

Response to Reviewers

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Title: A mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints

We would like to thank both reviewers for their useful comments and suggestions.

Response to Reviewer #1 (Responses in **BOLD**, additions to text in **BOLD ITALICS**)

1) “mechanistic” in the title is misleading, since at least a.) the emission factors, b.) pulsing, c.) the differentiation between arid soils and elsewhere are all empirical. The title should be changed to something similar to “Major steps towards a mechanistic...”. This should be the case throughout the article.

**Title: “*Steps toward a mechanistic...*”.**

**P3556 L7: the word “mechanistic” was removed.**

**P3571 L16: “...designed to *better* represent...”**

2) Although the authors try to justify not using a canopy reduction, I cannot accept their argument. Chaparro-Suarez et al. (2011) say in their conclusions “If we transfer such indications from laboratory measurements to the natural environment we would conclude that forests do not release NO<sub>2</sub>”. The results of Raivonen et al. (2009) are, in my opinion, highly uncertain and speculative:

- Their deposition measurements exhibit a lack of data during daytime (see their Fig. 1).
- They measured at treetops only.
- The measured shoots were never exposed to rain, therefore nitrate on the leaf surface was not removed by rain.
- They give neither any uncertainty estimation nor detection limits of their instruments.

NO emitted from the soil is rapidly oxidized to NO<sub>2</sub> and this NO<sub>2</sub> is subject to wet and dry deposition within the canopy. I would recommend either applying the old canopy reduction as previously implemented in GEOS-Chem or even better integrate the deposition velocities derived by Chaparro-Suarez et al. (2011) into a new reduction factor for temperate forests. But applying a canopy uptake is in my opinion indispensable when comparing to satellite derived NO<sub>2</sub> columns. If you find something more convincing to ignore the canopy reduction, I would review the arguments again.

**We agree with the reviewer that the arguments presented in Chaparro-Suarez et al. (2011) are interesting—however we think it premature to assume that the NO<sub>x</sub> flux community has fully accepted those results. Incorporating them into a model is beyond the scope of this paper.**

**Our understanding of the literature describing the current CRFs is that the values chosen are not based on a mechanism. We think it better if the user of our model makes choices if and how to implement a CRF themselves, at least until a**

mechanistic model is developed. That said we recognize the reviewers' and probably many readers' interest in a comparison to models in a more standard formulation and have added text describing a calculation using standard values for CRFs. The change has a negligible effect on our comparisons to satellite data.

**P3560 L14:** “...Jacob and Bakwin (1991). *This canopy reduction factor is not mechanistic in nature; moreover, recent observations provide mixed evidence on the magnitude of such reductions and laboratory measurements of NO<sub>2</sub> compensation points in some cases suggest that NO<sub>2</sub> should be emitted from forest canopies at low NO<sub>x</sub> concentrations (Raivonen et al., 2009; Chaparro-Suarez et al., 2011). Given this uncertainty, we primarily use above-soil estimates from both models (YL95 and BDSNP) here, and stress that future users of the model should implement a canopy reduction scheme they find most appropriate for their application. However, to allow comparison with previous studies, we also implement the previous GEOS-Chem canopy reduction scheme and provide values in the text and Table 1 regarding the effects of canopy reduction. Figure 1a...*”

**P3566 L10:** “...soil moisture changes. *The canopy reduction scheme yields an above-canopy S<sub>NO<sub>x</sub></sub> of 9.0 Tg N yr<sup>-1</sup>; canopy reduction decreases emissions by ~10-15% in grasslands and up to ~85% over forested regions.*”

**P3568 L13:** “...without canopy reduction and focus our analysis on locations where the canopy effects are small (e.g. grasslands, agriculture), giving us greater confidence in the comparison between the model and observations.”

**Table 1:** included “9.0” in the appropriate location (above-canopy emissions from this study).

Minor comments:

1. Also mention the smooth temperature dependence between 0C and 30C in the abstract.

**P3556 L14:** “...smooth function of soil moisture *as well as temperature between 0 and 30°C.*”

2. How good is the representation of soil moisture in GEOS-Chem compared to the real world?

Soil moisture in GEOS-Chem has not yet been evaluated, partly because until recently, there was no good comprehensive data set against which to test the model. With the advent of the ESA SMOS satellite instrument soil moisture dataset, validation will hopefully occur soon, but we suggest that doing so is outside the scope of this paper. We have added some text to address the uncertainties associated with the use of this parameter.

**P3562 L14:** “...2000). *We note that it is uncertain how well  $\theta$  represents real-world water-filled pore space because the parameter has not yet been validated. However, the*

*use of this parameter in the soil parameterization still represents movement towards a more mechanistic approach, and we recommend validation of  $\theta$  as future work.”*

3. Page 3557, line 9: change “...to predicting atmospheric composition and to understanding...” to “...to predict atmospheric composition and to understand...”.

