Multi-satellite sensor study on precipitation-induced emission pulses of NOx from soils in semi-arid ecosystems by J. Zörner et al.

Reply to anonymous referee #1

We would like to thank reviewer #1 for the very constructive and encouraging comments and suggestions. Our replies as well as the changes made to the manuscript are provided below.

Blue: Reviewer comment
Black: Author's reply
Red: Modified text in manuscript

General comments:

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(1) a calculation of the budget at the seasonal scale in TgN for the Sahel. The evaluation of NO fluxes from soils is a good point, but it would have been interesting to know the overall budget at the regional scale. All the necessary information is available (fluxes, area) for this calculation. If this calculation cannot be provided please explain why precisely.

Such a calculation certainly helps to put the total amount of emitted N from soils in a perspective to other N sources and the total N budget. Therefore, we included an additional subsection to the discussion part and updated the other parts of the manuscript correspondingly. Furthermore, we added average time series for NO_2 VCDs, precipitation, fire and lightning to the manuscript to further illustrate variations in background NO_2 VCDs as well as potential causes.

Addition to introduction page 4 line 6:
In contrast to previous satellite studies, our study makes a clear distinction between (i) pulsed emissions which show strong gradients on a day-to-day scale triggered by a singular precipitation event and (ii) background emissions which are not directly affected or could not be unambiguously related to a strong precipitation pulse. This facilitates the assessment of the contribution from single sNO_x pulses additionally to background levels.

Addition to the end of section 5.1:
Our study focuses on the quantification of pulsed soil emissions and determines the NO_2 enhancement on Day0 and the following days with respect to a sophistically determined background. However, the seasonal pattern of the determined background, i.e. the NO_2 enhancement at the onset of raining season (compare Jaeglé et al., 2004), clearly indicates that it is mainly driven by microbial emissions from soils as well: from pulsed emissions discarded by our strict selection criteria or continuous emissions during wet season. Note that the seasonal pattern of NO_2 over the Sahel, as shown in Fig. 7, can neither be explained by biomass burning nor lightning. Fig. 13c of the manuscript shows that the background NO_2 VCDs are about $0.9 * 10^{15}$ molec cm^{-2}. This is about $0.17 * 10^{15}$ molec cm^{-2} higher than background in winter. Thus, in addition to the pulsed emissions quantified above, a mean background of $0.17 * 10^{15}$ molec cm^{-2} can be attributed to soil emissions as well. These estimates are based on TMPA precipitation data. For other precipitation products (CMORPH or PERSIANN), results change only slightly (see Appendix E).

In summary, we discriminate between soil emissions within: (a) 1-3 days (initial peak), (b) 14 days, and (c) several months (background during the wet season). The separate quantification of soil emissions belonging to these three categories might also be adopted in model parametrizations of soil emissions. However, further research needs to be conducted on how these emission categories vary for different regions worldwide.

We introduced a new subsection: 5.2 Seasonal soil nitrogen emissions in the Sahel
In this section we quantify the total soil emissions, both due to pulsed emissions and background, for the Sahel region. For the pulsed emissions on Day0 (category a) and the following 2 weeks (b), the fluxes estimated above are multiplied by the area of the investigated region (0-30°E, 12-18°N). The statistics of

our analysis in the Sahel suggest that on average one large pulsing event (after 60 days of drought) occurs within a single pixel in the April-May-June period. Scaling up the Day0 emissions results in 1.2 GgN and 12 GgN, considering the lower and upper flux estimates estimated above. Analogously, the emissions over the following two week period add up to 8.8 GgN. Total emissions due to pulsing add up to about 10.1 to 20.8 GgN. As mentioned above, the observed increase of the background in the AMJ-period of $0.17 * 10^{15}$ molec cm^{-2} is mainly driven by microbial emissions from soils as well. When integrated over the complete April-May-June period, this enhancement of the background emissions corresponds to 46.4 GgN (again based on a NO_x lifetime of 4 hours). Consequently, the pulsing events contribute about 21-44% additionally to total soil emissions for the Sahel and dominate the local NO_x concentrations on the particular days.

Jaeglé et al. (2004) determine top-down total soil emissions from GOME-2 measurements of about 400 GgN for North Equatorial Africa (0-18°N) in June alone. Our estimated total soil emissions of nitrogen (56.5-67.2 GgN for AMJ) are smaller, but are determined for a smaller region as well which makes a direct comparison difficult.

