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Atmospheric
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Discussions

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.

J.-T. Lin et al.

jlin5@seas.harvard.edu

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This paper makes a valuable contribution to estimating NOx emissions using multiple satellite retrievals. A new methodology is developed to combine tropospheric NO2 column retrievals from GOME-2 and OMI. The algorithmic development is laudable. Nonetheless major issues need to be resolved prior to publication 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’ NOx 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 NOx 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).

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The comparison in Figure 2 indicates a large difference between the retrieved and simulated tropospheric NO2 column concentrations. This difference needs to be better explained. The conclusion that a priori bottom-up emissions are basically correct is surprising in spite of this difference.

Response: An important cause of the difference is the probable positive bias in both retrievals. 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 ~2.5x10ˆ15 molec/cm2 (By Bremen University) to ~5.1 x10ˆ15 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 ~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.

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As discussed in Sect. 2.1.1, systematic errors in the GOME-2 and OMI retrievals are expected to correlate positively with each other, since the two retrievals are derived with a very similar methodology (same stratospheric correction, NO2 profiles from the same TM4 model, cloud corrections based on same assumptions, and same radiative transfer model to calculate AMFs). An analysis of spatiotemporal correlation shows that the monthly mean NO2 VCD has a large spatial correlation between GOME-2 and OMI, with a R2 of 0.81 over East China (Fig. 1b). Additionally, the regional mean VCDs over East China from GOME-2 and OMI vary temporally with each other with a day-to-day correlation coefficient of 0.74 for July 2008 (Fig. 3a).

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 mode-retrieval difference shown in Figure 2. Also, potential systematic errors in model simulations and emissions may contribute to the model-retrieval difference. The text has been revised to reflect this point (Sect. 3).

We have re-structured the manuscript to better elaborate the retrieval errors and their impacts for both inverse modeling approaches.

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A possible explanation for the model-satellite discrepancy in Figure 2 is implied by criticizing the Martin et al. method and speculating that nighttime evolution of NOx impacts that method. Is there any direct evidence that nighttime evolution of NOx causes the bias in Figure 2?

Response: We don't believe the nighttime evolution of NOx is an important factor for the difference in Figure 2. 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. This issue has been clarified.

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What may be happening is that systematic errors in the OMI and GOME-2 retrievals contribute to the discrepancy in Figure 2, but some of those errors cancel in the Lin et al. method. A more formal calculation of the error in the difference between the two retrievals could elucidate that effect. As written it is concerning that the expected errors in the top-down estimate are smaller than the expected errors in the satellite retrievals.

Response: We agree. More analyses of retrieval errors have been included in Sect. 2.1.1. Also, we have re-structured the manuscript to better elaborate the retrieval errors and their impacts for both inverse modeling approaches.

The systematic error in the difference between the two retrievals could be inferred from case 6-8. Assuming a systematic error of 32% for GOME-2 and 15% for OMI, the systematic error in the difference between the two retrievals could be estimated by comparing the best estimate with case 7 (where GOME-2 and OMI are reduced by the corresponding amounts for purposes of top-down calculation). Case 7 suggests a budget of 7.3 (5.8) TgN/yr for China (East China), in good agreement with the best estimate of 6.8 (5.5) TgN/yr. This test alone would suggest the systematic error in the difference between the two retrievals is relatively small. Overall, cases 6-8 suggest that the systematic error in the difference between the two retrievals is likely at a magnitude of 17% or less, which consequently results in a likely underestimate by the same amount in our best top-down estimate. More detailed discussions on the effect of systematic errors on our top-down estimate are presented in Sect. 5.

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The retrieved tropospheric NO2 column concentrations are higher than the simulated values for East China. Yet the top-down estimate for East China is actually lower than the prior emission budget. The description in section 4 implies that the most important cause of this discrepancy in sign is that the top-down estimate is based on changes in the retrieved columns. Thus more confidence is placed in the difference in retrievals

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between the two instruments than in the retrieval from either instrument alone. This may offer reduction in systematic errors that are common to the two instruments, but other errors arise in the comparison of two different instruments. The authors could consider reformulating the methodology to use more information from the absolute observations from each instrument, in addition to their difference.

