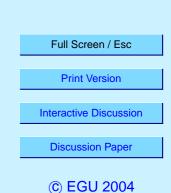**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Using GOME NO$_2$ satellite data to examine regional differences in TOMCAT model performance" *by* N. H. Savage et al.

N. H. Savage et al.

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We agree with many of remarks from both referees. I will reply to each point individually but I wish to point out that this was a first attempt at using satellite data for validation of the TOMCAT model. In contrast several of the major general criticisms made by these referees would require separate studies in themselves, which we consider to be beyond the scope of this paper. Clearly this manuscript is in an emerging area of research i.e. the use of global remote sensing data with global models. It is an area where much work remains to be done. However, without studies such as that presented in this manuscript, it is not possible to proceed further. This issue has now been addressed in the introduction. In addition in the limited time available to reply to referees comments it is not possible to do much in the way of additional model runs or analysis. Despite this, major changes have been made both to the structure and the

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content of the manuscript and we have addressed as many of the referees comments as possible.

H. Eskes

General comments

1) "Quantifying uncertainties related to aspects of the transport and chemistry would be very valuable. I realise that this is non-trivial and often beyond the scope of this paper. However, rough error estimates are possible for some aspects, e.g. the photolysis rate difference between clear sky and a climatological cloud cover can be quantified."

The issue of quantifying model uncertanties is a very important one which one which there is too little work in general. However, even for the photolysis rates this is not a simple issue. Although it is comparatively simple to calculate the change in the photolysis rates at a given location with and without climatological cloud, it would be necessary to find the change in the total NO2 column with and without the cloud and thus requires a sensitivity study of some kind.

2) "The paper presents inter-comparisons of monthly means. A better approach is the comparison of individual pixels with collocated model values (which can then be presented in a monthly-averaged way). This is especially important for time-varying aspects such as the outflow over the oceans which is strongly dependent on the changing wind direction and convection. Because the number of GOME observations in the monthly mean is often small and far from uniformly distributed in time (depending on the clouds) this sampling issue may introduce considerable differences between model and GOME monthly averages. The authors note that such a space-time sampling of the model at the observation locations would be valuable. Therefore it is not clear to me why the authors have not chosen this approach."

Although we recognize the need for space time sampling, there are some practical constraints. Space-time sampling of the model was not employed in this study because

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it was not possible given the human and computer resources available. Such an approach would require a substantial amount of time to write and test a program to read in the information on the positions of GOME data, interpolate the model to the points in space and time where the observations are made and output them. In addition this would generate far larger output files requiring more work to interpret these files. This approach would also cause increases in model run time and storage requirements. Clearly this is a desirable next step but it was not possible at the current time.

3) "One step further is the inclusion of the averaging kernels in the comparison to also account for the vertical sensitivity of the measurements."

The GOME DOAS retrieval used here does not generate averaging kernals. We consider the determination of the slant column to be a well defined and constrained mathematical problem. The derivation of air mass factors relies on knowledge of the surface spectral reflectance, molecular and particle scattering. This requires some apriori assumptions, which have been explained briefly in this article and in depth elsewhere.

4) "The article tends to attribute the model-GOME differences to model shortcomings. It is important to stress that also the GOME retrievals are characterised by uncertainties that are not small."

This was indeed a major weakness in the previous version of the manuscript so the emphasis has now been changed.

Specific comments

1) Abstract: "... modelled columns are too large ... " Should this be replaced by " ... modelled columns are larger that the GOME retrievals ..."? In other words: is it justified to attribute the difference to TOMCAT, and is the error bar of the GOME retrieval (systematic error) significantly smaller than the difference observed?

The abstract has been changed as suggested

2) 2572, l9: " ... this might reduce OH concentrations thus reducing the lifetime of

NOx." Please explain the mechanism here. What about the direct reaction with OH to form HNO3?

This was a mistake and has been corrected to read "thus increasing the lifetime of NOx."