Addition to the end of the abstract and the conclusion section:
With respect to the seasonal NO_x budget, we assess a contribution between 21 to 44% from these rain-induced intense pulsing events to total soil NO_x emissions in the Sahel.

Addition to section 4.4.1 Lightning NO_x:
Fig. 7 depicts daily time series for NO_2 VCDs, precipitation and lightning counts averaged for the years 2007 to 2010. The seasonal evolution of the number of lightning strikes closely follows the precipitation patterns. Fig. 7 also illustrates that lightning is not a governing source of NO_x in the Sahel as no correlation between lightning strikes and NO_2 VCDs can be found, although a direct proportionality would be expected. Precipitation also does not correlate well with the observed seasonal cycle in NO_2. This is, however, expected as microbial emissions of NO_x from soils are not a linear function of soil moisture content or precipitation.

Addition to section 4.4.1 Fire:
The seasonal cycle in fire counts, depicted in Fig. 7, shows highest activity in the Sahel in October and November for the years 2007 to 2010, while average NO_2 VCDs are highest in summer.

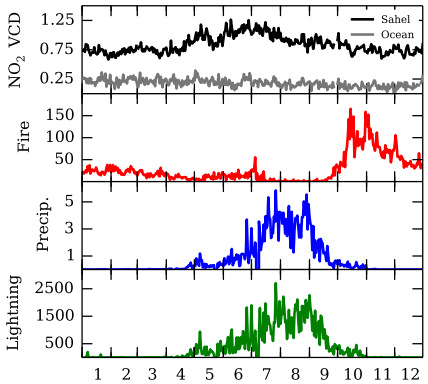

Figure 7: Daily time series for the Sahel region (0-30° W, 12-18° N) averaged for the years 2007, 2008, 2009 and 2010. The first row of each panel shows mean NO_2 VCDs from OMI in molecules cm^{-2} (black) and a clean ocean reference (grey, 130-150°W, 12-18°N). The second row shows the number of active fire counts in the Sahel from MODIS. The third row shows average precipitation from the TMPA/TRMM product in mm. The fourth row shows the number of lightning strikes detected by WWLLN.

(2) References on soil NOx emissions are a bit old, except Hudman et al. (2012). New references could be included, see suggestions below.

The introductory section is updated accordingly. Proposed changes to the manuscript are provided in the answer to question #10.

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(3) Too little explanations are given on processes responsible for soil NOx emissions. References are suggested below.

Proposed changes to the manuscript can be found below in the document (see reply to question #6).

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(4) The Western Sahel is not included in the study, the reasons why are not clear and must be detailed.

We agree with the Referee that a region West of our main study region also shows enhanced NO_2 columns in response to the first rain (Fig. 4a). Thus, we added the argumentation on why we focus on the Central and Eastern part of the Sahel to the beginning of section 4.2.

We restrict our detailed analysis to the Central and Eastern part of the Sahel region (0-30° E, 12-18° N), similar as in previous studies (i.e., Jaeglé et al. 2004, Hudman et al. 2012). The western part of the Sahel shows a slightly weaker NO_2 response to rain pulses, which might be related to different inter-annual variability patterns and seasonal cycles of precipitation regimes (Lebel and Ali, 2009).

Specific comments:

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(5) Introduction Line 7, page 2: NOx is also removed by NO2 deposition on vegetated surfaces.

We added the following reference and changed the manuscript accordingly:
Ganzeveld LN, Lelieveld J, Dentener FJ, Krol MC, Bouwman AJ, Roelofs G-J. 2002 Global soil-biogenic NOx emissions and the role of canopy processes. J. Geophys. Res. 107, ACH9-1–ACH9-17, doi:10.1029/2001JD001289

Line 10, Page 2: Furthermore, NO_x is also removed by NO_2 deposition on vegetated surfaces (Ganzeveld et al. 2002).

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(6) Line 23 page 2: explanations of nitrification and denitrification processes are a bit confused. Please refer to Pilegaard et al., Phil Trans. R.. Soc., 2013.

We updated this paragraph accordingly and also added corresponding references.

Emissions of NO_x from natural and anthropogenically influenced soils are mainly driven by microbial activity within the top soil layer and associated chemical reactions (Conrad, 1996). Primarily, two important groups of micro-organisms, nitrifiers and denitrifiers, are involved in processes related to the turnover of nutrients in the soil (Pilegaard et al., 2013, Behrendt et al. 2015). They are directly responsible for the corresponding processes of: (i) nitrification, the biological oxidation of nitrogen compounds, typically the oxidation of soil ammonium (NH_4^+) to nitrate (NO_3^-) and (ii) denitrification, the reduction of nitrate by microbes to gaseous products, i.e. N_2O and finally N_2. NO is a gaseous by-product of both processes and once released reacts with ambient O_3, to form NO_2 and oxygen (O_2) within minutes.

