

## 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<sub>x</sub> 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<sub>x</sub>-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<sub>x</sub> 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<sub>x</sub>-Fer simulated by CLASSIC, OCN and ORCHIDEE were applied instead of the model ensemble mean. We explicitly showed the TBM variabilities in SNO<sub>x</sub>-Fer and their impacts on O<sub>3</sub> and CH<sub>4</sub> 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<sub>x</sub>-Fer estimating methods rather than the TBM spread in this study. The NMIP spread of SNO<sub>x</sub>-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<sub>x</sub> 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<sub>x</sub> emissions and also O<sub>3</sub> and CH<sub>4</sub>.

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 O<sub>3</sub> 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 approch	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 <sub>x</sub> 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 <sub>x</sub> -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 <sub>x</sub> -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 <sub>x</sub> -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<sub>x</sub>-Fer and impacts on O<sub>3</sub>:

#### ‘4.2 The seasonal cycle of SNO<sub>x</sub>-Fer and the associated impact on O<sub>3</sub> concentrations

Figure 4 shows the seasonality of SNO<sub>x</sub>-Fer in four agricultural hotspot regions among different SNO<sub>x</sub>-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<sub>x</sub>-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<sub>x</sub>-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<sub>x</sub>-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 O<sub>3</sub> 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 O<sub>3</sub> shows during the north-hemispheric summertime (June–August) for most of the estimating approaches in three temperate regions, when the absolute O<sub>3</sub> 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 O<sub>3</sub> enhancement in both Western Europe and Eastern Asia (Figs. 5b and 5c). The weak sensitivity of O<sub>3</sub> to NO<sub>x</sub> 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 O<sub>3</sub> to SNOx-Fer could also depend on regions (e.g. O<sub>3</sub> enhancement also peaks during spring in Linear\_7525 in Eastern U.S., Fig. 5a), spatial simulating resolution or different modelling chemical mechanisms. The O<sub>3</sub> 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 O<sub>3</sub> enhancement during May to July relative to February to April.

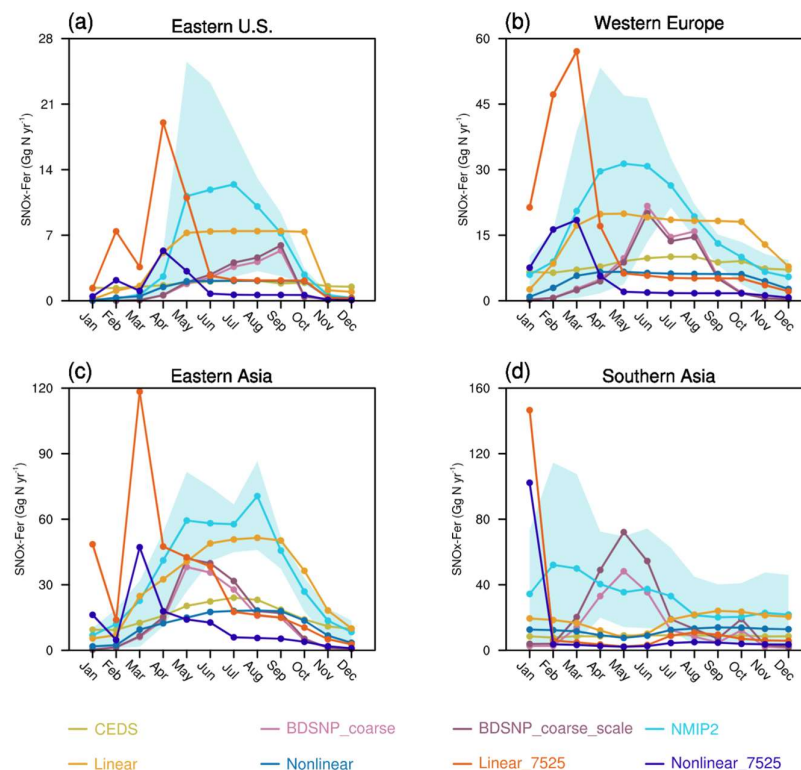


Figure 4. The monthly regional SNOx-Fer (Gg N yr<sup>-1</sup>) in the (a) Eastern U.S., (b) Western Europe, (c) Eastern Asia and (d) Southern Asia with different SNOx-Fer estimating approaches. The cyan-blue shades indicate the spreads among three different TBM models (CLASSIC, OCN and ORCHIDEE) in the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

