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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.

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We thank the Referee for the detailed critical evaluation of our paper. All of the Referee's concerns have been very carefully addressed during the revision process. In our earlier interactive comment (Berezin et al., 2013), we had already responded to some of the critics. In particular, we had regretted that the review does not take into account the paper's main robust results. The substantial additional analysis performed during the revision process confirmed the adequacy, robustness and self-consistency of the results of our study. Below we describe our point-to-point responses to the Referee's comments.

1. Referee's first specific comment concerns the difference between our method and the methods employed in the studies exploiting correlation between atmospheric concentrations of CO_2 and co-emitted species. We agree that this difference is important and that it was not sufficiently explained in the reviewed manuscript. Accordingly, we have restructured the Introduction and added a necessary comment clarifying this difference. In regard to Referee's suggestion to change the structure of the paper entirely by presenting first the NO_x emission trends and then our CO_2 emission trend estimates, we would like to note that the logic of the paper reflects our desire to put more emphasis to the main results of our study by presenting them in Section 4 in the first place. The main results are then followed by their analysis (including analysis of the trends in the NO_x emissions) and discussion of uncertainties. We believe that such structure is sufficiently logical and quite consistent with the main goals of our study.

2. Referee's second comment concerns conditions that have to be met in order to use species correlation to constrain the emissions of one of them. We agree that these conditions are not fully satisfied in our case (in particular, NO_2 columns and CO_2 concentrations can hardly significantly correlate on the temporal and spatial scales addressed in our study) because of short lifetime of NO_2. And this is indeed the reason why we have not used the correlation analysis in our study. In our case the relationship between CO_2 and NO_x emissions is not constrained by observations but is derived from the bottom-up inventories. In this sense, our method provides a kind of hybrid estimates rather than "classical" top-down estimates. This point is further clarified in the Introduction of the revised manuscript.

3. The main point of the third comment is that disregarding seasonal changes in NO_x lifetime due to seasonal changes in NO_x emissions may cause systematic biases in our estimates. The Referee suggests including seasonality in emissions into the modeling analysis in order to reduce such probable systematic biases. While we recognize that our estimates of the seasonal cycle of emissions may be uncertain, we doubt that a way to tackle this uncertainty by employing the available bottom-up information on seasonal

ACPD
13, C3752–C3757, 2013

variation of anthropogenic NO_x emissions (as it is suggested by the Referee) might be really efficient. Indeed, on the one hand, the bottom-up inventories indicate that such variation in China is very small (the ratio of the maximum to minimum emission rate 1.3 or less, as it is noted in Section 4.1), and therefore the results obtained with such variation would be inevitably very close to results in our case III of the estimation procedure, where the seasonal variation is assumed to be absent. On the other hand, the Referee recognizes that the available bottom-up information about the magnitude of the seasonal variation is uncertain. Although we do not claim that our estimates of the seasonal variation are more accurate than those based on the bottom-up information, by comparing the results for the two cases of our estimation procedure (such as the case I and the case III which has been re-defined in the revised version of the manuscript to better address this concern of the Referee), we made sure that our "baseline" estimates of NO_x emission trends are not sensitive to the assumptions about the seasonal cycle. Note that the impact of chemical nonlinearities on the relationship between NO_x emissions and NO_2 columns is not found to be significant in the considered situation (please see also our response to the next comment of the Referee) both on the annual and monthly scales. Note also that Eqs. 4 and 5 are reformulated in the revised version for a more general case where NO_x emission specified in a model may vary from month to month and the possible difference between the estimated and real NO_2 lifetime is taken into account. The discussion in Section 4.1 is also re-arranged in order to better address the role of the seasonal variations and its uncertainties on our estimates.

4. The major point of the fourth comment is that the changes of the NO_x lifetime may cause high systematic biases in the derived top-down emission. To take into account this effect the Referee suggested "to run at least two years of simulations to derive the range of NO_x lifetime changes during the study period: one using 1996 emissions and one using 2008 emissions, then linearly interpolate the lifetime in between". We did this exactly by using the EDGAR v 4.2 data. The difference between the updated estimates presented in the revised and reviewed versions of the manuscript is almost negligible. This result is quite consistent with our discussion of nonlinearities in Section