**P3557 L9: “...to predict atmospheric composition and to understand the...”**

4. Additional reference that you might want to include in the introduction to highlight the effects of (soil) NO<sub>x</sub> on ozone, OH and aerosols could be Dentener and Crutzen (1993), Andreae and Crutzen (1997), Martin et al. (2003) and/or Steinkamp et al. (2009) and for the earth’s nitrogen cycle it could be Galloway et al. (2004) and/or Phoenix et al. (2006). van der A et al. (2008) also fits well into the introduction, since this shows a dominant soil source of NO<sub>x</sub> in parts of the Sahel region.

**P3557 L9: “...rates (Galloway et al., 2004; Phoenix et al., 2006). Understanding...”**

**P3557 L11: “...climate (Dentener and Crutzen, 1993; Andreae and Crutzen, 1997; Martin et al., 2003; Steinkamp et al., 2009)...”**

**P3557 L25: “...fertilizer (van der A, 2008)...”**

**Added to references:**

**Andreae, M. O. and Crutzen, P. J.: Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry, *Science*, 276, 1052–1058, 1997.**

**Dentener, F. J. and Crutzen, P. J.: Reaction of N<sub>2</sub>O<sub>5</sub> on tropospheric aerosols - impact on the global distributions of NO<sub>x</sub>, O<sub>3</sub>, and OH, *J. Geophys. Res.*, 98, 7149–7163, 1993.**

**Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S. P., Asner, G. P., Cleveland, C. C., Green, P. A., Holland, E. A., Karl, D. M., Michaels, A. F., Porter, J. H., Townsend, A. R., and Vorosmarty, C. J.: Nitrogen cycles: past, present, and future, *Biogeochem.*, 70, 153–226, 2004.**

**Martin, R. V., Jacob, D. J., Yantosca, R. M., Chin, M., and Ginoux, P.: Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols, *J. Geophys. Res.*, 108, 19, 2003.**

**Phoenix, G. K., Hicks, W. K., Cindersby, S., Kuylenstierna, J. C. I., Stock, W. D., Dentener, F. J., Giller, K. E., Austin, A. T., Lefroy, R. D. B., Gimeno, B. S., Ashmore, M. R., and Ineson, P.: Atmospheric nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in assessing N deposition impacts, *Global Change Biology*, 12, 470–476, 2006.**

**Steinkamp, J., Ganzeveld, L., Wilcke, W., and Lawrence, M.: Influence of modeled soil biogenic NO emissions on related trace gases and the atmospheric oxidizing efficiency, *Atmos. Chem. Phys.*, 9, 2663–2677, 2009.**

**van der A, R. J., Eskes, H. J., Boersma, K. F., van Noije, T. P. C., Van Roozendaal, M., De Smedt, I., Peters, D. H. M. U., and Meijer, E. W.: Trends, seasonal**

**variability and dominant NO<sub>x</sub> source derived from a ten year record of NO<sub>2</sub> measured from space, J. Geophys. Res., 113, 2008.**

5. What scientific questions do you address with your new implementation, please be more precise at the end of the introduction.

**P3558 L14: “...thus affect S<sub>NO<sub>x</sub></sub>. *This more mechanistic approach improves the time resolution of modeled soil emissions, implying that the model can better reproduce daily variability and allow the study of important processes such as daily ozone response and a more accurate assessment of the atmospheric lifetime of the emitted NO<sub>x</sub>. Both quantities are strongly non-linear functions of NO<sub>x</sub> and as a result will be systematically biased if the temporal patterns of pulsed emissions are represented as a continuum emission on a month long or seasonal time scale. After describing...*”**

6. Consistently abbreviate Yienger and Levy (1995) as YL95.

**P3559 L13: “...of the YL95 empirical scheme, which...”**

7. Mention that the model setup and description of the satellite data is in the appendix

**P3558 L18: “...satellite observations. *Descriptions of the model setup and satellite product retrievals can be found in the appendix.*”**

8. Page 3559, line 5: DNDC (Butterbach-Bahl et al., 2009) does account for pulsing.

**Based on our reading of Butterbach-Bahl (2009) DNDC does not explicitly account for precipitation-induced pulsing, although as a process-based model it may reproduce pulsing as described in the manuscript. However, the figures in the paper only indicate that the model reproduces pulses after fertilizer application; no information is provided on precipitation events but nearly all observed pulses that are reproduced in the model occur following fertilizer applications. Given the lack of discussion about precipitation-induced pulses we alter the text as follows:**

**P3559 L6: “...do not *explicitly* account for pulsed emissions, *although the process-based DNDC model may reproduce these pulses. Pulsed...*”**

9. Page 3560, line 3: add “and soil moisture state.” behind “. . . distinguish between vegetation type”.

**P2560 L3: “...vegetation type and soil moisture state. Soils...”**

10. Page 3562, equation 3: where does the number 0.103 come from.

**P3562 L8: “*The exponential dependence on temperature is identical to that for wet soils in YL95, where 0.103 is the weighted average of temperature dependencies for several biomes. The parameterization...*”**

11. Page 3562, line 16: It should also be mentioned that at high values of water filled pore space diffusion is hardly possible.

