Response to Anonymous Referee #1 comments to paper "The contribution of soil biogenic NO emissions from a managed hyper-arid ecosystem to the regional NO_2 emissions during growing season"

The authors would like to thank Anonymous Referee #1 for his/her constructive and detailed comments as well as his/her very helpful suggestions. In our revised MS, we have performed three major corrections: (1) complete restructuring of the original MS, (2) consideration of the partitioning between NO and NO_2 for the top-down estimates (see Fig. 11), and (3) comparison of bottom-up estimates (see Fig. 11) on top-down estimates taking into account the emission ratio between satellite overpass (at 13:00 LT) and average diurnal emission.

We addressed the individual comments (in bold **Times Roman***) for each reviewer as indicated below (in italics).*

Response to the summary and general comments:

1. In my opinion however, this article only marginally fits within the scope Atmospheric Chemistry and Physics. It is quite local and very technical in nature, and more importantly is primarily focused on the application of land surface remote sensing and the development of a "General Tool" for ArcGIS. Of the generous 41 pages of ACPD text (not to mention the 22 figures), I estimate 3 or 4 of these pages are directly relevant to implications for general atmospheric chemistry (though the actual impacts on chemistry are never explored). While I believe there could be an audience in ACP for this manuscript, I wonder why it is not better suited for Biogeosciences, Geoscientific Instrumentation, or Geoscientific Model Development as examples. I welcome discussion from the authors (and the editor's discretion) on this issue.

Our work is an interdisciplinary study. As is often the case with interdisciplinary work, in our study different methods are used. Thus, it is difficult to find the 'perfect' scientific journal that exactly matches the scope. The major outcome of our paper is that in arid regions soil emissions can dominate the NOx emissions during the growing season and that in general soil emissions from such regions are systematically underestimated. This is an important finding for atmospheric chemistry. Thus, in our opinion, the paper fits well to ACP. To balance the different parts of the paper in a better way and to make the main focus more clear, in the revised MS we moved several parts (including the description of the GGTP model to supplement).

2. If the paper is to be published in ACPD, I advise a significant revision and restructuring of the manuscript……………………………………………………………..
I would additionally suggest restructuring the article to better streamline the material……………………………………………………………………………………….
Given that ACP allows for deviation from the traditional "Intro / Methods / Results Conclusions" headings, my suggestion to improve readability and clarity would be to reorganize all the material (methods and results) into the following sections: (1) Soil sampling and lab measurements (with results); (2) Development and application of GGTP using Landsat observations and lab results (with validation and the resulting 2-D distribution of biogenic soil NO emissions); (3) Scaling of bottom up biogenic NOx inventory to monthly means, and the results; (4) Development of a bottom-up anthropogenic inventory; (5) Discussion of soil vs. anthropogenic contributions based on these bottom-up estimates; (6) Development of top-down estimate and

comparison with bottom-up inventories.

The authors would like to thank Anonymous Referee #1 for his/her very helpful suggestions. With regard to restructuring of the manuscript, the suggestions of Referee #1 and Referee #2 significantly overlap with following aspects:

- *Scaling of bottom up biogenic NOx inventory*
- *bottom-up anthropogenic inventory*
- *bottom-up soil vs. anthropogenic contributions*
- *top-down estimates*
- *bottom-up total soil emissions vs. top-down estimates*

Given the restructuring of the revised MS, most part of the description of the GGTP model was transferred to the supplement. With consideration of the suggestions of both Referees and the guidelines of manuscript preparation of ACP (only three levels of sectioning are allowed), the revised MS has now the following sections:

Section 1: Introduction

Section 2: Materials and methods

2.1 Site description and soil sampling

2.2 Remote sensing and accompanying data

2.3 Bottom-up calculation of biogenic NO emission estimates

2.3.1 Laboratory determination of land use type specific net potential NO fluxes

2.3.2 Determination of land use types and corresponding soil surface temperatures from Landsat Imagery

Land use classification

Land surface temperature T_s

2.3.3 Temporally high resolution data

Soil temperature, T_{soil}

Gravimetric soil moisture θ_g

Fertilizer factor FF

2.3.4 Monthly soil biogenic bottom-up emissions of Tohsun oasis

2.4 Bottom-up anthropogenic NO_2 emission estimates

2.5. Top-down total NO_2 emission estimates from satellite observations

2.5.1 Satellite derived tropospheric VCD_{NO2}

2.5.2 Monthly total NO_2 emissions of Tohsun oasis (top-down)

Section 3: Results and Discussion

3.1 Bottom-up soil biogenic and anthropogenic emissions from Tohsun oasis

3.1.1 Laboratory derived net potential NO fluxes

3.1.2 Land use type specific net NO fluxes based on soil temperature, soil moisture content, and enhancement by fertilizer application

3.1.3 Monthly soil biogenic emissions of NO and HONO from Tohsun oasis (bottom-up)

3.1.4 Monthly anthropogenic emissions from Tohsun oasis (bottom-up)

3.1.5 Soil biogenic vs. anthropogenic emissions of Tohsun oasis

3.2 Top-down satellite derived total NO_2 emissions from Tohsun oasis

3.2.1 Spatio-temporal variation of the tropospheric VCD_{NO2} measured from satellite

3.2.2 Monthly total NO_2 emissions from Tohsun oasis (top-down)

3.3 NO_X emissions of Tohsun oasis: bottom-up vs. top-down

Section 4: 4 Summary

3. Another problem I have is that there is little-to-no mention about uncertainties in the

HONO emissions that have been estimated. It is not clear to me whether HONO release was measured directly in the lab incubation experiments presented here. If not, then I think a more significant treatment of the uncertainty in the estimate is required. Are the HONO emissions an estimated fraction of what was measured in the lab? Or added to the amount measured in the lab based on a scaling function from the literature? Scaling up the HONO emissions to monthly means implies that they are driven by identical functions as the soil NO emissions (i.e. same dependence on soil temperature, moisture, fertilizer application). Has this been shown to be true? Or is it assumed? If the latter, what is the rationale? Given that the calculated HONO emissions can be on the order of half of the total biogenic emissions, if these were not directly measured in the lab by the present authors, the uncertainty associated with these estimates must be discussed further.

