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***Interactive comment on* “Constraint of anthropogenic NO_x emissions in China from different sectors: a new methodology using multiple satellite retrievals” by J.-T. Lin et al.**

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In this paper, Lin et al. report on a new method to estimate anthropogenic emissions of NO_x by using the differences of satellite measurements of NO₂ taken at different times of the day. The method is applied to data taken by GOME-2 and OMI in July 2008 over China, and very good agreement is found between the derived emission strengths and the a priori emissions used which were taken from the INTEX-B studies. Application of a method relying on a linear relation between emissions and satellite derived columns leads to much larger emission estimates. In a series of sensitivity studies, the authors explore the uncertainties of the inversion and find individual uncertainties of the order

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of 10% to 15%.

The paper reports on an interesting and novel approach to estimating NO_x emissions from satellite measurements of NO₂. It is clearly structured and well written and fits well into the scope of ACP. However, I do have major concerns with respect to the results presented and a number of points need to be clarified before the paper can be published in ACP.

General Response: We thank the constructive comments of the reviewer.

We have revised and re-structured the manuscript to clarify several points. First, a major cause of the model-satellite difference may be the positive biases in the retrievals (Sect. 2.1.1, Sect. 4). Second, the retrievals errors are positively correlated (Sect. 2.1.1). Third, our approach is based on the difference of the two 'retrieved' NO_x columns such that it is less susceptible to systematic retrieval errors provided that the retrieval errors are consistent (Sect.4). Fourth, the Martin et al. method is based on a proportional relationship between daily mean NO_x emissions and NO₂ column at a particular time of day, thus its top-down results are subject to systematic retrieval errors without any 'screening' as in our approach (Sect. 4.1). Fifth, the inclusion of nighttime evolution mainly affects the spatial distribution of top-down emissions when retrievals in the morning are used; while the emission budget over a large area like China is not affected significantly (Sect. 4.1).

My main concern lies with the contradiction between the results shown in Fig. 2, which clearly show an underestimation of the NO₂ column by the model and the conclusion of the inversion that the emissions used are basically correct. To me, such a difference between model and measurements can be explained in three ways: 1) the satellite data are wrong, 2) the emissions used in the model are wrong, or 3) the chemistry and/or transport in the model are wrong. In practice, all three effects will contribute to the differences but the usual approach is to assume that mainly emissions are wrong and

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need to be updated. My question to the authors is how they explain this discrepancy if they conclude that the emissions are correct.

Response: In general, all these three factors contribute to the model-retrieval difference. Particularly for this study, an important cause of the difference results from systematic retrieval errors.

As shown in Sect. 2.1.1 of the revised manuscript, van Noije et al. (2006) compared the KNMI retrieval method for GOME with the other two independent methods at Bremen University (Richter and Burrows, 2002; Richter et al., 2005) and Dalhousie University/SAO (Martin et al., 2003). They found that the VCD in July 2000 retrieved by the three methods ranges from $\sim 2.5 \times 10^{15}$ molec/cm² (By Bremen University) to $\sim 5.1 \times 10^{15}$ molec/cm² (by KNMI) over northern East China (110°E–123°E, 30°N–40°N). Assuming the mean of VCD retrievals from the three methods as the true VCD, the KNMI retrieval for GOME would be overestimated by $\sim 32\%$ for July 2000. The GOME-2 retrieval is expected to contain the same level of error due to the highly comparable retrieval method. In addition, a number of recent studies (Boersma et al., 2009b; Hains et al., 2009; Huijnen et al., personal communication, 2009; Lamsal et al., 2009a,b; Zhou et al., 2009) have suggested that the KNMI OMI retrieval is biased positively, most likely with a magnitude of 0–30% irrespective of season.

A simple calculation reducing the GOME-2 retrieval by 32% and OMI by 15% (mean of 0–30%) leads to adjusted retrievals about 11% higher than model results (see Figure S1 below). Therefore it is concluded that systematic errors of retrievals are most likely the main cause of model-retrieval difference shown in Figure 2.

These analyses have been added to the revised manuscript (Sect. 2.1.1 and Sect. 3).

