

July 6, 2025

Dear Editor,

We have received the comments from the reviewers of the manuscript. We greatly appreciate your consideration and the reviewers' comments. Below are our responses and the revisions that we have made in the manuscript.

Thank you for your efforts on this manuscript. We look forward to hearing from you.

Best regards,

Guohui Li, PhD

Response to Reviewer #1

We thank the reviewer for the helpful comments and insightful suggestions to improve the manuscript. We have revised the manuscript following the suggestion, as described below.

I appreciate the authors' thorough efforts to address my previous concerns. I have only one minor suggestion remaining.

Lines 184–201: I recommend reorganizing this paragraph to improve the logical flow when introducing OMI and IASI and explaining how the satellite data were processed. At present, the section is somewhat unclear. For example, while both satellites are described as operating in sun-synchronous polar orbits with local overpass times around 13:45 (local time, LT) (once daily) and 9:30 am / 9:30 pm (twice daily), respectively, over North China, the text does not explicitly indicate which overpass time refers to which satellite and which pass time was used for the IASI-NH₃ data in this study. Additionally, there is some redundancy, such as the repeated mention of the cloud fraction threshold (<30%) in both Line 187 and Line 194.

Overall, I consider the manuscript suitable for acceptance after minor revisions.

Response: We have revised this paragraph to improve the logical flow in Lines 189-208: *“Satellite-derived tropospheric NO₂ columns are from OMI hosted by the Aura satellite that is launched by the National Aeronautics and Space Administration (NASA). The Level-3 product, where pixel level data of good quality are binned and “averaged” into 0.25°×0.25° grids, was retrieved and analyzed in the present study. The satellite operates in a sun-synchronous polar orbit and has a local overpass time of around 13:45 (local time, LT) in North China. The dataset is for all atmospheric conditions, and for sky conditions with cloud fraction less than 30% (https://cmr.earthdata.nasa.gov/search/concepts/C1266136111-GES_DISC.html). The dataset has a spatial resolution of 13 km × 24 km, with a temporal coverage of 2005-2022 (Lamsal et al., 2021). Note that the number of pixels included in NO₂ retrievals changes over*

time because of the increase in the number of pixels affected by the row anomaly issue, making the data unsuitable for trend analysis and possibly introducing uncertainty in seasonal averages. The Level-2 product of NH_3 columns is employed, which is from the Space Administration and the Infrared Atmospheric Sounding Interferometer (IASI) hosted on the MetOp series of satellites. The satellite also operates in a sun-synchronous polar orbit and has a local overpass time of around 9:30 am and 9:30 pm in North China (twice a day). We construct a $0.125^\circ \times 0.125^\circ$ mesh grid and calculate the average of the NH_3 columns from IASI within each grid cell (Clarisse et al., 2023). Low-quality satellite data are filtered out due to the interference of clouds. To cover all the domain (Figure 1), the data used in this study are averaged into seven-day mean datasets of NO_2 and NH_3 columns with a non-overlapping 7-day window during 2007-2021. The data are interpolated into the model grids using bilinear interpolation.”

Response to Reviewer #2

We thank the reviewer for the helpful comments and insightful suggestions to improve the manuscript. We have revised the manuscript following the suggestion, as described below.

The authors have conducted a very thorough revision of the manuscript in response to reviewer comments, and I thank them for their efforts. I do still think a few things need some additional consideration, the most important of which are the interpretation of the NH_3 time series, the NO_x titration discussion, and the BDSNP default fertilizer assumptions. All line numbers refer to the tracked changes version of the manuscript. Please ignore any highlighting below, which is copied from the tracked changes version of the manuscript.

General comments:

(1) NH_3 time series: Figure S4 and S5 (the NH_3 time series): I mentioned this in part in the first review, but I think the NH_3 time series are extremely important for interpreting the NO_2 peaks—so much so that I continue to (strongly) argue for overlaying the NH_3 time series over the NO_2 time series in Figure 2 of the main manuscript. I'd also love to see a version of Figure 6 for the entire spring (March through June).

The issue is that NH_3 peaks are occurring in June, after the first topdressing application, with *much* smaller peaks associated with the planting fertilizer application. This is consistent with a split application in which considerably more fertilizer is applied at topdressing than at planting. But it differs from the NO_2 time series, which has a larger peak in March than in June.

To me, the NH_3 time series looks like a very convincing signal of fertilizer emissions. The essential question here is why the two time series don't have similar dynamics. Even if no clear explanation is found, it needs to be acknowledged that the ammonia time series raises some questions about whether the March NO_2 peak does indeed represent a fertilizer pulse.