3) 2574, bottom: It is mentioned that daily NO2 fields of SLIMCAT are used. I assume that these are daily-average fields instead of local-time fields. One important point here is that day-time NO2 has quite a different profile than night-time NO2 in the stratosphere, the latter peaking at higher altitudes. Since dynamics-related features are strongly height-dependent this may introduce error patterns in the troposphere as well. Please provide a remark on this.

The daily NO2 fields from SLIMCAT used to correct for the longitudinal variation of the stratospheric NO2 fields are model output for the time of GOME overpass. At high latitudes, the satellite has several overpasses per day, and this is not taken into account. However, for the latitude range used here (+/- 60 degrees), the difference between model time and satellite overpass is small. This information has been added to the text.

4) 2575, l15: Tomcat profiles are used to calculate the air-mass factor: " .. the vertical profile of NO2 used is taken from the daily results of the TOMCAT model run .. ". Please provide more details how this is done. Does this mean the AMF is calculated for each individual GOME pixel separately based on a collocated model profile? Or does it mean something else?

TOMCAT profiles have been used for the airmass factor calculations in the following way: For each day, the NO2 profile for each grid cell has been used to derive an airmass factor using the surface elevation, surface reflectivity, and aerosol type for this cell. The GOME data have then been converted from slant to vertical tropospheric columns using the airmass factor of the cell closest to the centre of the GOME pixel.

5) 2575, l27: " .. data were selected to be cloud free .." Which cloud algorithm is used here?

The cloud algorithm used is a simple threshold algorithm based on the average intensity of the spectral region used for the NO2 retrieval (425 - 450 nm). Bright surfaces such as ice and snow are also flagged as cloudy.

6) I assume there is no cloud correction applied in the retrieval for cloud fractions between 0 and 10%. Please state that this is the case.

This is correct and has been added to the text.

7) 2576: Please provide more information how TOMCAT describes the daily cycle. E.g. discuss the development of the boundary layer in the morning, the use of meteo (on 6h basis?), and the daily variation of emissions (if present)?

This has now been added to the manuscript.

8) 2579, l25 and 2580, l3: It is mentioned that Velders and Lauer find model values that are much higher compared to GOME. Is this due to differences between the models, or to differences between the GOME retrievals? Please provide some information on the differences in the GOME data sets. How large are these? A brief summary of retrieval details would be interesting as well. How do the models inter-compare (on average).

We agree that it is would be very valuable to compare the retrievals and the models in the manner suggested. However, it is very difficult to make an assessment of how model results or observations compare without access to both sets of data. Simply comparing plots of data can be very misleading due to differences in contour levels etc. It would indeed be worth comparing multiple models and GOME retrievals, However that is a major exercise and beyond the scope of this paper.

9) 2580: It is not very useful to have forward referencing to sections 4.3 and 4.4 - please remove and discuss aspects at one place only.

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Deleted

10) Table 2: The authors may consider to provide a map to replace the table. This will be helpful for the reader.

Done

11) Table 3 caption: mention if gradient is the ratio of observation divided by model, or model divided by observation.

Done

12) Fig 4: text in figure mentions June 1999 instead of June 1997 !?

This error has been corrected.

13) The large seasonality of the comparison over Europe is quite mysterious to me!?

To us as well! This is one of several areas where a whole study is deserved, ideally with NO2 profile data and other models.

14) Emissions should be well characterised here I would think.

I agree

15) Convection is mentioned as one explanation, however this is not fully convincing to me. I would challenge the authors to address this point in more detail. Can there also be retrieval aspects playing a role? Please provide a more extensive discussion of the comparisons between models concerning the seasonal cycle over Europe, perhaps including results of regional models.

This is a complex area. Part of the issue may be the relationship between retrieval and modelled NO2 profile. Other issues are numerous. The reason for suggesting convection in particular is that, as described in the manuscript, we believe that the model-GOME comparison is particularly sensitive to the way in which the model distributes NO2 in the vertical. To investigate this further would require a paper in itself with de-

tailed model validation against NO2 profiles. However the amount of NO2 profile data is itself very limited and this is again beyond the scope of this paper. A discussion of the comparisons between models would require access to other modelled \chem{NO_2} columns, permission to use these and a much broader collaboration. While this would be a very worthwhile study we do not currently have these data or the resources to conduct such a comparison. The suggestion of using regional as well as global model data is certainly a valuable and will be considered for the future.