Oswald et al. (2013) found, with regard to tropospheric chemistry, that HONO emissions were one of the most relevant sources of reactive nitrogen particularly in arid areas. Two of the co-authors of our present manuscript (T. Behrendt and F.X. Meixner) have also been co-authors of the of that publication: Oswald, R., ***Behrendt, T.****, Ermel, M., Wu, D., Su, H., Cheng, Y., Breuninger, C., Moravek, A., Mougin, E., Delon, C., Loubet, B., Pommerening-Röser, A., Sörgel, M., Pöschl, U., Hoffmann, T., Andreae, M.O.,* ***Meixner, F.X.****, Trebs, I.: HONO emissions from soil bacteria as a major source of atmospheric reactive nitrogen, Science, 341, 1233-1235, 2013. T. Behrendt and F.X. Meixner have been directly involved in the laboratory experiments for the determination of HONO emissions and its relation to biogenic NO emissions. For these laboratory experiments, a total of 17 soil samples were used, which has been gathered globally from arable and arid lands. Five of these soil samples (S13–S17, s. Oswald et al. 2013) originated from the immediately neighboring region of our study area. Due to the same climate conditions, identical irrigation regimes and fertilizer applications, these soils have similar properties to those used in this study. For the laboratory experiments of Oswald et al. (2013), a state-of-the-art laboratory dynamic chamber system has been applied, which was just a next-laboratory version of that used our present study. HONO concentrations were measured by a long path absorption photometer (LOPAP) and NO concentrations by a chemiluminescence detector (identical to that used in our present study). Calculation of net potential HONO and NO fluxes by Oswald et al. (2013) was identical to that used in our present study. The result from Oswald et al. (2013) is that optimum emissions of NO and HONO have comparable magnitude and occur at similar soil moisture contents. Considering the comments of Anonymous Referee #1, in our revised MS the following was added:*

<u>Page 15, lines 9-20</u>*: "Since the HONO emissions were not directly measured within the present study, the land use specific constant scale factor namely the ratios of HONO to NO releases for the optimum conditions to estimate the HONO emissions ((*$F_{N,opt}$*(HONO) to* $F_{N,opt}$*(NO)) were adapted from the study of Oswald et al. (2013). This is justified by: (a) the strong assumption that the soil from our study emits the same magnitude of HONO as the soil by Oswald et al. (2013) because they have the same soil properties, and they experienced the same irrigation regimes and fertilizer applications, and (b) the soil release of HONO and NO by Oswald et al. (2013) has been measured also in the Max Planck Institute for Chemistry by a next-laboratory version of that dynamic chamber system which was used to measure the soil release of NO in the present study. Consequently, corresponding ratios for optimum condition*

of HONO and NO emissions (i.e., $F_{N,opt}$(HONO) to $F_{N,opt}$ (NO)) have been used to calculate monthly HONO emissions from Tohsun oasis from the measured NO emissions.".

<u>Page 27, lines 12- 19</u>: *"Nevertheless, the optimum soil water content (SWC_{opt}) has a strong influence on the ratio of $F_{N,opt}$(HONO) to $F_{N,opt}$ (NO). We used this ratio for the calculation of monthly HONO emissions by scaling corresponding NO emissions. However, particularly for cotton soil, the value of the optimum gravimetric soil water content for NO (2.1 %) is slightly lower than the corresponding value for HONO (3.6 %, s. Oswald et al., 2013). This may result in a certain error of the calculated HONO emissions, which unfortunately can not be quantified more precisely at the moment".*

<u>Page 27, line 24 -26</u>: *"Nevertheless, the qualitative and, in particular, the quantitative agreement between bottom-up and top-down estimates indicates that our assumptions are largely justified."*

4. The other prominent issue I have is the reference to "NO2 emissions" throughout the manuscript. It is confusing for an atmospheric chemistry audience whether or not the authors have accounted for both NO and NO2 in equilibrium, or if all NOx emissions are being reported as mass NO2 for some conventional reason (I presume the authors rarely intend to mean primary NO2 emissions?). In parts, it seems like the authors assume for simplicity that all NO is converted to NO2. Since the paper is primarily focused on biogenic emissions of NO and HONO, I don't understand why the authors have chosen to express everything as NO2 (instead of, say, just simply mass nitrogen). If this is because satellite NO2 columns are being used in the top-down estimate, the actual NOx emissions (in order to compare with the bottom up inventory) still depend on the ambient NO:NO2 ratio. The issue of NO and NO2 in pseuo-photostationary equilibrium is mentioned, but not dealt with in the paper. Doesn't this ratio depend on season and time of day, and won't that impact how emission inventories are estimated?

Anonymous Referee #1is right, that all NOx emissions are being reported as mass NO_2. For that purpose soil biogenic emissions of NO and HONO (given as mass) were multiplied by the respective ratios of the corresponding molecular weights (M_{NO2}/M_{NO}, M_{NO2}/M_{HONO}). The reason for this procedure is that the reported mass becomes independent from the partitioning ratio between NO and NO_2. We added this information to the manuscript in sec. 2.3.4.

With regard of above comments of Referee #1, in our revised MS following sentences were added:

<u>Page 15, lines 25-31</u>: *In the present study, the total bottom-up biogenic emission estimates (NO and HONO) were expressed as NO_2- partly because the reported mass of emitted nitrogen compounds becomes independent from the partitioning ratio between NO and NO_2. Additionally, the anthropogenic and top-down emissions estimates are primarily derived for NO_2. In this study, the mean primary NO_2 emissions were not considered and all NOx emissions are being reported as mass of NO_2. For that purpose soil biogenic emissions of NO and HONO (given as mass) were multiplied by the respective ratios of the corresponding molecular weights (M_{NO2}/M_{NO}, M_{NO2}/M_{HONO}).*

While addressing this point of the reviewer it turned out that for the interpretation of the satellite results we indeed forgot to account for the partitioning between NO and NO_2. We are very thankful to the referee to pointing our attention to this point. We added the following information to section 3.2.2 (page 24, lines 28-31):

"In addition, a correction for the partitioning between NO and NO_2 in the atmosphere has to be made. For that purpose we multiply the NO_2 values by typical ratios between NOx and

NO_2 of about 1.3 (see e.g. Seinfeld and Pandis, 2012)." We also updated figures 11 accordingly (see response of comment 30).

