General response: We thank the two reviewers for useful comments and suggestions. We have revised the manuscript to accommodate the suggestions. In particular, we extend substantially the analyses of a priori and a posteriori lightning and soil emissions. We discuss the temporal and spatial relationship between anthropogenic and lightning/soil emissions to better justify our inverse modeling approach. We extend the evaluation for the effect of potential errors in the seasonality (timing and magnitude) of lightning and soil emissions, separately and in combination, on the inversion results; for this purpose we add nine additional sensitivity tests. We analyze the effect of potential errors in model convection on the inversion as well. We extend the discussion on the spatial pattern of top-down soil emissions and comparisons with recent bottom-up estimates.

Reviewer 1

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 (see 29808/20-21).

Response: We have significantly extended the analyses of lightning and soil emissions and uncertainties.

The uncertainty of 48% (now 47%) for a posteriori lightning/soil emissions is derived for emission budgets for East China as a whole. Uncertainties for individual locations may be much larger, as partially addressed by the spatial spread of the standard deviation of top-down emissions among the sensitivity tests (Fig. 11 in the revised manuscript). Information is not available on location-specific errors in the CTM, retrievals and a priori emissions, preventing us from conducting a complete analysis of location-specific errors in the a posteriori emissions. We have clarified this issue in the revised manuscript. In addition, on the 0.25 ° lat x 0.25 ° long high-resolution grid, maximum anthropogenic emissions are often not collocated with maximum lightning/soil emissions, and the spatial correlation between anthropogenic and natural emissions are less than 0.36 in all months. Moreover, although anthropogenic emissions are the dominant source of NOx over East China on the annual basis, lightning and soil emissions together can be as large as 25% of anthropogenic emissions in July. Locally, the natural contribution can be much larger. Finally, the contribution of a given amount of lightning emissions to NO2 column (considering averaging kernel) is larger than the contribution of the same amount of anthropogenic emissions (see Sect. 4.4).

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!

Response: We have significantly extended the analyses of lightning and soil emissions, their seasonal and spatial patterns and uncertainties, and impacts on the inversion results. We add a total of ten sensitivity tests to help address these issues.

Note that, as the inversion is done on a gridbox-by-gridbox basis, the spatial patterns of different emission sources do not have significant impacts on the inversion results. Nonetheless, we have added the respective analysis for a better understanding of these emission sources and showed a spatial correlation of less than 0.36 between anthropogenic and natural emissions.

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

Response: We have added a detailed analysis of the impact of potential errors in model convection on the inversion results. Specifically, assuming an increase by 50% in model convection of anthropogenic NO2 would result in reductions of about 4-5% in the top-down lightning and soil emissions.

I recommend publication in ACP after the auther has adressed 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.

Response: As mentioned above, we have significantly extended the analyses of lightning and soil emissions, their seasonal and spatial patterns and uncertainties, and impacts on the inversion results.

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?)

Response: The procedure consequently changes the swath width in use to about 800 km so that global coverage is achieved roughly about every three days. We have added this information in the revised manuscript. A test to address the impact on the inversion results was already included in the original manuscript.

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?

Response: For consistency with satellite retrievals, model VCDs in each day are obtained by regridding modeled NO_2 at each vertical layer to 0.25 °long x 0.25 °lat, sampled from gridboxes with valid satellite retrievals, and applied with the averaging kernel from DOMINO-2. The daily data are averaged then to obtain monthly mean values for each gridbox. This information has been clarified in the revised manuscript.

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 modeling 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 form 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 reslts to table 3. Without such a sensitivity study, the resulting uncertainty of the derived LNOx production is definitely too low!

Response: We have added an extensive analysis of lightning emissions in the revised manuscript. We include an evaluation on the seasonality of air temperature and precipitation in GEOS-5 for a better understanding of the modeled seasonality of convection and consequent lightning activities. We have added more tests to evaluate the impacts of potential errors in the seasonality of lightning on the inversion results. We further analyze the impact of potential errors in model convection on the inversion results.

As detailed in the revised manuscript, GEOS-Chem constrains monthly climatological lightning flashes on the gridbox-by-gridbox basis, using data from the OTD/LIS satellite measurements, but allows for interannual variability. We had attempted to use the ground-based lightning network to evaluate model lightning flashes. However, unlike in the U.S., such network is still in its early stage and lacks sufficient coverage. In addition, the data are not publically available. This is part of our motivation to use satellite NO2 data to constrain lightning emissions. While such approach is difficult and may contain large uncertainties, we think it is useful to improve our understanding of the contribution of lightning to NOx over China.

