the $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values correspond to the $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values of the sample. The peak area for the samples was around 10, while the peak areas for the two blank measurements were only 0.371 and 0.336, indicating an influence on the isotope values of less than 5%, which is negligible and thus not considered.

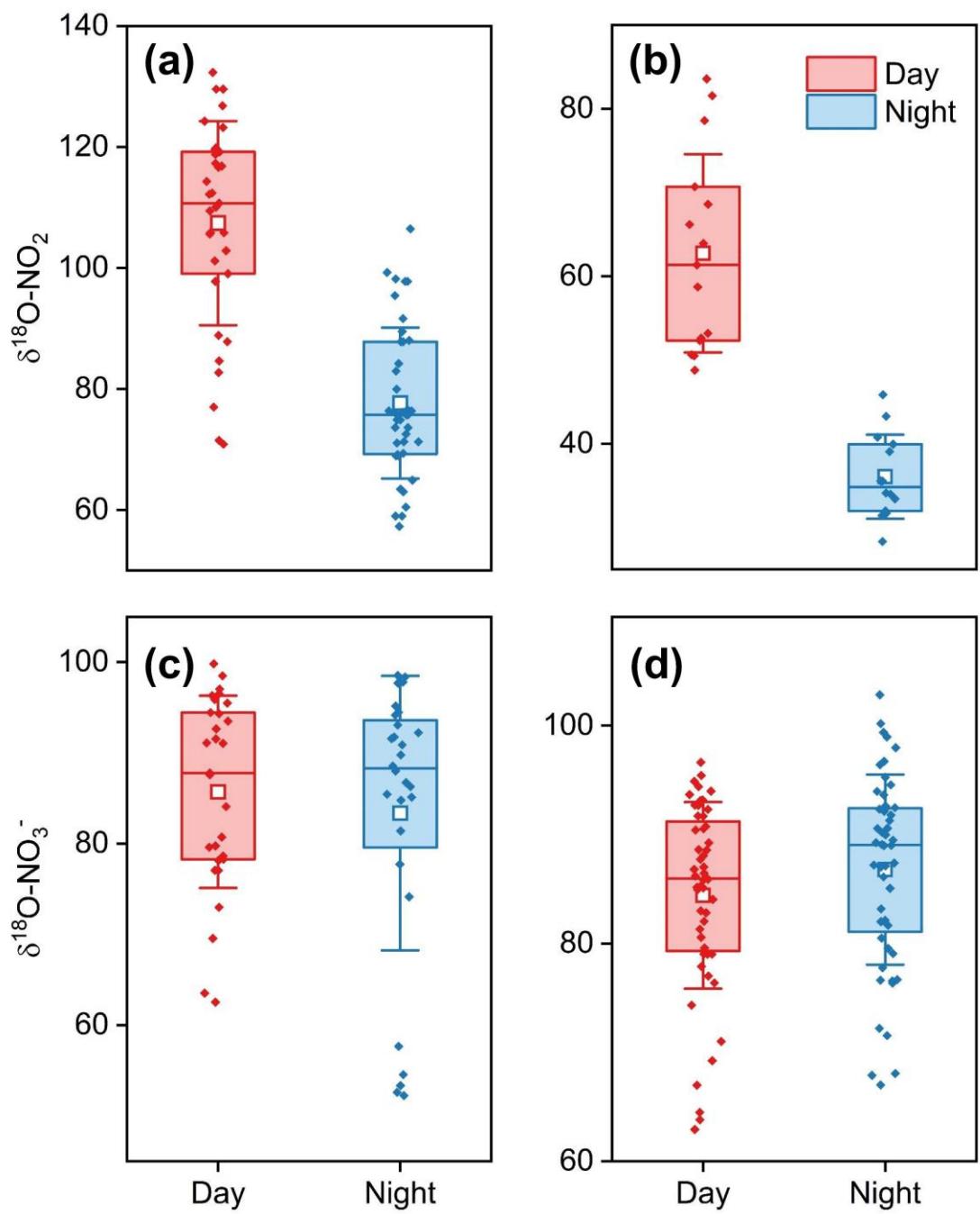


Figure S1 The diurnal values of atmospheric $\delta^{18}\text{O-NO}_2$ in Hefei winter (Zhang et al., 2025) (a) and in Nanchang summer (Cao, 2022) (b), and $\delta^{18}\text{O-NO}_3^-$ in Tianjin winter (Feng et al., 2020) (c) and in Nanjing winter (Zhang et al., 2022) (d).

