#### **Reply to Reviewer#1 and the editor:**

We thank the reviewers for taking the time to assess the manuscript (*egusphere-2024-359*) and providing helpful comments and suggestions to improve the manuscript. Below we address the reviewers' comments, with the reviewer comments in *italic and black*, and our responses in blue. We have revised the manuscript accordingly and mentioned the line number in the **tracked version of the manuscript**.

This manuscript focuses on the effects of soil nitrogen emission (SNOX and SHONO) on ozone pollution over two typical regions (BTH and FWP) in China. The authors found that SNOx and SHONO emissions account for a large proportion (nearly 50%) of total anthropogenic NOx emissions. Therefore, it can increase MDA8 O3 by 17 % and nitrate concentrations by 42% in the BTH. The results suggest that the presence of soil nitrogen emissions can offset the efforts in controlling anthropogenic emissions, which can provide theoretical implications on ozone pollution management in key regions of China. Overall, the manuscript is well written, although minor changes are needed to further improve it.

Thanks for the positive comments. Our item-by-item responses can be found in below.

### Comments:

### It is not clear how MEIC emissions are converted to model-ready formats.

Thank you for the suggestion regarding the conversion of MEIC emissions to model-ready formats. The MEIC inventory used in this study has a spatial resolution of  $0.25 \,^{\circ} \times 0.25 \,^{\circ}$  and is constructed on an equal latitude-longitude grid. The model domain, however, employs a Lambert projection, which results in a misalignment between these two grid systems. To address this issue, we implement a spatial interpolation method to reallocate emission fluxes to the model grid. Here is a detailed description of our method:

Under the Lambert projection, the model grids are rectangular, while the MEIC grids are deformed, approximating a trapezoid shape. For calculating the emission of each model grid, we determine which MEIC grid may fall in that grid, based on their central latitude and longitude coordinates, and then apply the principle that the ratio of emissions is equivalent to the ratio of areas between the model grid and MEIC grid. The area of the MEIC grid is denoted as A and its corresponding emission is denoted as E, and the area of the model grid is denoted as a, thus the emission fluxes of model grid e are expressed as  $e = E \times a / A$ . However, if the spatial resolution of the simulation domain is coarser than MEIC, the model grid often falls within multiple MEIC grids and this method would have errors. We thus divide the coarser model grid into n  $\times$  n finer subgrids. Since the spatial resolution of the nested domain is 9 km in this study, we choose n as 9. We apply the aforementioned method to calculate the emissions of

each finer model subgrid, and then sum up the emissions of  $n \times n$  finer subgrids to obtain the total emissions of the model grid. This method ensures a more accurate allocation of MEIC emission to the model grid, despite different spatial resolutions of the simulation domain.

The description of the above conversion method has also been added in the revised version as follows:

Page 8, Lines 144-146: "Due to the differences in spatial resolution and map projection between the MEIC inventory and model grid, we applied a spatial interpolation method to convert the MEIC inventory to the model-ready formats. The descriptions are detailed in Text S1."

Please see the detailed description in Text S1 in the revised Supplement.

### Is there any way to validate the emissions of NOx and HONO using BDISNP?

We admit that the optimal validation method is to compare the simulated and observed soil emission flux of NO<sub>x</sub> and HONO. Due to the influence of various factors, the observed soil nitrogen emissions reported in the literature couldn't align well with the simulated SNO<sub>x</sub> and SHONO. For example, many field observation experiments are conducted to study the impacts of anthropogenic fertilizer application and irrigation on soil Nr emissions, thus the measured soil Nr emission data are not representative in comparison to the model results (Li, 2013; Tang et al., 2020; Xue et al., 2019). Additionally, the observed SNO<sub>x</sub> and SHONO in the literature are often specific to certain regions and are based on a relatively limited number of observation sites (Wu et al., 2019; Wu et al., 2022). Therefore, it is difficult to compare the scattered and location-specific observations of soil Nr emissions reported in the literature (from different regions and periods) with the UI-WRF-Chem model results. To evaluate the simulation performance of the BDISNP scheme, we use satellite observations that have large spatial coverage and compare them with the simulated concentrations of NO<sub>2</sub>, HONO, O<sub>3</sub>, and nitrate in the atmosphere with and without the implementation of the soil Nr emission scheme against the observations. From Figure 2 to 4, the results show that the simulations with the implementation of the BDISNP scheme are in better agreement with TROPOMI NO<sub>2</sub> VCD, surface HONO, O<sub>3</sub>, and nitrate than the Default. Similar approach of validation was used in our past work as well (Sha et al., 2021).

# In Figure 2, please show statistical values of the comparison. It is hard to say which is better in comparison to the TROPOMI results.

We have added the normalized mean bias (NMB) and spatial correlation coefficient (R) in Figure 2, and also modified the discussions in the revised version as follows:

Page 14, Lines 283-286: "Overall, Base shows the improved performance in

simulating NO<sub>2</sub> VCD in comparison to Default with a decreasing bias from -30% (-21%) to +4% (+17%) and an increasing spatial correlation coefficient (R) from 0.62 (0.50) to 0.65 (0.54) in the study region (cropland)."