**P3562 L16: “...N<sub>2</sub>O and N<sub>2</sub>; diffusion of emitted gases through the soil pores is also limited. S<sub>NO<sub>x</sub></sub> dependence...”**

12. Page 3560, line 9; Page 3565, lines 14ff and Fig. 3: In the previous implementation 2.5% of applied fertilizer were emitted, whereas in BDSNP are 0.68% were emitted. However, in Fig. 3 the emissions of BDSNP are higher than the original GEOS-Chem emissions. Please explain this phenomenon.

**Here we note that we misstated something. We scaled emissions to yield 1.8 Tg N of fertilizer emissions, consistent with literature estimates. That scale factor was 0.68% of available N. However, because of pulsing and soil N buildup, the fraction of total fertilizer N emitted is not this value (this was misstated). In total, 1.5% of applied fertilizer is emitted. The rest of the discrepancy in totals between new and old scheme is due to differences in the fertilizer datasets. We have modified the text to clarify.**

**P3556 L16: “...fertilizer N input (1.5% of applied N) and...”**

**P3565 L18: “...scheme, we scale the emission rate,  $\bar{E}$ , so that the total global above-soil NO<sub>x</sub> emissions due to fertilizer matches observed estimates of fertilizer emissions of 1.8 Tg N yr<sup>-1</sup> from Stehfest and Bouwman (2006). Figure 3...”**

**P3571 L24: “...N input (1.5% of applied N)...”**

13. Page 3560, line 18: Change “Chaparro-Suarez et., 2011” to “Chaparro-Suarez et al., 2011”

**P3560 L18: “...Chaparro-Suarez et al., 2011...”**

14. Page 3564, line 7 and Fig. 4: Please mention and if necessary discuss the variability in the calculated mean of the start/end of the growing season. It would be also nice to add a picture of the calculated growing season length.

**Although we understand that it is difficult to derive the growing season length from these images, at this stage we have not included a picture of the growing season length as this value is not directly used in the model and may be a distracting addition, and can be approximated from the figures already displayed.**

**P3564 L10: “...(Ganguly et al., 2010). The standard deviation for this average ranges from 10-40 days for most locations except some tropical forests and deserts where the seasonal variation in EVI is low. However, with the exception of a few tropical areas, fertilizer application rates in these areas are extremely low (Potter et al., 2010)**

*indicating that this variability is not a major source of bias in total emissions. We apply...*

15. Same paragraph: What happens in region that have two growing seasons a year, or for example regions in China with a rice-wheat rotation?

**P3564 L14:** “...1998). *We note that no adjustment has been made for regions with two growing seasons or crop rotations in this version; future work will consider how to treat these regions.*”

16. Page 3566, section 4: Where are the soil NO emissions more sensitive to the emission factors by Steinkamp and Lawrence (2011) and where to the new algorithm of BDSNP? Is a new algorithm justified? This could, in my opinion, be a highlight of the manuscript.

**We have attempted to highlight that the pulsed parameterization is the core advancement of our model. The emission factors of Steinkamp and Lawrence (2011) were an important contribution in the effort to improve the fidelity of models to observations, but that is not an improvement we are contributing. Steinkamp and Lawrence (2011) have discussed the effects of their new emissions factors thoroughly. The mechanistic improvements we make with respect to soil moisture, pulsing, and fertilizer imply that we should see significant spatial and temporal differences between the prior parameterizations (including ones using emission factors from Steinkamp and Lawrence (2011)) and the one described here; as a result the entire model is sensitive to the new parameterization even if the yearly total emissions are only changing with the biome emission factors.**

17. Page 3566, line 21: Also displacement of NO enriched air in the soil by water after a rainfall event can lead to pulsing, as well as, but rarely, Chemodenitrification. Since these processes also play a role, they should be mentioned.

**P3566 L21:** “...(Serca et al., 1998). *Other mechanisms leading to NO pulses with first rainfall include the displacement of NO-enriched air by water, as seen with CO<sub>2</sub> and N<sub>2</sub>O (Clough et al., 2000; Huxman et al., 2004), as well as chemodenitrification, the process by which NO<sub>2</sub><sup>-</sup> is chemically oxidized to NO (Davidson, 1992a). A similar...*”

**Added to references:**

**Clough, T. J., Sherlock, R. R., and Cameron, K.C.: Entrapment and displacement of nitrous oxide in a drained pasture soil, Nutr. Cycl. in Agroecosyst., 57, 191-193, 2000.**

**Davidson, E. A.: Sources of nitric oxide and nitrous oxide following wetting of dry soil, Soil Sci. Soc. Am. J., 56, 95-102, 1992a.**

**Huxman, T. E., Snyder, K. A., Tissue, D., Leffler, A. J., Ogle, K., Pockman, W. T., Sandquist, D. R., Potts, D. L., and Schwinning, S.: Precipitation pulses and carbon fluxes in semiarid and arid ecosystems, Oecologia, 141, 254-268, 2004.**

18. Page 3567, line 1: The convective precipitation (see page 3569, line 8) is not mentioned here, which lead to 50% increase in the emissions.