Text S2

In most studies, the tropospheric $\delta^{15}\text{N-NOx}$ was often assumed as 0‰ following Walters and Michalski (2016), Luo et al. (2023) and Deng et al. (2024). In addition, the tropospheric $\delta^{18}\text{O-H}_2\text{O}_{(\text{g})}$ in Beijing in winter was determined as -27.9‰ in Wen et al. (2010), and in Qingdao, it was determined as -18.6‰ in Wang et al. (2022). The tropospheric $\delta^{18}\text{O-NOx}$ ranged from 112‰ to 122‰ (Michalski et al., 2014; Walters and Michalski, 2016). The f_{NO_2} values in Beijing and Qingdao were 0.655 (Luo et al., 2023) and 0.786 (Lian et al., 2022) in winter, respectively.

$$\begin{aligned}\delta^{15}\text{N} - \text{NO}_3^- &= \gamma \times [\delta^{15}\text{N} - \text{NO}_3^-]_{\text{OH}} + (1 - \gamma) \times [\delta^{15}\text{N} - \text{NO}_3^-]_{\text{N}_2\text{O}_5} \\ &= \gamma \times [\delta^{15}\text{N} - \text{HNO}_3]_{\text{OH}} + (1 - \gamma) \times [\delta^{15}\text{N} - \text{HNO}_3]_{\text{N}_2\text{O}_5}\end{aligned}\quad (S1)$$

$$\begin{aligned}\delta^{18}\text{O} - \text{NO}_3^- &= \gamma \times [\delta^{18}\text{O} - \text{NO}_3^-]_{\text{OH}} + (1 - \gamma) \times [\delta^{18}\text{O} - \text{NO}_3^-]_{\text{N}_2\text{O}_5} \\ &= \gamma \times [\delta^{18}\text{O} - \text{HNO}_3]_{\text{OH}} + (1 - \gamma) \times [\delta^{18}\text{O} - \text{HNO}_3]_{\text{N}_2\text{O}_5}\end{aligned}\quad (S2)$$

$$\begin{aligned}[\delta^{15}\text{N} - \text{HNO}_3]_{\text{OH}} &= \delta^{15}\text{N} - \text{NO}_2 \\ &= 1000 \times \left[\frac{(^{15}\alpha_{\text{NO}_2/\text{NO}} - 1)(1 - f_{\text{NO}_2})}{(1 - f_{\text{NO}_2}) + (^{15}\alpha_{\text{NO}_2/\text{NO}} \times f_{\text{NO}_2})} \right] + \delta^{15}\text{N} - \text{NOx}\end{aligned}\quad (S3)$$

$$[\delta^{15}\text{N} - \text{HNO}_3]_{\text{N}_2\text{O}_5} = 1000 \times (^{15}\alpha_{\text{N}_2\text{O}_5/\text{NO}_2} - 1) + \delta^{15}\text{N} - \text{NOx} \quad (S4)$$

$$\begin{aligned}[\delta^{18}\text{O} - \text{HNO}_3]_{\text{OH}} &= \frac{2}{3} \times [\delta^{18}\text{O} - \text{NO}_2]_{\text{OH}} + \frac{1}{3} \times [\delta^{18}\text{O} - \text{OH}]_{\text{OH}} \\ &= \frac{2}{3} \times \left[\frac{1000 \times (^{18}\alpha_{\text{NO}_2/\text{NO}} - 1) \times (1 - f_{\text{NO}_2})}{(1 - f_{\text{NO}_2}) + (^{18}\alpha_{\text{NO}_2/\text{NO}} \times f_{\text{NO}_2})} + [\delta^{18}\text{O} - \text{NOx}] \right] \\ &\quad + \frac{1}{3} \times \left[(\delta^{18}\text{O} - \text{H}_2\text{O}_{(\text{g})}) + 1000 \times (^{18}\alpha_{\text{OH}/\text{H}_2\text{O}_{(\text{g})}} - 1) \right]\end{aligned}\quad (S5)$$

$$[\delta^{18}\text{O} - \text{HNO}_3]_{\text{N}_2\text{O}_5} = \delta^{18}\text{O} - \text{NO}_2 + 1000 \times (^{18}\alpha_{\text{N}_2\text{O}_5/\text{NO}_2} - 1) \quad (S6)$$

$$1000(^m\alpha_{x/y} - 1) = \frac{A}{T^4} \times 10^{10} + \frac{B}{T^3} \times 10^8 + \frac{C}{T^2} \times 10^6 + \frac{D}{T} \times 10^4 \quad (S7)$$

