

Interactive comment on "The contribution of soil biogenic NO emissions from a managed hyper-arid ecosystem to the regional NO₂ emissions during growing season" by B. Mamtimin et al.

Anonymous Referee #1

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In this article, the authors present novel laboratory and modeling work to estimate the contribution of soil biogenic NO emissions to the total NOx budget in the Tohsun Oasis region of China. This is performed by upscaling lab incubation measurements of soil samples from the study area to the regional level by combining Landsat remote sensing products with in-situ timeseries of soil temperature and moisture. A bottom-up estimate of anthropogenic emissions is calculated in order to determine the relative contributions of the biogenic emissions to total NOx, and satellite-derived NO2 columns are used to corroborate the inferred importance of the biogenic signal.

Summary comments:

C11153

This manuscript presents nice evidence of the importance of biogenic NO emissions in the region to the overall NOx budget, leading to the interesting result that biogenic emissions from managed drylands during the growing season can potentially exceed anthropogenic emissions. It may certainly be of interest to the community, highlighting the uncertainty of soil NOx contributions due to a lack of observations and widely differing global inventories. The observations and results presented herein make a decent contribution to the literature which merit publication.

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.

General comments:

If the paper is to be published in ACPD, I advise a significant revision and restructuring of the manuscript. It was at times difficult to read. The largest issue for me is that the methods section is extremely long and technical, and focuses minimally on advances relevant to atmospheric chemistry/physics. I think most of the material from Sections 2.4.1 to 2.4.6, 3.2 and 3.4 could be moved to a supplement (including the associated figures). If the methods presented herein are particularly novel, then given its length I would have suggested their publication in a different journal for a better audience. However, it appears to me that the authors are mostly implementing methods previously established by others in the literature, and so these could be summarized much more succinctly with the details moved to a supplement.

I would additionally suggest restructuring the article to better streamline the material. There is a (meritoriously) wide combination of data products, methods, and calculations used - but each end up divided into multiple sections. To me, this results in awkward separation of all the different steps into too many subsections. For example, the Landsat data is first introduced in Section 2.2, then details of their retrieval are given throughout Section 2.4, then the spatial and temporal scaling is alluded to in Section 2.5.2, then the exact assimilation is explained in Section 2.5.3 - all the while other sections regarding lab work field, validation, and bottom-up/top-down inventories are scattered in between. 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.

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

C11155

in the lab by the present authors, the uncertainty associated with these estimates must be discussed further.

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?

Finally, I encourage the authors to draw careful attention to many grammatical errors in the manuscript. I identify some below, among my technical/specific comments.

Technical/Specific comments:

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"?

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.

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.

p. 34535, l. 3: "which photo-stationary equilibrates with NO2" is not grammatically clear.

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.

p. 34535, l. 20: Is there a connection between "bushy" and "dryland farming"?

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

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.

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

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?

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

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?

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

C11157

authors more clearly state how the different areas were selected?

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

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?

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?

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.

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.

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

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

p. 34551, l. 20: "of sufficient quality" – How exactly was this determined?

p. 34553, l. 21: Insert a comma between "NO flux" and "theta(x,y)"

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".

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.

p. 34557, I 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?

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

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?

p. 34558, I. 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

C11159

Tohsun County?

p. 34559, I. 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.

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

p. 34560, I. 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?

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?

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

p. 34562, I 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.

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

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.

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?

p. 34571, l. 26: "interstingly" to "interestingly"

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?

p. 34572, I. 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.

p. 34574, I. 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."

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

Figures: It is my opinion that Figures 3, 4, 5, 6, 7, 9, 11, 12, 15, and 16 could all be moved to a supplement.

C11161

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 34533, 2015.