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(7) Line 31 page 2: you should also mention HONO emissions from semi arid soils, see Oswald et al., Science 341, 1233, 2013. "N-fixing microbial species occur": not clear enough

We added HONO emissions to the beginning of the paragraph. Furthermore, we concretized the statement on enhanced nitrogen gas emissions under the presence of N-fixing organisms.

Findings from Oswald et al. (2013) suggest that gaseous nitrous acid (HONO), which is rapidly photolyzed

to NO, is also emitted from soils.

Emissions of nitrogen-containing gases, such as NO, N_2 and N_2O increase dramatically in soils with enhanced nitrogen availability due to the presence of N-fixing microbial species and plants (Virginia et al., 1982, van Groenigen et al., 2015).

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(8) Line 6 page 3: Soils emission depend also on pH, N content (not only N input).

We added these two properties and updated the paragraph accordingly.

Soil emissions of trace gases depend on a wide range of ambient environmental conditions such as soil type, soil moisture, temperature, pH-Value and nitrogen content (Conrad, 1996; Ludwig et al., 2001; Meixner and Yang, 2006; Oswald et al., 2013).

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(9) Line 8 page 3: In the Sahel, the presence of cattle is an important provider of organic fertilization. This should be mentioned. See for example Delon et al. (2010), already referenced in your paper.

We agree with the referee that this type of fertilization should be mentioned. Therefore, we inserted the following statement:

In remote regions like the Sahel, where synthetic fertilizer is limited, manure plays a prominent role in the fertilization of agricultural fields and can contribute significantly to the input of organic nitrogen into the soil (Schlecht and Hiernaux, 2004; Delon et al., 2010).

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(10) Line 11 page 3: Add some recent publications of pulsing. Such as Kim et al., Biogeosciences, 9, 2459–2483, 2012, Wang et al., Volume 6(8), Article 133, Ecosphere, 2015.

We like to thank the referee for the suggested literature on NO_x emissions from soils and updated the paragraph accordingly.

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(11) Line 6 page 4: Section 3 is mentioned, but sections 1 and 2 should be mentioned first.

We inserted the following note at the beginning of the paragraph:

The paper is organized as follows: in section 2, all data products used within this study are presented.

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(12) Line 7 page 4: "This approach..." should be "In section 4, this approach..."

Done. The new sentence reads:

In section 4, this approach is then applied to areas with different spatial extents.

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(13) Line 10 page 4: precise which governing parameters you refer to.

Done. The new sentence reads:

In a second step, we focus on Africa and the Sahel region, in specific, and separate the analysis for different seasons. For this region, we investigate fundamental relationships between soil emissions and some of their governing parameters, i.e. soil moisture content, temperature, air humidity.

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(14) Line 11 page 4: "also" is not at the right place in the sentence.

The updated sentence now reads:

Within this analysis possible interferences from other parameters are also investigated, and detailed sensitivity studies are conducted.

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(15) Line 19 to 26 page 4: the way of presenting the different points ((i) (ii) (ii)) is not easy to read. Making proper sentences would be more readable.

We updated this paragraph as suggested. The edited lines now read:

Tropospheric VCDs are usually derived in a multi step process (e.g. Boersma et al., 2004, 2007; De Smedt et al., 2008, 2012). First, total slant column densities (SCDs) are retrieved, i.e. the integrated concentrations along the effective light path, by fitting the measured spectrum with a model taking into account all other absorbers in the atmosphere. Second, tropospheric SCDs are derived by subtracting the stratospheric column (NO_2) or a latitude-dependent bias estimated over the Pacific (HCHO). Third, the tropospheric SCDs are then translated to tropospheric VCDs.

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(16) Line 19 page 6: "Relative fluxes": relative to what?

We deleted the word "relative" and exchanged it with are more concise description of the relation.

The processes of nitrification and denitrification, which govern sNOx fluxes, are closely related to the soil water content (Meixner and Yang, 2006).

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(17) Line 27 page 6: reformulate sentence beginning with "The data sources..."

The updated sentence now reads:

The data sources include active (scatterometer) and passive (radiometer) microwave observations acquired preferentially in the low-frequency microwave range.