Response to the Technical/Specific comments:

1. p. 34534, l. 11-13: "The results show that the soil biogenic emissions of NO2 during the growing period are (at least) equal until twofold of the related anthropogenic sources." Do the authors mean "to" instead of "until"?
"until" has been changed for "to" in revised MS.

2. p. 34534 l. 17-18: "The resulting total NO2 emissions show a strong peak in winter and a secondary peak in summer, providing confidence in the method" It's not clear from the information in the abstract why this provides confidence in the method.
We changed the sentence into (Page 2, lines 22-25): "The resulting total NO_2 emissions show a strong peak in winter and a secondary peak in summer, providing confidence in both completely independent methods."

3. p. 34535, l. 1-2: "The present evolution of anthropogenic as well as biogenic NOx sources triggers a potential increase of global tropospheric O3 concentrations". The meaning of this is not clear; please rephrase.
In revised MS (page 3, line 5-8) the sentence was added: "Ozone is usually generated in polluted, industrialized regions, where ambient levels of NO_X are high. Moreover, the intensification of agriculture is also associated with increased NO emissions, which also causes an increase of available nitrogen in the atmosphere (Denman et al., 2007)".

4. p. 34535, l. 3: "which photo-stationary equilibrates with NO2" is not grammatically clear.
The sentence was changed to"which reaches a photo-stationary equilibrium with NO_2 within a few minutes".

5. p. 34535, l. 7-9: "Other globally important sources are soil biogenic NO emission (10–40 %), biomass burning (13–29 %) and lightning (5–16 %)." Please offer references to these estimates, or make it clear these are referring to citations from the preceding sentence.
The corresponding references are given in the previous sentence. We added "and" to connect both sentences.

6. p. 34535, l. 20: Is there a connection between "bushy" and "dryland farming"?
"bushy" has been corrected for "bushy vegetation".

7. p. 34535, l. 27: "convincingly" – This subjective qualification seems awkward to me given that the authors are referring to some of their own work.
- *We deleted 'convincingly'*

8. p. 34537, l. 25: "s. Fig 2" Are the authors abbreviating "see" to "s."? This is done in other places throughout the manuscript, while in some places they write "see Fig xx". At first I thought they were referring to a supplement.
The authors like to thank Anonymous Referee #1 for his/her comment concerning our spelling variation. All "s." in MS have been changed to "see".

9. p. 34538: "and 1 September mm" should be "and September 1 mm"?

- corrected, thanks.

10. p. 34537-34538: Here there are 3 paragraphs about the site/region, then only two sentences about the actual soil sampling: : : I had many questions: How much soil is sampled? How deep? Is the vertical structure kept, or does the soil get mixed? Is only one sample from each site taken, or duplicates? How is it removed and subsequently treated? How is it stored, and for how long, until lab experiments were performed?

With regard of above comments of Referee #1, in our revised MS the following sentences were added:

<u>Page 6, line 14-21</u>: "Since the primary production and consumption zones for NO are located within a very shallow layer at the soil surface (1 to 5 cm, e.g. Meixner and Yang, 2006), only soil samples of 5 cm depth of the top soil were taken. At each of the sampling sites ten soil samples (approx. 150 g) with stainless steel soil cores (5 cm length, 2.5 cm radius) were taken (randomly from a 10x10m^2 area); these were then combined to constitute a representative soil sample of approximately 1.5 kg in mass. These soil samples were stored at 4°C for a maximum 3 months until analysis, since the microbial alterations are not expected within storage for up to 3 months at 4° C (Stotzky et al., 1962)".

11. p. 34539: l. 6: Change "begin" to "beginning" (and likewise in other instances)

"begin" has been changed to "beginning"

12. p. 34539, l. 19: Can the authors demonstrate there is no significant trend in the NO2 columns in the region during from 2006-2010, allowing them to use the mean instead of just data from 2010?

We have investigated the seasonal patterns of satellite-derived NO_2 column densities (2006-2010) and monthly mean tropospheric VCD_{NO2} during 2006–2010 over the selected areas (Fig. 9 and Fig 10 in the revised manuscript). Thereby, the authors want to demonstrate the importance of representativeness of satellite-derived NO_2 and the true seasonal effect on tropospheric NO_2. That is because a mean value of longer periods leads to significantly reduced uncertainties and it can be better demonstrate the representativeness of satellite-derived NO_2. As can be seen from Fig. 10, there exist some inter-annual variability but no systematic trend of VCD_{NO2} (2006-2010) within the study area and its surrounding area.

Tropospheric NO_2 column densities for the year of 2010 were used for top-down estimates of total NO_2 emissions and comparison with total bottom-up emission estimates (biogenic + anthropogenic).

Following sentences have been added in the revised MS:

<u>Page 18, lines 6-11</u>: "To check the representativeness of satellite-derived NO_2 on the .regional scale, we investigated the inter-annual variability of tropospheric VCD_{NO2} from 2006 to 2010. That is because a mean value of longer periods leads to significantly reduced uncertainties and it can be better demonstrate the representativeness of satellite derived NO_2. Evidence of clear seasonal pattern in the long-term observations can reflect the true seasonal effect on tropospheric VCD_{NO2}, which is of substantial importance with respect on the intended comparison "bottom-up" versus "top-down"".

