

Response to Referee #1

Title: Uncertainties in fertilizer-induced emissions of soil nitrogen oxide and the associated impacts on ground-level ozone and methane

MS number: egusphere-2025-1416

Authors: Cheng Gong, Yan Wang, Hanqin Tian, Sian Kou-Giesbrecht, Nicolas Vuichard, and Sönke Zaehle

Summary: This paper assesses the emissions of soil NO_x from several different emissions parameterizations, and the global impact of these emissions on atmospheric chemistry. Overall, the large range of differences in the emissions (both in terms of magnitude and spatial pattern) impacts surface ozone and methane concentrations. I agree with the author's conclusions that this is an important issue to consider and they raise some interesting points. However, this study felt very cursory - results were presented and discussed in very short text. Adding some depth and further discussion to the paper would strengthen the manuscript.

Response:

We appreciate reviewer's acknowledgment on the importance of our work. Our initial manuscript was concise to maintain the focus on the uncertainties in SNO_x-Fer, while we apologize if this brevity resulted in the omission of scientific details. We are grateful for the reviewer's invaluable comments and glad to add additional details and discussions in the revised manuscript. Please find our point-to-point response below:

Major points

Overall, more detail and depth to the paper is needed to make this a valuable contribution to the literature. For example:

Section 2.4: The text mentions that the TBM simulations are only the model mean – why wouldn't you be interested in including the different members? Isn't that variability part of examining the physical processes that drive the NO_x emissions? At a minimum, the NMIP model spread be shown in Figure 2 instead of just the model mean.

Response:

To address this comment, we implemented three additional GEOS-Chem simulations in which the standalone SNO_x-Fer simulated by CLASSIC, OCN and ORCHIDEE were applied instead of the model ensemble mean. We explicitly showed the TBM variabilities in SNO_x-Fer and their impacts on O₃ and CH₄ in all plots with the exception of the spatial pattern plot (Figs. 3 and 6), because we prefer to focus on the differences among the different SNO_x-Fer estimating methods rather than the TBM spread in this study. The NMIP spread of SNO_x-Fer was shown in SI Fig. S3, the same as our original manuscript.

Section 4.2: The impacts on ozone are focused on the summer, yet soil NO_x often peaks in the spring in the northern hemisphere. Have the authors evaluated other seasonal aspects of impacts on ozone? More detail about seasonal changes in the emissions and the subsequent ozone changes would enhance the paper.

Response:

While we have added a seasonal analysis in the revised manuscript to address this comment, we want to clarify that one important limitation of the data availability is that there is no monthly (or even daily) grid-level data of agricultural fertilizer application available. The HaNi dataset we used in this study, as well as the equivalently up-to-date fertilizer dataset (Adalibieke et al., 2023) only provide annual fertilizer application rates globally. This results primarily from the difficulties involved in capturing the diverging local fertilizer management strategies (e.g. when to apply fertilizer and how to distribute the total loadings into several fertilizing times). As a result, there are limitations to confidently assess the season cycles of SNOx-Fer impacts.

We discuss this uncertainty in more details in this revised manuscript. Table 1 summarized how different methods distribute the annual fertilizer across the year. In particular, we added two additional experiments namely Linear_7525 and Nonlinear_7525, which follows the Linear and Nonlinear EF approaches, respectively, but sets 75% of the annual fertilizer applied in the first month of growing season and the rest 25% evenly applied in the rest growing months, the same distributing method as the BDSNP scheme (Hudman et al., 2012). These two new simulations could help us better understand how the seasonal cycle of fertilizer application affects soil NO_x emissions and also O₃ and CH₄.

We have added method descriptions in Sect. 3 accordingly:

‘In order to further examine the seasonality of SNOx-Fer and the associated impacts on ground-level O₃ in agricultural hotspot regions, we investigate how different SNOx-Fer approaches distribute the annual fertilizer seasonally (Table 1). The HaNi dataset, as well as the equivalently up-to-date fertilizer dataset (Adalibieke et al., 2023), only provide annual fertilizer application rates given the lack of specific information to distribute the N fertilization seasonally. The CEDS, BDSNP and NMIP2 models approaches have their own specific monthly distribution, while the monthly distribution of fertilizer application in the linear and nonlinear EF are arbitrarily assumed to be even during growing season. Here, we added two additional GEOS-Chem sensitivity experiments for the linear and non-linear approach, named Linear_7525 and Nonlinear_7525, which apply the seasonal pattern of the BDSNP scheme (Hudman et al., 2012), assuming that 75% of the annual fertilizer is applied in the first month of growing season and the rest 25% evenly applied in the rest growing months.’

Table 1. Summary of the sensitivity experiments in GEOS-Chem and the methods used by different SNOx-Fer estimating approaches to distribute the annual N fertilizer into monthly.

SNOx-Fer estimating approach	Experimental name in this study	Emissions of SNOx-Fer	Fertilizer monthly distribution
None	Zero	Zero	None
Emission Factor (EF)	Linear	Linear EF	Evenly distributed during the growing season
	Nonlinear	Nonlinear EF	
	Linear_7525	Linear EF	75% of the annual fertilizer is applied in the first month of growing season, while the rest 25% is evenly distributed in the rest growing months
	Nonlinear_7525	Nonlinear EF	

Emission inventory	CEDS	CEDS agricultural NO _x sector	Not clear
	BDSNP_coarse	GEOS-Chem default BDSNP with resolution of 2°×2.5°	
BDSNP	BDSNP_coarse_scaled	BDSNP scaled with the interannual variations of HaNi fertilizer loadings with resolution 2°×2.5°	75% of the annual fertilizer is applied in the first month of growing season, while the rest 25% is evenly distributed in the rest growing months
	BDSNP_fine (offline)	GEOS-Chem default BDSNP with resolution of 0.5°× 0.625°	
	NMIP2-OCN	OCN simulated SNO _x -Fer	Distributed the annual N fertilizer loadings into four equal doses in the first half of the growing season
Terrestrial biosphere models (TBMs)	NMIP2-CLASSIC	CLASSIC simulated SNO _x -Fer	Evenly distributed throughout the year in the tropics (between 30S and 30N); Evenly distributed from spring equinox to fall equinox between 30N (30S) and 90N (90S)
	NMIP2-ORCHIDEE	ORCHIDEE simulated SNO _x -Fer	Not clear*
	NMIP2	TBMs ensemble mean	

*It is hard to contact all of the model contributors in the summer vacation season. The ORCHIDEE detail will be updated in the next revision, which will not influence any of the results of this study but only provide more information.

We added a new section to demonstrate the seasonal cycles of SNO_x-Fer and impacts on O₃:

‘4.2 The seasonal cycle of SNO_x-Fer and the associated impact on O₃ concentrations

Figure 4 shows the seasonality of SNO_x-Fer in four agricultural hotspot regions among different SNO_x-Fer estimating methods. In the temperate regions like Eastern U.S., Western Europe and Eastern Asia, the TBM ensembles NMIP2 shows very strong seasonal variations, which reaches highest during May to July in Eastern U.S., April to June in Western Europe and May to August in Eastern Asia, respectively. The seasonality of the linear and nonlinear EF methods is strongly dependent on the assumption of fertilizer applying time (Table 1), where the monthly SNO_x-Fer emissions are at similar levels during the growing season for the Linear and Nonlinear experiments, but peak in a pronounced manner in the north-hemispheric spring time (around February to April) in the Linear_7525 and Nonlinear_7525 cases. Although the BDSNP applies the same assumption of fertilizer applying time as Linear_7525 and Nonlinear_7525, the SNO_x-Fer in BDSNP peaks much later (September to October in Eastern U.S., June to August in Western Europe and May to June in Eastern Asia). This arises because the EF methods estimate SNO_x-Fer instantaneously in response to the fertilizer application, but the BDSNP scheme cumulates N fertilizer with a 4-months time window (Eq. 3). It is also very important the BDSNP

includes the regulation of soil temperature and moisture on SNOx-Fer, both of which also have strong seasonality, but the EF methods do not. Furthermore, in the tropical regions of Southern Asia, the NMIP2, Linear_7525 and Nonlinear_7525 experiments estimate the peak SNOx-Fer in the beginning of the year, while the SNOx-Fer of BDSNP reaches highest in May due to the N cumulation assumption (Fig. 4d). The rest methods, including the emissions inventory CEDS, the Linear and Nonlinear EF method, show very weak seasonality of SNOx-Fer in Southern Asia.

