Interactive comment on “Satellite constraint for emissions of nitrogen oxides from anthropogenic, lightning and soil sources over East China on a high-resolution grid” by J.-T. Lin

Anonymous Referee #1

Received and published: 2 January 2012

The paper "Satellite constraint for emissions of nitrogen oxides from anthropogenic, lightning and soil sources over East China on a high-resolution grid" by J.-T. Lin is an extensive study of NO2 from OMI over China. Using the GEOS-Chem model, the author derives estimates of anthropogenic, soil, and lightning NOx source strengths by an inversion method.

The paper is clearly written and the method is well described. Several uncertainties of the assumptions made are investigated and discussed. But still, I am not convinced that it is possible to estimate lightning and soil emissions of NOx with an uncertainty as low as 48% over China, which is by far (>90%) dominated by anthropogenic emissions.
My main concern is that crucial uncertainties are not discussed in this study. Both lightning and soil NOx production are still highly uncertain - the total amount, but the temporal and spatial patterns as well. Thus, an important assumption of the method, i.e. that GEOS-Chem is doing right in modelling spatial and temporal patterns of lightning/soil NOx correctly, can not be considered as given and has to be discussed!

My conclusion from the presented study would be that the a-priori model setup misses some source with a summer maximum; this does not prove that LNOx/soil NOx is too low in the a-priori. It could also indicate, for instance, that convection of anthropogenic NOx is underestimated!

I recommend publication in ACP after the author has addressed these issues. The uncertainties of amount, spatial, and temporal patterns of LNOx and soil emissions, and the consequences on the inversion uncertainties, have to be clearly pointed out.

Comments

1. 29812/17: By skipping 15 pixels east and west, the swath width is drastically reduced. Thus, the considered dataset does not have daily global coverage any more. Please add a discussion of this issue (what is the resulting swath width?)

2. Section 3.1: Please explain how the monthly means of GEOS-Chem VCDs are derived. Are they only calculated for coincident satellite observations and averaged afterwards? Or do they comprise a full month?

3. The description of the implementation of LNOx is far too short; the author mentions an "adjustment for horizontal distribution", citing a paper which is just submitted; this information is not sufficient.

Please give details of LNOx implementation and discuss the consequences of the choice of e.g. the convection parameterization scheme on this study (see e.g. Tost et al., Atmos. Chem. Phys., 10, 1931-1951, 2010, for the uncertainties in modelling C13766
convection; the production of LNOx per flash for CG/IC is also still highly debated).

Did you compare the modelled GEOS-Chem lightning with lightning measurements? I would expect that, even if the modelled lightning is somehow tuned to a lightning climatology, the individual monthly means still could differ considerably. In such a case, the inversion can not work!

From the title of the Murray et al. manuscript, I assume that the release of LNOx is somehow linked to LIS/OTD lightning measurements. However, the spatial patterns of LNOx in Fig. 9 look quite different from the LIS/OTD climatology (http://thunder.msfc.nasa.gov/images/HRFC_AnnualFlashRate_0.5.png). The latter shows a clear increase towards the South. Please clarify.

I propose to repeat the inversion study for July using different LNOx setups and add the results to table 3. Without such a sensitivity study, the resulting uncertainty of the derived LNOx production is definitely too low!

4. Also the description of soil NOx in GEOS-Chem is quite short. Please discuss, how reliable/uncertain the parameterizations are (based on publications in the 1990s) with respect to e.g. spatial distribution and temporal patterns like pulsing, and the consequences for the inversion. Please also specify the settings for the Hudman et al. (not yet submitted!?) implementation.

5. Section 4.1.1: the sum of \((\Omega_r-\Omega_p)/\sigma)^2\) is just \((\varepsilon/\sigma)^2\) (Eqs. 1 and 2) and does not contain \(k\) ny more!? 

6. 29817/6-12: The constraints for \(k\) are reasonable on average; however, given the high uncertainties of spatial and temporal patterns of LNOx as well as soil NOx, it could easily happen that \(k_l\) is off by an order of magnitude for some grid cells. Please discuss the effect of the choice of the thresholds on the inversion (e.g. for 0.1 and 10).

Fig. 1: It would be helpful to include NO2 VCDs (e.g. annual mean) to this map.

Fig. 3: Why is the right column missing for April, October, and annual mean? Please
Fig. 8: \( k_\perp \) is in saturation (\( \approx 5 \)) for several, sharply localized grid cells. Is this meaningful? Please extend the discussion of these spatial patterns. E.g.:

- Is there any reason why soil or lightning NO\(_x\) should be that strongly underestimated in the a-priori at e.g. 38\(^\circ\)N, 115\(^\circ\)E?

- Did you check the daily OMI measurements? I suspect that some of the spots could be caused by just one high OMI pixel.

- How would the figure look like for thresholds of 0.1/10 for \( k_\perp \)? Would the \( k_\perp \) increase to 10 for these spots? Would the overall soil/lightning emissions change?

Minor issues:

a) 29809/7: add "e.g." in the reference list.

b) 29810/13: start a new paragraph for the lightning topic.

c) 29813/20: start a new paragraph for the soil emissions.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 29807, 2011.