<u>Page 18, lines 26-28: "</u>Top-down NO_2 emissions from Tohsun oasis (ng m^{-2} s^{-1}, in terms of NO_2) were calculated from all VCD_{NO2} -whose pixels cover the area of Tohsun oasis and where each represents the particular monthly mean of 2010-"

<u>Page 23, lines 25-27</u>:"Comparing the mean distribution (2006-2010) of the different seasons, it is clearly visible that for all seasons enhanced values are found over the area of Tohsun oasis (compared to the surrounding desert)".

13. p. 34539, l. 20: “Four different areas” – Do these correspond to Figure 20? If so, can this be stated here? Or can the areas be drawn in Figure 1 as well? Otherwise can the authors more clearly state how the different areas were selected?

The four different areas correspond to the Figure 20 of original MS (in revised MS Fig. 3). It is not possible to indicate these areas also in Fig. 1 because this figure covers only a small part of the area shown in Fig. 20. With regard of above comments of Anonymous Referee #1, in our revised MS the following sentences have been added:

Page 18, lines 15-23: “Satellite observations reflect contributions from different emission sources. In principle the retrieval of NO_2 emissions over a city or oases is similar. However, usually the emissions from cities are higher and the corresponding enhancements can be better identified compared to the background. In order to establish the relationship between biogenic NO_2 and satellite-derived NO_2, it is necessary to understand their spatial patterns with respect to the locations and shapes of the potential sources. Thus, four different areas (see Fig. 3) were selected to represent (1) typical agricultural areas (study area) as biogenic source, (2) mixed land use areas (agricultural & small urban) as biogenic and anthropogenic sources, (3) large urban areas as anthropogenic source, and (4) desert area as background source “.

14. p. 34539: What are the sources of the land use map and traffic map referred to here?

„2010“ changed to „Ma, 2010“

15. p. 34541: If this particular method has been used for the past two decades as the authors say, maybe this section can be abbreviated to simply the final paragraph? I actually found myself asking other questions more specific to this implementation that could have been covered: What is the geometry of the dynamic chambers – is there a specific reference from the above list which uses an identical chamber? Is the area that the soil takes up in the dynamic chamber the same as the area of the soil sampled? (i.e. is the thickness of the sample kept the same?) And most important, is there a specific reference that shows that these laboratory methods are equivalent/identical to an in-situ dynamic chamber method in the field?

”The laboratory determination of land use type specific net potential fluxes” (revised MS sec. 2.3.1) is one of the most important parts of our study. As is evident from our study, the laboratory method was successfully performed as one of the essential methods for bottom-up emission estimates. We, therefore, believe that the method has to be addressed in the paper to ensure the description of the methods section in a holistic way. However, in our revised MS the method was described as clear and simple as possible since the laboratory method has been used for the past two decades.

In the revised MS the following information has been added:

Page 8, lines 12-21: The laboratory incubation method is usually based on a dynamic chamber system which consists of gas dilution, thermostat valve, thermostat cabinet, and analysers (Behrendt et al. 2014). The dynamic chamber was made of Plexiglas with a diameter and height of 9.2 cm and 13.6 cm, respectively. Five chambers were used for soil samples while one was kept empty as a reference and the thickness of the sample in the dynamic chamber kept the same (approx. 5 cm). Laboratory measured NO fluxes have been repeatedly shown to be in good agreement with those measured in the field (Mayer et al., 2011; van Dijk et al., 2002, Ludwig et al., 2001).The wetted soil samples were placed into five chambers while one was kept empty as reference chamber. Then, the net potential NO release rates were measured until the soil was completely dried out. Laboratory measured NO fluxes using the incubation method have been repeatedly shown to be in good agreement with those measured in the field (Mayer et al., 2011; van Dijk et al., 2002, Ludwig et al., 2001).

16. p. 34541: Here I am a bit confused about the estimated HONO release. Is HONO ever measured in these particular lab incubation experiments? Or is HONO assumed to be a certain fraction of the total NO that is measured? Or is the HONO estimate added to the NO measured based on the scale factors in the literature? If they are not directly measured, what is the uncertainty associated with these estimates?
The HONO emissions were not directly measured within the present study and the ratios of HONO to NO fluxes were adapted from the study of Oswald et al. (2013). In our revised MS, we have added a section related to the HONO estimates (sec. 2.3.4, pages 14, 15) and the uncertainty associated with these estimates were discussed in section 3.3 (page 27).

17. p. 34542, l. 24: "in particular" – This makes it sound like there are other schemes or calculations that are required (and that have been implemented), but that aren't being described here.
these were described by the sub-schemes from S1.1 to S1.9 in the supplement. In the revised MS (in supplement page 2, line 9), "see S1-S9" was added.

18. p. 34542: Development of GGTP – Given the aim and focus of the ACP journal, I think all of sections 2.4.1 to 2.4.6 (and accompanying figures) should be in supplemental information; this is an extremely long part of the paper. Moreover this is all a description of land surface products and calculations. Since I am not a land surface remote sensor, I found these descriptions enlightening, but not exactly germane to the ACP focus. From Sections 2.4.1 to 2.4.6, the authors are implementing calculations or methods that have been published and accepted elsewhere. In my opinion, a summary of the sections (e.g. 1 sentence each?) seems like it would suffice, with all the material moved to a supplement. Likewise, Section 3.2 and Section 3.4.
The authors would like to thank Anonymous Referee #1 for his/her suggestions. In our revised MS, we have considered the above comments of Referee #1. Given the restructuring of the revised MS, the section 2.4.1, 2.4.6, 3.2 and 3.4 were replaced to supplement (S1.1, S1.6, S1.7 and S1.8).

19. p. 34543, l. 3: "causing that scanning patterns exhibited wedge-shaped" – Replace "that" with "the"?
"that " was replaced by "the".

20. p. 34549, l. "the level at which plants will irreversible" – I think there are some words missing in this sentence.
this sentence changed to " will become irreversible"

21. p. 34551, l. 20: "images of sufficient quality" – How exactly was this determined?
With the "images of sufficient quality", is actually meant here the cloud free images. In revised MS (page 11, line 28 in supplement), "(cloud free observations)" was added.

22. p. 34553, l. 21: Insert a comma between "NO flux" and "theta(x,y)"
many thanks, the comma was added.