ACPD
13, C3752–C3757, 2013

5, as well as with the bulk majority of numerous previous studies (many of which are cited in the manuscript) where satellite NO_2 measurements were used to estimate NO_x emissions and their trends. Note that our simulations performed with the original EDGAR v.4.2 emissions for 1996 and 2008 (where not only NO_x emissions but also VOC emissions were different), and additional simulations where the VOC emissions were swapped between 1996 and 2008 datasets indicate that a small nonlinearity in the response of NO_2 columns to NO_x emission changes is almost completely compensated both on the monthly and annual scales by corresponding changes in VOC emissions. (Specifically, the increase of NO_x emissions lead to the increase of NO_2 lifetime for the annually average columns, but the increase in VOC emissions is found to result in the lifetime decrease). As it explained in Section 5, we have performed additional test of nonlinearity by simulating the dependence of NO_2 columns on the baseline (2008) NO_x emissions scaled within the scale factor ranging from 0.3 to 1 and found that nonlinearity is small. We are puzzled why the Referee thinks that these scaling factors "do not seem to be correct", because such scaling covers the range of the NO_x emission changes in our estimates and significantly exceeds the ratio of the 2008 to 1996 NO_x emissions ($\sim$1.8) according to EDGAR v.4.2.

5. The Referee expressed a concern about a possible impact of uncertainties in soil emissions on the derived top-down trend of anthropogenic NO_x. In response to this Referee's comment, we first would like to note that possible uncertainties in natural emissions constitute only a part of uncertainties in the simulated background NO_2 columns. Other uncertainties may be e.g. due to inaccuracies in boundary conditions (taken in our case from a global model), in vertical transport scheme, chemistry and deposition. The different factors may enhance or compensate each other. Accordingly, we believe that it would be more reasonable and straightforward to try to evaluate the effect of the total uncertainties in the background NO_2 columns on our NO_x emission trend estimates, rather than to discuss the contributions of uncertainties of the individual factors mentioned above. This evaluation and corresponding discussion are further improved in the revised manuscript. In particular, we explain why we think that

ACPD
13, C3752–C3757, 2013

the considered cases I, II and III of our estimation procedure represent the range of possible uncertainties associated with the background NO_2 columns. In response to Referee's question about the magnitude of NO_x soil emissions specified in our model, we note in Section 5 of the revised manuscript that the total annual NO_x emissions calculated in CHIMERE with the MEGAN inventory data in eastern China are 0.37 Tg N/yr. This number is in excellent agreement with the recent top-down estimate of $0.38 \pm 65\%$ Tg N/yr reported by Lin (2012) and also in agreements with the bottom-up estimate of $0.50 \pm 25\%$ Tg N/yr calculated in Lin (2012) by using the improved inventory by Hudman et al. (2012). Accordingly, we believe that the simulated contribution of soil emissions to the background NO_2 columns is sufficiently adequate.

6. As it is suggested by the Referee, in addition to the NO_x-to-CO_2 emission conversion factors from the EDGAR inventory, we considered the conversion factors calculated using data of a regional inventory. Specifically, we adopt the NO_x-to-CO_2 emission ratio from the REAS (v.1.11 and v.2.1) emission databases for the period from 1996 to 2008. We found the results obtained with the different NO_x-to-CO_2 emissions ratio (see Case IV in Fig.4 of the revised paper) to be in agreement with the main conclusions of our study, although the CO_2 emission trends estimated with the REAS data is found to be somewhat smaller than those obtained with the EDGAR data.

7. The Referee found that "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". More specifically, in Referee's opinion our results mean "a factor of 2 emission differences between the bottom-up and top-down CO_2 emissions in 2008". This point has already been thoroughly addressed in our previous comment (Berezin et al., 2013). In particular, we have argued that the Referee's conclusion does not follow from our analysis and that "only relative changes (not absolute values) of emissions are evaluated in this study; thus the differences between the top-down and bottom-up emission estimates cannot be unambiguously attributed to certain years". This feature of our analysis is emphasized throughout the paper (in Introduction, in

ACPD
13, C3752–C3757, 2013

Section 4.1, and in the caption of Figure 4). The reported uncertainties in bottom–up inventories for CO_2 in China are mentioned in Introduction, as requested by the Referee.

The responses to minor comments:

1. A change is made in Introduction to avoid possible misinterpretation of our intentions.

2. As it was mentioned before we don't exploit any correlations between NO_2 columns and CO_2 concentrations in our analysis. The paper by Brioude et al. is mentioned as an example of a recent study where CO_2 emissions are estimated by using the observation of co-emitted species. The respective clarification is made in the Introduction.

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