**P3567 L3: “...50% increase in emissions *with subsequent convective precipitation* (Hudman et al., 2010)...”**

19. Page 3567, line 6: The 0.5 Tg(N) yr<sup>-1</sup> from atmospheric deposited nitrogen are responsible for 5% of the total emission. The spatial contribution might not be evenly distributed over the whole globe. Please discuss this a little further. And it should be emphasized more in the abstract and conclusions. And this should be one scientific question (see point 5).

**The authors disagree that this is a scientific question addressed by their model. While this value has not been estimated by previous parameterizations, the estimate presented here is not constrained by observations, nor is it a particularly sophisticated treatment. We think to focus on this element too strongly detracts from the more significant improvements made elsewhere to the parameterization. We have included some text further discussing the spatial distribution of the emissions from deposition.**

**P3567 L6: “...to the emissions. *These emissions from deposited N are largest in regions with high anthropogenic NO<sub>x</sub> emissions, notably locations in northeast China and in India, and can contribute significantly (>5 ng N m<sup>2</sup> s<sup>-1</sup>) to the total emissions in these areas.*”**

20. Page 3567, line 16: The difference between the arithmetic and geometric mean does not say anything about the variability. This is a large logic mistake.

**P3567 L16: “...illustrating the large tail in the measurements, which were shown to fit a log-normal distribution. We use the geometric mean here as it is the more appropriate metric to represent log-normal distributed data, and is most consistent with...”**

21. Page 3568: Section 5.1 mentions OMI in the title but not in the text anymore. Please rename the section.

**P3568 L6: “Sensitivity of NO<sub>2</sub> Columns to S<sub>NO<sub>x</sub></sub>”**

22. Section 5.1 and 5.2: Please clarify, that you used daytime values of GEOS-Chem only to calculate the model statistics. This is too important to be mentioned in the appendix only.

**P3568 L16: “...without S<sub>NO<sub>x</sub></sub>. *To validate with data from OMI, the model is sampled between 12:00-15:00 LT corresponding to the OMI overpass time. During the...*”**

**P3570 L5:** “...interannual variability. *The model is sampled daily between 12:00-15:00 LT to correspond with the OMI overpass time. In GEOS-Chem...*”

23. Page 3568 and Fig 5: A “soil column” is in my vocabulary a column within the soil. Please find another term like “soil derived NO<sub>2</sub> column”, this can be abbreviated since it is used more often.

**P3568 L14:** “...mean ratio of *soil-derived NO<sub>2</sub> column* ( $C_{SNO_x}$ ) to total column, where  $C_{SNO_x}$  is defined as the difference between a simulation with and without  $S_{NO_x}$ . During the onset of the summer monsoon over the northern equatorial tropics,  $C_{SNO_x}$  is predicted to...”

**P3568 L24:** “...standard deviation in  $C_{SNO_x}$  to the standard deviation in the total column without  $S_{NO_x}$ . This measure can be used to diagnose the contribution of  $S_{NO_x}$  to observed column variability. Over the African Sahel *variability in  $C_{SNO_x}$  is 5x greater...*”

**P3591 Fig. 5 caption:** “...the ratio of the *soil-derived NO<sub>2</sub> column* ( $C_{SNO_x}$ ) standard deviation to the simulated tropospheric NO<sub>2</sub> column standard deviation without soil NO<sub>x</sub> emissions.  $C_{SNO_x}$  is defined...”

This was also changed in Figure 5 itself.

24. Page 3569, line 14: Change “...deviations from...” to “...deviations for June of each year from...”

**P3569 L14:** “...deviations *for June of each year from ...*”

25. Page 3570, line 2: Although the relative anomaly is the same for DP\_GC and BEHR, the absolute difference seems to be large. Please add one sentence as explanation.

**P3570 L3:** “...BEHR retrieval. *We note that the difference in the total anomaly is related to differences in the stratospheric subtraction and profile shapes used in the retrievals, and also partly due to a discrepancy in the direction of the anomaly in the eastern portion of the region of analysis where the anomaly is less controlled by soil emissions (see Fig. 5). We compare...*”

26. Page 3570, line 18: See point 17.

**P3570 L18:** “...The first rains of the wet season *release large pulses of gaseous NO, due to reactivation of water-stressed bacteria, displacement of NO-rich air, and chemodenitrification (Davidson, 1992a,b; Clough et al., 2000; Huxman et al., 2004; Jaeglé et al., 2004)...*”

27. Page 3571, line 9: What are the 0.025mm? Soil water or precipitation, and what was the old value?



This is in regards to the dry spell criteria used to determine the first rain. In the case of the OMI column data, the first rain is the first day of at least 2mm precipitation following at least 60 days of <2mm precipitation (using TRMM total rainfall). This criteria was mentioned in the Figure 7 caption but we neglected to put it in the text, so we add it here. The criteria had to be reduced to 0.025 mm for the model because of the change in resolution (0.25 by 0.25 degrees vs. 2 by 2.5 degrees).

**P3571 L2:** “...first rainfall, determined as the first day with precipitation >2mm (as measured by TRMM) following a dry spell (at least 60 days of precipitation <2mm day<sup>-1</sup>). Ratios are...”

28. Page 3571, line 26: Do your regions of dominating soil NO emissions match to results of van der A et al. (2008)? And why or why not?