16) Fig 6 and fig 7: It looks like TOMCAT has negative model columns ?! Is this true?

Yes. An explanation of why has now been added to the model processing section. The model has been processed in the same way as the retrieval - a 'clean sector mean' has been subtracted from all data. This means that where the model has a lower concentration than in the clean sector the calculated column is negative.

17) Sec 4.3: The long-range transport comparison may be misleading due to availability of a limited number of cloud-free GOME pixels. As mentioned above, a better approach is a comparison of equal place/time collocated model with GOME. For studies of export over the Atlantic ocean this may be crucial, especially when outflow and cloud occurrence is somehow correlated. Please add a remark on the additional uncertainty that is introduced by the (monthly-mean) inter-comparison approach.

Done

18) 2587: Discussion of retrieval errors. Although I do not like to promote our own work, I think a reference to Boersma et al. (JGR 109, doi:10.1029/2003JD003962, 2004) would be appropriate here.

Yes, it is. We have included it in the text and added a few sentences.

19) 2587, bottom: Good point. There are relations between clouds, frontal systems and the absence of GOME observations. This shows again that comparing individual measurements with model values collocated in space and time is more convincing.

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I agree but see above comments about the reasons why we were unable to do this.

20) 5.3.1, horizontal transport. I do not understand how fig 10 proves that horizontal transport will not be the main source of error. Please explain or weaken the conclusion.

I have added an additional sentence of explanation and weakened the conclusion.

Anonymous Referee 1

Major remarks

1) "The authors introduced an updated version of the TEM by taking into account the horizontal pattern of stratospheric ozone, based on results from the SLIMCAT model. I think it would be worth investigating the effect of such an approach and to quantify the differences. This could also become an important contribution to this paper (if proven to enhance the quality of the TEM) and should be mentioned then in the abstract."

As the focus of this paper is on the comparison of a Chemistry-Transport model to GOME data it is not appropriate to discuss improvements in the retrieval here. This would be dealt with more appropriately in a separate paper.

2) "There is a principle problem in the discussion of the results. The authors mention that there are uncertainties in the absolute values of the NO_2 GOME columns. This has to be taken into account already during the discussion of the results (see 3 "+" below): + Differences between Lat x Lon plots from GOME and Model data should be compared to GOME uncertainties, to see where differences are significant (Fig. 1-3). + The correlations should take into account a range of GOME data, instead of a single value, which then would lead to ranges of gradients, which then perhaps are not anymore significantly different from 1 ? (Fig. 4,6,7) + Using an appropriate test should prove, which of the differences in the seasonal cycle are really significant. (Fig. 5 and 9). Taking only the variability in the area into account already leads to the impression that the differences are hardly significant. (e.g. mean GOME data are only 1 std. dev. lower than model data, An additional GOME uncertainty would probably lead to no

significant differences in Fig. 5. A point-by-point inter-comparison would then help to minimise the variability. "

Difference plots have now been added to figure 1-3 and also minimum and maximum estimates for the gradients calculated. More work has been done throughout the manuscript on the issue of the significance of differences between the model and GOME data. See text for more details.

3) "The GOME data rely on TOMCAT NO2 profiles, which has been discussed in the discussion. This is a crucial point and has to be clarified and quantified in more detail. E.g. How much changes the GOME column when different TOMCAT vertical profiles would be taken into account. This has to be included in the uncertainty discussion and should be included in the part satellite data."

We thank the reviewer for this comment and this issue is now referred to in the satellite section.

4) "The comparison concentrates on the year 1997 only although more satellite and model data are available. The authors are aware of the speciality of this year, namely the El Nino event, which has a large impact on the meteorology and biomass burning. Two points have to be mentioned: (i) How representative are those results for other "normal" years? (ii) What impact has the difference in the biomass burning patterns between the emissions data used in the model and in reality on the findings of the comparison? I suggest to extend the comparison."