The seasonality of ground-level monthly MDA8 O₃ changes in response to the SNOx-Fer in general aligns with the monthly variations of SNOx-Fer among different estimating approaches (Fig. 5). The strongest enhancement of regional MDA8 O₃ shows during the north-hemispheric summertime (June–August) for most of the estimating approaches in three temperate regions, when the absolute O₃ concentrations also reaches highest. However, it should be noted that spring-peak SNOx-Fer in the Linear_7525 and the Nonlinear_7525 cases does not lead to high O₃ enhancement in both Western Europe and Eastern Asia (Figs. 5b and 5c). The weak sensitivity of O₃ to NO_x during springtime is likely the result of the seasonal variations in other emissions (e.g. biogenic volatile organic compounds (BVOCs)), which alter the chemical sensitivity regime. The responses of O₃ to SNOx-Fer could also depend on regions (e.g. O₃ enhancement also peaks during spring in Linear_7525 in Eastern U.S., Fig. 5a), spatial simulating resolution or different modelling chemical mechanisms. The O₃ enhancement in Southern Asia is generally similar during north-hemispheric spring and summer time for all of the SNOx-Fer estimating approaches (Fig. 5d), except for the BDSNP scheme, which stimulates significantly higher O₃ enhancement during May to July relative to February to April.

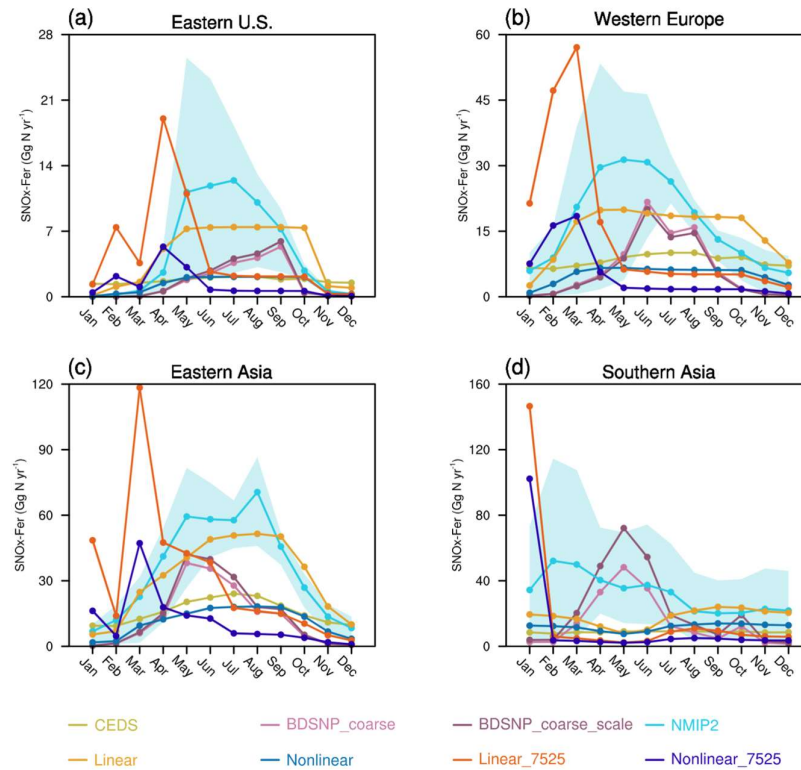


Figure 4. The monthly regional SNOx-Fer (Gg N yr⁻¹) in the (a) Eastern U.S., (b) Western Europe, (c) Eastern Asia and (d) Southern Asia with different SNOx-Fer estimating approaches. The cyan-blue shades indicate the spreads among three different TBM models (CLASSIC, OCN and ORCHIDEE) in the NMIP2 ensemble.

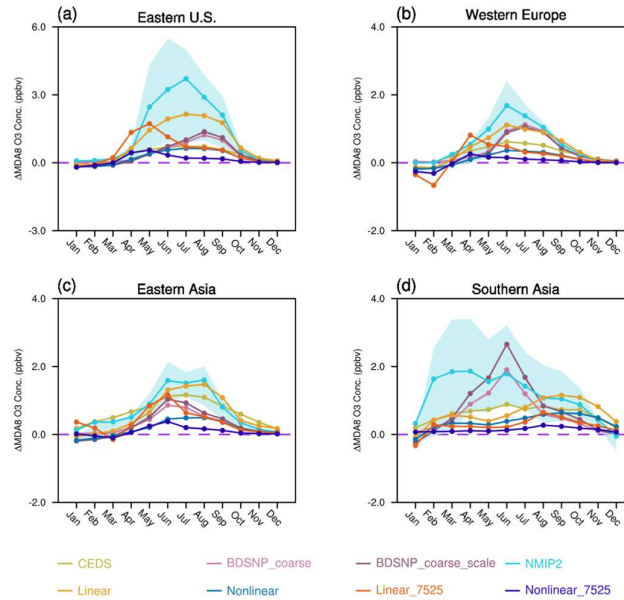


Figure 5. The regionally-averaged monthly MDA8 O₃ changes (ppbv) induced by SNOx-Fer in the (a) Eastern U.S., (b) Western Europe, (c) Eastern Asia and (d) Southern Asia with different SNOx-Fer estimating approaches. The cyan-blue shades indicate the spreads among three different TBM models (CLASSIC, OCN and ORCHIDEE) in the NMIP2 ensemble.

We further added discussions on the uncertainties induced by the fertilizer applying time as well as the definition of growing season:

‘The seasonality of SNOx-Fer and the associated impacts on surface O₃ concentrations are also important but poorly constrained. The most difficult challenge is to precisely estimate the monthly (or even daily) N fertilizer loadings in the global scale. Because the N fertilizer data underlying the gridded products is derived from the annual statistics by the Food and Agricultural Organization (FAO) (<https://www.fao.org/faostat/en/#data>), the HaNi dataset applied this study, as well as the equivalently up-to-date fertilizer dataset (Adalibieke et al., 2023), only provides gridded, annual fertilizer application rates. In the EF approaches, the growing season is determined only by temperature and greenness in this study, which could result in a mismatch with the real crop or pasture calendar, especially ignoring the multiple-harvest crops per year. A refined calendar could further improve the prediction of SNOx-Fer seasonality. Furthermore, the NO_x-VOCs-O₃ chemical sensitivity regimes could be determined by not only soil NO_x emissions, but also other anthropogenic and biogenic emissions of NO_x and VOCs, as well as the climate seasonal variations. Therefore, the seasonal cycles of the enhancement of O₃ concentrations may not strictly follow the variations in SNOx-Fer, as our Linear_75 sensitivity experiment implies in Western Europe and Eastern Asia (Figs. 5b and 5c).’

Section 4.3 – there are some interesting conclusions about the impacts of soil NOx on methane, yet very little in depth discussion on Figure 4 and the implications. How much OH is changing due to these changes and can it be tied to some of the hotspots in Figure 2? Right now, the discussion on this result (which is one of the larger implications of the paper) is less than 10 lines of text! Can the authors provide any insight into the model behavior and spatio-temporal implications of these results?