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(18) Line 13 page 7: Sentence beginning with "The Moderate..." is difficult to understand.
Please reformulate

Done. The updated sentence now reads:

The MODIS global monthly fire location product MCD14ML (Terra and Aqua combined, Giglio et al., 2006) is used to filter out locations affected by fires.

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(19) Line 17 page 7: Specify "others". Specify the time period and the time resolution used.

Done. The updated paragraph now reads:

In order to understand the prevailing meteorology and filter for special circumstances in the Sahel region, modelled data of air temperature, pressure, humidity as well as wind fields are taken from the ECMWF ERA-Interim analysis (Dee et al., 2011). The model data is acquired at a spatial resolution of 0.25° and a temporal resolution of 6 hours over the period from 2007 to 2010. The data is publicly available via http://apps.ecmwf.int/datasets/.

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(20) Line 5 page 8: the time period is precised here, it should be precised earlier.

We added this information in the introduction.

(ii) high spatial resolution, which is both achieved by expanding the time span of the study to several years (2007 to 2010) enabling an investigation of single grid pixels of 0.25° with reasonable statistics.

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(21) Line 16 page 8: "in the Sahel and shorter" should be "in the Sahel to shorter periods"

The edited sentence now reads:

However, the length of drought phases are quite different for semi-arid areas in the world, varying from very long (several months in winter) in the Sahel to shorter periods (several weeks to months in summer) in South West Africa.

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(22) Line 20 page 8: precise that background level is precipitations < 2 mm during 60 days.

We realize the concern of the referee. However, we think this issue is already sufficiently explained by the sentence before.

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(23) Line 3 page 9: Mention that fig 3 will be described below in the Results paragraph.

We thank the referee for this note as it reveals a typo in the manuscript. Figure 3 should not be mentioned in the methodology. Instead, we refer to figure 2 in the paragraph before. We updated the corresponding part to:

In the example shown in Fig. 2 a 0.25° x 0.25° pixel is chosen which provides a complete NO_2 time series over 10 days.

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(24) Line 15 page 9: Ad "the" between "Although" and "best". This sentence is confusing, in the sense that you write that analysis cannot be not in the Tropics, Northern America, Europe, South Asia? Please explain.

We changed this sentence to improve the readability:

For most regions in the world enough data points are found for our analysis; exceptions are regions with no pronounced seasonality in rainfall (e.g., tropical rainforests, North America, Europe) and regions where rain occasionally falls during the dry season (Southeast Asia). Our algorithm is not optimized for those regions.

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(25) Line 23-24 page 9: Sentences need to be reformulated.

We changed this sentence to:

The corresponding results for NO_2 VCDs observed by GOME-2 and SCIAMACHY are similar to Fig. 3d, but are more affected by noise due to poorer statistics (see appendix D).

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(26) Line 2 page 10: Transports are mentioned to explain NO2 VCDs enhancements. Neither transports nor industries and traffic were mentioned in the introduction as possible sources. Why should transport explain enhanced emissions at the first day of rain?

We do not refer in this context to transportation related to traffic by ships or cars but to long-range transport of air masses. This potential influence was not mentioned in the introduction as this effect was not expected to be relevant for this study. However, for some grid pixels in proximity to coastal areas we find slightly enhanced NO_2 VCDs. This could be possibly explained by advection of polluted air synced with the moving precipitation system around the first day of rainfall (i.e. a change of wind direction and speed favouring local enhancements in NO_2 VCDs). For clarification we replace "transport" with "advection". The updated sentence now reads:

However, over the Mediterranean sea and in proximity to coastal regions over oceans small-scale enhancements in NO_2 VCDs are detectable which might be related to advection.

(27) Line 7 page 10: the dry season in the Sahel lasts nearly 8 months. Was the month of July tested as part of the months when the first day of rain occur? Sometimes when the wet season is late, the first day of rain occurs only in July. See Lebel et al., Journal of Hydrology 375 (2009) 52–64.

For the sensitivity studies as well as the emission calculation we only investigated the months April, May and June. Fig. 1 depicts the number of triggered rain events that fulfil a required threshold of 2 mm after 60 days of drought for individual pixels in the Sahel for the April-May-June period. It can be seen that within the 4 years (2007 to 2010) we investigate in this study, the first rain events of the wet season in the Sahel region all occur within these months.