In several places, the results from this inversion approach are contrasted to what the authors call the Martin et al. method. It is suggested that the latter is less accurate,

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mainly because night time chemistry could affect the morning measurements and also because the diurnal variation of the emissions has to be prescribed. In my opinion, the paper does not show that the new method is more accurate than the Martin et al. approach. The fact that the retrieved emissions are significantly different between the two methods does not imply that the Martin et al. inversion is wrong. Also, the difference between the Martin et al. inversions of GOME-2 and OMI does not prove that the method fails, in particular considering that a change of only 13% in the GOME-2 columns would bring the results in good agreement and the authors show that GOME-2 NO₂ used is about 10% larger than SCIAMACHY NO₂. In any case, the Martin et al. inversion based on OMI data should not suffer from night time effects, and still strongly disagrees with the results obtained with the new method. I think that the authors need to explain why their method leads to different results and why they believe these results are more accurate than previous approaches.

Response: As discussed before, an important cause of the model-satellite difference may be the positive biases in the retrievals, which are positively correlated (Sect. 2.1.1). By utilizing the difference between the two retrievals, our approach reduces the effect of systematic retrieval errors on top-down emissions, because part of the errors can be canceled out by the differentiation. Therefore it is relatively less sensitive to systematic retrieval errors given that the errors are consistent (like in this case). By comparison, the Martin et al. method assumes a linear relationship between retrievals and top-down emissions, it thus does not provide any ‘screening’ for retrieval errors. In other words, systematic errors in retrievals will fully propagate to top-down emissions. Therefore, while the Martin et al. method is not necessarily incorrect, it is relatively more sensitive to systematic retrieval errors. An important contribution of this study is to introduce an approach that is less susceptible to systematic retrieval errors.

Indeed the difference between the Martin et al. inversions of GOME-2 and OMI does not necessarily prove that the method fails. However, it does raise concerns with the uncertainties of top-down emissions estimated by the Martin et al. method from indi-

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vidual studies (e.g., Martin et al., 2003; Jaegle et al., 2005; Martin et al., 2006; Wang et al., 2007; Zhang et al., 2007), particularly when the purpose of the top-down studies is to improve the bottom-up estimate. In addition, the difference for July 2008 between GOME-2 and OMI inversions may happen to be at the lower end of their differences. Our results for January 2009 show that top-down emissions relative to GOME-2 are 30% larger than that relative to OMI; and both estimates are much higher than the bottom-up estimate (Lin et al., 2009). In contrast, by combining both retrievals, our approach is able to reconcile such inconsistency, leading to top-down emissions much closer to our understanding from the bottom-up perspective.

We agree that the inclusion of nighttime evolution mainly affects the spatial distribution of top-down emissions when retrievals in the morning are used; while the emission budget over a large area like China is not affected significantly. In addition, the inclusion will not affect significantly the top-down calculation based on OMI and the Martin et al. approach. We have clarified this point in the revised manuscript (Sect. 4.1).

One aspect of this study is that not only emissions, but also their diurnal variation is retrieved. However, judging from Fig. 4, emissions in the time between the overpasses of GOME-2 and OMI are nearly constant. Thus, the method comes down to comparing the change in the satellite columns to the change in NO_x lifetime as predicted by the model. I'm surprised that under these conditions, the retrieval has enough information to invert 1) emissions, 2) their diurnal change and c) their distribution over 4 emission sectors from just two measured numbers. Considering the lack of difference in emissions between the two overpass times of the satellites, I would have expected that only one piece of information can be derived. Please explain where this information is coming from.

Response: The derivation of top-down emissions for the four individual sectors is done through iteration (Eqs. 7-11). The temporal profile of each sector, VAs_i, is prescribed

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and kept unchanged throughout the iteration (Figure 4). In particular, it is obtained from independent sources and NOT derived from the current approach.

Also, partitioning anthropogenic emissions to individual sectors (Eqs. 10-11) requires additional independent information about the uncertainty of emissions in each sector, U_s , which is also prescribed and NOT derived from our approach. More discussion on VAs_i and U_s can be found at the end of Sect. 2.3.