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Response: Particularly for KNMI retrievals which most likely contain consistent and positive systematic errors, our approach provides an important improvement over the Martin et al. method by analyzing the difference between retrievals such that top-down emissions can be less susceptible to such systematic errors. A key factor here is that the systematic retrieval errors be consistent. Inconsistent errors may lead to biases based on our approach. We include a detailed discussion for this situation in Sect. 5, which suggests that inconsistencies in spectral fitting, cloud algorithms, and albedo effects may lead to top-down emissions underestimated by $\leq$17% (mostly likely $\leq$10%). We are considering revising the methodology to deal with such situation, which will be the topic of future papers.

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Table 1 should contain a more complete description of level-2 NO2 retrievals: such as spectral window, cloud parameters, and surface reflectivity.

Response: Done.

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The discussion of retrieval errors includes many important topics. The use of the same surface reflectivity database for both satellites should be added. This is concerning due to different spectral windows in the retrievals, diurnal variation of surface reflectivity, and BRDF effects which vary with sun-satellite geometry.

Response: The use of the same surface reflectivity database for both satellites has

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been included. Currently the same albedo is being used in GOME-2 and OMI retrievals (TOMS/GOME surface reflectivity). To some extent this is preferable, because the surface reflectivity has been derived under the same assumptions, from the same satellite instruments, at the same wavelengths and the same spatial resolution. On the other hand some uncertainties arise as the TOMS/GOME set holds for a mid-morning viewing geometry, not necessarily valid for OMI. The KNMI-team is currently implementing the OMI-derived surface reflectivity [Kleipool et al., 2009], which would constitute an obvious improvement for OMI retrievals, but not necessarily improve the consistency between GOME-2 and OMI, because the OMI surface reflectivity database has a higher spatial resolution and is not based on a minimum Lambertian Effective Reflectivity approach (like TOMS/GOME) but rather on the mode of reflectivities. A detailed comparison of mid-morning and early afternoon reflectivities, their spatial resolutions, and methods would be recommended, but is beyond the scope of this study.

The effect of inconsistency in systematic errors due to various factors (spectral window, etc.) has been analyzed in Sect. 5.

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The Lin et al. method depends on accurately modelling the growth of the PBL depth in morning to represent the diurnal variation in the NO2 column. Please discuss.

Response: In this study, the non-local scheme by Holtslag and Boville (1993) is adopted by GEOS-Chem to calculate the temporal variation of PBL mixing. As shown in Sect. 2.1 of the revised manuscript, this scheme has been shown to simulate fairly well the vertical distributions of NOx and ozone in the lower troposphere (Lin et al., 2009) and the diurnal variation of surface ozone concentrations over the U.S. (Lin et al., 2008b; 2009). However, it may still result in some errors in the modeled PBL mixing (due to errors in meteorological fields, etc.). The effect of errors in the PBL mixing is estimated to be 4% for July 2008, as discussed in Sect. 5.

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The different cloud pressures reported for the different products (FRESCO & O2-O2) are concerning if two retrievals are being compared. Whether the cloud is reported in the PBL or just above could have a systematic effect on the inferred diurnal variation. Please discuss.

Response: We agree that the cloud pressures differ between the two cloud schemes. Boersma et al. (2007) found that cloud pressure relative to OMI (using the O2-O2 band) is ∼60 hPa larger than that for SCIAMACHY (using the FRESCO scheme), since the O2-O2 band is more sensitive to the lower troposphere. For GOME-2, errors in cloud pressure are estimated to contribute only ∼2% to the VCD error, since the cloud top is typically above the lowest troposphere where NO2 concentrates. For OMI, clouds are lower and closer to the polluted layer, and errors in cloud pressure can lead to ∼15% error in the VCD retrieval. This discussion has been included in the manuscript.