As it is, I'm not sure we understand what's happening in this system. Possibilities that come to mind that might be worth exploring to explain the differences in the two time series could include some combination of 1) a fossil fuel source of NO_x in March, 2) elevated background soil NO_x emissions in March associated with the 'spring thaw' period in which soil microbes become active during a period where there is no competition for N from plants, and after the winter during which N is expected to accumulate in soils (this is separate from any emissions associated with a freeze/thaw transition), 3) differences in fertilizer type (i.e., fertilizers with different potentials for ammonia volatility) between March and June, and 4) differences in application method (e.g., banding or deep soil placement vs broadcasting, particularly if without incorporation) in March and June, with differences in fertilizer amounts in March and June potentially contributing in cases 3 and/or 4.

And it's important to remember that BDSNP is going to be heavily—and it seems inappropriately—biased towards having a fertilizer-induced emission peak **only** at planting/green-up. But you do have good correspondence between BDSNP and OMI in 2020 (Fig 3), which is a bit of a mystery to me (and a real limitation of having only a single year of simulations).

If it is not possible to fully resolve this issue in this manuscript, in future work, it might be interesting to include a dynamic bi-directional NH_3 scheme along with BDSNP to explore the fertilizer pulsing question.

Response: We have included the NH_3 time series over the NO_2 time series in Figure 2 and updated the figure caption accordingly. Figure S4 has been removed in the revised version because it presents the same message as the added time series in Figure 2. Accordingly, we have included discussion on the discrepancy between the NO_2 and NH_3 time series in Lines 272-278: *“We note that the two time series don't have similar seasonal dynamics, which may raise some questions about whether the March NO_2 peaks represent fertilizer pulses. This*

discrepancy could be due to several factors: (1) contributions from fossil fuel-derived NO_x, (2) elevated background soil NO_x emissions during the “spring thaw” period, (3) differences in fertilizer type, as fertilizers vary in their potential for ammonia volatilization, and (4) variation in application methods, e.g., banding or deep soil placement vs broadcasting, especially when the latter is done without incorporation.”

The uncertainties in the BDSNP scheme have been discussed in the revised manuscript, which is referred to the response to Comment 3. Regarding the future implication, we have included discussion in Lines 599-602: *“The discrepancies between NO₂ and NH₃ column densities suggest substantial differences in the soil emission mechanisms of NO_x and NH₃, especially after fertilization. Future studies could incorporate a dynamic bidirectional NH₃ scheme alongside the BDSNP scheme to further investigate the nature of fertilizer-induced emission pulses.”*

We have also updated Figure 6 showing the results for the entire simulation period (February through April, 2020). The description of the results in the manuscripts have been updated accordingly in Lines 379-386: *“When there are no soil NO_x emissions from agricultural fertilization, the simulated NO₂ concentration is significantly lower than the observed by 9.4 μg m⁻³ during February through April, 2020. While considering these emissions, the mean bias (MB, Text S2) between the simulation and the observation decreases to 2.6 μg m⁻³, and the index of agreement (IOA, Text S2) also increases from 0.48 to 0.78. Similarly, the simulated NH₃ concentration is in good agreement with the observed when the soil NH₃ emission related to agricultural fertilization is involved, e.g., the MB decreases from -12.0 ppb to -4.4 ppb, and the IOA increases from 0.56 to 0.64 (Figures 6c and 6d).”*

