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Comment

Interactive comment on “Multiannual changes of CO₂ emissions in China: indirect estimates derived from satellite measurements of tropospheric NO₂ columns” by E. V. Berezin et al.

E. V. Berezin et al.

konov@appl.sci-nnov.ru

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We thank the Referee for the generally positive evaluation of our paper and the useful comments. All of Referee's comments and suggestions are carefully addressed in the revised manuscript. Below we describe our point-to-point responses.

Referee: *Top down estimated CO₂ emissions. The question is whether the approach that has been followed is a top-down method for estimating CO₂ emissions. . .*

We agree with the Referee and have made necessary changes in the revised manuscript. Specifically, we refer to our estimates of CO₂ emission trends as a kind of

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hybrid estimates based on combination of both top-down and bottom-up approaches. Note that our estimates of NO_x emission trends can still be regarded as typical top-down estimates.

Referee: Uncertainty estimates. It is mentioned that the three cases bracket the lower and upper limits of the NO_x emission uncertainty. Besides the fact that it is unclear why these scenarios represent upper and lower limits (the alternatives to the base scenario lead to very similar results), what matters in the end is to bracket the uncertainty in the inferred CO₂ trend. The question is how significant the uncertainty in the conversion factor may be. Potentially it could be pretty large given the revolutionary nature of the economic developments that are taking place. This issue is recognized as an important source of systematic uncertainty that "is impossible to evaluate". I'm not quite convinced by this argument and believe several inventories are available that should allow the authors to derive such an estimate.

Indeed, the uncertainties in the inferred CO₂ emission trends due to uncertainty in the assumed NO_x-to-CO₂ conversion factor were not taken into account in the reviewed manuscript. Based on the arguments expressed in Section 5, we expected that such uncertainties could not affect the major conclusions of our study. To get a better idea about the magnitude of the uncertainty in the conversion factor, in addition to the conversion factors from the EDGAR inventory, we considered the NO_x-to-CO₂ emission ratio from the REAS emission inventory. The differences between the different estimates of the conversion factors are not large but not negligible. Importantly, the results obtained with the different NO_x-to-CO₂ emission conversion factors (these results are shown as Case IV in Fig.4) fall into the range of uncertainties bracketed by the results for the cases I and case III.

Identifying definite quantitative bounds for possible systematic uncertainties in our top-down estimates is perhaps irresolvable task. Such situation is quite typical for inverse modeling studies involving complex models. Instead, it is possible to evaluate the impact of different factors on our estimates and in this way to get an idea about the

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overall uncertainty. In our study we follow this way, and we recognize that the statements concerning bracketing the uncertainties were not sufficiently accurate in the reviewed manuscript. The corresponding changes are made in the revised manuscript. Nonetheless, our subjective judgment is that the four cases considered in the revised manuscript indeed span the range of possible uncertainties in our estimates. Specifically, as it is discussed in Section 4.1, the magnitude of the estimated trends in NO_x emissions increases with the increase in the background NO_2 . If the background NO_2 would be considerably larger than that calculated by the model, then the magnitude of the NO_x emission trends in summer would become larger than in winter, and this would be, in our opinion, quite unrealistic result (considering the dynamics of the different emission sectors). On the other hand, the case III addresses the unrealistic situation where the background NO_2 columns are zero, and the corresponding results could therefore be considered as the lower bound for the NO_x emission trends. These points are clarified in Section 4.1 of the revised manuscript.

Referee: Photochemistry. Although the authors investigate the linearity of the relation between total column NO_2 and NO_x emissions in CHIMERE they do not estimate how the lifetime of NO_x may have changed due to increasing levels of other pollutants. This would not be difficult to assess and would contribute greatly to the overall uncertainty assessment, which is currently described in detail in the discussion section but not sufficiently quantified.

We have taken into consideration Referee's suggestions and performed additional simulations where the VOC emissions were swapped between the 1996 and 2008 EDGAR v4.2 datasets (along with the baseline simulations with the 1996 and 2008 EDGAR v4.2 emissions as explained in our response to the comments of the first anonymous referee). The results indicate that changes in VOC emissions compensate (almost completely) a small nonlinearity ($\sim 10\%$) in the response of NO_2 columns to NO_x emission changes both on the monthly and annual scales. More specifically, the increase of NO_x emissions leads to the increase of NO_2 lifetime for the annually average columns, but

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the increase in VOC emissions is found to result in the lifetime decrease (indeed VOC provide the source of OH which removes NO₂ from the atmosphere). These results are discussed in Section 5 of the revised manuscript.

Referee: *Page 262, line 12: The suggestion is made that the reduced repeat cycle of SCIAMACHY compared with GOME is due to its smaller footprint, but this is not the case (it has probably to do with the alternate nadir-limb sounding in SCIAMACHY).*

We agree that it would be incorrect to compare characteristics of the two satellite instruments in this way because the objectives of these instruments were different. The corresponding correction is made in the revised manuscript.