Response:

The OH changes are shown in SI Fig.S4 in our original manuscript, which is in general consistent with the SNO_x-Fer hotspots in Fig. 2. Meanwhile, CH₄ has a much longer lifetime (~9 years) than O₃ and is thus spatially more evenly distributed. As a result, we believe it makes more sense to focus on the global averaged concentration rather than the spatial differences, because the latter are smoothed by atmospheric transport. As we showed in SI Fig.S3, the variations in SNO_x-Fer did not significantly alter the CH₄ global distribution and hotspot regions, but only change the averaged concentration.

We added the description about the spatial distribution as below:

‘Figure 7 shows that N fertilizer-induced soil NO_x induced the reduction of global averaged CH₄ concentrations ranging from 6.7 ppbv (0.4%) to 16.6 ppbv (0.9%) in 2019 by increasing atmospheric OH concentrations (Fig. S5), spatially aligned with the distributions of SNO_x-Fer among different estimating approaches (Fig. 3). Because CH₄ has a significantly longer atmospheric lifetime than either OH or NO_x, the spatial differences in the impacts of SNO_x-Fer on CH₄ concentrations are insignificant (Fig. S4). As a result, we only focus on the globally averaged changes in CH₄ concentrations. The magnitude of our estimate is consistent with recent estimates of around 17.4 ppbv by Gong et al. (2024)...’

We further added the discussions on the effects of NO_x on CH₄ concentrations:

‘The impacts of the changes in short-lived air pollutants on global CH₄ budget have attracted increasing attention in recent years (Peng et al., 2022; Zhao et al., 2025), where NO_x is one of the most important drivers. However, it should be noted that the sensitivity of CH₄ lifetime to NO_x emissions varies substantially among atmospheric chemistry models from -25% to -46% in response to the total NO_x changes from pre-industrial to present-day period (Thornhill et al., 2021). Because few studies investigated how NO_x from agricultural sources affects CH₄, it is difficult to assess if the overall impacts of SNO_x-Fer on CH₄ presented in this study based on the GEOS-Chem model are underestimated or overestimated, even though certain uncertainties are expected. Nevertheless, our results indicate that SNO_x-Fer could be one uncertain but important source in calculating future changes of the global CH₄ budget, the importance of which could be increasing with future continuing reduction in fossil-fuel NO_x emissions (Rao et al., 2017).’

The global resolution of the chemistry model is 2x2.5 degrees which is relatively coarse. Could you discuss the implications of this resolution for the modeled NO_x emissions?

Response:

We agree that the resolution will be a factor for soil NO_x emissions. However, we would like to clarify that most of the SNO_x-Fer estimating approaches, except for the BDSNP, are implemented at fine resolutions and then interpolated into the 2 by 2.5 degrees grid of the atmospheric model (Linear and nonlinear EF method: 0.5° * 0.5° consistent with the HaNi dataset; CEDS: 0.5° * 0.5°; OCN: 1° * 1°; ORCHIDEE and CLASSIC: 0.5° * 0.5°). For BDSNP, we added one offline simulation with 0.5°*0.625° resolution and found it slightly enhanced the global SNO_x-Fer estimates, while the difference is less than the variations among different methods (Fig.1). We added discussions on the resolution of BDSNP in estimating SNO_x-Fer:

‘For the modelling of SNO_x-Fer, on the one hand, recent developments of the parameterization of BDSNP in CTMs focused more on the soil NO_x responses to changing temperature or soil moisture (e.g. Wang et al., 2021; Huber et al., 2023), while the accuracy of the soil N availability has been less

investigated. Even with the scaled N fertilizer loadings to interannually vary the N availability, BDSNP still showed weaker increasing trend of SNOx-Fer in response to the N fertilizer enhancement relative to the empirical EF methods and the TBM simulations of NMIP2 in the past decades (Fig. 1). Nevertheless, it should be noted that the BDSNP scheme is also sensitive to the spatial resolution, where the coarse resolution may miss small-scale hotspots and thus underestimate the global SNOx-Fer, as the BDSNP_fine experiment shows in Fig. 1. On the other hand, terrestrial N availability is a key concept in the development of TBMs, as the process-based TBMs need detailed description of the N cycle to understand nutrient limitation levels and associated C-N coupling....'

For the atmospheric transport and chemical processes, higher resolution is always better but also needs more computational resources. It normally takes hundreds of days to finish one-year GEOS-Chem simulation with $0.5^\circ * 0.625^\circ$ resolution, which appears unnecessarily expensive to support the conclusions of this study.

Minor points

Line 79: The authors characterize the BDSNP model that fixes the fertilizer emissions to 1998, and does not represent interannual variability in fertilizer application. My understanding is that this is not what most interactive models use – they do vary the fertilizer application, and the authors may want to include other applications of this model that do include the interannual variability.

Response:

Thank you for this constructive suggestion. In the BDSNP of GEOS-Chem, there is a one-year daily grid-level data prescribing the soil NOx emissions induced by the N availability items (N_{avail} in Eq. 2), while the scaling factor $\bar{E}$ is tuned with a simulation in 2000. Because the N_{avail} is linearly correlated with the annual fertilizer loading (Eq. 3), it is possible to directly scale this N availability data based on the HaNi dataset on each grid. The new grid-level N_{avail} in the yr year is calculated by:

$$N_{avail}(i, j, yr) = N_{avail}(i, j, 2000) * \frac{Fertilizer_{HaNi}(i, j, yr)}{Fertilizer_{HaNi}(i, j, 2000)} \quad (1)$$

Where $Fertilizer_{HaNi}(i, j, yr)$ represents the total N fertilizer loadings in HaNi dataset at the grid of i latitude and j longitude in the yr year. By applying this modification, we re-run the GEOS-Chem model to examine how SNOx-Fer in BDSNP changed with the interannually varied N fertilizer. Figure 1 shows the modified BDSNP estimates the enhancement of global SNOx-Fer from $0.90 \text{ Tg N yr}^{-1}$ to $1.50 \text{ Tg N yr}^{-1}$ over 1980-2019, the magnitude of which, however, is still slower than most of other estimates.

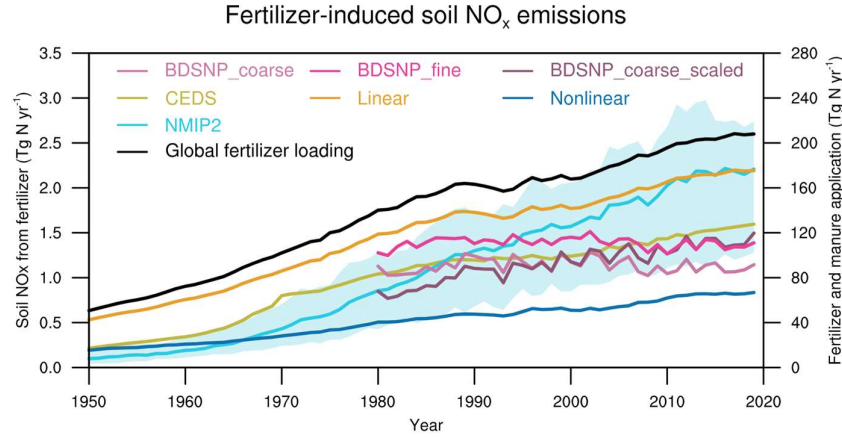


Figure 1. Global estimates of N fertilizer-induced soil NO_x emissions by different approaches. The black line (right Y axis) indicates global annual-mean N synthetic fertilizer and manure inputs over 1950-2019 assessed from the HaNi dataset. The rest lines (left Y axis) indicate the N fertilizer-induced soil NO_x emissions over 1950-2019 estimated by different approaches, including emission inventory (CEDS), linear and non-linear EF, the widely-used CTM parameterization with coarse resolution (2°×2.5°, BDSNP_coarse), fine resolution (0.5°×0.625°, BDSNP_fine) and interannually varied N availability (BNDSP_coarse_scaled), and the TBM ensembles (NMIP2). The light cyan shadows indicate the spread across three different TBMs in NMIP2.