## Figure 6, the panel (c) is not correctly named.

Thanks for the reminder, the title of panel (c) in Figure 6 has been corrected in the revised manuscript.

In Conclusion, "leads to a slower increase rate of T2 compared to scenarios without soil Nr emissions", I suggest add the values of T2 increase rate in parentheses (??°C).

We have added the values of T2 increase rate in the revised version as follows:

Page 26, Lines 547-551: "The presence of soil Nr emissions, acting as precursors of O<sub>3</sub> and SIA, has a suppressing effect on efforts to mitigate summer O<sub>3</sub> pollution, particularly in the BTH region, and also leads to a slower increase rate of T2 (0.098 °C) in July compared to scenarios without soil Nr emissions (0.14 °C) when anthropogenic emissions are excluded."

Line 332-351, the authors found that soil NOX lead to OH decrease in their study, which is contrary to other studies that showed soil NOX can increase OH. I suggest add some explanations.

Thanks for the suggestion. We have added the discussions in the revised version as follows:

Page 19, Lines 395-403: "The discrepancy between our findings and those of other studies regarding the impact of  $SNO_x$  on  $\cdot OH$  levels could be attributed to the abundance of ambient  $NH_3$  in China during summer, where soil emissions may lead to a significant increase in nitrate, and the increased aerosols can affect the concentrations of  $\cdot OH$  through photochemical reactions (Wang et al., 2011; Xu et al., 2022). Additionally, after taking into account the  $SNO_x$  in the model, the environment may shift to a relatively  $NO_x$ -saturated regime, thus the termination reaction for  $O_3$  production could be  $NO_2$  and  $\cdot OH$  to generate  $HNO_3$  (Chen et al., 2022; Wang et al., 2023b)."

Line 363-365, "The distribution of sensitivity of O3 to precursor emission in FWP regions are more complex with a mix of three O3 formation regimes, which is attributed to the large population, regional urbanization and industrialization". The explanations are broad, I suggest the authors give more specific explanations or cite some relevant papers, otherwise no need to explain this phenomenon.

We have modified the explanations in the revised version as follows:

Page 20-21, Lines 419-430: "Figure 6 illustrates that the majority of BTH region has  $H_2O_2/HNO_3$  values lower than 0.35 in Base simulation, indicating a VOCs-sensitive regime or NO<sub>x</sub>-saturated regime in July. In contrast, the distribution of sensitivity of O<sub>3</sub> to precursor emission in FWP regions is more complex with a mix of three O<sub>3</sub> formation regimes. The spatial patterns of O<sub>3</sub> formation regimes presented in this study are consistent with the previous studies based on satellite observations and model simulations during summer seasons, despite using a different method (Wang et al., 2019; Wang et al., 2023b). This agreement across multiple approaches strengthens the confidence in the spatial patterns of O<sub>3</sub> formation regimes in the key regions of China."

## **Corrections:**

*The reference "TROPOMI ATBD of the total and tropospheric NO2 data products", please check it.* 

We have revised this reference to "van Geffen, J. H. G. M., Eskes, H. J., Boersma, K. F., and Veefkind, J. P.: TROPOMI ATBD of the total and tropospheric NO<sub>2</sub> data products, Report S5P-KNMI-L2-0005-RP, version 2.2.0, 2021-06-16, KNMI, De Bilt, The Netherlands, http://www.tropomi.eu/data-products/nitrogen-dioxide/ (last access: 7 March 2022), 2021." (Page 34, Lines 761-765)

In Line 471, "we admit that uncertainties in both soil Nr and anthropogenic emissions", add "exist" after " uncertainties"

Thanks, we have corrected it in the revised version. (Page 26, Lines 553)

In Figure 2 caption, note that (e) and (f) should be (b) and (c)

Thanks, we have corrected it. See Figure 2 in the revised version.

Line 228, "(Zhan et al., 2021)", maybe Zhang et al?

We forgot to cite this literature, and have added "Zhan, Y., Xie, M., Gao, D., Wang, T., Zhang, M., and An, F.: Characterization and source analysis of water-soluble inorganic ionic species in  $PM_{2.5}$  during a wintertime particle pollution episode in Nanjing, China, Atmos. Res., 262, 105769, 10.1016/j.atmosres.2021.105769, 2021." in the reference list. (Page 37, Lines 855-858)

*Line 391-394, no need to entirely list values under 20%, 40%, 60%, 80%, and 100%. Maybe three percent (20%,60%,100%) are enough.* 

We have revised it to "Specifically, in the BTH region, MDA8 O<sub>3</sub> decrease by 1.3% (1.8  $\mu$ g m<sup>-3</sup>), 6.3% (8.7  $\mu$ g m<sup>-3</sup>), and 17.4% (24.0  $\mu$ g m<sup>-3</sup>) with anthropogenic NO<sub>x</sub> emission reductions by 20%, 60%, and 100%, respectively, in the present of soil Nr emissions." (Page 22, Lines 457-460)