**P3569 L4:** “*van der A et al. (2008) use satellite data from the GOME and SCIAMACHY instruments to identify regions where soil NO<sub>x</sub> emissions dominate the observed NO<sub>2</sub> column. There are several broad similarities between the regions they identify as major soil NO<sub>x</sub> sources and the locations we identify above (e.g. the Sahel, northern Great Plains), but also some differences (e.g. southern Great Plains). We note that van der A et al. (2008) identify locations as soil-dominant according to the seasonal cycle, as soil emissions peak in the summer while areas not dominated by soil emissions experience wintertime peaks in NO<sub>2</sub> column densities. We instead identify locations where soil emissions are a large percentage of the total column and dominate the day-to-day variability in the measured column over a specific time interval (AMJ/JJA). It is possible that soil emissions may contribute a large fraction of NO<sub>2</sub> to the total columns during summertime in locations identified as e.g. anthropogenic-dominated sources by van der A et al. (2008), and may also dominate daily variability in those locations, accounting for the differences observed. Regardless, the results presented by van der A et al. (2008) increase our confidence that the regions selected for validation here should be highly influenced by soil NO<sub>x</sub> emissions.*”

29. Please remove the borders of the US states in the global maps.

**We acknowledge the reviewer’s comment that state borders do not belong on a global countries map, but because we validate over the Great Plains region which is shown in greater detail with the state outlines in another figure (and is of interest to those in the US) we think it is best to keep the border lines on the map for reference’s sake.**

30. Please remove the county borders (or whatever it is in Fig. 6). US states are sufficient.

**The lines are latitude/longitude gridlines and have been removed.**

31. Page 3586: Steinkamp and Lawrence (2011) also included the above canopy fluxes of the arithmetic mean (8.61 Tg(N) yr<sup>-1</sup>). Please list this in your table.

**Table 1: added S&L 2011 above canopy fluxes for both arithmetic (26.7) and geometric (8.61) means. Table footnote 7 (RE: the arithmetic/geometric values) has been adjusted to include the above-canopy values.**

32. Page 3590, Fig. 4: Is there really a straight line of growing season start in the Amazon basin?

**Some smoothing across latitude had to be used here to fill in areas that did not have adequate growing season start and end dates (see response to issue #14 above). This is an artifact of the smoothing. However, there is very little fertilizer applied in this region so this artifact should not significantly affect our results.**

33. Page 3593, Fig. 7b: A continuous colorbar for nominal data is not suitable, please change the colorbar.

**We admit to being somewhat confused by the reviewer's request. The color bars for both Figure 7a and 7b have clearly delineated cutoffs (i.e. are not continuous) and Figure 7b does not present nominal data. We have made no changes to these figures at this stage.**

34. Page 3593, Fig. 7b: "... following at least 60 days of  $<2 \text{ mm day}^{-1}$ ." Is "smaller than" correct, if this should depict the beginning of the wet season? Is it a running mean or must each day fulfill the condition?

**Please see response to issue #27. This depicts the start of the wet season following a dry season, which is the 60 days of  $<2 \text{ mm day}^{-1}$ . In other words, the first day that it rains  $> 2\text{mm}$  in a grid box we consider the first rain.**

**Figure 7 caption: "...OMI overpass time (13:30UTC - 13:30UTC) following a dry spell (at least 60 days of  $<2\text{mm day}^{-1}$ )..."**

Response to Reviewer #2 (Responses in **BOLD**, additions to text in **BOLD ITALICS**)

1) First of all, the authors acknowledge that they test the above-soil NO<sub>x</sub> emissions, without taking into account the effect of any vegetation shielding or assimilating NO<sub>x</sub> emitted by soils. Because their focus is on the U.S. Mid-West and the Sahel, the canopy reduction of soil NO<sub>x</sub> emissions, may not be as important as over dense forests, but I agree with the other reviewer that one cannot simply gloss over it by stating that there is debate on the canopy effects. This is indeed the case, and therefore I suggest the authors at least try to also include estimates of how much soil NO<sub>x</sub> reaches the atmosphere, after accounting for canopy reduction. For instance Delon et al. [2009] test their soil NO<sub>x</sub> emissions with and without canopy reduction. Then, even over areas with modest vegetation or leaf area, there is strong evidence that vegetation has a shielding influence on the soil/air exchange of NO<sub>x</sub> [Pang et al., 2009], suggesting that canopy reduction might also be important for regions like the U.S. Midwest and the Sahel. In fact, I think

the authors detract from their achievement by not accounting for canopy effects. To illustrate this, it is still difficult to compare their soil NO<sub>x</sub> emissions to the state-of-science above-canopy estimates from 4.7-13 TgNyr<sup>-1</sup>.