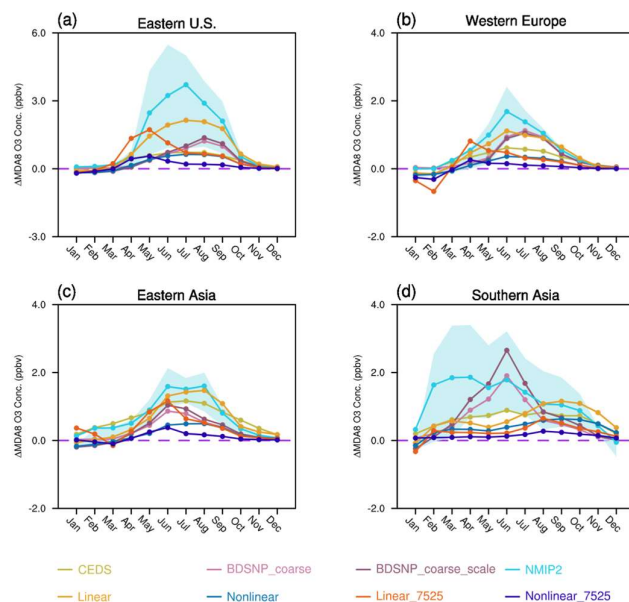


Figure 5. The regionally-averaged monthly MDA8 O<sub>3</sub> changes (ppbv) induced by SNOx-Fer in the (a) Eastern U.S., (b) Western Europe, (c) Eastern Asia and (d) Southern Asia with different SNOx-Fer estimating approaches. The cyan-blue shades indicate the spreads among three different TBM models (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 O<sub>3</sub> 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<sub>x</sub>-VOCs-O<sub>3</sub> chemical sensitivity regimes could be determined by not only soil NO<sub>x</sub> emissions, but also other anthropogenic and biogenic emissions of NO<sub>x</sub> and VOCs, as well as the climate seasonal variations. Therefore, the seasonal cycles of the enhancement of O<sub>3</sub> 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 NO<sub>x</sub> 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<sub>x</sub>-Fer hotspots in Fig. 2. Meanwhile, CH<sub>4</sub> has a much longer lifetime (~9 years) than O<sub>3</sub> 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<sub>x</sub>-Fer did not significantly alter the CH<sub>4</sub> 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<sub>x</sub> induced the reduction of global averaged CH<sub>4</sub> 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<sub>x</sub>-Fer among different estimating approaches (Fig. 3). Because CH<sub>4</sub> has a significantly longer atmospheric lifetime than either OH or NO<sub>x</sub>, the spatial differences in the impacts of SNO<sub>x</sub>-Fer on CH<sub>4</sub> concentrations are insignificant (Fig. S4). As a result, we only focus on the globally averaged changes in CH<sub>4</sub> 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<sub>x</sub> on CH<sub>4</sub> concentrations:

‘The impacts of the changes in short-lived air pollutants on global CH<sub>4</sub> budget have attracted increasing attention in recent years (Peng et al., 2022; Zhao et al., 2025), where NO<sub>x</sub> is one of the most important drivers. However, it should be noted that the sensitivity of CH<sub>4</sub> lifetime to NO<sub>x</sub> emissions varies substantially among atmospheric chemistry models from -25% to -46% in response to the total NO<sub>x</sub> changes from pre-industrial to present-day period (Thornhill et al., 2021). Because few studies investigated how NO<sub>x</sub> from agricultural sources affects CH<sub>4</sub>, it is difficult to assess if the overall impacts of SNO<sub>x</sub>-Fer on CH<sub>4</sub> 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<sub>x</sub>-Fer could be one uncertain but important source in calculating future changes of the global CH<sub>4</sub> budget, the importance of which could be increasing with future continuing reduction in fossil-fuel NO<sub>x</sub> 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<sub>x</sub> emissions?*

**Response:**

We agree that the resolution will be a factor for soil NO<sub>x</sub> emissions. However, we would like to clarify that most of the SNO<sub>x</sub>-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<sub>x</sub>-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<sub>x</sub>-Fer:

‘For the modelling of SNO<sub>x</sub>-Fer, on the one hand, recent developments of the parameterization of BDSNP in CTMs focused more on the soil NO<sub>x</sub> 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.