Additional detail for the different emissions methods used would be helpful. Specifically:

Line 107: Please clarify the 1.1% - this is the emissions of nitrogen are 1.1% of the mass of fertilizer applied? This could be explained more clearly.

Response:

The meaning of EF has been explained in the introduction: ‘...the most straightforward and widely-used method is applying the emission factor (EF), which indicates the proportion of N from fertilizer application emitted as NO_x’. Here we added a simple explanation to clarify:

‘... where the value of 1.1% (1.1% of N in the fertilizer will be emitted as NO_x; named as EF_{linear} hereafter)’

Line 113: For the non-linear approach, could you briefly explain what Equation 1 is based on?

Response:

The nonlinear EF is developed in Wang et al. (2024), where the cropland soil NO_x emission data in 223 field experiments with at least three N-input levels are collected. Here is the method description in Wang et al. (2024) :

‘First, the EF for every non-zero N application rate was calculated as follows: $EF(\%) = (E_N - E_0) / N \times 100$, where E_N (kg N ha⁻¹) is NO emissions for non-zero N input, E_0 (kg N ha⁻¹) is NO emissions for zero N input (control), and N (kg N ha⁻¹) is N application rate. Then, the linear relationship between

EF and N application rate in each experiment was described by the following expression: $EF (\%) = EF_0 + \Delta EF \times N$, where EF_0 and ΔEF (% per kg N ha⁻¹) are the intercept and slope of the effect function, respectively. The ΔEF of this relationship could be deemed as the degree of nonlinearity of the NO emission response to the N rate: zero ΔEF means that NO emissions grow linearly with the N rate (i.e., the constant EF), a positive ΔEF indicates that NO emissions increase faster than a linear response with N rate (i.e., the increasing EF), and a negative ΔEF indicates that NO emissions increase slower than a linear response (i.e., the decreasing EF).

ΔEF and EF_0 are calibrated as 0.008 and 0.22 in Wang et al. (2024) respectively, which are the values we used in this study. Such method is widely used in estimating not only soil NO_x emissions, but also soil N₂O (Shcherbak et al., 2014) and NH₃ emissions (Jiang et al., 2017).

The HaNi dataset could be explained more – for example, there are specifics on the different types of nitrogen loadings, yet it is unclear how these are going to directly impact the resulting emissions.

Response:

The different types of N fertilizer have no influences on SNOx-Fer with the EF methods – they are summed up as one single N inputs. However, the synthetic fertilizer and manure, as well as the N forms (NH₄⁺ and NO₃⁻) are explicitly distinguished in terrestrial biosphere models because they belong to different inorganic N pools. We added explanations in the EF method (Sect 2.1) as below:

‘The annual N inputs from HaNi dataset, which are summed by all N forms of synthetic fertilizer and manure, are evenly applied in the months of growing season....’

And add explanations in the TBM method (Sect. 2.4) as below:

‘For each TBM model, anthropogenic fertilizer application are estimated by the HaNi dataset (Tian et al., 2022), where the fertilizer types (NH₄⁺ and NO₃⁻; synthetic fertilizer and manure) are explicitly distinguished in the model.’

The title for Section 2.2 could be more clear – perhaps name the emissions inventory you are using?

Response:

We have revised the title as ‘The emissions inventory CEDS’

Line 136, similar to the comment above – the EF value is 0.7% of what?

Response:

We have added the explanation as ‘... a constant EF value of 0.7% (0.7% of N in the fertilizer will be emitted as NO_x)’

Line 176- probably more important to list the name of the gas-phase chemical mechanism used, as well as how aerosols are treated in the model than to refer to the KPP.

Response:

The chemical mechanism of GEOS-Chem is independently developed referring to the kinetics and products based on JPL recommendations (Bates et al., 2024), as a result it cannot be simply represented by mechanism names used in other climate-chemistry models like CB6, SAPRC99. We have clarified it as well as the aerosol schemes as below:

‘The atmospheric gas-phase chemistry is independently developed referring to the kinetics and products based on JPL recommendations (Bates et al., 2024) and solved by the Kinetic Pre-Processor (KPP). Aerosol thermodynamic equilibrium is calculated by the ISORROPIA II package (Fountoukis and Nenes, 2007).’

Figure 1 – it is very difficult to read the yellow line and text, please update. Also, if the NMIP2 run is an ensemble, then could the spread be shown instead of just the model mean.

Response:

We have updated the Fig.1 accordingly.

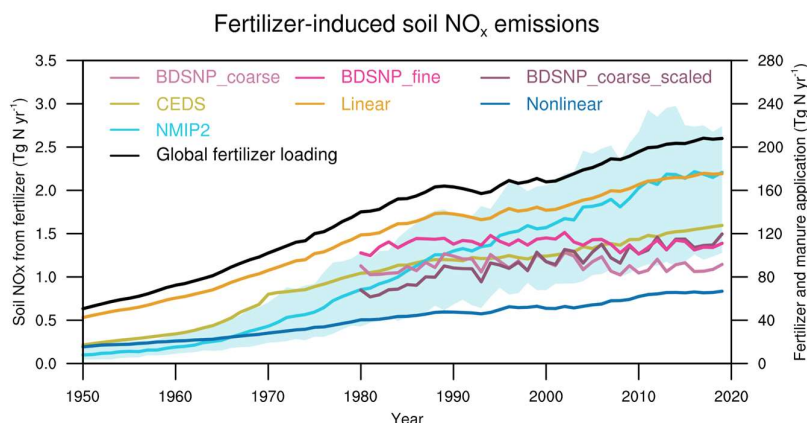


Figure 1. Global estimates of N fertilizer-induced soil NO_x emissions by different approaches. The black line (right Y axis) indicates global annual-mean N synthetic fertilizer and manure inputs over 1950-2019 assessed from the HaNi dataset. The rest lines (left Y axis) indicate the N fertilizer-induced soil NO_x emissions over 1950-2019 estimated by different approaches, including emission inventory (CEDS), linear and non-linear EF, the widely-used CTM parameterization with coarse resolution (2°×2.5°, BDSNP_coarse), fine resolution (0.5°×0.625°, BDSNP_fine) and interannually varied N availability (BNDSP_coarse_scaled), and the TBM ensembles (NMIP2). The light cyan shadows indicate the spread across three different TBMs in NMIP2.

Figure 2 – If I am understanding this correctly, Figure 2a is only used to calculate emissions for Figure 2d. Figures 2b, 2c, 2e and 2f each use their own version of fertilizer. The caption states that 2a is from the 2019 HaNi dataset, but are the emissions shown only for that representative year (or all years averaged together from Figure 1?)