After the emissions for each sector are derived, total anthropogenic emissions for the i -th hour can be calculated as the sum of emissions from all sectors for that hour. The daily mean top-down anthropogenic emissions are the average of emissions at individual hours.

While a number of interesting sensitivity studies are presented to investigate the robustness of the approach, the most obvious one is not shown: To reduce the a priori emissions by 30% and let the retrieval bring them up again to the original values which were confirmed by the current inversion. Can this be done?

Response: Thanks for your suggestion. This test has been added in the revised manuscript. It leads to top-down anthropogenic emissions of 5.2 (6.4) TgN/yr for Each China (China total), close to our best estimate of 5.5 (6.8) TgN/yr.

In the discussion of uncertainties, the authors emphasize that using the difference of measurements at different times of the day instead of the absolute column cancels many errors in the retrievals. However, I'm not convinced that this actually is an advantageous situation for error propagation for several reasons: 1) taking the difference of two rather similar values will reduce the size of the signal increasing the importance of absolute errors while at the same time introducing two instead of one random error, 2) measurements from two instruments often have systematic biases which are a prob-

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lem in this approach, 3) systematic errors can arise from diurnal changes in satellite sensitivity (clouds, aerosols, BRDF, boundary layer height, NO₂ temperature), and 4) any uncertainties in the modelled diurnal variation of the NO_x lifetime will have a large impact on the retrieved columns.

Response: We do believe that our approach is less susceptible to systematic retrieval errors than the Martin et al. method. By utilizing the difference of the retrievals, it reduces the effect of systematic retrieval errors given that the errors are consistent. Systematic errors in the retrievals are estimated to be the main cause of the fact that top-down emissions derived by the Martin et al., method are too high. As shown in our results and analysis, these errors have a much smaller effect on our calculation. We are fully aware that the two retrievals may have different magnitudes of systematic errors (Sect. 2.1.1, Sect. 5). In addition, random errors may affect the top-down calculation as well. We have included three tests to analyze the effect of such situation to our top-down estimates (Sect. 5). However, it is found that the likely differences in systematic (and random) errors of the two retrievals lead to biases in our top-down emissions by less than 17% (most likely within 10%).

Errors in the NO_x lifetime calculated by TM4 (the CTM for deriving the KNMI retrievals) and other factors would lead to errors in the TM4-modeled vertical profile of NO₂, which would affect the magnitude of retrieved NO₂ column density. It is estimated that such error is about 10% of the retrieved columns (e.g., Martin et al., 2006; Wang et al., 2007). In this study, the averaging kernel is applied to GEOS-Chem simulated NO₂ concentrations at different altitudes to generate the GEOS-Chem columns ‘as seen by the satellite retrievals’. Subsequently, the ratio of the retrieved and the GEOS-Chem modeled NO₂ columns, instead of their absolute values, is used for top-down calculations. Therefore the effect of potential errors in the ‘a priori’ NO₂ profile simulated by TM4 on the top-down emissions is removed, although the retrieved and the GEOS-Chem modeled NO₂ columns may still contain inaccuracies. We have clarified this point throughout the revised manuscript.

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Detailed comments:

p19210 and later: The authors apply the averaging kernels to the modelled columns and state that retrieval errors from inaccuracies in the a priori profile are thereby removed. However, in my opinion the basic uncertainty in the retrieved tropospheric column cannot be removed as the true atmospheric profile of NO₂ is unknown. All you can do is to replace the profile assumptions made in the retrieval by the profile assumptions based on the GEOS-chem model which improves consistency but does not guarantee accurate columns. If GEOS-chem predicts the wrong NO₂ profile, the column and therefore the emission will still be wrong.

Response: As shown before, the averaging kernel is applied to GEOS-Chem simulated NO₂ concentrations at different altitudes for generating the GEOS-Chem columns ‘as seen by the satellite retrievals’. Subsequently, the ratio of the retrieved and the GEOS-Chem modeled NO₂ columns, instead of their absolute values, is used for top-down calculations. Therefore the effect of potential errors in the ‘a priori’ NO₂ profile simulated by TM4 on the top-down emissions is removed, although the retrieved and the GEOS-Chem modeled NO₂ columns may still contain inaccuracies. We have clarified this point throughout the revised manuscript.