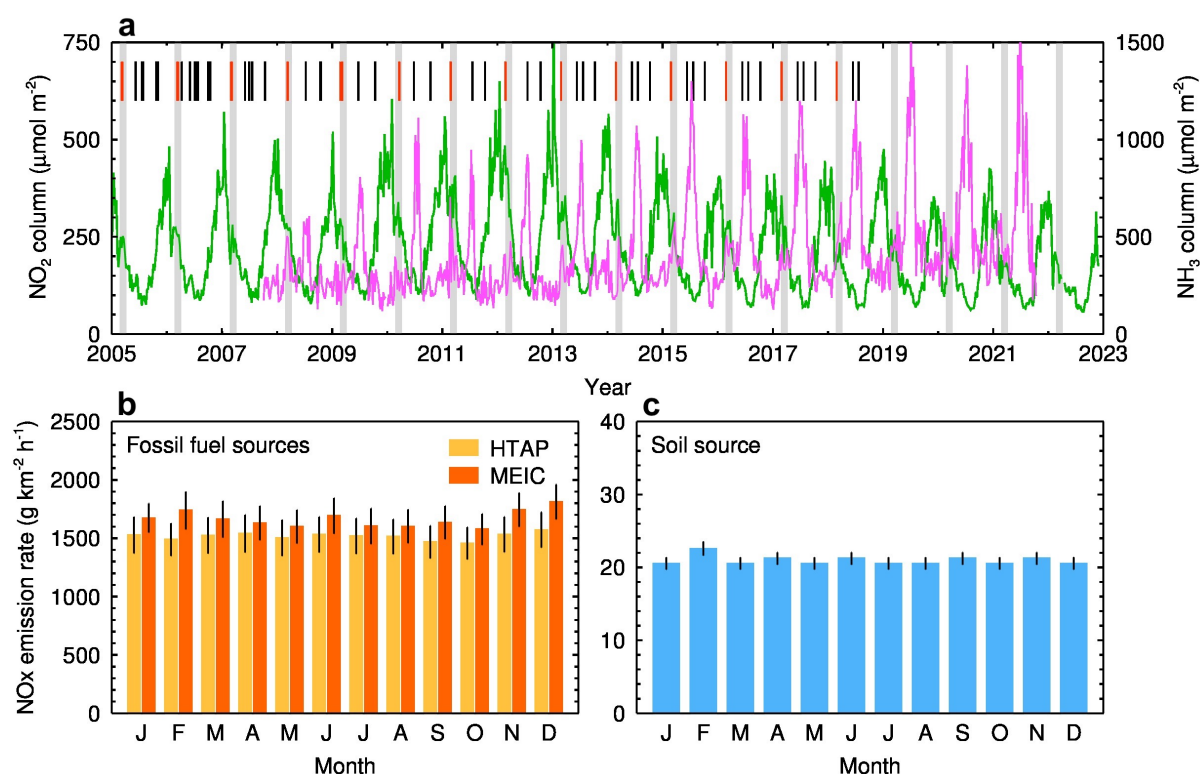


Figure 2. NO_2 column pulses in March, NH_3 column variation, and NO_x emissions from fossil fuel and soil sources over the NCP. (a) Long-term variation of seven-day mean tropospheric NO_2 column observed by OMI during 2005-2022 (green) and NH_3 column retrieved from IASI during 2007-2022 (pink). Intersections of the gray bars and the green lines denote a sub-peak of NO_2 column occurred in each March, and the short bars represent the timing record for agricultural fertilization at Fengqiu station in the NCP, of which the red ones indicate the fertilization period in early spring. (b) Monthly mean NO_x emission rates with $\pm 1\sigma$ standard deviation (SD) in two sets of anthropogenic emission inventories, the HTAP v3 (2005-2018, orange) and MEIC v1.3 (2008-2017, red). (c) Same as (b), but for NO_x emission rates from soils in the HTAP v3 inventory (2005-2018).