The data on the NASA website are total flash rates in each 0.5° gridbox, not on the per unit of area basis. On the per unit of area basis, the flash rate in the north will be increased relative to that in the south. In addition, the NASA data are for climatological annual mean, not data for July 2006. The occurrences of lightning are concentrated on July and August in the north but are more evenly distributed from April to August in the south, following the movement of the summer monsoon. Furthermore, our emissions include information on the NO yield from each flash, which is assumed to be 500 moles per flash north of 35° N and 260 moles per flash south of 35° N. We have double checked our flash rate data and find no errors.

We have also considered other lightning schemes, which however had been found not to perform better than the Price et al. scheme used here (Hudman et al., 2007). In part for this reason, the GEOS-Chem model had discontinued the use of other lightning schemes but rather use the OTD/LIS measurements to constrain the climatology of lightning flashes for individual gridboxes derived from the Price et al. scheme.

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) whith 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.

Response: We have extended significantly the analysis of soil emissions, including spatial and temporal patterns and comparisons with recent bottom-up emissions by Steinkamp and Lawrence (2011) and Hudman et al. (2012). We conducted in the original manuscript some evaluation of the effects of potential errors in soil emissions using several sensitivity tests, including but not limited to a test using the new bottom-up emission dataset by Hudman et al. (2012) to adjust the soil induced VCDs of NO2 prior to the inversion process. In the revised manuscript, we have extended the evaluation by including more sensitivity tests on the impacts of the seasonality (timing and magnitude) of soil emissions. We have given specifications of Hudman et al. scheme; see Sect. 5.1.2.

5. Section 4.1.1: the sum of ((Omega_r-Omega_p)/sigma)ˆ2 is just (eps/sigma)ˆ2 (Eqs. 1 and 2) and does not contain k ny more!?

Response: We have revised the manuscript to clarify this issue. Specifically, we revise Eq. 1 to reflect that, in combination with Eq. 2, the coefficient 'k' in Eq. 2 is an estimate of the theoretical coefficient 'K' in Eq. 1. As such, Omega_r-Omega_p contains the coefficient 'k', where Omega_r is retrieval (observation) and Omega_p is its prediction.

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).

Response: A larger range allowable for k_l (e.g., 0.1 – 10) results in more extreme values of k_l in some sparse locations whose k_l is already large under current constraint. Results in these locations may not be all physical, as expected from simplifications in the inversion algorithm. This however has negligible impacts on the top-down emission budgets for East China. We have clarified this point in the revised manuscript (Sect. 4.2). See below for more comments.

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

Response: Done.

Fig. 3: Why is the right column missing for April, October, and annual mean? Please add.

Response: We chose July and January as extreme cases (e.g., with shortest and longest lifetime of NOx) to evaluate the top-down emissions. We did not attempt to conduct the associated simulation for the entire year.

Figure 3 has been changed to Fig. 4.

Fig. 8: k_l is in saturation (==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 NOx should be that strongly underestimated in the a-priori at e.g. 38_N, 115_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_l? Would the k_l increase to 10 for these spots? Would the overall soil/lightning emissions change?

Response: We have extended the analysis of the spatial pattern of top-down emissions in comparison with the a priori data in the revised manuscript (Sects. 4.2 and 5.1.2).

The large values of k_l and resulting top-down emissions in southern Hebei and along the northern coasts of the Bohai Sea are explained in detail in the revised manuscript as contributed in part by an underestimate in fertilizer-associated emissions in the a priori dataset. Our results are consistent with recent bottom-up estimates by Steinkamp and Lawrence (2011) and Hudman et al. (2012).

The value of k_l also has spike values at spotty locations in other parts of East China where natural emissions are normally very low. These spikes are likely artificial results of the inversion algorithm, errors in GEOS-Chem, and/or errors in the satellite product; they however have negligible impacts on the emission budgets over East China.

Daily data in DOMINO-2 vary significantly due to the short lifetime of NOx, particularly for values at individual locations. It is difficult to determine whether the signal is realistic for individual days. That’s why we focus on monthly mean data. Monthly mean data may still be affected by errors in individual days, though. We have added in the revised manuscript that many spotty spikes may be associated with errors in the satellite product, errors in the CTM and/or simplifications in the inversion process.

