Dear Editor,

Please find attached our responses to the comments by the reviewers. We have addressed all comments in our response, and indicated changes in the revised manuscript with a blue and red color for comments by Reviewer 1 and 2, respectively.

The most important changes to the revised manuscript are that we now improved our discussion of the beta values, which account for non-linear responses of NO_2 columns to changing NO_x emissions. Our discussion, and additional calculations presented in the Supplementary Material, allow for a better comparison with beta values presented by Lamsal et al. (2011). Furthermore we have adapted our manuscript in line with the recommendations and comments of the reviewers.

We would appreciate your consideration of our revised manuscript for publication in Atmospheric Chemistry and Physics and look forward to your response to the changes we have made.

Kind regards,
Geert Vinken

Atmos. Chem. Phys. Discuss., 14, C5656–C5658, 2014
www.atmos-chem-phys-discuss.net/14/C5656/2014/

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Worldwide biogenic soil NO_x emissions inferred from OMI NO_2 observations" *by* G. C. M. Vinken et al.

G. C. M. Vinken et al.

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We thank reviewer #1 for the review and constructive comments provided. Please find our detailed replies to the comments below. We adapted our manuscript in line with these recommendations. We marked updates in our manuscript corresponding with this review with a blue text colour.

Comments

1) The paper should include a more thorough discussion of how it excludes the possibility of biases that are correlated with soil NO_x emissions. Could burning of Agricultural wastes or large scale fires that occur with similar timing and be misinterpreted as soil NO_x?

In this study we attribute observed NO_2 enhancements to soil NO_x emissions based on the modelled contribution of a particular source to the NO_2 column, and do not attribute based on timing of emissions (as was done in e.g. Ghude et al., 2013). Our filtering minimises the influence of other strong NO_x sources on the NO_2 column over our selected regions, and as a result the bias from collocated non-soil emissions correlated should be small. For example, the influence of fires is minimised, by screening out situations with fires according to (daily) GFED data. Also, we reduce absolute biases in either model or observations (due to other emission sources) by fitting an offset in our regression. We have extended the discussion of our filter in the manuscript (Section 3.1).

2) The paper overstates the case for large uncertainties in emissions in the literature. Figure 1 shows that with the exception of 1 paper the mean values reported by 16 papers over the last 20 years are quite similar. This paper should lay out the current challenge which is to narrow the uncertainty of this more recent range - or explain why it is still reasonable to consider the outliers in the literature as likely.

Indeed, reporting the full range might have overstated the uncertainty. We have adapted this in the manuscript.

3) Hudman et al. report substantial interannual variability in specific regions. In light of those analyses how representative of global average behavior is an analysis of a single year?

Indeed, Hudman et al. (2012) report on interannual variability for the Mid-Western USA. In our work, we constrain emissions in 11 independent regions on different continents and hemispheres. We acknowledge that while the variability for one particular region may be substantial, the use of 11 independent regions will dampen the possible influence of variability on our estimate of the global total. Extending our work to cover more years would be useful to improve the soil NO_x parameterisation, but is beyond the scope of the current study.

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 14683, 2014.

Atmos. Chem. Phys. Discuss., 14, C5659–C5667, 2014
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We thank reviewer #2 for reviewing our paper and the provided comments and detailed specific comments. Please find a detailed discussion on the comments below. We adapted our manuscript in line with these recommendations. We marked updates in our manuscript with a red text colour.

General Comments

1) The β values shown in Table 2 are much higher than the values reported in Lamsal et al. (2011). One possible explanation put forward is the boundary effects (p. 14696, l. 15), however, I doubt it in view of the short NO_x lifetime over the regions considered. The authors should prove their point (maybe by using regions of different sizes) or

remove the argument. Comparison is made difficult because Lamsal et al. provided annual averages for β, and the values given here are reported mostly for summer due to the filtering scheme. I would strongly recommend providing annual averages for the purpose of comparison. In lines 25–26 of p.14696, agreement is said to be good with Lamsal et al. in low NO_x areas, however, over the few regions where comparison is possible (Midwest US and Spain/France) the values reported by Lamsal et al. are about a factor of 2 (or more) lower than in this study. The authors should clarify the reason of these discrepancies.