### **References:**

- Chen, W., Guenther, A. B., Jia, S., Mao, J., Yan, F., Wang, X., and Shao, M.: Synergistic effects of biogenic volatile organic compounds and soil nitric oxide emissions on summertime ozone formation in China, Sci. Total Environ., 828, 154218, 10.1016/j.scitotenv.2022.154218, 2022.
- Li, D.: Emissions of NO and NH<sub>3</sub> from a Typical Vegetable-Land Soil after the Application of Chemical N Fertilizers in the Pearl River Delta, PLoS One, 8, e59360, 10.1371/journal.pone.0059360, 2013.
- Sha, T., Ma, X., Zhang, H., Janechek, N., Wang, Y., Wang, Y., Castro García, L., Jenerette, G. D., and Wang, J.: Impacts of Soil NO<sub>x</sub> Emission on O<sub>3</sub> Air Quality in Rural California, Environ. Sci. Technol., 55, 7113-7122, 10.1021/acs.est.0c06834, 2021.
- Tang, K., Qin, M., Fang, W., Duan, J., Meng, F., Ye, K., Zhang, H., Xie, P., Liu, J., Liu, W., Feng, Y., Huang, Y., and Ni, T.: An automated dynamic chamber system for exchange flux measurement of reactive nitrogen oxides (HONO and NO<sub>x</sub>) in farmland ecosystems of the Huaihe River Basin, China, Sci. Total Environ., 745, 140867, 10.1016/j.scitotenv.2020.140867, 2020.
- Wang, N., Lyu, X., Deng, X., Huang, X., Jiang, F., and Ding, A.: Aggravating O<sub>3</sub> pollution due to NO<sub>x</sub> emission control in eastern China, Sci. Total Environ., 677, 732-744, 10.1016/j.scitotenv.2019.04.388, 2019.
- Wang, S., Xing, J., Jang, C., Zhu, Y., Fu, J. S., and Hao, J.: Impact Assessment of Ammonia Emissions on Inorganic Aerosols in East China Using Response Surface Modeling Technique, Environ. Sci. Technol., 45, 9293-9300, 10.1021/es2022347, 2011.
- Wang, W., Li, X., Cheng, Y., Parrish, D. D., Ni, R., Tan, Z., Liu, Y., Lu, S., Wu, Y., Chen, S., Lu, K., Hu, M., Zeng, L., Shao, M., Huang, C., Tian, X., Leung, K. M., Chen, L., Fan, M., Zhang, Q., Rohrer, F., Wahner, A., Pöschl, U., Su, H., and Zhang, Y.: Ozone pollution mitigation strategy informed by long-term trends of atmospheric oxidation capacity, Nat. Geosci., 17, 20-25, 10.1038/s41561-023-01334-9, 2023b.
- Wu, D., Horn, M. A., Behrendt, T., Müller, S., Li, J., Cole, J. A., Xie, B., Ju, X., Li, G., Ermel, M., Oswald, R., Fröhlich-Nowoisky, J., Hoor, P., Hu, C., Liu, M., Andreae, M. O., Pöschl, U., Cheng, Y., Su, H., Trebs, I., Weber, B., and Sörgel, M.: Soil HONO emissions at high moisture content are driven by microbial nitrate reduction to nitrite: tackling the HONO puzzle, ISME J., 13, 1688-1699, 10.1038/s41396-019-0379-y, 2019.
- Wu, D., Zhang, J., Wang, M., An, J., Wang, R., Haider, H., Xu-Ri, Huang, Y., Zhang, Q., Zhou, F., Tian, H., Zhang, X., Deng, L., Pan, Y., Chen, X., Yu, Y., Hu, C., Wang, R., Song, Y., Gao, Z., Wang, Y., Hou, L., and Liu, M.: Global and Regional Patterns of Soil Nitrous Acid Emissions and Their Acceleration of Rural Photochemical Reactions, J. Geophys. Res.: Atmos., 127, 10.1029/2021jd036379, 2022.
- Xu, W., Zhao, Y., Wen, Z., Chang, Y., Pan, Y., Sun, Y., Ma, X., Sha, Z., Li, Z., Kang, J., Liu, L., Tang, A., Wang, K., Zhang, Y., Guo, Y., Zhang, L., Sheng, L., Zhang, X., Gu, B., Song, Y., Van Damme, M., Clarisse, L., Coheur, P.-F., Collett, J. L., Goulding, K., Zhang, F., He, K., and Liu, X.: Increasing importance of ammonia emission abatement in PM<sub>2.5</sub> pollution control, Sci. Bull., 67, 1745-1749,

10.1016/j.scib.2022.07.021, 2022.

Xue, C., Ye, C., Zhang, Y., Ma, Z., Liu, P., Zhang, C., Zhao, X., Liu, J., and Mu, Y.: Development and application of a twin open-top chambers method to measure soil HONO emission in the North China Plain, Sci. Total Environ., 659, 621-631, 10.1016/j.scitotenv.2018.12.245, 2019.