We do not expect results for every individual gridbox to be physical due to simplification in the algorithm and errors in model and satellite data. We have explained in the revised manuscript that many isolated spikes of k_l may not be physical but have negligible impacts on the top-down emission budgets for East China. A larger range allowable for k_l (e.g., 0.1 – 10) results in more extreme values of k_l in some spotty locations with very low emissions, but has negligible impacts on the top-down emission budgets for East China.

Minor issues:

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

Response: Added.

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

Response: Done.

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

Response: Done.

Reviewer 2

The manuscript by Lin is a thorough effort to provide a comprehensive estimate of NOx emissions in China for the year 2006. The author brings together state-of-science models and datasets, and by combining the modeling results in an appropriate manner with the satellite measurements, the author is able to draw conclusions on total NOx emissions over China. The manuscript is a straightforward extension of previous work, that is appropriately cited, but it is at the same time innovative and new in the sense that this is the first inversion over China that uses the high-resolution (0.5 deg x 0.67 deg) nested-grid GEOS-Chem model, in combination with a recently improved OMI satellite dataset. The description of the multivariate regression has much improved compared to an earlier version that I saw.

Response: We thank the reviewer for useful comments. In addition to the use of new satellite product and high-resolution model, we also extend previous studies to separate anthropogenic from natural emissions by taking advantage of their differences in seasonality. The multi-step approach (not just a simple multivariate regression but also the supplementary procedures) is crucial for the top-down constraint, particularly when the inversion is done gridbox by gridbox.

My main concerns with the manuscript are related to the use of the GEOS-Chem model here. A number of important GEOS-Chem characteristics go undiscussed.

Response: We have added much more detailed analysis of the model, particularly on lightning and soil emissions.

* First of all, which mixing scheme has been used, the original 'instant mixing' scheme or the recently implemented non-local mixing scheme? This is an important issue as it affects vertical distributions of NO2 (important when using the kernel and when dealing with lightning NO2 aloft) and O3 (important in view of potential non-linearities in the inversion method).

Response: We have clarified that the non-local mixing scheme is used in the CTM. We in fact implemented the non-local scheme in the model (Lin and McElroy, 2010). We agree that the simulation of vertical mixing is important.

* Secondly, it should be made clear right from the beginning what the emitted totals are for anthropogenic, biomass burning, lightning, and soil NOx in GEOS-Chem. Then, with respect to the lightning NOx production, the authors cite an unpublished and unaccessible paper by Murray et al. I have some idea of the method by Murray et al., who appear to be using LIS and OTD to provide horizontal constraints on the lightning flash frequency. Since LIS coverage extends to only 30-35 deg (latitude), and the OTD mission ended in 2000, the horizontal redistribution of flashes over much of China is climatological constraint at best. The author should provide more information on how the lightning NOx production has been done in this version of GEOS-Chem.

Response: We have added a detailed analysis of lightning and soil emissions, including their descriptions, emission budgets over East China and spatial and seasonal patterns, in comparison with anthropogenic emissions (Sect. 3.1). Emissions from biomass burning are negligible for East China (Wang et al., 2007; Lin et al., 2010a) due to the relatively low combustion temperature; the total over East China is only about 0.013 TgN for 2006 based on the GFED2 data.

* Also, to build confidence that it is actually possible to simultaneously obtain information on lightning and soil NOx patterns vs. anthropogenic NOx patterns, the author should give an idea on the orthogonality of these patterns. Unlike the other reviewer, I believe it is possible to obtain information on lightning and soil NOx emissions over China, as long as these emission categories are sufficiently orthogonal (not just in time but also in space) from the anthropogenic emissions. However, I agree with the other reviewer that the patterns shown in Figures 9 and 10 look pretty questionable with apparent hotspots of top-down lightning and soil NOx emissions that happen to coincide with China's highly populated regions. The authors should clarify all these issues, and put forward a more convincing case as to why the top-down constraints are also meaningful for lightning and soil NOx.

Response: We have extended substantially the analyses of the seasonal and spatial patterns of anthropogenic and lightning/soil emissions to better support our inversion approach and results.