**Please see the response to Reviewer #1 (major issue #2).**

2) Another concern is how the authors dealt with another variable source of tropospheric NO<sub>x</sub>: lightning. The same precipitation events that trigger soil NO<sub>x</sub> pulsing are likely to have generated a number of lightning flashes leading to NO<sub>2</sub> that then will be detected by OMI. How have the authors avoided this interference in the satellite signals? Simply filtering out measurements with cloud radiance fractions < 0.5 is not enough since the clouds may have moved out of the footprint before the OMI measurement. In that respect, it is far from reassuring that the authors report that they compare to GEOSChem simulations without lightning (P3570, lines 3-5), because it appears to affect 'interannual variability'. Interannual variability of what? The authors should clarify all this, and convince us why model (no lightning contribution) and OMI (soil + lightning NO<sub>2</sub> contributions) can be compared.

**P3570 L5: “...climatology (Murray et al., *submitted to J. Geophys. Res.*, 2012). This scaling does not reproduce interannual variability in observed lightning from satellites, leading to June anomalies in modeled NO<sub>2</sub> columns that do not match the observed anomalies. This effect was discussed in depth by Hudman et al. (2010), who also used the National Lightning Detection Network to demonstrate that lightning over the Great Plains region was anomalously low in 2006, implying that the observed anomaly cannot be due to increased lightning emissions of NO. We acknowledge that lightning emissions are a source of uncertainty in this comparison between satellite observations, which include lightning-emitted NO<sub>2</sub>, and modeled NO<sub>2</sub> column anomalies. ...”**

3) The evaluation of the success of the pulsing scheme (section 5.3) is ramshackle. It is unclear to me how the satellite NO<sub>2</sub> columns have been averaged over the Sahel. It cannot be just the overall average over the Sahel region, since different locations have different days of first rainfall, and, therefore, different days of pulsing, as shown in the lower panel of Figure 7.

**P3571 L2: “...following first rainfall, determined as the first day of precipitation >2mm (as measured by TRMM) following a dry spell (at least 60 days of precipitation <2mm). All column data are averaged daily to 0.25°x0.25° resolution where cloud radiance fraction <50%. The first day of rain is identified and ratios for the day of first rain and subsequent days are taken against the average NO<sub>2</sub> column in the 5 days preceding first rainfall in each box; these data are then averaged across all 572 boxes that meet the dry spell criteria. The mean column over the region in the five days preceding first rain is  $9.5 \times 10^{14}$  molecules cm<sup>-2</sup>. Mean (Median)...”**

In order to generate a Figure 8, the authors must have repeated their calculations for successive days, but then how did they account for the fact that yesterday's NO<sub>x</sub> pulse

from one region affects today's background for an adjacent region where first rain occurred one day later?

**We did not explicitly address the question of whether emissions in one location might affect another downwind and we are not sure exactly how to do that quantitatively without adding a variety of new tracers to the model. However we believe the effects are small. First, the short lifetime of NO<sub>x</sub> (~1 day) means that transport of a NO<sub>x</sub> pulse will only have a significant influence within about one day (50-75km) of emission. This is a range that is small compared to the GEOS-CHEM grid cells. Second, transport effects are present in both model and satellite, so the comparison between the two remains valid.**

Any information on the absolute levels (in 10<sup>15</sup> molec. cm<sup>-2</sup>) is lacking, we only see ratios in Figure 8, so it is impossible to judge how strong the NO<sub>2</sub> signal was, and how that relates to the satellite detection limit. Furthermore, any information on the number of samples, and on error statistics is lacking from this part.

**In our opinion, the exponential decay shown in the figure gives a clear indication that there is a significant signal on day one and day two reaching the regional mean on days 3 and 4 after a rain event. The difference between the mean and median shown was intended to give the reader a sense of the statistical significance of the result.**

**See above (the text added to provide more information about the averaging) for text added to address this issue.**

There is no discussion of interference from lightning,

**P3571 L8: "...the BDSNP. We note that we do not consider the impact of lightning on the observed NO<sub>x</sub> column, but the observed increases occur on the day of first rain only, not subsequent rains, indicating that the observed increases are likely due to pulsing and not lightning. For comparison..."**

and last but not least, the introduction of a new product (Standard Product) to validate soil NO<sub>x</sub> pulses, while not including the DP\_GC product, is questionable. In section 5.2 the authors argue for the use of both the BEHR and DP\_GC products 'as a measure of uncertainty in the NO<sub>2</sub> anomaly', but in section 5.3 using just the SP is apparently deemed sufficient. The argument that DP\_GC is not available on 0.25 deg x 0.25 deg can be easily overcome by downscaling the SP to whatever resolution the DP\_GC has. Evaluating soil pulsing from both SP and DP\_GC would be very valuable as a measure of uncertainty, and, on top of that also, provide important information on the consistency of the retrievals.

**The two different time scales for variation discussed in the two sections justify our distinct assessments of how to approach characterizing the uncertainty in the columns derived from the absorption spectrum recorded by OMI. On the seasonal**

time scale, the various products available have quite different columns, primarily because they make different choices of how to calculate the a priori profile. These differences are negligible when comparing one day to the day immediately following. As a result, comparing different products provides some measure of how well we understand the NO<sub>2</sub> column on a seasonal basis—as we do for the anomaly in the U.S. Midwest. In contrast, on a daily basis, the different products vary primarily in their method for stratospheric subtraction; on an average scale we expect this to change very little from day to day. Also, SP cannot be downscaled (nor can DP\_GC at 2x2.5 degrees be used) as the pulsing is a highly localized phenomenon (see Fig. 7b for spatial variability in first rain) and so the signal is lost at the lower resolution. This effect is not an issue in the model as the first rain occurs at the same time throughout a single model grid box.