P19210: problem in comparing cloud retrieval schemes. While I agree that there is currently no comparable cloud retrieval algorithm available for the two instruments, this could be done in principle as there is no reason why the O₂-O₂ approach should not be applied to GOME-2 data.

Response: We agree that a better consistence between the two retrievals can be achieved by reconciling the current cloud schemes, and we are looking forward to seeing this coming. Meanwhile, this work is out of the scope of the present study. The

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effect of the likely differences in systematic errors between the two retrievals (due to cloud schemes here and many other factors) on our top-down estimates is analyzed in detail in Sect. 5 using three sensitivity tests.

P19212, I12 and following: something is wrong with the sentences in this section, please check

Response: We have re-structured Sect. 2.1.1 to better analyze the retrieval errors.

P19213: This derivation of the method implicitly makes the assumption that NO_x in the complete tropospheric column can be treated as one quantity with one (effective) lifetime. However, this is of course not really true and NO_x in each altitude layer has its own lifetime. Newly emitted NO_x is injected in the lowest layers where NO_x lifetime is short and thereby also changes the effective column lifetime depending on model assumptions on vertical mixing. I think this point deserves some discussion.

Response: Indeed an effective lifetime has been assigned to each column at each hour in this study, as a common practice in such studies (Martin et al., 2003; Jaegle et al., 2005; Martin et al., 2006; Wang et al., 2007). In reality NO_x has different lifetime at different altitudes. However, most of NO_x are located in the PBL where the lifetime is more consistent, especially over China where the amount of NO_x from lightning in the upper troposphere is relative small as compared to the amounts in the U.S. and the tropics due to less deep convection (Lei Murry, personal communication). As in previous studies, assuming an effective lifetime for the whole column here is not thought to be an important source of errors for top-down estimates. For example, a test doubling lightning emissions leads to a 15% decrease of top-down anthropogenic emissions.

P19216 and later: GOME-2 swath is 1960 km, not 1500 km.

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Response: The number has been corrected.

P19218 and 19219 exponent missing for NO2 column value

Response: Corrected. There mistakes were caused by file conversion.

P19221 I find this discussion of the apparent contradiction between model/measurement column differences on the one hand and constant emissions on the other hand unconvincing. The fact alone that emission estimates depend on column amounts in a non-linear way is not an advantage and also does not explain the differences, in particular if there are no changes in emissions relative to the a priori and thus no nonlinearities to be expected. The sentence “That higher retrievals may not lead to higher top-down emissions is a unique characteristic of our approach that is worth highlighting.” in my opinion describes the problem of the approach and not its advantage.

Response: As clarified before, an important cause of the model-retrieval difference is estimated to be systematic retrieval errors. These errors do not affect significantly our top-down estimate, since they are positively correlated and can be canceled partially through the differentiation. Therefore we do believe that our approach is less susceptible to systematic retrieval errors than the Martin et al. method. We have clarified this point the revised manuscript (Sect. 4, Sect. 4.1).

Linking retrievals to top-down emissions in a non-linear way, by itself, may not necessarily be an advantage, and we did not claim that it was without context. However, by employing the difference between retrievals with consistent systematic errors (which consequently leads to the specific non-linear relationship), our approach is less susceptible to systematic retrieval errors.

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P19226 Here the importance of nighttime effects is highlighted again. However, I do not understand how more efficient transport at night could impact on the emission estimates for a large country like China or even for East China. Please explain or remove.

Response: The inclusion of nighttime evolution mainly affects the spatial distribution of top-down emissions when retrievals in the morning are used; while the emission budget over a large area like China is not affected significantly. In addition, the inclusion will not affect significantly the top-down calculation based on OMI and the Martin et al. approach. We have clarified this point and moved related discussion to Sect. 4.1 in the revised manuscript.

Fig. 1: Please add how sampling was done – were the two datasets average independently or were only pixels used where both instruments had measurements on the same day?

Response: The monthly data are average over days when both instruments had measurements. This has been clarified in the revised manuscript.

Fig. 2: it would be good to add the figures for the simulation using the a posteriori emissions also, in particular as they use different soil emissions.