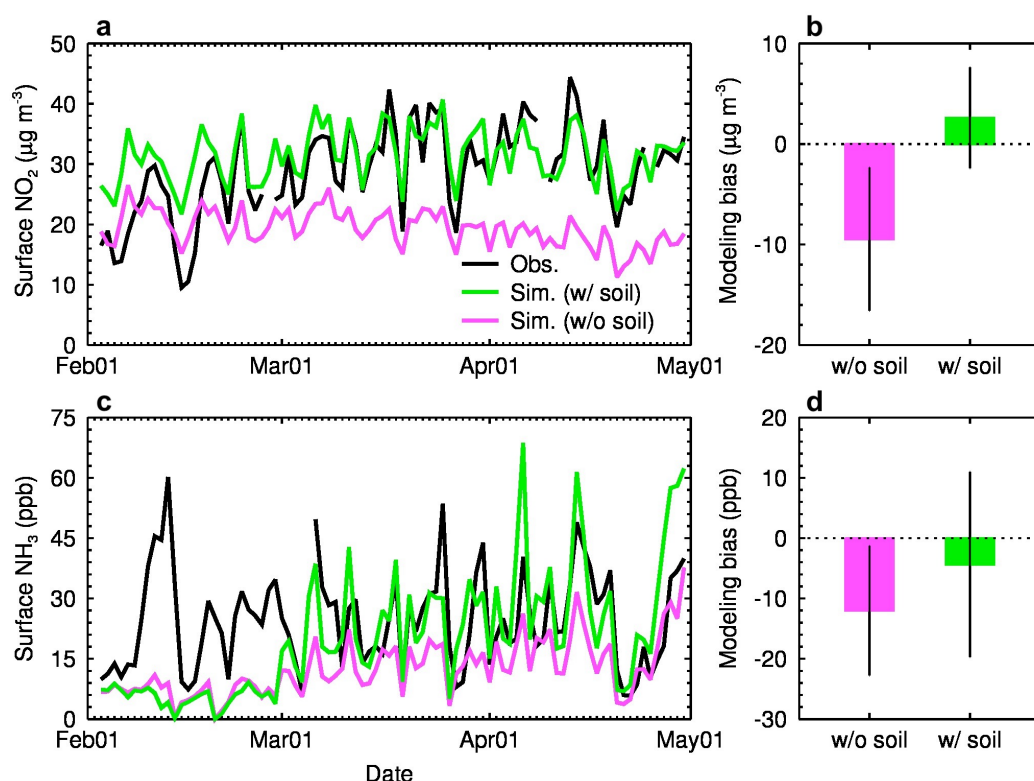


Figure 6. Contribution of soil emissions from agricultural fertilization on surface NO_2 and NH_3 during February through April 2020 over the NCP. (a-b) Change in surface NO_2 concentration with (green) and without (pink) soil NO_x emission from agricultural fertilization, the black line in (a) is for observed surface NO_2 concentration. (c-d) Same as (a-b), but for NH_3 . The error bar in (b and d) denotes $\pm 1\sigma$. NO_2 observations are averaged over the 141 monitoring stations in the study area from the CNEMC network. NH_3 observations are from the rural Xianghe station (Figure 1). According to in-situ measurements on NO_2 and NH_3 , the units for NO_2 and NH_3 concentrations are $\mu\text{g m}^{-3}$ and ppb, respectively.

(2) NO_x titration discussion: In general, I'd really like to see more discussion of this and better contextualizing--it's a very interesting result ($15.6 \mu\text{g m}^{-3}$ reductions) that needs more interrogation. Attributing NO_x titration occurring in March but not in the summer to solar insolation is not convincing as currently argued. I would have thought NO_x titration of ozone to be more related to VOC: NO_x ratios than insolation, and I wouldn't expect daytime insolation

in March to be all that different in summer at 33N-40N, and the argument being made here is that the insolation is different enough that instead of causing a $15.6 \mu\text{g m}^{-3}$ reduction in daytime ozone in March, NO_x emissions in the NCP cause an increase in daytime ozone concentrations in summer. Left out of the discussion is how large the increase in VOC emissions is between March (when it is presumably negligible) and summer (when it is presumably at its annual peak), and whether that's enough to shift the ozone regime—to me that sounds like a more likely culprit than insolation. The model could help answer these questions. Patterns in ozone measurements at the surface or from space that support these dynamics would also be helpful for supporting and understanding the model results.

Huang et al. 2023 (<https://doi.org/10.5194/acp-23-14919-2023>) might be a good place to start (but not end) the discussion: it includes evidence for soil NO_x leading to ozone reduction over the NCP in section 3.3, e.g., “With a 25 % reduction in soil NO emissions, there was a widespread small decrease in monthly average MDA8 ozone concentration (ΔMDA8 : $-1.5 \pm 0.9 \mu\text{g m}^{-3}$), except across the NCP, where ozone showed a slight increase (up to $1.3 \mu\text{g m}^{-3}$) in the Shandong and Henan provinces.” I think that reference also describes the NCP as VOC limited, which is important to point out.

Purely optional, but it may also be interesting to discuss a bit more the implications of agricultural emissions for ozone and air quality goals in urban areas, especially as fossil fuel NO_x emissions decrease.

Response: We have revised the discussion from the aspect of O_3 formation sensitivity in Lines 449-453: “*Continuous agricultural NO_x (mainly NO) emissions inhibit the O_3 formation, which indicates that the NO_x abundance is excess for O_3 formation over the NCP. Huang et al., (2023) similarly reported that variations in O_3 concentrations were inversely related to the changes in soil NO_x emissions in the NCP. Lu et al., (2021) also reported a NO_x -saturated O_3 formation regime in the NCP.*” and in Lines 494-498: “*During early spring, a large amount of agricultural*

NO_x (mainly NO) emission causes a NO titration effect during daytime, decreasing O₃ concentrations, when the O₃ chemistry is under the VOC-sensitive (NO_x-saturated) or the transitional regimes (Figure S6) (Sillman, 1995). In contrast, the O₃ formation chemistry in summer shifts from VOCs-sensitive to NO_x-sensitive (Sha et al., 2021; Wang et al., 2022a)."