Table S1 Values of $\delta^{18}\text{O}$ from atmospheric components

Components	Values (‰)	References
O_3	From 80 to 130	Michalski et al., 2011
O_2	23.5	Kroopnick and Craing; 1972
H_2O (g) in Beijing winter	-27.9	Wen et al., 2010
H_2O (g) in Qingdao winter	-18.6	Wang et al., 2022
·OH in Beijing winter	From -72.4 to -64.9	$\delta^{18}\text{O-OH} = \delta^{18}\text{O-H}_2\text{O}_{(\text{g})} + 1000(\text{18\alpha_{\text{X/Y}}} - 1)$
·OH in Qingdao winter	From -61.2 to -57.8	(Walters and Michalski, 2016)

Table S2 $^{15}\alpha_{A/B}$ and $^{18}\alpha_{A/B}$ regression coefficients as a function of the temperature (150 K \leq T \leq 450 K) (Walters and Michalski, 2015, 2016)

		A	B	C	D
$^{15}\alpha_{A/B}$	N_2O_5/NO_2	0.69398	-1.9859	2.3876	0.16308
	NO_2/NO	3.8834	-7.7299	6.0101	-0.17928
$^{18}\alpha_{A/B}$	NO/NO_2	-0.04129	1.1605	-1.8829	0.74723
	$\cdot OH/H_2O_{(g)}$	2.1137	-3.8026	2.5653	0.5941
	N_2O_5/NO_2	-0.54136	0.13073	1.2477	-0.1272

Table S3 Equations for calculating the statistical evaluation indices

Statistical index	Formula
1. Mean Bias	$MB = \frac{1}{N} \sum_{i=1}^N (Sim - Obs)$
2. Root Mean Square Error	$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (Sim - Obs)^2}$
3. Index of agreement, IOA	$IOA = 1 - \frac{\sum_{i=1}^N (Sim - Obs)^2}{\sum_{i=1}^N (Sim - \overline{Obs} + Obs - \overline{Obs})^2}$
4. Normalized Mean Bias	$NMB = \frac{1}{N} \sum_{i=1}^N \left(\frac{Sim - Obs}{Obs} \right)$
5. Normalized Mean Error	$NME = \frac{1}{N} \sum_{i=1}^N \left \frac{Sim - Obs}{Obs} \right $
6. Correlation coefficient (R)	$R = \frac{1}{N} \sum_{i=1}^N \left[\frac{(Sim - \overline{Sim})(Obs - \overline{Obs})}{S_p S_o} \right]$ $S_p = \left[\frac{1}{N} \sum_{i=1}^N (Sim - \overline{Sim})^2 \right]^{\frac{1}{2}}$ $S_o = \left[\frac{1}{N} \sum_{i=1}^N (Obs - \overline{Obs})^2 \right]^{\frac{1}{2}}$

Table S4 Sources of nitrate observation data for the winter of 2013 and the winter of 2018 in the NCP

City	Winter, 2013	Winter, 2018
Beijing	Song et al. (2019)	Fan et al. (2020)
Tianjin	Yao et al. (2020)	Observation
Shijiazhuang	Wang et al. (2016)	Zhou et al. (2020)
Jinan	Cheng et al. (2021)	Observation
Zhengzhou	Wei et al. (2019)	Dong et al. (2020)
Qingdao	Observation	Observation
Yantai	/	/

The NO_3^- observation data collected during the winter of 2018 for Tianjin were sourced from direct observations by the group of Li Xiaodong at Tianjin University (sampling site: Building 19 rooftop, Tianjin University; coordinates: 39.11°N, 117.16°E). The NO_3^- observation data collected during the winter of 2018 in Jinan were sourced from observations by the group of Xue Likun at Shandong University (sampling site: Jinan City Environmental Monitoring Station; coordinates: 36.66°N, 117.05°E). For Qingdao, NO_3^- observation data for both the winter of 2018 and the winter of 2013 were derived from our own observations.