Response: An important cause of the model-retrieval difference is from systematic retrieval errors. Using the a posteriori emissions to drive GEOS-Chem simulations is not expected to reduce the difference significantly, as the retrievals are still biased. Therefore we do not think it is particularly helpful to run the model with a posteriori emissions.

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Fig. 4: Please indicate the time of satellite overpasses in the figure

Response: Done.

Fig. 5: Too small at least in my printout

Response: It seems to be fine with our printer.

Fig. 7: I'd suggest to include the figure with the emissions from this study for direct comparison.

Response: Done (now changed to Fig. 5).

Fig. S1. Corrected tropospheric NO₂ column concentrations (10^{15} molec/cm²) for July 2008 retrieved by (a) GOME-2 and (b) OMI, and corresponding GEOS-Chem simulations in (c) for 10:00am and (d) 2:00pm, respectively. This figure is the same as Fig. 2, except that GOME-2 and OMI retrievals in (a-b) are reduced by 32% and 15%, respectively.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 19205, 2009.

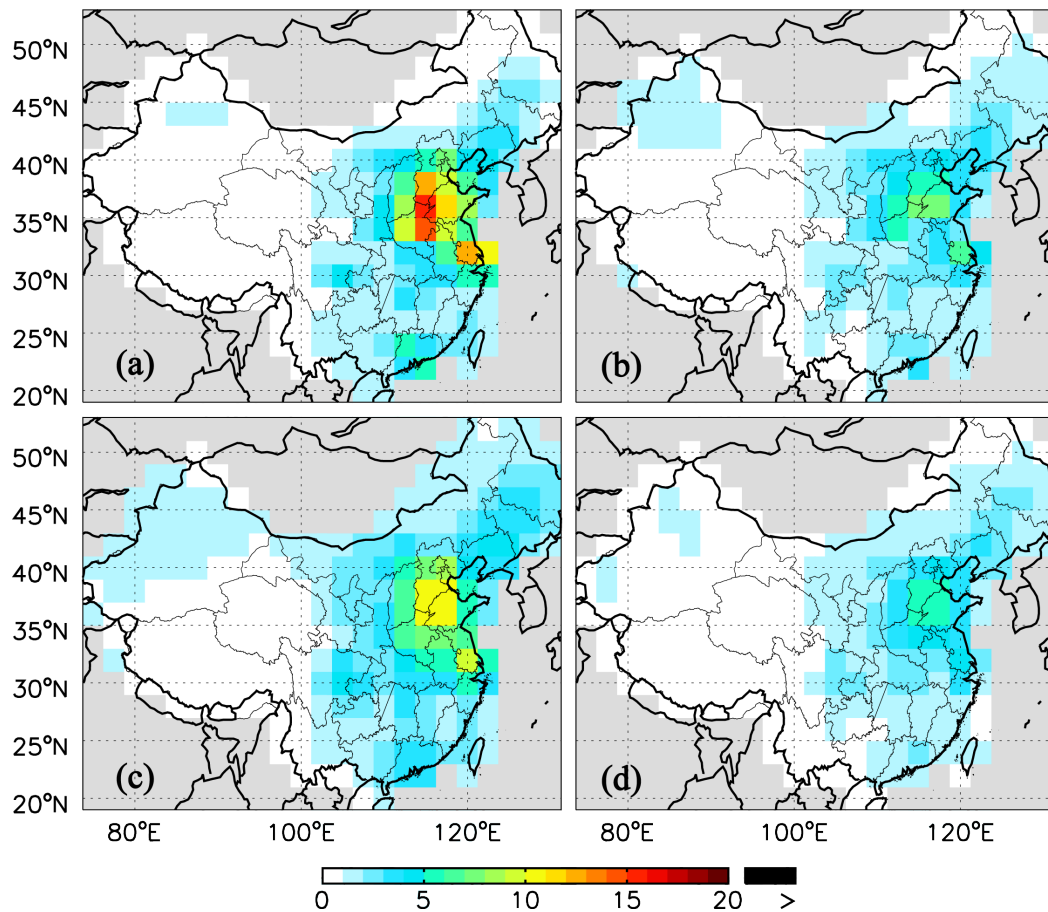
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