Atmos. Chem. Phys. Discuss., 13, C492–C499, 2013
www.atmos-chem-phys-discuss.net/13/C492/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Multiannual changes of CO_2 emissions in China: indirect estimates derived from satellite measurements of tropospheric NO_2 columns" *by* E. V. Berezin et al.

Anonymous Referee #1

Received and published: 10 March 2013

Review of "Multiannual changes of CO2 emissions in China: indirect estimates derived from satellite measurements of tropospheric NO2 columns" by Berezin et al.

This manuscript discusses the trend of anthropogenic CO2 emissions in China derived in a 'top-down' fashion using multi-year satellite retrievals of tropospheric NO2 columns. The study time period is 1996-2008, based on tropospheric NO2 columns derived from the GOME and SCIAMACHY satellite instruments. A chemical transport model is used to relate the NO2 columns to anthropogenic NOx emissions. The main conclusion is that both the top-down derived trend of anthropogenic CO2 emissions and anthropogenic NOx emissions exceed the bottom-up estimates and the authors

essentially attribute most, if not all, of the discrepancies to the bottom-up emission method, especially the energy statistics. The merit of this study is that it explores the potential of using satellite-derived NO2 columns to constrain anthropogenic CO2 emissions, a direction not investigated before. But I found several of the important underlying assumptions are not justified or incorrect and the main result is not reasonable, as stated below.

Major issues:

1. In the introduction, the authors discuss the potential of using species correlations to constrain CO2 emissions. Although this study appears to be using NOx and CO2 relationship to constrain CO2 emissions, it is fundamentally different from those species correlation top-down studies discussed in the introduction in that it does not rely on the observed NO2 to CO2 relationships as top-down constraints. Instead, it is a mere two-step analysis: first the authors derived top-down NOx emission trend over China using satellite NO2 columns, second the authors apply the CO2 to NOx emission ratios from a bottom-up inventory (EDGAR) to that top-down NOx emission trend to derive the top-down CO2 emission trend. The first step is not new as there have been numerous previous studies on using satellite NO2 columns to constrain Chinese NOx emissions. So the manuscript is just one step forward in applying the NOx to CO2 emission ratio to the derived top-down NOx emission trend. In that sense, I suggest the authors to re-structure the paper to clarify their approach in the introduction and the methodology. In particular, I suggest the authors to put the discussion of the top-down NOx emission trend before that of the CO2 emission trend, as this forms the basis to their calculated the top-down CO2 trends, and discuss in more detail the difference of their top-down NOx emission trends compared to previous studies such as Zhang et al. (2007) and Lamsal et al., (2011). In discussing the top-down CO2 trend, the authors should put more emphasis on the sensitivity of their results on the choice of NOx to CO2 emission ratio (to be commented in details later).

2. This is a further comment to the authors in order to elaborate my previous com-

ments on species correlation. If species correlations are to be used to constrain the emissions of one or another, two conditions need to be met: (1) the two species are co-emitted from common sources; and (2) the atmospheric transport/processes/other-emissions will not significantly distort their correlations at the time when observations are taken. For example, CO2 and CO correlations meet the two criteria and CO2 and CO have been demonstrated to exhibit good correlations by atmospheric observations and models when combustion sources dominate in the CO2 sources. As a consequence, there have been quite a few previous studies using observed CO and CO2 correlations to constrain the anthropogenic portion of CO2 emissions (as referenced in the introduction). Although the authors establish the first criteria for NOx and CO2 relationship (that is, the two species are co-emitted from combustion), they did not discuss the second criteria. Many species are emitted from fossil fuel combustions along with CO2, but not all of them can be used to constrain CO2 emissions. Do the authors have evidence from observations and models that atmospheric NO2 columns and CO2 concentrations are well correlated on the temporal and spatial scale of their interest (i.e., monthly time step over China)? As NOx is very short lived (lifetime of hours to days) compared with CO2, they may not exhibit strong correlations in the atmosphere. If such a correlation is lacking, the methodology of using observed NO2 columns as a proxy for CO2 emissions cannot be established. I understand that suitable observations may not be available to the authors, but they have a chemical transport model to test the correlations. Driven by the same bottom-up emission inventory of NOx and CO2 (e.g., from EDGAR as used in the manuscript), do the CTM simulated NO2 columns correlate with the simulated CO2 columns? Since the authors do not use observed CO2 to NOx ratio as constraints, the suggested correlation analysis may be irrelevant and in this case no need to reply this comment.

3. pg 268-269, method: I don't agree with the discussion of equation 3. The lifetime of NOx needs to be calculated with seasonally varying anthropogenic NOx emissions in order to improve the accuracy of top-down emission estimate and to reduce systematic biases. The reason is that NOx lifetime depends on NOx emissions. In the

main loss reaction of NOx: NO2 + OH — > HNO3, OH will be titrated in the conditions of high NOx emissions which leads to increased NOx lifetime. This is well known in the NOx chemistry. We know for sure that anthropogenic NOx emissions vary with season in China with higher emissions in winter and lower emissions in summer, only that the magnitude of the seasonal variation is uncertain. This bottom-up information (i.e., seasonality in emissions) should be included into the modeling analysis in order to reduce the systematic biases in the top-down analysis. I acknowledge that the authors discussed this issue as a source of uncertainty later in the paper and argued it is small, but the different trend in wintertime and summertime NOx emissions found in the manuscript (e.g. Fig 5) warrants an in-depth analysis of this issue. Also, the author acknowledged that their winter-to-summer ratio of NOx emissions is substantially larger than both the bottom-up and previous top-down studies (pg 274, line 25-), which also suggest the seasonality of lifetime calculated using annual-mean emissions is not correct and introduces systematic biases. This is a bias that can be corrected easily so the authors should do it.