Response:

The HaNi fertilizer dataset is used in not only NMIP2 TBM ensembles, but also the Linear and Nonlinear EF calculation. CEDS and BDSNP have independent fertilizer sources, while the scaled interannually-varied BDSNP, which we added in the revised manuscript (see above), also uses the HaNi fertilizer data. Meanwhile, all the SNOx-Fer emissions are in the year of 2019. To clarify, we split Fig.2a and the other SNOx-Fer emissions figures and add more clarification in the figure captions:

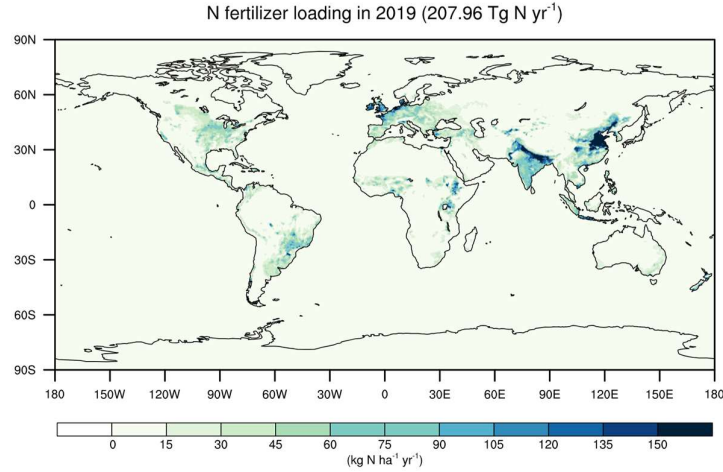


Figure 2. The global spatial patterns of N synthetic fertilizer and manure application in 2019 from the HaNi dataset.

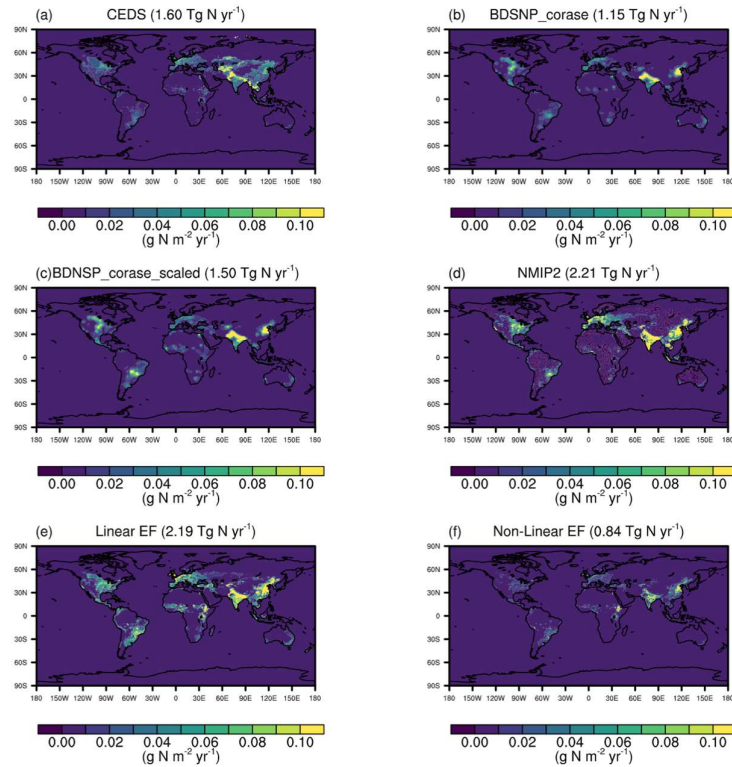


Figure 3. The N-fertilization induced soil NO_x emissions estimated by different approaches in 2019. (a) - (f) The soil NO_x emissions induced by N fertilizer estimated by the CEDS agricultural sector, the default BDSNP scheme in GEOS-Chem with coarse resolution (2°×2.5°), the coarse-resolution BDSNP scheme in GEOS-Chem by interannually scaling the N availability using the HaNi dataset, the NMIP2 ensemble, the linear EF and non-linear EF, respectively. The global total budget of each estimate is given in the sub-titles.

Lines 230-232: This explanation is rather confusing – more expansion on these conclusions would be helpful as it seems to be an important part of the conclusions.

Response:

We have revised the explanation as below:

‘The SNO_x-Fer estimates by NMIP2 ensemble are higher in agricultural hotspots (Table 2), but lower in regions with less synthetic fertilizer application, e.g. in part of the Africa and South America (Figs. 3d and 3e), relative to the Linear EF approach. Because plants and microbes are supposed to have high priority to assess additional N in N-limited regions, which leads less N loss as the gas forms. However, in N-saturated regions, the applied N fertilizer excessive for the living biomes, yielding a higher sensitivity of soil NO_x emissions to N fertilizer application (Du and De Vries, 2025). Such N dynamics have been included in the C-N fully-coupled TBMs, but fail to be represented by the linear EF approach.’

Line 247 – more discussion on the seasonal dynamics would be helpful here. The text states that soil NO_x peaks in the summer, which it might if only temperature were a factor. But other studies that link to fertilizer application are more closely tied to spring soil NO_x peaks. Could the authors expand this discussion about the seasonal dynamics?

Response:

We have added a new section 4.2 to discuss the seasonal cycle. Please see our responses above.

Line 283: what meta-analyses are being referred to here? Also, can you be more specific about what emission inventory you are using?

Response:

Both the linear and non-linear EFs are from meta-analysis. We have added the references here, as well as the inventory name:

‘In this study, we integrated knowledge from meta-analyses (Hergoualc’h et al., 2019; Wang et al., 2024), the emission inventory CEDS, parameterizations in CTMs and the TBM ensembles to better quantify the uncertainties in N fertilizer-induced soil NO_x emissions and the associated impacts on global O₃ and CH₄ concentrations.’

Lines 303-304: I think the authors have missed some of the work that have modified the BDSNP for different environmental drivers (e.g., Wang et al. 2021 for temperature, and Huber et al. 2023 for soil moisture). This should be included as well.

Response:

Here we are discussing about the uncertainties remaining in the data-driven method, e.g. the linear and non-linear EF from meta-data. For the modelling uncertainties, we have already mentioned the recent modifications in BDSNP in the next paragraph of original manuscript: '...recent developments of the parameterization of BDSNP in CTMs focused more on the soil NO_x responses to changing temperature or soil moisture (e.g. Wang et al., 2021; Huber et al., 2023)...'

Line 328: uncertainties in....? (I think you want to say emission inventories but it would be helpful to be clear)

Response:

We have revised the sentence as:

'To summarize, with a comprehensive investigation of different approaches to describe SNO_x-Fer, our results revealed the uncertainties in quantifying SNO_x-Fer and associated important implications in simulating regional air quality and the global greenhouse gas CH₄.'

Reference:

- Adalibieke, W., Cui, X. Q., Cai, H. W., You, L. Z., and Zhou, F.: Global crop-specific nitrogen fertilization dataset in 1961-2020, *Scientific Data*, 10, 10.1038/s41597-023-02526-z, 2023.
- Bates, K. H., Evans, M. J., Henderson, B. H., and Jacob, D. J.: Impacts of updated reaction kinetics on the global GEOS-Chem simulation of atmospheric chemistry, *Geoscientific Model Development*, 17, 1511-1524, 10.5194/gmd-17-1511-2024, 2024.
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Response to Referee #2

Title: Uncertainties in fertilizer-induced emissions of soil nitrogen oxide and the associated impacts on ground-level ozone and methane

MS number: egusphere-2025-1416

Authors: Cheng Gong, Yan Wang, Hanqin Tian, Sian Kou-Giesbrecht, Nicolas Vuichard, and Sönke Zaehle

Comments:

The manuscript titled “Uncertainties in fertilizer-induced emissions of soil nitrogen oxide and the associated impacts on ground-level ozone and methane,” written by Gong et al., quantifies the uncertainties in soil NO_x emissions induced by N fertilizer application (SNO_x-Fer) using different estimation approaches and investigates the associated impacts on the simulation of global O₃ and CH₄ concentrations. Overall, this manuscript is well-structured, and the conclusion is important. However, I would like to raise two major concerns and several minor suggestions for improvement.

Response:

We appreciate reviewer’s acknowledgment on the importance of our work and the constructive comments to help further improve this work. The manuscript has been revised accordingly. Please see our point-to-point response below.