**P3570 L28:** “...used was provided at *too low a resolution to observe the highly localized pulses; however, use of a different retrieval is not predicted to produce significantly different results for an analysis like this where we examine variability from day to day. This variability results primarily from differences in stratospheric subtraction which do not vary much on a daily basis. Figure 8...*”

P3558, L7: I suggest the authors clarify here what seasonality they refer to here.

**P3558 L7:** “...b) *growing season start and end dates derived from data obtained by the Moderate Resolution Imaging Spectrometer (MODIS) are used...*”

P3566, L8: I suggest to add that this concerns the N available in soils apart from fertilizer use.

**We believe the reviewer meant P3565 rather than P3566 as there is no text regarding N available in soils in the given location. The biome emission factor does, in fact, include nitrogen from fertilizer emissions. In YL95 the emission factor for agriculture is adjusted from the emission factor for grassland with the fertilizer emissions. In our parameterization it incorporates the previously discussed available N from fertilizer and wet/dry deposition.**

**P3565 L8:** “...N available in soils *and incorporates the available N from fertilizer and deposition. We choose...*”

P3567, L25: OMI footprints come in km<sup>2</sup>.

**P3567 L25:** “...OMI, 13 x 24 km<sup>2</sup> nadir...”

P3570: although the anomalies are the same (+11%), the absolute anomaly in soil N emissions estimated from the BEHR retrieval is more than 1.5 times stronger than estimated from the DP\_GC retrieval. Why has the Standard Product not been used as a third retrieval to evaluate the absolute anomaly?

**Since BEHR is a derivative of the SP and has demonstrated improvements we do not believe the SP provides additional independent information. We also direct the reviewer to our response to Reviewer #1, minor comment #25.**

P3570, L26 – P3571, L1: what is the statement that ‘use of a different retrieval is not predicted to produce significantly different results’ based upon? There are quite a number of indications in this paper even (Fig. 6) that retrievals are still quite different.

**We refer the reviewer to our response to their comments on the use of OMI data in the evaluation of the pulsing scheme.**

P3575, L7: the authors should clarify what cloud product has been used here.

**P3575 L7: “...than 50% as determined by the OMI Cloud Data product (OMCLD02)...”**

P3575, B1. Was a cross-track bias correction not used for the Standard Product?

**Cross-track bias corrections were used for both SP and DOMINO. The DP\_GC algorithm included additional destriping as residual striping was still visible in the DOMINO product.**

**P3575 L17: “...model. A cross track bias correction is performed as described by Celarier et al. (2008)...”**

**P3575 L21: “...DOMINO algorithm, after cross track bias correction (Boersma et al., 2007), the stratospheric...”**

**P3575 L25: “...2° x 2.5° and includes an additional cross-track bias correction...”**

P3575, L22: when referring to the stratospheric slant column from TM4, it would be appropriate to cite Dirksen et al. [2011] who provide a full description and validation of stratospheric NO<sub>2</sub> from the DOMINO and Standard products. This also relates to citing errors for the stratospheric contribution (B4), where Dirksen et al. [2011] provide updated numbers based on intercomparisons and validation.

**P3575, L22: “...3° x 2° (Dirksen et al., 2011). Surface...”**

**P3576, L11: “...slant column density, and  $<0.3 \times 10^{15}$  molec. cm<sup>-2</sup> for the stratospheric subtraction for both products (Dirksen et al., 2011). Tropospheric AMF...”**

**References:**

**Dirksen, R. J., Boersma, K. F., Eskes, H. J., Ionov, D. V., Bucsela, E. J., Levelt, P. F., and Kelder, H. M.: Evaluation of stratospheric NO<sub>2</sub> retrieved from the Ozone Monitoring Instrument: Intercomparison, diurnal cycle, and trending, J. Geophys. Res., 116, D08305, doi:10.1029/2010JD014943, 2011.**

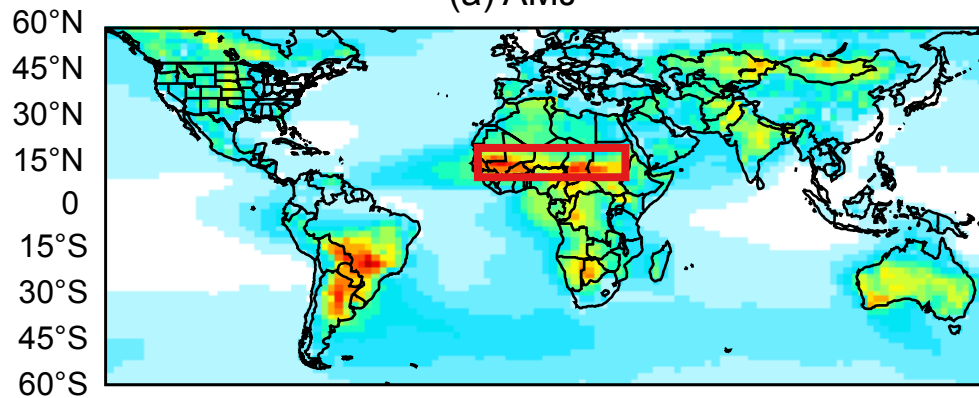
P3575, L23: the surface albedo in DOMINO is from the combined TOMS/GOME database.