We have discussed the implications of agricultural emissions for ozone and air quality goals in Lines 529-536: *"Additionally, the ongoing stringent control measures on emission sources significantly reduce anthropogenic emissions in urban areas, thus the impact of agricultural fertilization on urban air quality is becoming more pronounced (Figure S7). Since soil NO_x emission is sensitive to soil temperature, as global warming is ongoing, routine events like agricultural fertilization will continue to have amplified impacts on air quality with the joint help of atmospheric dispersion/transport and chemical transformation processes (Bennetzen et al., 2016; Ma et al., 2022; Tubiello et al., 2013). These impacts are not confined in agricultural areas alone, but extend to surrounding cities."*

The References section has been updated accordingly.

(3) BDSNP default fertilizer assumptions: In my original point 6, I raised some questions about fertilizer applications. Thank you for clarifying that the default BDSNP implementation was used. I think some additional text is needed to acknowledge important limitations of that implementation. *"Though the 75/25 treatment is the most typical global farming practice (Matson et al., 1998), it may probably introduce extra biases in a specific region."*

While it is true that Hudman et al. included this statement in their 2012 paper, it is incorrect. 75/25 is not typical global farming practice. Matson et al. 1998 is not an appropriate reference to support this statement; it is a paper focused on a site in Mexico and makes no statements about global fertilization applications; the 75/25 split is an estimate for local farmers in Mexico in 1994. Typical practice would place more fertilizer in a topdressing application, and less at

planting—e.g., GGCM, the largest global crop modeling exercise, uses a 20/80 split (see 7th paragraph under “GGCM phase 3 crop modelling protocol” in the Methods of <https://doi.org/10.1038/s43016-021-00400-y>). I did a quick search on fertilizer splits, and found this statement (but I expect if you simply contact an agricultural extension agent you can get an answer): “ ‘One base and one topdressing’ mode is currently the most common form of fertilization for maize spring in Northeast China^{24,25,26}, with 40% of nitrogen fertilizer is applied as a base fertilizer, and 60% is applied as top dressing during the growing period.” <https://doi.org/10.1038/s41598-023-38724-3>. I would also report what the fertilizer split was at Fengqiu Station.

Also it’s worth noting that BDSNP treats the topdressing not as a single application, but is spread out over a period of weeks or months, which is also perhaps not an ideal assumption that will affect results.

I think the best path forward for the authors is to simply state that they used the default BDSNP fertilizer & global emission assumptions, and that these assumptions (in this case, primarily the 75/25 split and not applying the topdressing in a single application) may not accurately the fertilizer applications in China. As the authors note in part in lines 159-165, the offline fertilizer files and emissions tuning factor end up being fine (though not necessarily for the right reason). The authors could also argue after line 165 that the BDSNP tuning, which was done for year 2000 agricultural emissions estimated by Stehfest & Bouwman, is not so far off from recent estimates of global emissions, possibly on the high side (e.g., Table 2 in DOI: 10.1111/gcb.16193, and <https://egusphere.copernicus.org/preprints/2025/egusphere-2025-1416/egusphere-2025-1416.pdf> argues that current-day estimates range between 0.84 and 2.2 Tg N yr⁻¹, though note this manuscript has not been peer reviewed).

But the 75/25 split (including the assumption that the 25% is applied evenly throughout the remainder of the growing season) is something that needs to be acknowledged – you could

simply state something like ‘using a 20/80 split in fertilizer applications, as is commonly used in crop modeling (Jägermeyer et al. 2021 <https://doi.org/10.1038/s43016-021-00400-y>), or a 40/60 split, as has been reported to be common in Northeast China (<https://doi.org/10.1038/s41598-023-38724-3>) would be expected to result in differences in the magnitude and timing of emissions compared to the default BDSNP scheme. Among other impacts, at planting there is no canopy interception of emitted soil NO_x, which would result in substantially larger emissions to the atmosphere under the default BDSNP 75/25 split than in a 20/80 split. In addition, because BDSNP applies the 25% topdressing application evenly over the growing season following the 75% basal application, it is less likely to produce sizable pulses of emissions.’

(As an aside, something that may be of interest to the authors: it may be that as a consequence of having unvarying fertilizer applications and constraining global fertilizer emissions to 1.8 Tg N, BDSNP is unable to reproduce historical trends or much in the way of interannual variability in emissions: <https://egusphere.copernicus.org/preprints/2025/egusphere-2025-1416/egusphere-2025-1416.pdf>)

Response: We have included descriptions and discussions regarding the uncertainties of the BDSNP scheme in Lines 165-170: “*It should be noted that we use the default BDSNP fertilizer and global emission assumptions, and these assumptions (primarily the 75/25 split and not applying the topdressing in a single application) may not accurately reflect the fertilizer applications in China. The BDSNP tuning, which was done for year 2000 agricultural emissions estimated by Stehfest and Bouwman (2006), is close to recent estimates of global emissions, possibly on the high side (Gong et al., 2025; Wang et al., 2022b).*”, in Lines 355-362: “*We should note that the 75/25 treatment is an estimate for local farming practice in Mexico in 1994 (Matson et al., 1998), which probably introduces biases in other regions.*