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How is the averaging kernel treated? Does Omega_r continue to depend on the TM4 NO2 profile? If it does, that could be a source of systematic error in Figure 2 and in the application of the Martin et al. method used for comparison.

Response: We have shown in the manuscript that the averaging kernel is applied to the model NO2 column the remove the effect of a priori profile on top-down emissions (Sect. 2.1.1, Sect. 2.3).

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Sec. 2.3. What is the diurnal variation in tau_a? A plot would be helpful.

Response: As shown in the revised manuscript (Sect. 2.3), the lifetime ranges from 3 – 5 hours during 10:00am-2:00pm, depending on location and time.

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How is tau_a actually calculated? Eq. 2 does not clearly explain. Loss of NOx to PAN could lead to an underestimate of effective tau_a if the PAN rapidly regenerates NOx. It may be better to treat NOx and PAN as a chemical family.

Response: As shown in the revised manuscript (Sect. 2.3), the lifetime is derived for every hour by applying Eq. (2) to GEOS-Chem modeled NOx column densities and emissions for each hour.

The lifetime here is the 'effective' lifetime of NOx in the column, as a result of all chemical and physical processes in the column, including the NOx-PAN conversion. However, during 10:00am-2:00pm, the loss of NOx is mainly through the formation of HNO3. Combining NOx and PAN as a family is not thought to be appropriate because the lifetime of PAN is much longer than NOx so that it can be transported to longer distance, in violation of our assumption of negligible horizontal transport.

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Is tau the NOx lifetime in the column? Or something else?

Response: tau is indeed the 'effective' lifetime of NOx in the column. This is clarified in the revised manuscript.

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Sec 2.4 implies that the Lin et al. method does not require assumptions on emission diurnal variation. Page 19216, l12 indicates that the Lin et al. method assumes diurnal variation for 20 hours of the day. The Lin et al. method only determines diurnal variation for 4 hours of the day. Please clarify in sec 2.4.

Response: We have clarified this point. In addition, Sect. 2.4 has been changed to Sect. 4.1.

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The close agreement between the top-down and prior bottom-up emission estimates for China is presented as evidence for success of the Lin et al. method. Comparisons over the US or Europe would be more convincing since the bottom-up emissions are better known there.

Response: We did not have enough information (e.g., emissions, diurnal variations and uncertainties for recent years) for the U.S. and Europe at the time this study was conducted. We plan to analyze these two regions in the future. Nonetheless, based on our communications with experts on Chinese emissions of NOx (Qiang Zhang, Yu Zhao, Yu Lei and scientists at Tsinghua University) we are pretty confident with our estimate for emissions of NOx in China.

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Abstract, l17-20: Errors in the inversion are implied to be <15%. In fact, each sensitivity test addresses a source of error. Their combination should be presented here and in the conclusions.

Response: We have clarified the abstract and conclusion that each factor for model/methodology errors typically contributes to errors in top-down emissions by less than 15%. As discussed in Sect. 5 for Figure 8, the combined effect of those errors is difficult to estimate, as they may or may not be independent.

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Abstract: 9:30 and 1:30 are close to the equator crossing times. The times for China should be used here and on page 19216. They may be closer to 10:00 and 1:00.

Response: Changed.

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p19226, l23: Is horizontal transport really neglected? Doesn't GEOS-Chem account for transport? What about NOx from lightning?

Response: GEOS-Chem accounts for horizontal transport, while the top-down formulation does not. This small difference is expected to have a negligible effect on top-down estimates as NOx is destroyed quickly by photochemistry during 10:00am-2:00pm preventing it from being transported to a long distance relative to the size of the gridcell here (2x2.5 degree).