Major concerns:

I can tell by Figures 2, 3, 4, and Section 4.2 that, in general, regions with higher SNO_x-Fer have higher O₃ enhancement. Is this an approximately linear relationship? Does this relationship vary across different sensitivity experiments and different regions? Providing a more detailed analysis of the response of the O₃ simulation to NO_x estimations would further highlight the importance of this work. The same concern also applies to the OH/CH₄ simulation.

Response:

Thank you for this valuable point. The responses of O₃ to NO_x changes could vary a lot depending on the local NO_x/VOC ratios, the magnitude of NO_x perturbation and metrological variations. Therefore, it is not a simply linear relationship. We agree that the analysis you suggested could help us better understand O₃ in which region is more sensitive to the SNO_x-Fer changes, at least in the GEOS-Chem model. Here we further examined the monthly MDA8 O₃ changes in response to SNO_x-Fer across all simulated grids in four representative hotspot regions (Fig. R1). Our results show the sensitivities of O₃ to SNO_x-Fer changes are all positive, i.e. O₃ increase with enhanced SNO_x-Fer, which might because of the relatively coarse resolution, but vary a lot among different regions and different SNO_x-Fer estimating approaches. The Eastern U.S. in general has the strongest sensitivity, while sensitivities in the rest three regions are similar. Nevertheless, the R² of the linear regressions are not very high (0.04–0.68), indicating the strong non-linearity behind the O₃-NO_x-VOCs chemistry.

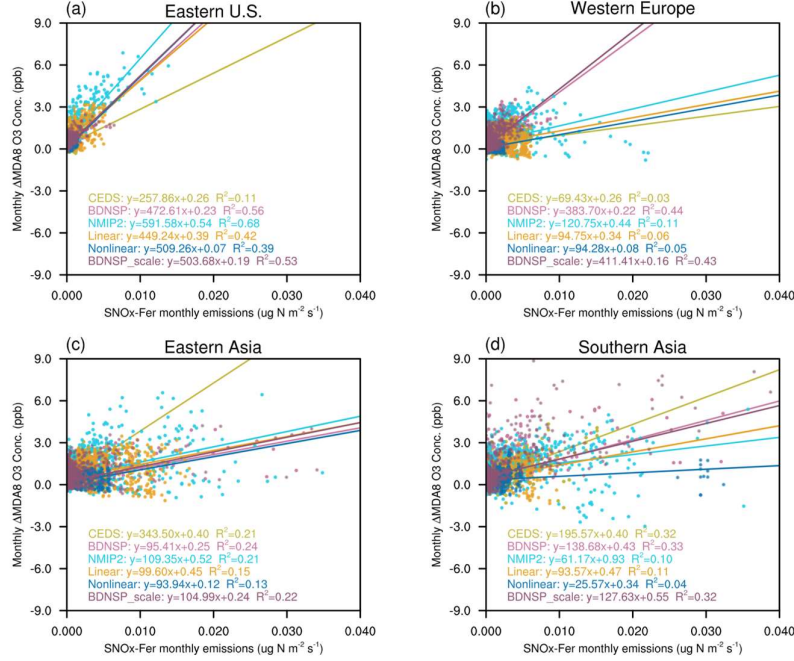


Figure R1. The sensitivity of changes in monthly MDA8 O₃ concentrations (ppbv) to the SNO_x-Fer changes (ug N m⁻² s⁻¹) among different approaches in four agricultural hotspot regions. Each dot indicates the monthly SNO_x-Fer emissions and associated monthly MDA8 O₃ changes induced by SNO_x-Fer on a simulated grid. The different SNO_x-Fer estimating approaches are indicated by lines with different colors.

Similar patterns are also found in the response of OH to SNO_x-Fer changes (Fig. R2). Furthermore, because CH₄ has much longer lifetime and thus the atmospheric transport could smooth the local CH₄ changes induced by varied OH, the regional CH₄ changes are more determined by the global signals rather than local SNO_x-Fer perturbation. Nevertheless, we could still find discrepancies in the responses of ground-level CH₄ to SNO_x-Fer among different regions and different SNO_x-Fer estimates (Fig. R3), indicating the strong non-linearity in CH₄-OH-NO_x chemistry.

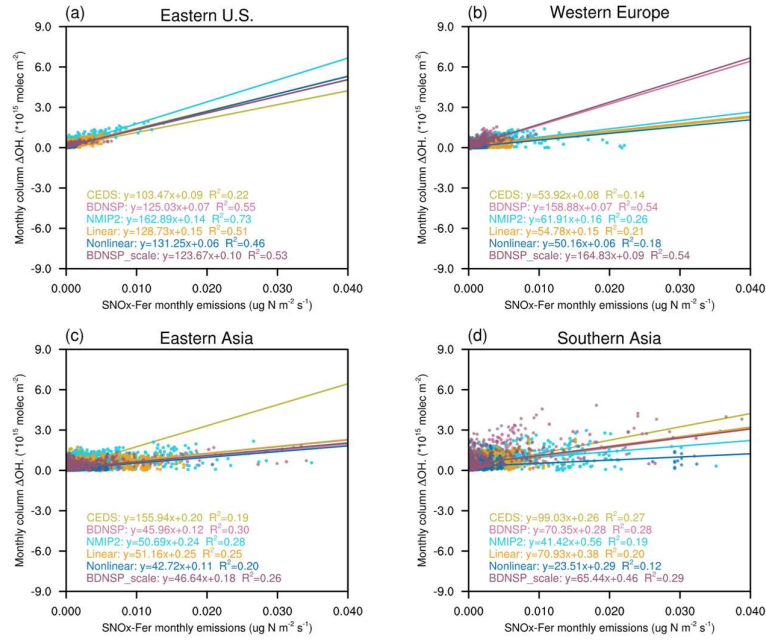


Figure R2. The sensitivity of changes in monthly column OH concentrations ($\times 10^{15}$ molec m^{-2}) to the SNO_x-Fer changes ($\mu g N m^{-2} s^{-1}$) among different approaches in four agricultural hotspot regions. Each dot indicates the monthly SNO_x-Fer emissions and associated monthly OH changes induced by SNO_x-Fer on a simulated grid. The different SNO_x-Fer estimating approaches are indicated by lines with different colors.

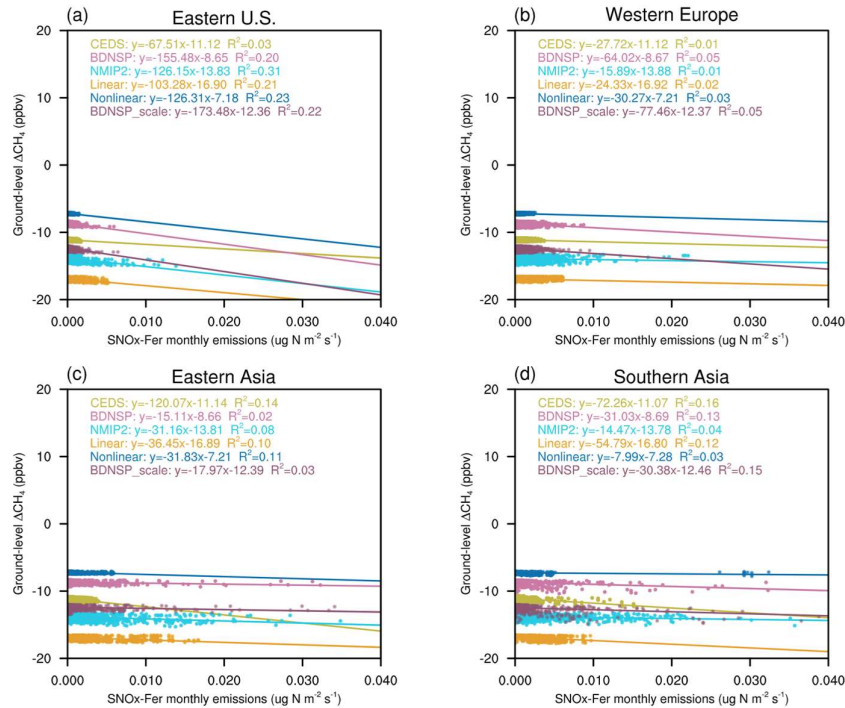


Figure R3. The sensitivity of changes in monthly ground-level CH₄ concentrations (ppbv) to the SNO_x-Fer changes ($\mu g N m^{-2} s^{-1}$) among different approaches in four agricultural hotspot regions.