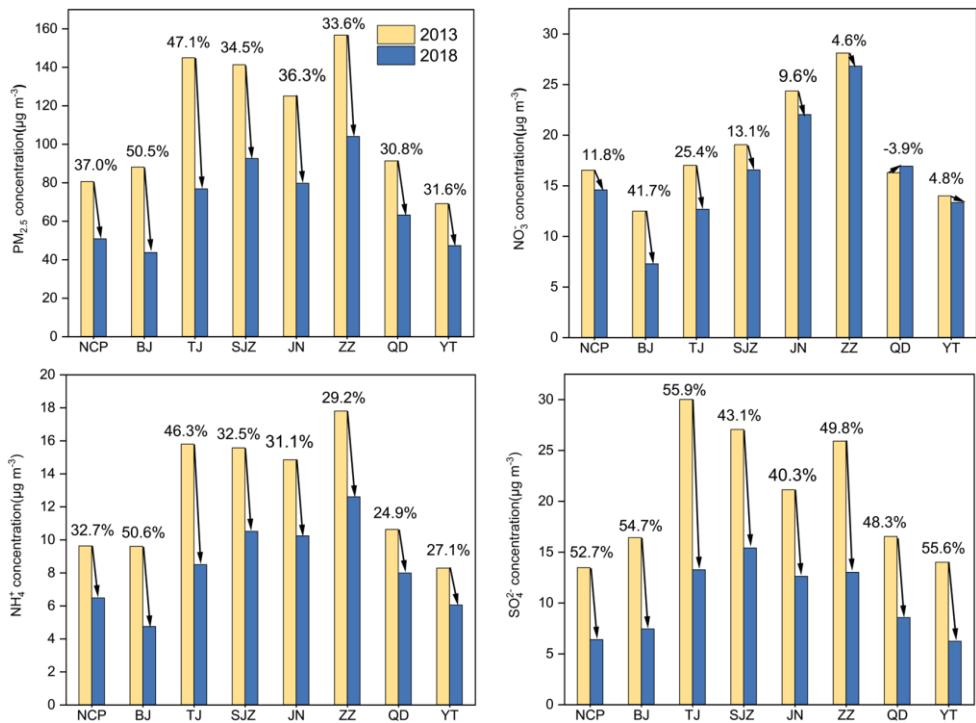


Figure S2 Concentrations of PM_{2.5} and its components in seven major cities in the NCP region during the winters of 2013 and 2018

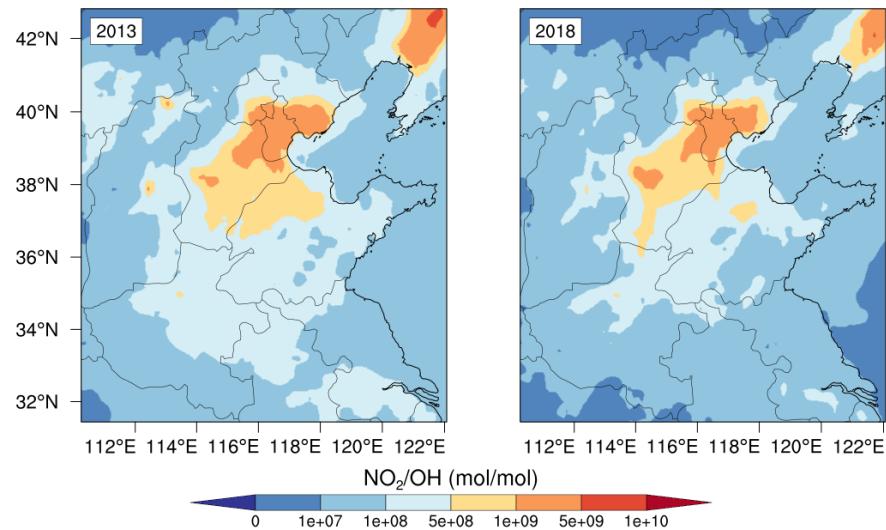


Figure S3 Spatial distribution of the NO₂/OH molar ratio in the NCP region during the winters of 2013 and 2018

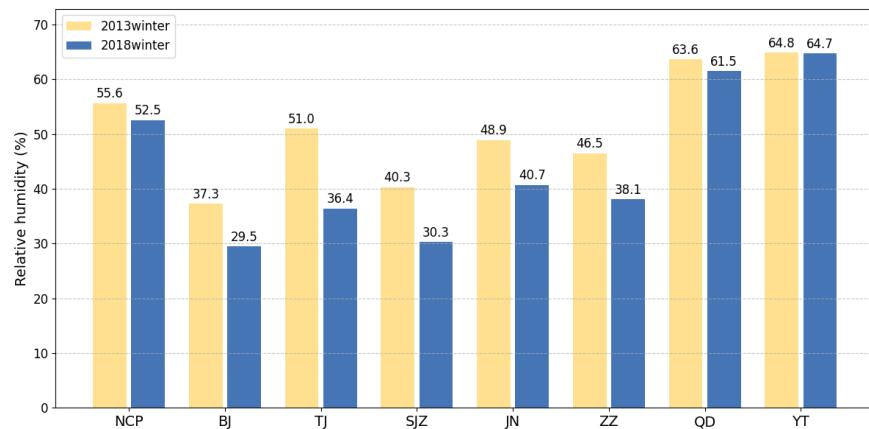


Figure S4 Relative humidity in the NCP and seven major cities (2013, 2018)

Reference

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