As analyzed in the original manuscript, Omega_m,a reach maximum values in winter and minimum in summer; while Omega_m,l and Omega_m,s exhibit an opposite seasonality. We exploit this feature to separate anthropogenic from natural emissions. Also for this purpose, we do not just simply employ a multivariate regression for individual gridboxes. Rather, we make a lot of efforts to implement supplementary procedures (Fig. 6) to better quantify the top-down emissions gridbox by gridbox.

We include analyses for spotty spikes of the scaling factor k_l. We note that some isolated spikes may not be physical and may result from simplifications in the inversion algorithm and errors in model/retrievals, but they have negligible impacts on the top-down emission budgets for East China. We have also extended the comparison with recent bottom-up estimates by Steinkamp and Lawrence (2011) and Hudman et al. (2012). In particular, the large top-down soil emissions in southern Hebei and along the northern coasts of the Bohai Sea, in comparison with the a priori dataset, are caused in part by the underestimate of fertilizer-derived emissions in the a priori dataset.

* One last issue I have is with the discussion of the linearity of NOx emissions vs. NO2 columns. In the initial method, the assumption is that NO2 columns respond linearly to changes in NOx emissions, however, the results indicate that the sensitivity is less than one. The authors should discuss the implication of this non-linearity on the ultimate conclusions of this study.

Response: The resulting uncertainties in the top-down and a posteriori emissions have been included in the revised manuscript.

Specific comments

P29808, L18-19: 'They are each less than 6% of anthropogenic emissions annually'. Later on, it becomes clear what the authors mean, but perhaps this can be rephrased here to make clear that lightning NOx and soil NOx each make up less than 6% of the anthropogenic NOx emissions.

Response: The sentences have been clarified in the revised manuscript.

P29810, L26-28: The statement that biomass burning emissions are unimportant over China came to me as a surprise. I've seen papers where biomass burning does make a significant contribution to e.g. HCHO emissions. I think the author should provide some more information on how small the contribution of biomass burning NOx emissions is, and give some references. This issue comes back at P29816, L22. Introduction: although it is clear, the introduction reads somewhat as a literature overview, and lacked some focus to me. Why is the author so interested in NOx emissions over China?

Response: Biomass burning is an important source of VOC and CO because of the large emission factors as a result of incomplete combustion. It is not important for NOx in East China due to the low emission factor. For 2006, the emission budget is only about 0.013 TgN based on the GFED2 data. We have clarified this point in the revised manuscript and included two references. We had included one reference in the original manuscript.

NOx is key constituents for tropospheric chemistry, air pollution and climate forcings. We had stated the importance of understanding NOx emissions over China and contributions from individual sources. In the revised manuscript, we add a note with references that anthropogenic emissions of NOx in China are significant and grow rapidly in recent years and have attracted a lot of attention in the international community.

P29812: it seems to me that the author should provide some more basis for assuming a larger retrieval error in winter than in summer. It is true that some satellite retrieval errors are probably larger in winter (lower solar zenith angles, thinner boundary layers so more impact of albedo and NO_2 profile errors), but aerosol errors could well be larger in summer, when AOT is generally higher. I think Fig. 2 is unnecessary (and holds very little information at all), and the author should give more justification for his (assumed) seasonal behavior of the retrieval errors.

Response: In the original manuscript, we had several references in stating that retrieval errors may be larger in winter than in summer. It is difficult to have a definite conclusion for China due to lack of independent and accurate measurements for error evaluation. For this reason, we had also tested in the original manuscript the impact of the assumed seasonality of retrieval errors on the inversion results. We found that whether or not the retrieval errors have seasonality has only a small impact on our inversion results.

We have removed Fig. 2 as suggested.

P29813, L11: 'contribute only to 6%', please remove 'to'.

Response: Removed.

Section 4.1.2: this part is not really clear to me. I appreciate the author's effort to communicate his method as complete as possible, but this appears more suitable for an Appendix. Instead of all this, it would be better to provide more detail on how lightning and soil NOx are modeled in GEOS-Chem.

Response: The present inversion methodology is innovative and crucial for the top-down results. Therefore a detailed description is important, and should be put in the main text. We have added extensive analyses of lightning and soil emissions in the CTM.

P29818, L1: 'many of the areas with stripe patterns in Fig. 7' Which patterns are referred to here?

Response: We have removed 'with stripe patterns' not to cause confusion.

Figure 3: titles are hard to read in the paper version.

Response: Titles have been enlarged. Figure 3 has been changed to Fig. 4.