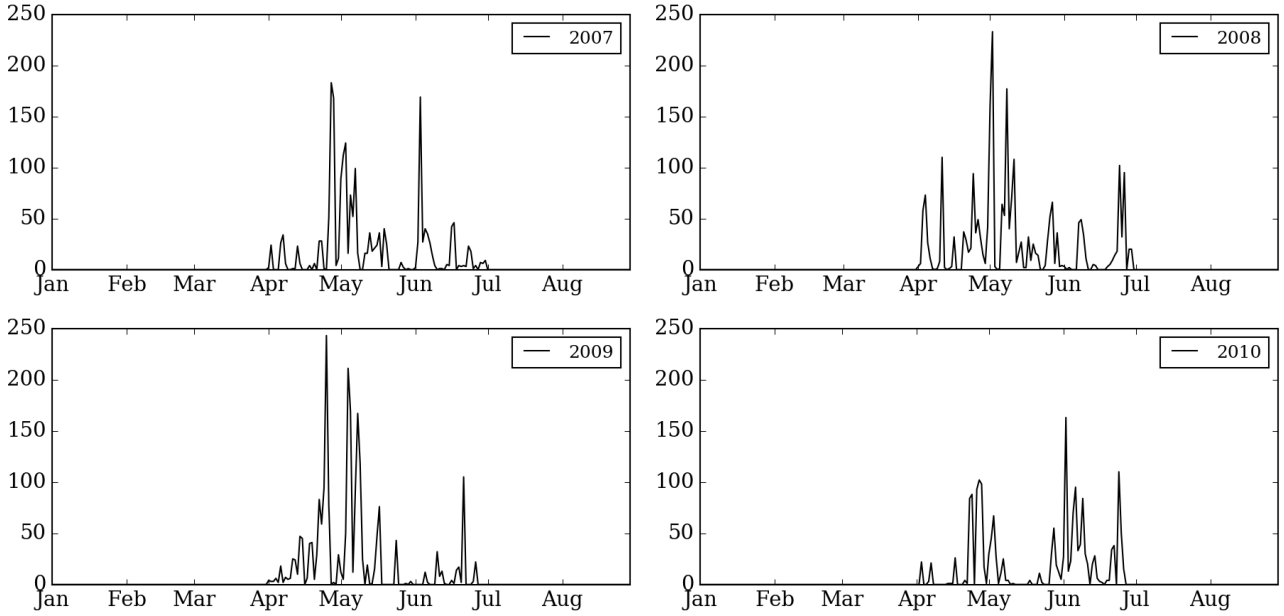

Figure 1: Number of triggered rain events that fulfill a required threshold of 2 mm after 60 days of drought for individual pixels in the Sahel for April-May-June.

(28) Line 14 page 10: add “next” after “gradually with the start of the...”

The modified text now reads:
The subsequent dry season begins in October and ends gradually with the start of the next wet season in April/May/June (AMJ-period).

(29) Line 19 page 10: Why did you exclude the western Sahel of the studied zone?

We provide our answer and corresponding changes in the manuscript in question #4 of this document.

(30) Lines 22-25: this paragraph should be in the methodology section.

We moved this paragraph to the end of the methodology section.

(31) Line 24 page 10: You mean that for a period less than 2 months, the N enrichment is not sufficient? Can you give references for that?

We did not intend to convey that the N enrichment for a period less than two months is too short. It is meant that during drought N enrichment dominates over N depletion in the soil. In appendix B, we find

pulsing events for even short periods of about one week. However, the observed enhancements in NO_2 VCDs on Day0 are lower for such cases. For clarification we revised this paragraph as follows:

The drought period of about two months is chosen as we find the highest response in NO_2 with this setting. In appendix B, the impact of drought lengths on the derived soil emission pulses is investigated.

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(32) Line 11 page 11: smaller instead of smallest.

Done.

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(33) Line 25 page 11: the expression "dry phases" is not understandable here. It is only understandable once reading the following sections. May be a word or two to explain could be useful.

We updated the sentence to:

In sections 4.4 and 4.5, this important finding is studied more in detail by analyzing the NO_2 levels after Day0 depending on wind conditions and the precipitation on Day1 and beyond.

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(34) Lines 27-30, page 11: do you think the enhancement of HCHO the day before Day0 has a link with air moisture? What are the processes that may be involved?

In our opinion, the presented HCHO data are dominated by noise and we do not attempt to interpret them.

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(35) Lines 6-7 page 12: industrial activities and strongly fertilized agriculture are hardly found in the Sahel even in the Southern part. You mean may be the southern part of West Africa?