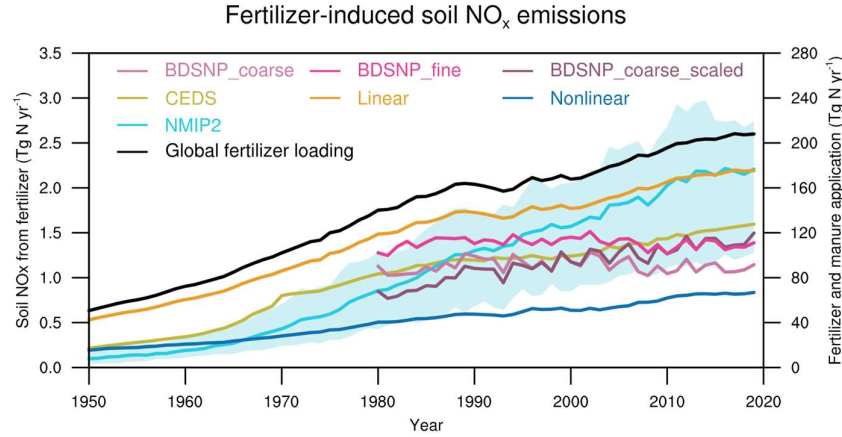


Figure 1. Global estimates of N fertilizer-induced soil NO<sub>x</sub> emissions by different approaches. The black line (right Y axis) indicates global annual-mean N synthetic fertilizer and manure inputs over 1950-2019 assessed from the HaNi dataset. The rest lines (left Y axis) indicate the N fertilizer-induced soil NO<sub>x</sub> 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<sub>x</sub>’. 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<sub>x</sub>; 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<sub>x</sub> 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<sup>-1</sup>) is NO emissions for non-zero N input,  $E_0$  (kg N ha<sup>-1</sup>) is NO emissions for zero N input (control), and  $N$  (kg N ha<sup>-1</sup>) 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  $\Delta EF$  (% per kg N ha<sup>-1</sup>) are the intercept and slope of the effect function, respectively. The  $\Delta EF$  of this relationship could be deemed as the degree of nonlinearity of the NO emission response to the N rate: zero  $\Delta EF$  means that NO emissions grow linearly with the N rate (i.e., the constant EF), a positive  $\Delta EF$  indicates that NO emissions increase faster than a linear response with N rate (i.e., the increasing EF), and a negative  $\Delta EF$  indicates that NO emissions increase slower than a linear response (i.e., the decreasing EF).

$\Delta EF$  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<sub>x</sub> emissions, but also soil N<sub>2</sub>O (Shcherbak et al., 2014) and NH<sub>3</sub> 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 (NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>) 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 (NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>; 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<sub>x</sub>)’

*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.

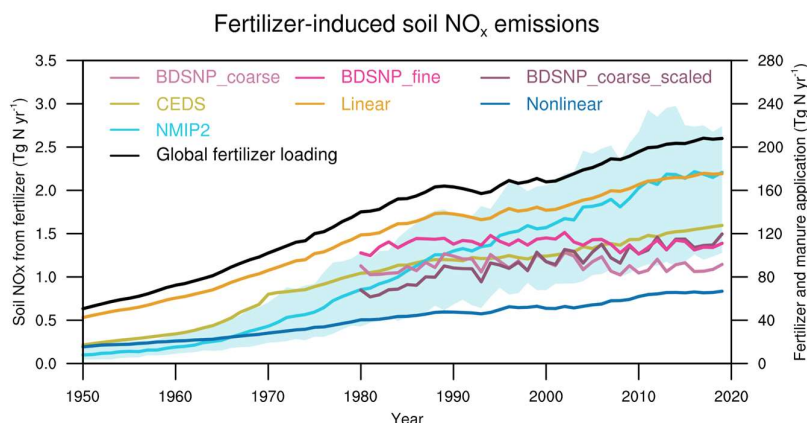


Figure 1. Global estimates of N fertilizer-induced soil NO<sub>x</sub> emissions by different approaches. The black line (right Y axis) indicates global annual-mean N synthetic fertilizer and manure inputs over 1950-2019 assessed from the HaNi dataset. The rest lines (left Y axis) indicate the N fertilizer-induced soil NO<sub>x</sub> 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:

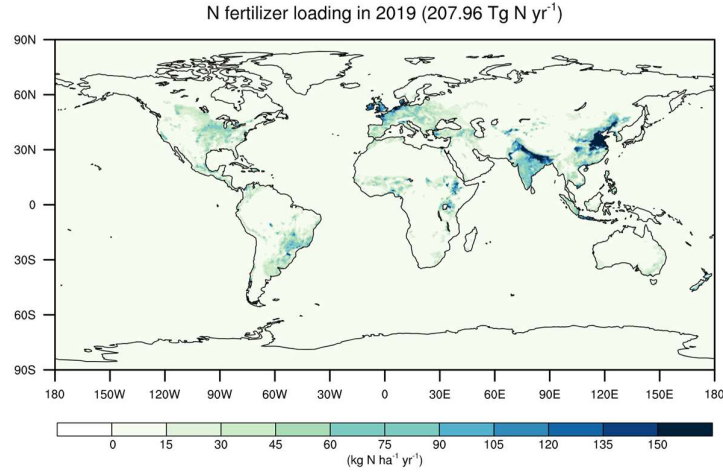


Figure 2. The global spatial patterns of N synthetic fertilizer and manure application in 2019 from the HaNi dataset.

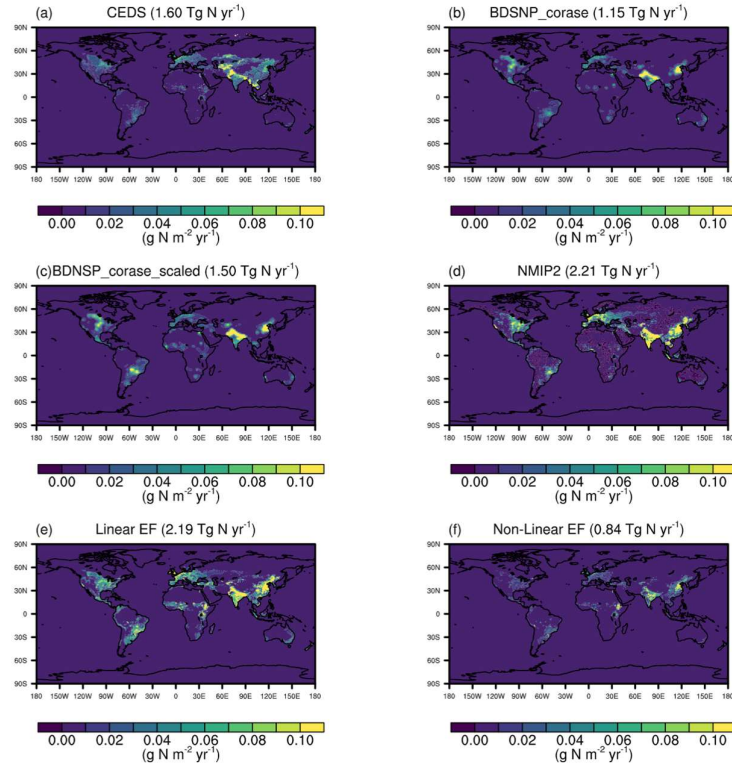


Figure 3. The N-fertilization induced soil NO<sub>x</sub> emissions estimated by different approaches in 2019. (a) - (f) The soil NO<sub>x</sub> emissions induced by N fertilizer estimated by the CEDS agricultural sector, the default BDSNP scheme in GEOS-Chem with coarse resolution (2°×2.5°), the coarse-resolution 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<sub>x</sub>-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<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub> emissions and the associated impacts on global O<sub>3</sub> and CH<sub>4</sub> 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<sub>x</sub> 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<sub>x</sub>-Fer, our results revealed the uncertainties in quantifying SNO<sub>x</sub>-Fer and associated important implications in simulating regional air quality and the global greenhouse gas CH<sub>4</sub>.'

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