Typical practice would place more fertilizer at topdressing and less at planting. For example, a 20/80 split was used by GGCMI, the largest global crop modeling exercise (Jägermeyr et al., 2021), and a 40/60 split was used for maize production in Northeast China (Zheng et al., 2023). It's also worth noting that the BDSNP mechanism treats topdressing as a series of applications spread out over several weeks or months, rather than as a single event, which could further influence the modeling results.” and in Lines 587-598: “We should not ignore the uncertainties regarding the BDSNP scheme. The default 75/25 split in fertilizer application may be not widely suitable for the globe. Using a 20/80 split, as is commonly used in crop modeling (Jägermeyr et al., 2021), or a 40/60 split, as has been reported to be common in Northeast China (Zheng et al., 2023), would be expected to result in differences in the magnitude and timing of emissions compared to the default scheme. Among other impacts, there is no canopy interception of emitted soil NO_x at planting, which would result in substantially larger emissions to the atmosphere under the default 75/25 split than in a 20/80 split. In addition, because BDSNP applies the 25% topdressing application evenly over the growing season following the 75% basal application, it is less likely to produce sizable pulses of emissions. Because fertilizer applications are kept constant and global fertilizer emissions are constrained to 1.8 Tg N, the BDSNP mechanism is unable to reproduce historical trends or capture significant interannual variability in emissions.”

(4) Regarding the row anomaly: the issue is that in the L3 product, the number of pixels changes over time because the pixels affected by the row anomaly (and excluded from the L3 product) changes over the OMI lifetime. This is the issue that I was asking be acknowledged/addressed. All that really needs to be done here is to acknowledge that the number of pixels included in NO₂ retrievals changes over time because of the increase in the number of pixels affected by the row anomaly, making the data unsuitable for trend analysis

and possibly introducing uncertainty in seasonal averages.

Response: Thanks to the reviewer for the suggestions. We have included clarifications regarding the row anomaly in Lines 197-199: *“Note that the number of pixels included in NO₂ retrievals changes over time because of the increase in the number of pixels affected by the row anomaly issue, making the data unsuitable for trend analysis and possibly introducing uncertainty in seasonal averages.”*

Specific comments:

(5) Line 85-86 “incorporating high fertilization rates according to the solar terms with excessive N fertilization.” What is meant by ‘according to the solar terms’ will not be understood by the majority of readers (including me) and will need to be explained. Perhaps it would be better to use the growing season or calendar year as context?

Response: In China, farmers plant and cultivate crops according to the traditional “Jieqi” calendar, which is a solar calendar that has been widely used throughout history. To avoid any misunderstanding, we have revised the sentence in Lines 84-86: *“The agricultural management in the NCP has been known for incorporating high fertilization rates with excessive N fertilization (Sun et al., 2022; Vitousek et al., 2009; Zhao et al., 2006).”*

(6) Line 87: “Thus, this region is primarily responsible for agricultural N-fertilizer consumption” This line is not clear as currently phrased. Should it be “This region is the largest consumer of agricultural fertilizer N in China”?

Response: Yes, we have revised the sentence in Lines 86-87: *“Thus, this region is the largest consumer of agricultural fertilizer N in China (Yu et al., 2022) ...”*

(7) Line 95: I still think this pulse is not unexpected: e.g., fertilizer pulses have been seen by

OMI, fertilizer pulses are typically observed in field measurements, and BDSNP is designed in a way that would result in fertilizer pulses of some duration (Hudman et al. 2012)

Response: To avoid any misinterpretation, we have revised the sentence in Lines 95-96: *“In this study, we present a pulse of atmospheric NO₂ column in early spring during the past two decades over the NCP.”*

(8) Line 196: “the trajectory NH₃ from IASI is integrated into each 0.125° × 0.125° grid cell with the average during 2007-2021” This description sounds inaccurate. I don’t think “trajectory” is the correct word here; is vertical column density intended? And I also think “integrated” is incorrect (at least, I do not understand what is meant by “integrated” here). Since these were L2 data, the method of regridding should be specified, including the specific screening criteria (i.e., % cloud cover, retrieval error, and whether exceptions to the retrieval error criteria were made for low concentrations). In addition, “the average during 2007-2021” is stating that the dataset is a single map of a long-term mean, but a time series is presented in the SI figures. Finally, the 0.125 x 0.125 resolution seems much too fine for daily IASI retrievals. This all actually sounds like the description for an oversampled dataset, not the time series presented in the SI.