4. The authors found a factor of three increases in NOx emissions from 1996 to 2008 (Fig. 5) using the same NOx lifetime calculated based on a particular year of emissions. Which year of emissions the authors used to derive the NOx lifetime? Given the dependence of NOx lifetime on its emissions (see above), NOx lifetime should be lower in later years with much higher emissions. Without taking this into account, there should be systematic high biases in the derived top-down NOx emission trend. This high bias can explain why the top-down trend deviated more from the bottom-up trend as years go by. Although the authors discussed briefly this bias, their scaling factors (0.3-1) do not seem to be correct (pg 282, line 13). As indicated in the paper, the authors apparently do not want to run multi-year simulations with year-to-year variations of NOx emission. I suggest the authors run at least two years of simulations to derive the range of NOx lifetime changes during the study period: one using 1996 emissions and one using 2008 emissions, then linearly interpolate the lifetime in between. This would not be a substantial computational effort. As the manuscript is mainly concerned

with emission trend, it is important to correct for this bias too.

5. The authors spend substantial efforts to discuss the impact of natural emissions and how to treat background NO_2 in the paper. It is disappointing they didn't even mention the magnitude of natural emissions (soil NO_x emissions) used in their model simulation. Previous top-down studies using satellite NO_2 columns have found soil NO_x emissions underestimated in China (e.g, Jaegle et al., 2005; Wang et al., 2007). Recently there have been several studies to improve the estimate of soil NO_x emissions (e.g., Hudman et al., 2012). Is the magnitude of soil NO_x emissions used in their model comparable to these recent studies? How to address the impact of uncertainties in soil NO_x emissions on the derived top-down trend of anthropogenic NO_x?

6. As indicated above, the CO_2 emission trend depends on the NO_x emission trend and the NO_x to CO_2 emission ratio. The authors adopt the NO_x to CO_2 emission ratio from EDGAR. There are many regional emission inventories on NO_x or CO_2 emissions from China and the authors referenced some in the introduction. Although these emission inventories may only be for a specific year, they can still be used to derive CO_2 to NO_x emission ratio for that specific year as compared to the ratio derived from EDGAR. That way, the authors have a better sense on the true uncertainties of their top-down emission trend of CO_2.

7. Finally, I found the discrepancy between the bottom-up and top-down CO_2 emission trend is too large to be explained by uncertainties or even statistical errors in the bottom-up inventory. Compared with emissions of air pollutants (CO, NO_x, e.g.), anthropogenic CO_2 emissions are more accurate given its primary dependence on the energy consumption (emissions of air pollutant emissions depend also on emission factor and end-of-pipe emission control measures). The uncertainty for Chinese anthropogenic CO_2 emissions is quoted as 15-20% (Gregg et al., 2008), compared with that of 6%-10% for global anthropogenic CO_2 emissions. In the introduction, the authors should reference these bottom-up emission uncertainties for CO_2. In the one study included in the introduction, Guan et al. (2012) reported a1.4 Gt emission gap

of anthropogenic CO_2 between national and provincial statistics in China, which is still less than 20% of Chinese total anthropogenic CO_2 emissions. However, in Fig 4, the derived top-down CO_2 trend is a factor of 2 higher than that of the bottom-up trend. If taken simply, this means a factor of 2 emission differences between the bottom-up and top-down CO_2 emissions in 2008. The authors attributed the differences to statistics used in the bottom-up inventory, but they do so in a very general way. It will be extremely difficult, if not essentially impossible, to find such a large gap in energy statistics to explain the missing anthropogenic CO_2 emissions in China. Global coal consumption and oil consumption are well constrained and there is no evidence in atmospheric CO_2 record to indicate such a large magnitude of missing CO_2 emissions from northern middle latitude. Therefore, I strongly suggest the authors revise their top-down method to derive NO_x emissions as suggested in my previous comments and to correct for the known high biases before jumping to conclusions and discussions of the derived CO_2 emission trends.

Minor comments:

1. In the introduction, the authors emphasize the advantage of using NO_2 column to identify emission hot-spots and thus the potential of constraining CO_2 emissions at a finer spatial and temporal scales. However, their model is run at 1 degree by 1 degree which is very coarse for regional scale studies. Also, most of the analysis conducted therein is still on the national and monthly scale, although the province-level map (Fig 9) is a good step forward. Therefore, that point made in the introduction is not justified. I suggest the authors change the introduction or focus more on the fine scale features in their analysis.

2. Pg 259, line 6: the authors mentioned the study of Brioude et al. (2012) using NO_y to estimate CO_2 emissions from Houston. I wanted to caution that NO_y is substantially longer lived than NO_x, as NO_y includes NO_x and all its oxidation products. This study cannot be used as a foundation to establish the correlation between NO_2 and CO_2.

Typos:

Pg 2, line 12: a semi-column (;) is need after the parenthesis. Pg 260, line 10: Eastern Asia to East Asia.

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 255, 2013.