NOx from lightning in the upper troposphere has longer lifetime than NOx in the PBL. However, the amount of NOx from lightning is relatively small as compared to NOx emitted at the surface, particularly for China. (Over the U.S. and the tropics the contributions of lightning become more important due to stronger convection and weaker surface NOx sources. Lee Murray, personal communication.) Our test suggests that doubling lightning emissions over China only lead to 15% reduction in top-down anthropogenic emissions.

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P19217, l16, add “our” before GEOS-Chem

Response: Done.

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P19218, l25, 2x10 molec/cm2 ???

Response: It should read 2x10ˆ16 molec/cm2. The error was due to file conversion.

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P19219, l2, check number 10 molec/cm2

Response: It should read 10ˆ15 molec/cm2. The error was due to file conversion.

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Fig. S1. Corrected tropospheric NO2 column concentrations (10ˆ15 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.

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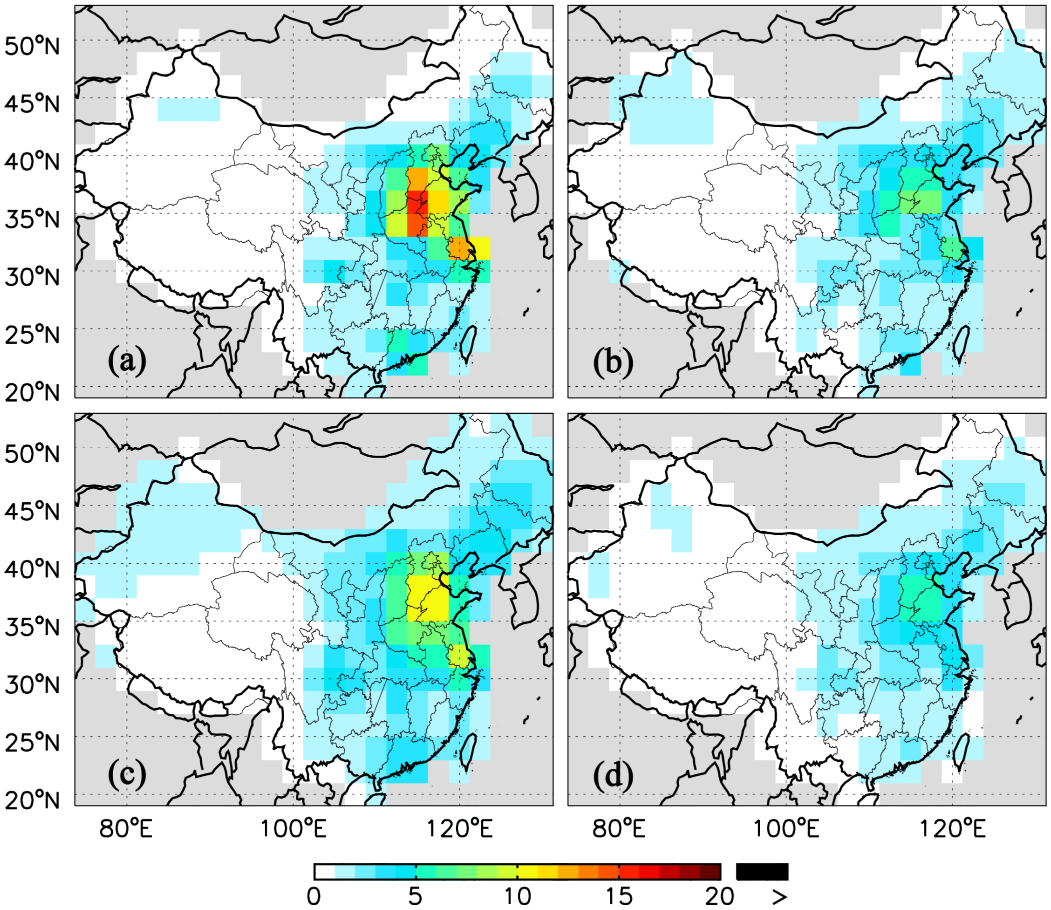

Fig. 1. Fig. S1.

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