23. Section 2.5: I find almost all of the initial discussion (until heading 2.5.1) unnecessary, and could be removed for brevity. It could be sufficient to simply say what you did (e.g."Mean monthly land use type specific soil NO emissions were averaged from data on the shorter time scales" and that "the NO and HONO emissions were reported in mass NO2".

In our revised MS, we have considered above comments of Referee #1. Given the restructuring of the revised MS, the initial discussion until heading 2.5.1 was removed and the mean monthly soil biogenic bottom-up emission estimates (NO and HONO) were described in section 2.3.4 (page 14-16).

24. **p. 34557, l. 10: Here the authors first mention "temporal scaling", but what is meant by this is not exactly clear. I think this is ultimately described in a later section, but this awkward division of methods makes it hard to follow.**

Here, additionally "with high resolution (<1h)" was added.

With regard to the above comments of Anonymous Referee #1, in our revised MS the following sentence has been added (page 13, lines 9-11 in supplement):
"Consequently, the mean monthly land use type specific soil NO emissions have to be averaged from data calculated by temporal up-scaling with high resolution (<1h)."

23. p. 34557, l 15: The authors use a constant value of soil moisture content for desert soil. Where is this number from? Is it an average of the data that was collected?

This information was already given in sections 2.2 and 2.4.5 of the original version of our MS. In the revised MS the related content can be found in section 2.3.3 as below:
Page12 lines 15-18: "Evaluation of the MSR® 165 data logger measurements (see sect. 2.2) at the site of the Tohsun County Meteorological station (bare desert soil) has demonstrated a temporally quite constant, very low gravimetric soil moisture content of 0.0028, which has been adapted for the entire growing season (April-September 2010) for the land use type "desert" of the Tohsun oasis".

24. p. 34558, l. 7-10: "result in FF = : : :" – Where did these numbers come from exactly? Lab incubation measurements in the present work? Or those from Fechner 2014 / Behrendt et al. 2014?

The fertilizer factor "FF" was calculated in the present work (sect. S3.3 in supplement). To calculate "FF", we used the dependency of NO-fluxes on the fertilizer amount (FA) which was obtained by Fechner (2014) from the laboratory measurement at the Max Planck Institute for Chemistry. In our revised MS the following sentences were added:
<u>*Page 13 lines 17-19*</u>*: "The impact of the fertilizer application on the NO-fluxes from arable soils of the Taklimakan oases was recently investigated by Fechner (2014) using the laboratory dynamic chamber system described by Behrendt et al. (2014)".*
<u>*Page 13, lines 29-30*</u> *: "dimensionless "fertilizer factor (FF)" applied to the standard net release $J_{NO}(\theta_{g,0},T_{soil,0})$ in Eq. (2)".*
<u>*Page 14, lines 6-9:*</u> *"With the assumption of an exponential decay of the fertilizer effects, corresponding functional relationships for the temporal behaviour of FF and $Q_{10}F$ have been developed (see sect. S3.3, supplement) and temporally high resolution data (30 min) of FF and $Q_{10}F$ were calculated".*

25. Section 2.5.3: I might have missed how the remotely sensed soil moisture index is ultimately used. Temporal scaling of temperature using the observations is described in detail here. How was satellite-inferred moisture used in the subsequent calculations?

The remotely sensed soil moisture index (SMI) which is calculated from Landsat images was used for the calculation 2-D distributions of biogenic soil NO emissions. Related information to this part is described in supplement S1.7 of the revised MS (page 6-8). The SMI are only representative for selected days. However, for any comparison of bottom-up and top-down estimates longer time scales (preferably monthly means) for the biogenic emissions have to be considered. That is because there exists a strong non-linear response of the net potential NO flux to soil temperature and gravimetric soil moisture, as well as to fertilizer applications.

Thus, the local chosen irrigation schedule, the two weeks regular temporal patterns, was used to determine the temporal variation of gravimetric soil moisture. For that, "drying-out" shape function was defined which was scaled by multiples of the soil specific physical quantity "field capacity (FC)". The calculation of the temporal variation of the gravimetric soil moisture for the bottom-up estimates are addressed in our revised MS in more detail (page 12, lines 15-30 and page 13, lines 1-14 in main text; S1.8 in supplement).

26. p. 34558, l. 10: As someone unfamiliar with the geography and development of the area, what evidence is there that Urumqi can substitute for the appropriate sectors of Tohsun County?

Anthropogenic emission estimates have to be calculated at the monthly level because these have to be comparable the biogenic ones. However, the data of fossil fuel consumption from the different economic sectors of Tohsun County were only available on an annual basis. The known mean monthly percentages of annual NO_2 emissions are only known for Urumqi. With regard to the above comment of Anonymous Referee #1, in our revised MS the following sentences were added:

Page 16, lines 20-25: "Mean monthly data of Tohsun oasis have been assimilated (down-scaled) by multiplicative consideration of known mean monthly percentages of annual NO_2 emissions of Urumqi (140 km NNW of Tohsun). This is justified by: (a) coal is the dominating fuel type of energy consumption in both areas (Mamtimin and Meixner, 2011; Pu, 2011), (b) identical arid climate conditions (resulting in identical heating periods), and (c) only from Urumqi there are known mean monthly percentages of annual NO_2 emissions".

27. p. 34559, l. 3-5: Are the authors assuming that the normalized diel variation is constant across different seasons? Perhaps I am not clear on the exact methodology applied here.

It is not assumed that the normalized diel variation is constant across different seasons. In the study, we used 6 Landsat Images to derive land use type specific surface temperature data. From this point data, the land use specific surface temperature for every individual day based on 3rd order polynomials was fitted (Fig. S8 in supplement). Then, in situ-measured original data (5 min) of every particularly day (Fig. S9 in supplement) have been normalized at 10:45 LT (Landsat overflight time) for the growing season (April-September, 2010). As a result, a data set of mean diel variation (30 min) has been created by averaging all respective data from 01 July-30 September (see Fig. 6). These methods were described more clearly in our revised MS (page 11-12 in revised MS and page 13-14 in supplement).