Each dot indicates the monthly SNO_x-Fer emissions and associated monthly CH₄ changes induced by SNO_x-Fer on a simulated grid. The different SNO_x-Fer estimating approaches are indicated by lines with different colors.

We have added discussions about this:

‘This range of responses leads to an enhancement in summertime surface MDA8 O₃ concentrations of 0.3 to 3.3 ppbv (0.2%-7.0%) in agricultural hotspot regions. The O₃ enhancement is highest in eastern U.S., while it is not only determined by the SNO_x-Fer emissions, but also the diverging sensitivities of O₃ to NO_x depending on different chemical regime in GEOS-Chem (Fig. S6). The varied SNO_x-Fer estimates also lead to a reduction in global CH₄ concentrations of 6.7 ppbv (0.4%) to 16.6 ppbv (0.9%) ...’

In Lines 125–129, the growing season is defined using monthly-mean 2-meter temperature and leaf area index instead of using some crop calendar datasets. While this approach is straightforward and climate-driven, it may oversimplify the actual crop phenology in diverse agricultural systems. Given that the rates of N inputs are set to zero during the non-growing season, this definition directly controls the temporal pattern of fertilizer application and thus significantly affects the estimates of fertilizer-induced NO_x emissions. If crop-specific growing seasons are not distinguishable in this study, the authors should at least discuss the potential implications of this assumption in the discussion section.

Response:

We acknowledge that reviewer raised an important issue when applying the EF method to assess SNO_x-Fer, while the rest SNO_x-Fer estimating approaches in this study are not dependent on this growing season definition. However, we are afraid that the more detailed crop calendar dataset (e.g. Minoli et al., 2019; Minoli et al., 2022) may still not be sufficient. It is not only because the consistent calendar datasets of pasture and rangeland are not available, but more importantly, the fertilizer management (e.g. how was the annual total fertilizer application distributed across months) should be the key factor that influence the seasonal cycles of SNO_x-Fer. However, to our knowledge, the dataset that records the seasonal fertilizer distribution is not available yet.

The definition of growing season used in this study is not only depending on the climate, but also relies on the realistic plant greenness index (e.g. LAI). Such method is also widely used to identify the phenology of agricultural land cover (e.g. used by the Food and Agriculture Organization of the United Nations <https://agriculture.africageoportal.com/datasets/d9944082e3c6421098464b1016fbae58/about>). As this study only focus on the annual and monthly (not day-to-day) variations of SNO_x-Fer, we believe that such simplified definition is sufficient to capture the dominant pattern.

We have explicitly addressed the seasonality analysis among different SNO_x-Fer approaches in the revised manuscript (Sect. 4.2 and Figs. 4-5. See the response to Reviewer #1). We discuss the uncertainties induced by the definition of growing season in the revised manuscript as below:

‘In the EF approaches, the growing season is determined only by temperature and greenness in this study, which could result in a mismatch with the real crop or pasture calendar, especially ignoring the multiple-harvest crops per year. A refined calendar could further improve the prediction of SNO_x-Fer seasonality.’

Minor points:

Are there any top-down methods for estimating NO_x emissions? If so, it would be beneficial for the authors also to describe it in the introduction, allowing for a more comprehensive review of the estimation approaches.

Response:

Top-down method could more precisely assess the total NO_x emissions from all sources. However, distinguishing different sources is always challenging. Although there are some studies are able to isolate soil NO_x emissions by assuming the fossil-fuel emissions inventory is accurate or only applying the retrieval in pixels without significant industrial activities (e.g. Bertram et al., 2005; Lin et al., 2024), it is still very difficult to further isolate the background and fertilizer-induced soil NO_x emissions.

We have added it as a discussion point in the revised manuscript:

‘... Last but not least, the top-down retrievals of NO_x emissions based on satellite NO₂ products could also have the potential to better constrain SNO_x-Fer, while gaps remained in how to precisely isolate the soil NO_x emissions (Bertram et al., 2005; Lin et al., 2024) and even the fertilizer contributions from the total NO_x sources. Synergizing the top-down NO_x retrievals with ultra-high resolutions, where it can be assumed that the atmospheric NO_x is dominantly affected by the soil sources in agricultural regions, with spatiotemporally detailed fertilizer management dataset could be one possible solution. However, more work is definitely needed to integrate such a big data in the future.’

Section 2.2: Consider also adding one or two sentences to describe why this specific inventory is chosen.

Response:

We have added the reason as:

‘We use the CEDS (Hoesly et al., 2018) for assessing the fertilizer-induced soil NO_x emissions in the emission inventories. CEDS is one of the most state-of-art emission inventories that comprehensively assess the sources of dominant air pollutants from pre-industrial period to present days, which has been used as the standard emission inventory to drive CMIP6 models.’

Figure 1: Consider merging (a) and (b) into a single figure using a secondary Y-axis for fertilizer input, which would help the reader better interpret the relationship between nitrogen inputs and SNO_x-Fer across approaches.

Response:

We have revised the Fig. 1 following both of your and the other reviewer’s comment as:

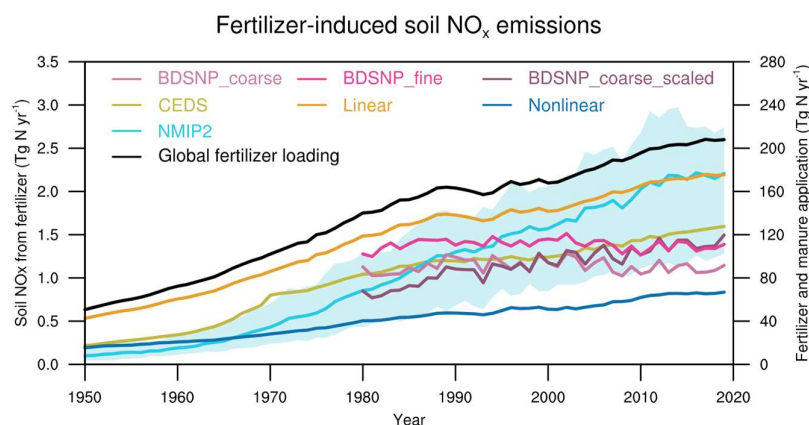


Figure 1. Global estimates of N fertilizer-induced soil NO_x emissions by different approaches. The black line (right Y axis) indicates global annual-mean N synthetic fertilizer and manure inputs over 1950-2019 assessed from the HaNi dataset. The rest lines (left Y axis) indicate the N fertilizer-induced soil NO_x emissions over 1950-2019 estimated by different approaches, including emission inventory (CEDS), linear and non-linear EF, the widely-used CTM parameterization with coarse resolution (2°×2.5°, BDSNP_coarse), fine resolution (0.5°×0.625°, BDSNP_fine) and interannually varied N availability (BNDSP_coarse_scaled), and the TBM ensembles (NMIP2). The light cyan shadows indicate the spread across three different TBMs in NMIP2.