**P3575, L22: “...Surface albedo is from the monthly  $1^\circ \times 1.25^\circ$  combined GOME/TOMS database (Boersma et al., 2004).”**

**Some small technical edits were also made to improve clarity in the text.**

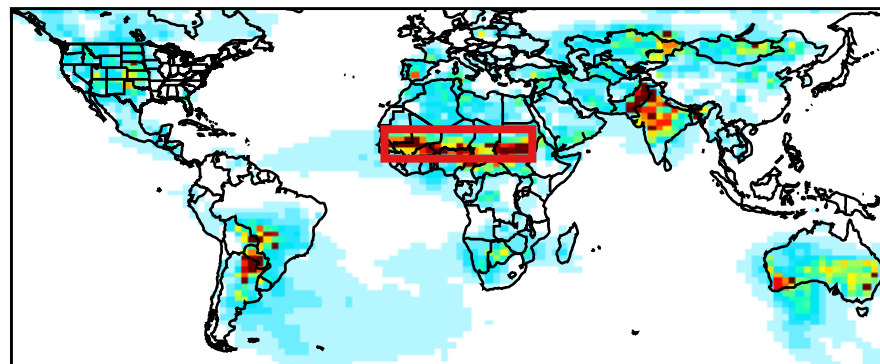
$C_{\text{SNOx}}$  / Total Column

(a) AMJ

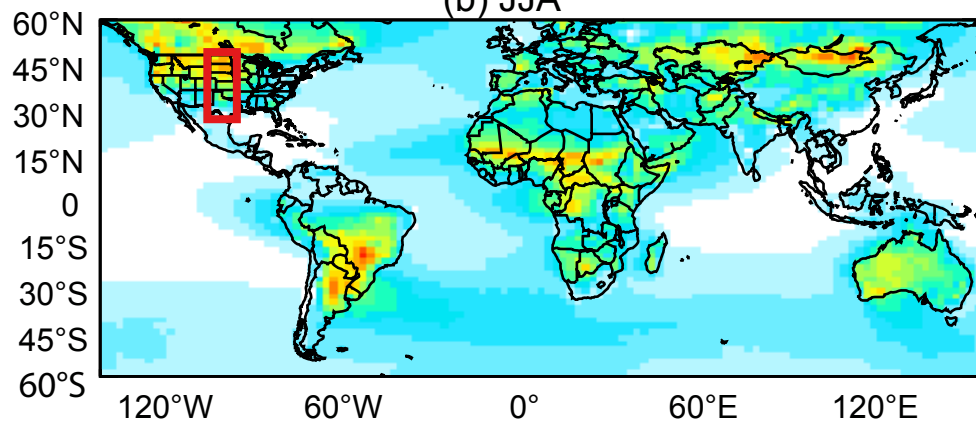


$C_{\text{SNOx}}$  Std Dev / No Soil Std Dev

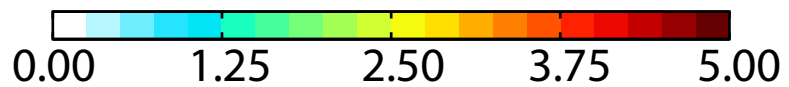
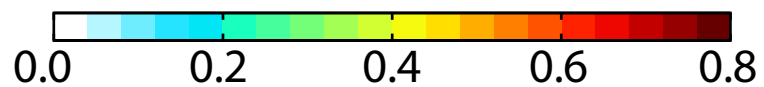
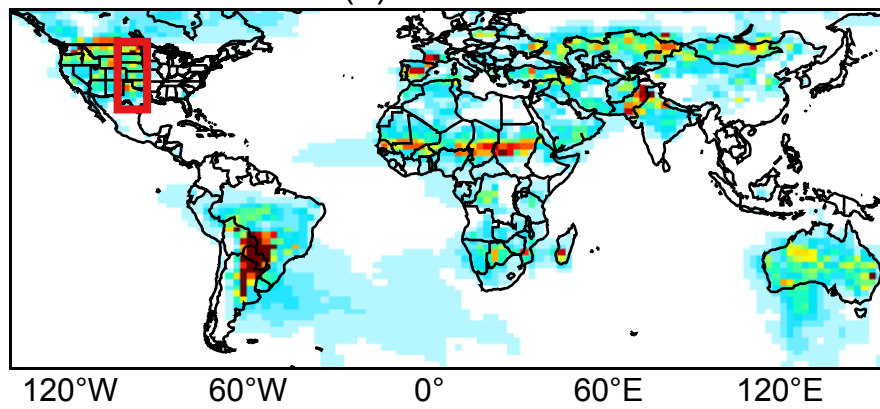
(c) AMJ



(b) JJA



(d) JJA



[ratio]