28. p. 34559, l. 5: How sensitive are the results to the assumptions about seasonal temperature evolution (i.e. other interpolation estimates besides the third order polynomial fit)? There is a lot of interpolation between Day 115 and Day 225.

We agree that the interpolation introduces uncertainties to the estimated temperatures. To estimate the corresponding uncertainties we compared results using polynomials of degree 2 and 3 for the fitted curve. We found that R^2 in the 2rd degree model was 89 % while in the 3rd degree model it is 98 %. Thus, the interpolation of the data just by the third order polynomial fit yielded the most consistent results. The corresponding temperature difference is found to be between 3-6°C.

In our revised MS the following sentences were added (page 26, lines 27-31; page 27, lines 1-4):

"The soil temperature at 30-minute resolution are calculated by using of satellite-derived surface temperature and in-situ measurements. The interpolation of the satellite-derived data was done by a 3rd order polynomial. From sensitivity analyses we found that the uncertainties of the derived temperatures are up to about 6°C. Substantial deviations of soil temperatures

(from the interpolated values) between Day 115 and Day 225 might be unlikely since surface temperatures in (hyper-) arid regions (even with (sparse) vegetation covering) are predominantly controlled by insolation only as long there is no impact of (convective) precipitation (indeed, Turpan's Meteorological Station has not reported any precipitation events during the entire growing period)".

29. p. 34560, l. 10-21: HONO emissions: This seems to assume that HONO follows the exact same emission parameterizations as NO – where has this been shown, or why should this be assumed?

Anonymous Referee #1is right: to calculate the monthly HONO emissions we used the land use specific constant scale factor namely the ratio HONO and NO releases for the optimum conditions (($F_{N,opt}$(HONO) to $F_{N,opt}$(NO)). The scale factor that we used is justified as follow:

- The studies by Su et al. (2011) and Oswald et al. (2013) pointed out that the HONO and NO emissions have to feature similar dependencies on soil properties, and there exist a co-emission of HONO with NO from the soil; so it could be expected that the HONO and NO emission processes follow similar parameters (Naegele and Conrad, 1990; Skopp et al., 1990, Gödde and Conrad, 2000; Oswalt et al., 2013).

- Furthermore, in laboratory experiments, Oswald et al. (2013) found that the reactive nitrogen is emitted as HONO from soils of this study area and that this is comparable to soil emissions of NO. Thus, this is additional loss term for fixed nitrogen in the soil beside of soil NO emission and additional source for reactive nitrogen in the atmosphere. To keep the estimation of monthly HONO emissions from cotton and grape soils simple, we applied just a scale factor, namely the ratio HONO and NO releases for the optimum conditions (($F_{N,opt}$(HONO) to $F_{N,opt}$(NO)).

The calculation of the monthly HONO emission are described more clearly in our revised MS (page 14-16 in main text). The resulting uncertainties were discussed in sec. 3.3 (page 27, lines 10-20).

With regard to the above comment of Anonymous Referee #1, in our revised MS the following sentences were added:

<u>Page 14, lines 26-30</u>: "the studies by Su et al. (2011) and Oswald et al. (2013) pointed out that the HONO and NO emissions have to feature similar dependencies on soil properties, and there exist a co-emission of HONO with NO from the soil; so it could be expected that the HONO and NO emission processes follow similar parameters (Naegele and Conrad, 1990; Skopp et al., 1990, Gödde and Conrad, 2000; Oswalt et al., 2013)".

<u>Page 27, lines 9-12</u>: "since the NO and HONO emissions have the similar dependence on soil moisture for soil properties in arid climate conditions (Oswald et al. (2013)), we used the land use specific constant scale factor namely the ratio of HONO and NO releases for the optimum conditions to estimate the HONO emissions (($F_{N,opt}$(HONO) to $F_{N,opt}$(NO))".

<u>Page 27, lines 16-19</u>: "this may result in a certain error of the calculated HONO emissions, which unfortunately can not be quantified more precisely at the moment. Nevertheless, these uncertainties do not question for the basic message of the study".

30. p. 34561: Where is the temporal/seasonal dependence of the NO to NO2 ratio considered/ accounted for in the top-down inventory? And would these emissions only be representative of satellite overpass time?

The temporal/seasonal dependence of the NO to NO_2 ratio was not considered in the top-down inventory. This can be explained as follow:

(a) the bottom-up emission estimates were calculated as NO_2 (in terms of ng m^{-2} s^{-1}) and the reported mass of emitted nitrogen compounds becomes independent from the partitioning ratio between NO and NO_2. Thus, the seasonal/temporal dependence of the NO to NO_2 ratio does not matter for the bottom-up emission estimates.

(b) Within the top-down emission estimates, a correction for the partitioning between NO and NO_2 in the atmosphere was performed. For that purpose, we multiplied the NO_2 values by typical partitioning ratios between NOx and NO_2 of about 1.3. These sentences can be found in sect. 2.3.4 (page 15) and sect. 3.2.2 (page 24).
In our revised MS the following sentences were added:
<u>Page 16, lines 1-5:</u> For the comparison of biogenic bottom-up emissions ($F_{biogenic}$) with top-down emissions ($F_{satellite}$) only the data around the time of the satellite overpass around 13:00LT was considered (bottom-up results for 12:45LT (12:30-13:00) and "13:15 LT(13:00-13:30)).To estimate the effect of the diurnal cycle of the emissions we calculated the ratio of the bottom-up emission at the satellite overpass (at 13:00 LT) and the daily average emissions Fig. 11 of the revised manuscript was updated accodingly.
Fig. S12 shows the monthly mean biogenic NO emissions during the vegetation period for daily average and noontime. Also the ratio is shown (right axis). The highest ratios are found for July and August (2.4) while it is only 1.01 for April.
These informations were added to sect. sect.3.3 (page 25, lines 14-15) and sect. 4 (page 28, lines 27-29 and page 29, lines 18-19) as well as in Fig S12 as follow (Fig. S12 was added in suplement): .