Section 4.2 and 4.3: When reporting changes in O₃ and CH₄ concentrations, consider also providing percentage changes rather than only providing the ppbv changes.

Response:

The percentage changes relative to the simulated concentrations have been added accordingly.

The HaNi dataset provided N inputs for cropland, pasture, and rangeland. Consider also providing the NO_x emissions from cropland, pasture, and rangeland in the Supplementary Information.

Response:

Added.

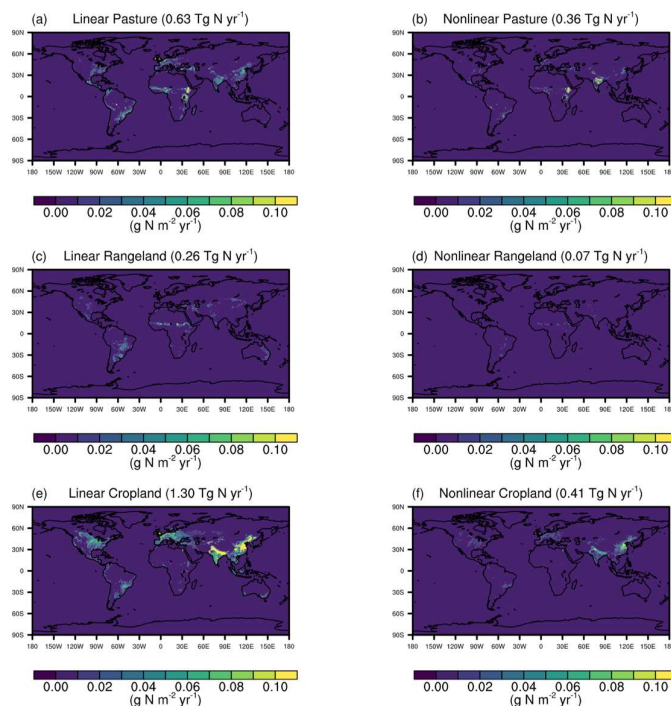


Figure S2. The spatial patterns of SNO_x-Fer in pasture, rangeland and crops estimated by linear EF and non-linear EF approaches in 2019. The global total budget of each estimate is given in the sub-titles.

Given the large differences in simulated surface O₃ concentrations across the different SNO_x-Fer estimation methods (e.g., Fig. 3 and 4), it would be valuable to include a brief comparison with surface O₃ observations. While a full validation is beyond the scope of this study, even a qualitative comparison could help indicate which NO_x emission estimation method may better reproduce observed O₃ levels in key agricultural regions.

Response:

We compared our simulated summertime monthly MDA8 O₃ concentrations against the ground-level observations in Eastern U.S. (<https://www.epa.gov/aqs>), Western Europe (<https://ebas-data.nilu.no/Default.aspx>) and China (<https://www.cnemc.cn/en/>) (Figure S7). The site-level observational O₃ concentrations are averaged on each simulated grid. Note that the differences in SNO_x-Fer estimates are not sufficient to explain the model systematic bias, but they are likely induced by uncertainties in other processes such as non-linear chemistry, transport and deposition.

We think it is an open question about how to properly evaluate which SNO_x-Fer approaches are accurate. As we also mentioned above, the high-resolution top-down NO_x retrievals could be another possible solution but there are also uncertainties in how to precisely isolate the SNO_x-Fer from the total NO_x emissions. We are glad to add discussions in the end to point out this question:

‘Beyond the uncertainties remaining in different SNO_x-Fer estimating approaches, an important but also difficult question is how to better evaluate the performances of each methods, especially in the regional and global scales. The first-hand meta-data collected from the field experiments is actually not an independent source, as it has been used to establish both of the linear and nonlinear EF methods. More importantly, most of the field experiments are manipulation experiments with artificial fertilizer gradients, which may not fully represent the real-world spatiotemporally varied SNO_x-Fer. Furthermore, we use O₃ data from the national or continental air quality observational networks to evaluate simulated

O₃ concentrations as a potential consistency check of the SNO_x-Fer (Fig. S7). However, the uncertainties in SNO_x-Fer are expected to be far less important relative to the uncertainties in the nonlinearity of atmospheric chemistry, emissions of BVOCs or the deposition processes, which together determined the biases between observational and simulated O₃ concentrations. As a result, it is inappropriate to determine the best SNO_x-Fer estimate as the one with the best statistic metrics in O₃ simulation. Moreover, most of the sites that monitor air pollutants are located in the urban regions, where the industrial impacts are far more important than the agricultural sources. A real-time O₃ observational network in the cropland or pasture would be crucial to advance the understandings in SNO_x-Fer and the associated impacts on air quality. Last but not least, the top-down retrievals of NO_x emissions based on satellite NO₂ products could also have the potential to better constrain SNO_x-Fer, while gaps remained in how to precisely isolate the soil NO_x emissions (Bertram et al., 2005; Lin et al., 2024) and even the fertilizer contributions from the total NO_x sources. Synergizing spatiotemporally detailed fertilizer management dataset with the top-down NO_x retrievals with ultra-high resolutions, where the atmospheric NO_x can be assumed to be dominantly affected by the soil sources in agricultural regions, could be one possible solution. However, more work is needed to integrate such a big data in the future.’

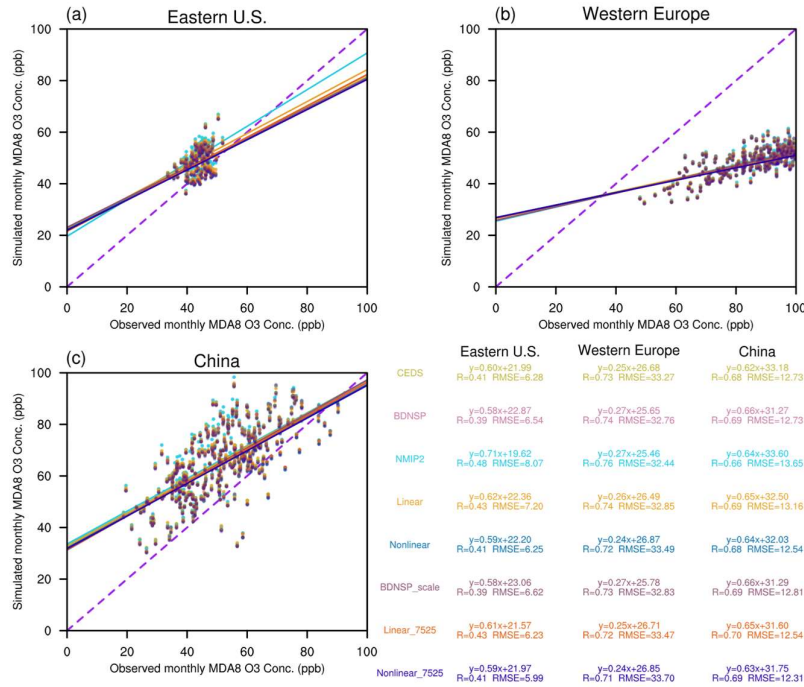


Figure S7. The comparison of monthly-averaged O₃ MDA8 concentrations between site-level observations and the GEOS-Chem simulations. The observational dataset in Eastern U.S., Western Europe and China are assessed via the Air Quality System (AQS, <https://www.epa.gov/aqs>), European Monitoring and Evaluation Programme (EMEP, <https://ebas-data.nilu.no/Default.aspx>) and China National Environmental Monitoring Centre (CNEMC, <https://www.cnemc.cn/en/>), respectively. Each dot indicates one simulated grid, where the observed O₃ concentrations are calculated by averaging all observational sites. The GEOS-Chem sensitivity experiments with different SNO_x-Fer estimating approaches are indicated by different colors.

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