We now provide annual averaged and unfiltered beta values in the Supplementary Material (Section S5), and extend the discussion on the differences with Lamsal et al. (2011) in section 3.2. We have done additional simulations, and find that differences are mostly driven by the application of the averaging kernel on GEOS-Chem simulated NO_2 columns in our study. Lamsal et al. (2011) do not apply the averaging kernel (L. Lamsal; personal communication), and we show in the supplementary material that this results in about 25% lower beta values. For clarity, we have changed the symbol β in Eq. 2 and 3 to β', and extended the discussion of β'. Other differences versus Lamsal et al. (2011) arise from our focus on low NO_x environments that are sensitive to OH-feedbacks, from our focus on selected months when beta values are higher (Tables 2, S4 and S5), and to a lesser extent from boundary effects (due to the absence of enhanced NO_x inflow from sources outside the region). We did also check the effect of boundary effects by calculating beta values for a smaller region (subset of the existing region) as you suggested, and found that this resulted in about 10% lower beta values. We added this to the discussion in the Supplementary Material.

2) The comparison with ground-based measurements is nice, however, the derived RSMD is not much reduced. The authors should provide comparison also with SCIAMACHY measurements using a priori and top-down emissions.

We agree that comparison with additional independent measurements would be valuable. However, comparison with SCIAMACHY is not done in this work as: 1) this would require an enormous effort to process and average the SCHIAMACHY NO_2 columns for 2005; 2) SCIAMACHY sampling is 6 times less than OMI, and has courser pixels than OMI, so it would be much less informative; 3) SCIAMACHY data would not offer a truly independent evaluation, as the retrieval is based on the same principles as OMI; and 4) soil NO_x emissions peak during mid-day (when temperature is highest), reducing the sensitivity of SCIAMACHY (morning) measurements to these signals.

3) A possibly important issue is the use (or not) of averaging kernels in the comparisons. Could you specify whether the DOMINO averaging kernels have been applied?

Indeed the use of averaging kernels is important in the comparison. In this study we have applied the averaging kernels provided along with the DOMINO retrieval to account for the vertical sensitivity of the satellite instrument. We clarified this in Section 2.1.

4) p.14691, line 21 : Why is the minimum number of observations taken to be only 3 per month and pixel, given the small size of OMI pixels? Would a higher threshold reduce the number of available data for comparisons?

We note that the minimum number of observations is 3 grid cells (not 3 individual measurements) per month, which includes many more than 3 OMI pixels (OMI pixels are as small as 13 x 24 km^2, much smaller than the GEOS-Chem grid cells of 250 x 200 km^2). For a grid cell to be included we require 75% of a grid cell to be covered by valid OMI observations, so we typically have at least 200 observations per grid cell per month. We have clarified this in the manuscript (Section 2.2).

5) p. 14695, line 17–23 : The small values of the slope in Australia likely mean that OH is very high in this region, whereas the high slopes in wintertime over India and Sahel are due to lower OH levels caused by less sunlight, not non-linearity. The feedbacks between NO_x emission and NO_2 lifetime do exist but are not the main factor determining the spatial and temporal variations in the lifetime. Note that NO_2 columns are similar over Australia and Sahel, despite having different slopes. Please adapt the

discussion, for example in line 23, replace "such nonlinearities" by "the variability of NO_2 column lifetime".

We agree with your comment, and have adapted the discussion in line with your recommendations.

Specific Comments

p. 14688, Consider citing here previous OMI-based studies, like the global studies of Miyazaki et al. (2012) and Stavrakou et al. (2013), and the regional study of Lin et (2010) over China.

We agree with this suggestion and have included references to these studies.

What is the diurnal profile of soil emissions in GEOS-Chem model?

Soil NO_x emissions in the GEOS-Chem model depend on soil moisture, temperature and biome type. In line with the diurnal profile of temperature, soil NO_x emissions peak during mid-day, and are lowest during the night and early morning.

What are the GEOS-chem choices for relevant reactions like $OH+NO_2$, HO_2+NO, HO_2 uptake on aerosol? Those reactions were shown lead to substantial uncertainties on top-down NO_x emissions, especially for natural sources (factor of 2 for soil emissions) (Stavrakou et al. 2013).

Indeed the choice for the $OH+NO_2$ reaction rate in the model is important when estimating NO_x emissions (and contributes to the uncertainty of the top-down emissions, also see our discussion on model uncertainties as a response to your later comment). GEOS-Chem mainly adopts the kinetic data from the Jet Propulsion Laboratory (JPL) (Sander et al., 2011), and GEOS-Chem does not include the (still controversial) HNO_3 formation channel that has been suggested in literature for the HO_2+NO reaction (Butkovskaya et al. 2005,2007,2009). The uptake coefficient of HO_2 on aerosols is from Mao et al. (2013). We added an additional sentence in Section 2.1, and added the reference to Mao et al. (2013).