Referee: *eq 1: Shouldn't rho simply represent how the footprint size varies along the swath? It is not clear why this would follow an exponential function. Else I would have expected SCIAMACHYs viewing angle to show up in the equation.*

To the best of our knowledge, there is no known "strict" way to make SCIAMACHY, GOME and model data quite compatible. In this study we use an approximate *ad hoc* method which was employed in our earlier inverse modeling study cited in the manuscript. One of the features of this method is that smoothing applies to the monthly or seasonally average fields (which we use in this study) rather than directly to the "orbital" data. For this reason, it cannot take into account any specific orbital parameters. The exponential (Gaussian) function is one of the simplest functions having a bell-like shape and thus allowing introducing an effective scale of extra smoothing of the GOME data with respect to the SCIAMACHY data. As it is noted in the reviewed manuscript the results concerning the total emission estimates for China are not sensitive to the configuration of this method. In fact, the results were found to be practically the same if no smoothing was applied. When emissions from smaller regions (such as China's provinces) are evaluated, the difference between the GOME and SCIAMACHY data becomes important. However, only qualitative (rather than quantitative) results for the provincial scale are discussed in our paper.

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Referee: *Page 265, eq 5: I find it much clearer to express the ratio of annual emissions as the mean of the monthly gradient ratios.*

We agree that it would be preferable to define the NO_x-to-CO₂ conversion emission conversion factors with the monthly rather than with annual temporal resolution. However, unfortunately only the annual data of the considered emission inventories are available so far.

Referee: *Page 269, line 2: The results may not be very sensitive to the treatment of the seasonal cycle of NO_x, but it is unclear why it would be “essential” to use seasonally constant emissions in the model.*

In fact, the use of seasonally constant emissions in the model is not essential but rather convenient. Eq. 4 is re-formulated for a more general case in the revised manuscript, but the use of the seasonally constant emissions allows us to significantly simplify the following-up Eq. 5 and corresponding analysis and interpretation of the obtained results.

Referee: *Page 269, line 26: It is unclear why setting C_b to zero addresses the possibility of similar trends in the background as in the fossil emissions*

In the reviewed manuscript we noted that setting C_b to zero addresses a situation where the relative changes in the background part of the measured NO₂ columns are the same as the relative changes in their anthropogenic part (including both seasonal and multi-annual changes). Indeed, if any relative variations in the background and anthropogenic part of NO₂ columns would be identical, then there would be no need to make any difference between them, and there would be no need to subtract the background from the total columns in order to evaluate trends in the anthropogenic emissions. Accordingly, in the discussed hypothetical situation we could safely assume that C_b is zero. The corresponding additional explanation is introduced in the revised manuscript.

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Referee: *Page 270, line 6: Why is eq 8 more sensitive to the reference year than 7, is it because it doesn't account for changes in the background?*

This is because the reference year defined only normalization in Eq.7 (which could not affect the estimation of exponential trends), but the NO₂ columns for the reference year k are directly involved in the estimate of emissions for the year j in Eq.8.

Referee: *Page 270, line 16: What has the exponential function been fitted to? Is there any a priori specified bound on the coefficients?*

The exponential function is fitted directly to the data provided by Eq. 5, 7 or 8 (in the revised manuscript). Any a priori constraints are not used. The corresponding definition is clarified in the revised manuscript.

Referee: *Page 273, line 15: The possible contribution of soil NO_x emissions should be quantified using available estimates.*

The requested numbers are added in Section 5 of the revised manuscript.

Referee: *Page 277, line 28: If the activity data of energy production generally do not capture rapid changes then where does the sharp transition in EDGAR around 2000-2002 come from?*

As it is explained in the reviewed manuscript (p.277, l. 15-18), there is an acceleration of the growth in the PEHP sector after 2001, and an upward tendency appears also in the MIC sector. The combined result of these changes is the appearance of the sharp bend of the total CO₂ and NO_x emissions in the period from 2001 to 2002. The appearance of the sharp bend may look not to be quite obvious, but this explanation is easily confirmed by direct simple calculations. It is necessary to take into account the difference in the magnitude of emissions from the major sectors.

Referee: *Page 279, line 24: What is meant by 'they'?*

"They" mean "our estimates". The corresponding statement is clarified in the revised

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version.

Referee: *Page 280, line 5: What is the significance of this statement? To me it seems low, given the small difference in correlation coefficients and the limited trend in the conversion factor.*

Yes, indeed. This statement is modified in the revised manuscript.

Referee: *Figure 3: The difference between the two figures is unclear (they look exactly the same).*

We are sorry for this unnoticed production error.

Referee: *Figure 10: The results for EDGAR are difficult to reconcile with what is shown in Figure 9. There emission ratios are typically around 2, whereas Figure 10 shows pretty flat lines for EDGAR.*

Actually, we could not find any contradiction between Fig. 9 and 10. Both figures show that the emission ratios in EDGAR data are about 2, and this is why the lines for EDGAR are "flat" in Fig 10.

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

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