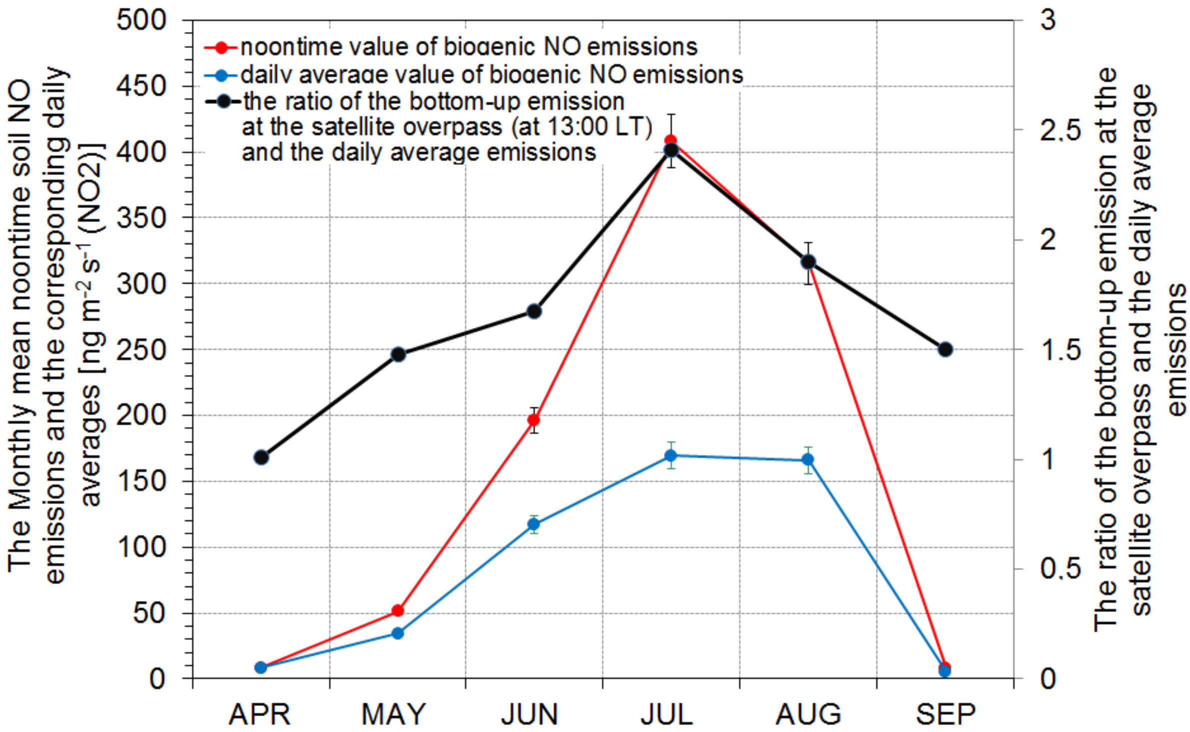

Fig. S12: Tohsun oasis biogenic NO emissions during the growing season for 2010: Monthly mean noontime emissions and the corresponding daily averages (all data are given in terms of mass of NO_2) and the corresponding ratio (right axis).

Thus, top-down emission estimates are only valid for the time of the satellite overpass (around 13:00 LT) and a few hours before. Thus we compared the top-down emissions to the results from the bottom-up emissions during the time of the satellite overlap (Fig. 11). Fortunately, the soil emissions during the satellite overpass time are strongest because of the high temperatures around noon (see sect. 3.3, page 26, and lines 23-26).

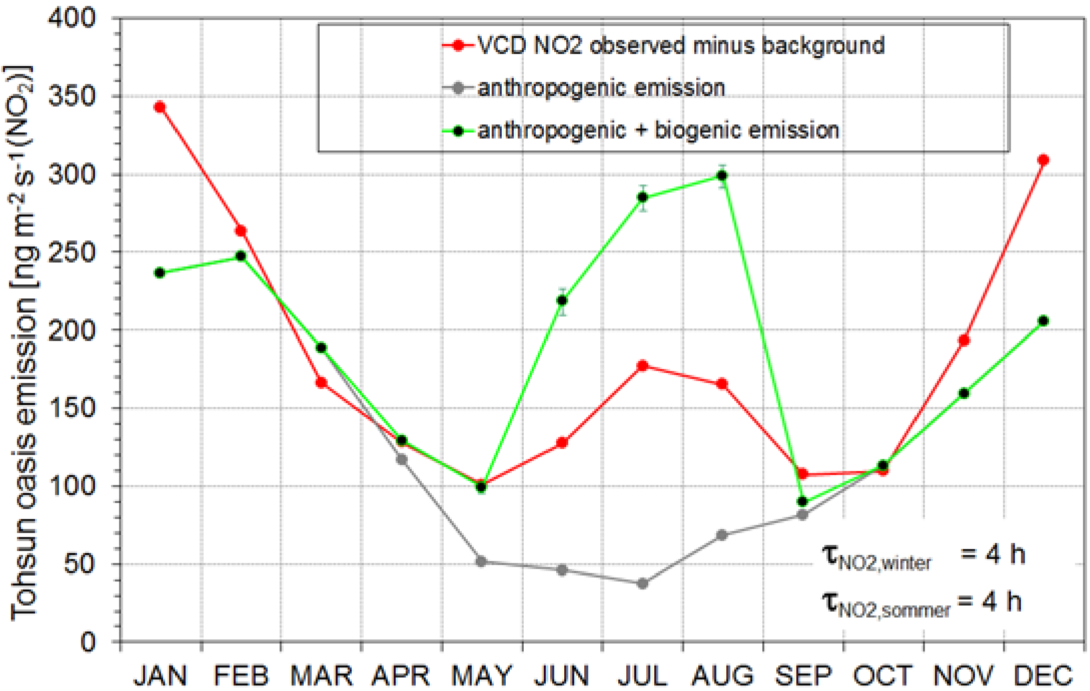

Fig. 11: Monthly mean top-down emissions (from satellite observations) are compared to bottom-up (soil biogenic & antropogenic) and antropogenic emissions (in terms of ng m^{-2} s^{-1}, NO_2). The biogenic bottom-up emissions around 13:00LT (at satellite overpass) are selected. They are systematically higher than the daily averages (see Fig. S12).