More details on Hudman et al. parameterization would be needed, as well as differences with Steinkamp et al.(2011), and discussion of the uncertainties.

Hudman et al. (2012) provide an extended discussion on their innovations to the work by Steinkamp and Lawrence (2011). Furthermore Maasakkers (2013) discusses the differences between these two parameterisations and the implementation in GEOS-Chem. The main improvements of Hudman et al. (2012) to the Steinkamp and Lawrence (2011) parameterisation are:

- Soil NO_x emissions are a smooth function of soil moisture as well as temperature
- Improved fertiliser and manure treatment
- Online calculation of wet- and dry-deposition of N
- Improved calculation of soil NO_x pulses by taking into account the length of the dry spell

p.14690, l.10 : Insert "that the" before "smallest".

We changed this in the revised manuscript.

p.14691, l.10 : Add "s" to "observation".

We changed this in the revised manuscript.

p.14694, l.2 : Replace "of" by "due to".

We changed this in the revised manuscript.

p.14694, l.11 : "the response of the modelled...with 1%" should read "the modelled NO_2 column obtained by increasing emission source i by 1%".

We changed this in the revised manuscript.

p.14694, l.13 : Replace "response to" by "obtained by".

We changed this in the revised manuscript.

p.14695, l.27 : Please specify the fitting period (month or year).

The fitting period is monthly, and we adapted this in the revised manuscript.

Figure 4 caption should more clearly explain the content of the plot. What represent the symbols?

We have clarified the caption to better explain the content of the plot (also in Figure 5).

p.14699, l.17–19 : Not clear. Rephrase.

We have rephrased this sentence in the revised manuscript.

p.14700, l.5 : Are those measurements daily averages?

Yes, the caption of Figure 9 indicates that the measurements from the different network were averaged from hourly, daily, or monthly observations.

p.14702, l.1 : The 25% model error seems arbitrary and overly optimistic, given the discussion provided in the cited studies.

The error estimates of modelled NO_2 reported in the literature are line with our estimates. For example, Martin et al. (2003) report 30%, Boersma et al. (2008) report 20%, Lin et al. (2012) report -10–20% (systematic) for east China, and Lin (2012) reports 30–40% for east China. We do not think that the factor of 2 in uncertainty reported by Stavrakou et al. (2013) is represents a true modelled NO_2 uncertainty, because in that study work extreme cases have been analysed, representative of a maximum NO_x loss, and a minimum NO_x loss scenario. We added these citations to Section 4.4.

p.14702, l.19 : To convince the reader that the error on β is 25%, differences with the results of Lamsal et al. must be elucidated.

See our response to General Comment 1.

p.14702, l.8-10 : I really do not see why the approach would be robust to biases in *either OMI or GEOS-Chem. Those biases will influence the values of the slope κ of the regression between OMI and GEOS-Chem and therefore the top-down emissions.*

Our method is robust to biases as a result of a constant offset in space or time (for example due to influence of non-soil NO_x emission sources). We account for these biases via the offset in the regression fit. We acknowledge that indeed our slopes are still sensitive to relative biases in OMI or GEOS-Chem NO_2 columns, and have adapted this in the manuscript.

p.14703, l.24 : How is it proved that NO_2 responds linearly to emission changes in anthropogenic source regions?

This is a result of the beta values being close to unity, as is shown in the response to General Comment 1.

p.14704, l.9-13 : This statement should be moderated since this study addresses only a small fraction of total soil NO_x emissions.

We included in this statement the fraction of soil NO_x emissions that we constrain.

p.14714. In the table caption please mention that the value of Hudman et al. is modified to account for CRF.

We added '(and applying the canopy reduction factor described in Sec. 2.1)' to the caption of Table 1.

There are some problems with the quality of the inset label in some of the figures, e.g. Fig. 3, 5, 6, 9.

We have increased the size of the inset labels where possible.

Consider removing Figure 8. It does not convey more information than already present in the text.

We acknowledge that Figure 8 does not provide new information. However, this Fig-

ure is a good illustration and summary of our study. It shows that we only constrain 13% of all soil NO_x emissions, and the effect of extrapolating this result to all soil NO_x emissions.

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