The original formulation indeed reflects the case for the more Western Part of the Sahel (which still overlaps with the Eastern region we investigate). Still, anthropogenic induced emissions are more frequent in the Southern part compared to the Northern part in the region which we investigate. Thus, we updated the sentence in the manuscript slightly:

Anthropogenic activity and related emissions such as domestic fires or fertilized fields are at a very low level, and originate mostly from the southern, more populated part of the Sahel (Delon et al., 2010).

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(36) Line 9 page 12: low nitrogen input and nitrogen content. The role of cattle should be developed in this paragraph. Mineral fertilizers are not widely used in the Sahel, while organic fertilization plays a non negligible role in sNOx emissions. See for example Schlecht and Hiernaux, Nutrient Cycling in Agroecosystems 70: 303–319, 2004.

We would like to thank the referee for the this remark and the suggested reference. In this section, however, we think that manure is not central to our analysis. Thus, we propose an additional paragraph in comment #9 in the introduction which covers the role of manure and its importance on the soil system.

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(37) Line 20 page 13: a short conclusion of possible under or overestimation of cloud effect on NO2 VCDs should be useful.

We updated the corresponding section and point out that the shielding effect dominates for cloudy conditions.

Addition the beginning of section 4.4.3:

We have investigated possible cloud effects on our results by analyzing the temporal evolution of the mean cloud fraction (CF), NO_2 VCDs, and NO_2 SCDs around the precipitation event. The latter was added

as it provides the actual measured signal without involving a tropospheric AMF, which is generally very sensitive to clouds.

Update to the last paragraph of section 4.4.3:
Interestingly, while there is a strong systematic enhancement of the FRESCO and OMCLDO2 cloud fractions, the NO_2 SCDs show no peak around Day0 for GOME-2 and OMI. This indicates that clouds effectively shield the pulsed soil emissions.

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(38) Line 9 page 14: ",thus," may be removed from the sentence.

Done.

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(39) Line 22 page 14: "at" instead of "on" the same latitude.

Done.

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(40) Line 16 page 15: "largest" instead of "larger".

Done.

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(41) Line 3 page 16: specify "Eastern" Sahel, because the analysis has not been made in Western Sahel.

As the global analysis, presented in Fig. 3, indicates enhancements not only in the Eastern Sahel, but also in the western and central part, we would like to keep this sentence.

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(42) Line 5 page 18: As indicated in Aghedo et al., Atmos. Chem. Phys., 7, 1193–1212, 2007, anthropogenic pollution is not likely to reach sahelian latitudes. An important added value could be brought here to this paper. As mentioned in the general comments, the budget over the whole studied area (i.e. Eastern Sahel) could be calculated in TgN for the studied period.

We added the budget calculation (as described at the top of this document) to the end of section 5.1 as well as to the conclusion section. We would like to thank the referee for pointing out that anthropogenic pollution probably does not reach the Sahel. Still, generally higher NO_2 VCDs are observed in the Tropics which might be due to biomass burning (anthropogenic influenced) that possibly could reach the Sahel. Therefore, we are quite cautious to exclude this potential interference in the NO_2 VCDs over the Sahel region and would like to leave this paragraph as is.

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(43) Conclusions Lines 7-13: difficult to follow with this a) to e) way of presenting the ideas. Proper sentences would be more readable.

We understand the concerns of the referee, but we still think that this way presents our main improvements over previous studies best.

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(44) Line 15 page 18: "maximum amount of precipitation"? do you mean the 2 mm threshold? In that case it is the minimum amount.

The word "maximum" is, indeed, ambiguous in that sense. Referring to the rain threshold "minimum and maximum amount of precipitation" is meant in this sentence. For simplicity, we deleted the word

"maximum" so that the phrase now reads:

(i) evaluate the impact of the a-priori assumptions on thresholds for daily rainfall, i.e. the amount of precipitation and the required duration

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(45) Line 17 page 18: "shown" instead of "showed".

Done.

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(46) Line 20 page 18: again see Oswald et al., 2013, where laboratory measurements made on semi arid soils are presented.

As we only want to indicate our main conclusions of our study we would like to stick to the current text. However, we added the contribution of HONO emissions (Oswald et al., 2013) to the introduction section as described above in the document.

Technical corrections

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(47) Line 11 page 3: "lab" should be laboratory

Done.

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(48) Line 13 page 6: Upper case to begin the paragraph is needed.

Done.

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(49) e.g. throughout the text should be in italics.

Done.