31. p. 34561, l. 2: I'm not familiar with a convention to capitalize Atmospheric Boundary Layer.

In revised MS "Atmospheric Boundary Layer" changed to "the atmospheric boundary layer (ABL)"

32. p. 34562, l 17: I would rephrase this to "At lab incubation temperatures of 25 degrees, the peak mean net potential fluxes in the Tohsun oasis for cotton, grapes, and desert were: : :" The current wording makes it sound like the peak emission occurs at 25 degrees.

This sentence is changed to "The highest mean net potential NO flux at 25°C occurred from the soil of cotton".

33. p. 34563, l. 4: What makes this remarkable? That it is such a small range?

Yes, that is small range of soil water content (between 0.01-0.03). Just this makes the net potential NO fluxes from desert soil remarkable. However, in revised MS "Remarkably" changed to "Emphasizing (page 19, line 29)".

34. p. 34566, l. 27-28: "NO from soil is largely controlled by soil moisture, temperature and fertilizer" – This is obvious, since these are the three inputs (besides land cover type) in the emission functions.

-In revised MS, this sentence was deleted (see S1.9 in supplement).

35. p. 34570, l. 16-21: These are details that should be moved to the methods section Section 3.8: Desert emissions from the GGTP calculation are predicted to be extremely minor, correct? But in Figure 12, the summer maximum in NO2 column over desert is about 30-50% of the maximum over the Tohsun Oasis. Does this suggest that the

desert biogenic soil NO emissions are underestimated your model?
In revised MS, these details have been moved to the methods section of 2.5.1.
Your comment about the maximum NO2 VCDs over desert refers probably to Fig. 21 of the original manuscript. With regard to the above comment of Anonymous Referee #1, in our revised MS the following sentence was added (sec 3.2.1 page 24, lines 11-18):
"Here, we have to mention that the desert VCD_{NO2} values are higher than in comparison to the NO emission of the GGTP model results (see supplement S1.9, Fig S6). This is indeed an interesting finding which can be attributed to the fact that in the desert region a small but still substantial diurnal variation in soil moisture is present. However, in our GGTP model we used a constant value for the soil moisture (0.0028); as a result, the diurnal cycle of soil moisture was not considered in the model calculation. It might indeed indicate that the desert soil emissions are underestimated. Alternatively, it might be partly also caused by inflow from NOx-reach air from the surrounding regions".

36. p. 34571, l. 26: "interstingly" to "interestingly"
many thanks, "interstingly" was changed to "interestingly".

37. p. 34572, l. 1: "Especially the good quantitative agreement was unexpected: : :" This sentence is awkward. Also, what quantitative measure/statistic was used to establish that the agreement is especially good?
We changed the sentence into: "Especially the quantitative agreement was better than expected taking into account the uncertainties of the bottom-up and top down emission estimates", (page 25, lines 27-29).

p. 34572, l. 5-25: Herein the authors list potential uncertainties, with the caution that the good agreement could be caused by cancellation of various systematic errors; mainly: air mass factor underestimating NO2 columns; NO2 lifetime overestimating true emissions; and uncertainty in irrigation cycles. But the authors have not expressed the rough magnitude of any of these errors, and whether or not they actually could cancel out to give the good quantitative agreement. Please elaborate and give quantitative estimates where possible. Moreover, as I mention above, there is no mention of uncertainties in the HONO emissions, if HONO release was not measured directly in the lab incubation experiments. Since they are potentially roughly half of the total NO emitted, the uncertainty in HONO emissions is necessary to estimate.
In revised MS, the most important errors sources are listed in the section of 3.3 (see page 25-27). Following important aspects were discussed:
Top-down emission estimates:
- *(a) satellite retrieval corcerning to air mass factor, NO2 profile, surface albedo;*
- *(b) assumption of the NOx lifetime of 4 hours;*
- *(c) satellite overpass time period.*

Bottom-up emission estimates:
- *(a) soil biogenic NO estimates;*
- *(b) soil biogenic HONO estimates;*
- *(c) anthropogenic NO_2.*

38. p. 34574, l. 8-10: While emissions estimate herein seems to be a good calculation, I personally think there is not enough certainty in the results to state unequivocally that soil emissions "are much more important contributors" to the regional budget; rather I suggest rephrasing this to something along the lines of, "We present evidence that soil emissions could be much more important: : : than thought before."

We changed the sentence "soil emission are much more important…" to "soil biogenic emissions of NO and HONO during the vegetation period could be much more important contributors to the regional NO_X budget", see page 29, line 22-24 in revised MS.

39. Finally, do the authors have any suggestions for future work that could corroborate this interesting result, or constrain any of the uncertainties further?

In future, we want to transfer this method to other semi-arid, arid, and hyper-arid regions to study consequences of ecosystem reactions forced by land use and/or climate change.

With regard to the above comment of Anonymous Referee #1, in our revised MS the following sentence was added (sec 3.3 page 27, lines 26-29, and page 20, lines 1-7):

The combination of independent methods applied in our present study was the first attempt to prove the comparison of soil NO emissions of different scales, ranging from the size of a soil sample to the size of a satellite pixel.

However, the following improvements in these methods for the future work might be necessary:

(a) *bottom-up biogenic emissions: derivation of individual NO and HONO net release rates from a series of simultaneous incubation experiments in the laboratory under well-defined conditions (full soil temperature and gravimetric soil moisture scales),*
(b) *temporally high resolution in-situ measurements of top soil temperature and gravimetric soil moisture by suitable (and simple) sensors at representative sites of all considered land use types (at least during the growing period),*
(c) *top-down emission estimates: temporally resolved consideration of the NO_2 lifetime.*

Reference:

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