Response: Since the 0.125° × 0.125° resolution is much too fine for daily IASI retrievals, we reconstruct the mesh grid at a 0.25° × 0.25° resolution. We have revised the sentence to make it more clear in Lines 203-208: *“We construct a 0.25° × 0.25° mesh grid and calculate the average of the NH₃ columns from IASI within each grid cell (Clarisse et al., 2023) ... the data used in this study are averaged into seven-day mean datasets of NO₂ and NH₃ columns with a non-overlapping 7-day window during 2007-2021”* When interpolating into the model grids, we specify that we use the bilinear interpolation method in Line 208: *“The data are interpolated into the model grids using bilinear interpolation.”*

(9) Line 209-212: In my previous comment, I was asking whether these measurements were made continuously, hourly, daily, or at some other interval.

Response: We have revised the sentences to make it more clear in Lines 216-219: “... *The sampling time is 1 min for these monitoring devices. Agricultural NH₃ concentration is monitored by a Picarro analyzer based on the principle of cavity ring-down spectroscopy (CRDS) at the rural Xianghe station (Figure 1), with a sampling frequency of 1 Hz. Hourly data are derived by averaging the high-frequency measurements.*”

(10) Lines 260-262: I don't think this statement is accurate—the NH₃ peaks are in June, not in March. Please see my first general comment above.

Response: The wheat-maize double-cropping system is predominate in the NCP, so fertilization occurs several times in the year. Although the main peaks of NH₃ occur in June, there are sub-peaks in March, which may also originate from fertilizer application. We have clarified this in Lines 268-272: “*Although the main peaks of NH₃ column occur in June, the sub-peaks of NH₃ column in March may provide favorable evidence that these NO₂ column sub-peaks are connected to agricultural activities because atmospheric NH₃ is largely originated from fertilizer application in agriculture.*” Regarding the discrepancies between NH₃ and NO₂ columns, we have included discussion in Lines 272-279: “*We note that the two time series don't have similar seasonal dynamics, which may raise some questions about whether the March NO₂ peaks represent fertilizer pulses. This discrepancy could be due to several factors: (1) contributions from fossil fuel-derived NO_x, (2) elevated background soil NO_x emissions during the “spring thaw” period, (3) differences in fertilizer type, as fertilizers vary in their potential for ammonia volatilization, and (4) variation in application methods, e.g., banding or deep soil placement vs broadcasting, especially when the latter is done without*

incorporation.”

(11)Lines 275-278: Same comment as for 260-262

Response: The wheat-maize double-cropping system is predominate in the NCP, so fertilization occurs several times during the year. The main peaks of NH_3 occur in June, and the sub-peaks in March. To avoid any misinterpretation, we have removed that sentence.

(12)Lines 298-301: I think it’s important to note here that BDSNP is going to be heavily (and arguably inappropriately) biased towards having a fertilizer-induced emission peak **only** at planting/green-up.

Response: We have included the text in Lines 315-317: *“We should also note that the BDSNP scheme would be heavily biased towards having a fertilizer-induced emission peak only at planting/green-up.”*

(13)Line 329: move the definition of IOA from line 361 to here.

Response: We have moved the definition of IOA from Line 361 to Line 345: *“indices of agreement (IOAs, Text S2)”*.

(14)Line 332 and following: I would add discussion here about the discrepancies between the IASI NH_3 peaks (which occur in June) and the OMI NO_2 peaks in March.

Response: The text here is to validate the model performance against measurements during simulation period in this study. Since the simulation period does not include June 2020, we have included discussion on the discrepancies between the IASI NH_3 peaks and the OMI NO_2 peaks in Lines 272-279: *“We note that the two time series don’t have similar seasonal dynamics, which may raise some questions about whether the March NO_2 peaks represent fertilizer pulses.*

This discrepancy could be due to several factors: (1) contributions from fossil fuel-derived NO_x, (2) elevated background soil NO_x emissions during the “spring thaw” period, (3) differences in fertilizer type, as fertilizers vary in their potential for ammonia volatilization, and (4) variation in application methods, e.g., banding or deep soil placement vs broadcasting, especially when the latter is done without incorporation.”