The reviewer is absolutely correct that issues of inter-annual variability of biomass burning and other processes are very important ones which cannot be addressed without a multi-annual integration of the model. However to achieve this the model would have to be run again for the whole period of interest with the GOME extraction routine turned on. Also the retrieval using TOMCAT data to calculate AMFs would have to be done for all years of interest and this has not yet been done. It is not possible achieve all this within the time limits for replying to referees comments.

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5) - Some parts of the interpretation of the differences are separated without further reason. E.g. the discussion of the radon simulations would help to understand the findings in section 4.

The radon section has been moved and the whole results section restructured.

Specific Remarks:

1) Abstract: Last sentence is very unspecific. And in principle I think this statement is correct. However, I do not see in this case what kind of insights are gained by the intercomparison, nor that a specific model deficiency has been identified and corrected.

This sentence has been deleted.

2) Introduction: 2572/8-10 Why does a OH reduction reduce the NOx lifetime?

This was a mistake and has been corrected.

Satellite data:

3) 2574/25ff As shown by the various references the NO2 data are very valuable for modelling groups. There has always been a discussion on the quality of the TEM data. It seems that the method described here improves the methodology. This should be quantified and discussed in this section. If the quality of this satellite product has been significantly increased this would be a nice outcome of the paper and also mentioned in the abstract.

See above.

4) 2574-2575: Nothing is said about the uncertainties of the satellite derived tropospheric columns. How good does it pick-up surface NO2, or is there a saturation effect? Is it possible to discuss some sort of error bars?

This information has now been added to the manuscript

5) 2575/15 What is the reason for choosing the year 1997 only? As far as I know GOME

data, SLIMCAT data and TOMCAT data exist for more than only this year. Taking several years into account would also give the opportunity to test the robustness of the results. Moreover 1997 was an El Nino year, which may limit the validity of the results, which also has been indicated by the authors in the following section. I suggest to expand the analysis to more years.

As referred to above this study was a preliminary one. This year was chosen because this study was part of the POET project and 1997 was the year which this study concentrated on. As mentioned before while a multiannual study would be very valuable it was not possible to achieve this within the framework of this paper.

TOMCAT model

6) Radon 2577/3 only short-range transport! (convection)

I do not understand this comment.

Processing

7) Table 1: Figure caption should be clearer. Correlation of what? How is the gradient defined? d(model)/d(satellite)? ....

Done

Results:

8) General remark: I would like to see some uncertainty/error estimate for the satellite data and difference plots for GOME-Model. These two information would give a better insight on the quality of the model, e.g. in cases where the GOME uncertainty is larger than the difference one cannot conclude that the model is doing a bad job. This also affects the correlation plots!!

There are now uncertainty estimates included in the satellite section.

9) 2579/12 the statement is too general. Please specify what you mean with generally

good agreement? It would be much easier to evaluate the quality if a difference plot would be included.

This sentence now reads "The modelled peak column densities are similar to the GOME column densities and are located in the same regions."

10) 2579/21 I am not sure how well the GOME data may resolve this feature when plotting them on the TOMCAT grid! Again a difference plot would help!

Difference plots are now included and it can indeed be seen that the difference here is small when plotted on the TOMCAT grid.

11) 2580/7ff The satellite data do not allow for directly measure emissions. Please rephrase the passages.

Done

12) One can see elevated NO_2, which is likely to result from biomass burning. What is the role of El Nino for biomass burning and how well are the regional displacement of biomass burning areas caused by El Nino simulated by the model? (or included in the emission data)? Here additional years would help to eliminate the El Nino effect.

Again to address this would require a multi-annual model integration. See above for remarks on this.

Correlations

13) 2581/15ff Since nothing is said about the GOME data quality the results are difficult to interpret. Winter in Europe may be dominated by low level clouds, which may reduce the number of measurements dramatically. If only data from a few days are available, then the results are hardly significant. In summer high NO_2 surface concentrations may not be seen by GOME due to saturation effects, which would bias the results to higher gradients.

a) lack of data in winter: This is certainly an issue, and there is nothing we can do

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about it. As weather in Europe is often cloudy in winter, few data points are available which reduces the representativeness of the data set. This is now referred to in the discussion.

b) saturation effects in summer: What exactly does the reviewer mean with saturation effects? There is a potential for underestimation of near surface NO2 under high aerosol loading situations, but we tried to account for this through the choice of aerosol type in the analysis.