(15)Line 339-341: “Though the 75/25 treatment is the most typical global farming practice (Matson et al., 1998)” Also discussed in a general comment. While it is true that Hudman et al. included a statement like this in their 2012 paper, it is not correct. Matson et al. 1998 is not an appropriate reference to support this statement; it is a paper focused on a site in Mexico and makes no statements about global fertilization applications; the 75/25 split is an estimate for local farmers in 1994. I do not think it’s possible to defend a 75/25 split globally, but I think you can simply acknowledge this as an issue and discuss how it may affect results (i.e., the point that BDSNP is going to be heavily (and arguably inappropriately) biased towards having a fertilizer-induced NO_x emission peak **only** at planting/green-up).

Response: We have included more discussion on the 75/25 treatment in the BDSNP scheme in Lines 355-362: *“We should note that the 75/25 treatment is an estimate for local farming practice in Mexico in 1994 (Matson et al., 1998), which probably introduces biases in other regions. Typical practice would place more fertilizer at topdressing and less at planting. For example, a 20/80 split was used by GGCMI, the largest global crop modeling exercise (Jägermeyr et al., 2021), and a 40/60 split was used for maize production in Northeast China (Zheng et al., 2023). It’s also worth noting that the BDSNP mechanism treats topdressing as a series of applications spread out over several weeks or months, rather than as a single event, which could further influence the modeling results.”* and in Lines 315-317: *“We should also note that the BDSNP scheme would be heavily biased towards having a fertilizer-induced*

emission peak only at planting/green-up.”

(16)Line 385-386: Change to “experiment that excludes soil sources of NO_x and NH_3 in the study domain” if that’s correct.

Response: We have revised the sentence in Lines 409-410: “*We perform a model experiment that excludes the soil sources of NO_x and NH_3 in the study domain to examine the impacts of soil emissions on regional air quality.*”

(17)Line 404-405 (Figure 4 caption): change “surface NO_2 and NH_3 during March 2020 over the NCP. (a and b)” to “a) surface NO_2 and b) NH_3 during March 2020 over the NCP.”

Response: We have revised the sentence in Lines 428-429: “*Figure 7. Direct impacts of soil emissions from agricultural fertilization on (a) surface NO_2 and (b) NH_3 during March 2020 over the NCP.*”

(18)Line 421: Li et al. 2017a only discusses NO_x titration at night. The interesting result presented in this manuscript is the daytime titration and the large reductions in daytime ozone concentrations. Huang 2023, discussed in general comment 2 above, seems like a better place to start.

Response: We have revised the discussion in Lines 449-453: “*Continuous agricultural NO_x (mainly NO) emissions inhibit the O_3 formation, which indicates that the NO_x abundance is excess for O_3 formation over the NCP. Huang et al., (2023) similarly reported that variations in O_3 concentrations were inversely related to the changes in soil NO_x emissions in the NCP. Lu et al., (2021) also reported a NO_x -saturated O_3 formation regime in the NCP.*” The References section has been updated accordingly.

(19)Line 435-440: I think just make it explicitly clear that this experiment doesn't include any changes in HONO emissions. Maybe "We note that soil nitrous acid (HONO) emission, which are not included in these modeling experiments, ...".

Response: We have revised the sentence in Lines 461-463: "*We note that soil nitrous acid (HONO) emission, which are not included in these modeling experiments, can also perturb atmospheric chemistry and the AOC (Feng et al., 2022; Tan et al., 2023) via providing NO and OH through photolysis.*"

(20)Line 464: See my comment to line 421--I believe Huang et al. 2023 actually includes evidence for soil NO_x leading to ozone reduction over the NCP.

Response: We have corrected the improper citations in Line 490: "... (*Tan et al., 2023; Wang et al., 2022*) ...".

(21)Line 515: Change "Changes in surface NO₂ and ..." to "Simulated changes in surface NO₂ and ..."

Response: We have revised the sentence in Lines 538-539: "*Figure 11. Simulated changes in surface NO₂ and related aerosol-chemistry products during March 2020 over the NCP.*"

(22)Figure S2: It took me a while to understand that the focus of these figures is the grey bars. I would change the title of the figure to "A re-presentation of the long-term NO₂ [tropospheric] column time series shown in Figure 2, but with June and October pulses highlighted using grey bars." Also, the 'short bars' are in Figure 2, not Figure 1, and you need to explicitly define here what the red bars represent in each panel—i.e., what fertilizer event they represent--since that is different from Figure 2.

Response: We have revised the caption of Figure S2 as "*Figure S2. A re-presentation of the*

long-term tropospheric NO₂ column time series shown in Figure 2a, but with (a) June and (b) October pulses highlighted using grey bars. The red bars show the fertilization events during sowing periods for maize and winter wheat in (a) June and (b) October, respectively.”

(23) Figures S4 & following: the figures are not properly numbered, starting with the second Figure S4.

Response: We have corrected that in the revised version.