14) 2581/23ff I would interpret Fig. 5 exactly the other way round! GOME data are in most cases within 1 std.dev. of the TOMCAT data, which rises the question whether the results are really statistically significantly different? And the bars are assuming the correctness of the GOME data! Taking some uncertainty of the measurements in addition into account a conclusion from this figure could be difficult.

These issues are now addressed. See text.

15) 2584/5 This information should be given in the satellite section, but in more detail, since it strongly influences the quality of the conclusion drawn from the intercomparison!

Moved.

16) 2584/7 This passage is confusing. The par starts with Figure 7 (January only), but then discusses all months, can one see that from the figure?

I have made this clearer. The plume can be seen in the global plots for January very clearly and to a certain extent in July and September as well.

17) 2584/13-15 NO2 columns are a product of different processes: emissions, chemistry, transport. How can one deduce from the fact that the modelled columns are similar to the GOME data that the emissions are correct. Why couldn`t it be that the emissions are way off but the chemistry and transport balance this? The discussion makes it quite clear that additional information on the export or chemistry is needed to

better interpret the results. Perhaps MOPITT CO figures could help?

I have weakened conclusions about the emissions. It would indeed be useful to compare to MOPITT data but due to issues related to the retrieval such as averaging kernels this would require another paper to make sensible use of this data. It would certainly be valuable to combine different satellite data products in the future.

18) 2585/5 approximately 75% of the No2 could be explained as far as I understood from this additional experiment. However, the huge reduction in the emissions nonlinearly affects the O3-Nox chemistry. Ozone production is probably totally reduced. So that also OH production is inhibited, which affects the NO2 lifetime.

The non-linearity is potentially a problem here, however if the OH concentration is reduced then in situ emissions would have a longer lifetime and so contributing a larger column in the middle of the Atlantic. This is now mentioned in the text

19) 2585/10 include the Figures you are addressing

done

20) 2585/13 I do not see the Asian plume. Please specify.

This reference to the Asian plume has been deleted as it was decided not to focus on it.

21) 2585/13ff Another possibility would be a too fast vertical transport of NO2, so that No2 is transported mainly at low levels to the W and in the model at higher levels to the E. In this case the African column wouldn't`t be different but NOx is transported away at different altitudes and therefore different directions. Here CO comparisons would lead to more insights. If CO also shows a plume in the GOME data. The NO2 plume is likely to arise from burnings.

This possibility is now refereed to in the text. CO is not available from GOME. See above for issues with using MOPITT data.

22) Figure 9 + 2586/10ff: Taking into account the std. dev. I think one hardly can derive a statistical significant difference in the seasonal cycle! Again more data would help!

This section has now been modified

23) Since a lot of emphasis is given to biomass burning, it would be helpful to discuss in more detail the used emission database in comparison to, e.g. ASTR fire counts, although of course from fire counts one cannot derive emissions directly, but it would give a better basis for the discussion of regional pattern and seasonal cycle.

The emissions description section now contains 2 plots of ATSR firecount data.

Discussion

24) 2587 Most of the information is already needed in the introduction of the satellite data and for the interpretation of the inter-comparison, to avoid mis-interpretations as discussed above.

This section has now been moved.

25) 2588/6-10 This point should be clarified better. Assuming correct emissions and problems in the transport, why should than be the concentration correct? Or in other words, why a gradient of 1 a indication for correct emissions given that you have discussed problems with the transport?

This section is now rewritten

26) 2588/11 I am not quite sure about this, because air conditioning consume a huge amount of energy, especially in N: America.

You are correct, as the next sentence states, the seasonal variation in industrial emissions is very small.

Finally we would like to thank both reviewers for their useful comments.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2569, 2004.