Atmos. Chem. Phys. Discuss., 9, C3194–C3206, 2009
www.atmos-chem-phys-discuss.net/9/C3194/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of tropospheric ozone columns derived from assimilated GOME ozone profile observations" *by* A. T. J. de Laat et al.

J. de Laat

laatdej@knmi.nl

Received and published: 23 July 2009

Response: We thank the referee for the constructive comments, and have prepared a detailed response.

Review of "Evaluation of tropospheric ozone columns derived from assimilated GOME ozone profile observations" by de Laat, van der A, and van Weele.

This manuscript describes the TORA method for producing tropospheric ozone column (TOC) estimates and presents results based on GOME ozone measurements from 1996-2001. This method uses a chemical transport model with analyzed winds and parameterized ozone photochemistry to produce stratospheric ozone profiles that,

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

when subtracted from a total ozone column observation, yield a residual tropospheric ozone column estimate. The chemical transport model is operated both in a "free running" mode and an assimilation mode whereby GOME ozone profile observations are assimilated using a Kalman filter technique. Time series of TORA TOC values are compared with independent ozone sonde measurements over a 6-year period from 1996-2001 when GOME profiles are of sufficient quality. The authors report that assimilation of GOME ozone profiles improves both TORA TOC and UT/LS estimates in the tropics but degrades TORA performance in the mid-latitudes. Overall the subject is appropriate for ACP. Tools for monitoring and analysis of global TOC are greatly needed, particularly as new remote sensing data sets become available.

However, I have several concerns regarding the basic methodology, the presentation of results, and the conclusions drawn from these results that should be addressed before I can recommend publication in ACP. I have listed each of these below, followed by a list of minor comments and corrections.

Comment: First, the basic approach and methodology presented in this article both seem a bit outdated. Aren't there more sophisticated approaches available for assimilating global total ozone and profile measurements that have been shown to be quite effective (e.g., Schoeberl et al., 2007; Stajner et al., 2008)? What does TORA offer that will be better for monitoring TOC than other methods?

Response: Schoeberl et al. [2007] and Stajner et al. [2008] both use stratospheric ozone profiles from MLS observations and total ozone from OMI. Schoeberl et al. [2007] assimilates the ozone profiles using a trajectory-based approach (rather than a transport model) and then subtracts the assimilated stratospheric ozone column to determine a tropospheric residual (like the TORA methodology). Stajner et al. [2008] assimilates both MLS ozone profiles and OMI total ozone columns, using a 3-D transport model and a linearized ozone chemistry scheme (like used for the TORA method) as well as a similar Kalman-filter type assimilation procedure. However, in their linearized ozone chemistry scheme they use "24-hour mean production, loss and depo-

ACPD
9, C3194–C3206, 2009

sition rates (PLD rates) from a full chemistry-transport model (GEOS-4, v 7.04)". They then analyze the tropospheric ozone column from the assimilation run – not a residual – which is different from TORA. In our simulations we do not update tropospheric ozone productions and loss rates, as we are predominantly interested in the assimilation of the stratospheric part, not the tropospheric part. The Stajner et al. [2008] comparison with ozone sonde observations (their figure 10) shows that the synoptic scale variability is already captured by the standard model without assimilation, a consequence of using the linearized ozone chemistry with updated and realistic tropospheric ozone tendencies. Unsurprisingly, as much variability in tropospheric ozone is related to dynamical processes of stratosphere-troposphere exchange, which is well captured by the linearized ozone chemistry (see our paper and de Laat et al. [2006] as well). The improvement by including the assimilation of ozone observations leads to somewhat ambiguous results. For certain locations biases improve, but not everywhere. RMS differences and correlations improve generally but the changes in correlations are modest for most locations as the model without assimilation already does a good job in reproducing ozone sonde observations. All in all, the TORA method lies in between the methods used by Schoeberl et al. [2007] and Stajner et al. [2008] as it uses a residual method with assimilation of ozone observations in a chemistry-transport model. However, we do use a different type of observations, with different sampling compared to MLS – ozone profiles from nadir UV/VIS spectra. We don't pretend that our method is better, rather that it is different using different type of observations. These type of ozone profile observations are available and will continue to become available, guaranteeing the creation of long time series. It is therefore important to use these observations and develop methods for assimilating them – even if their vertical resolution is not as good as that of MLS ozone profiles. Future efforts will start to aim at assimilating many different observations – limb, total ozone, ozone profiles from UV/VIS and IR – and this study helps in improving our understanding of what happens when assimilating these type of observations. For now we focus on well-characterized GOME observations, and it is valuable to investigate how far this method gets by using those observations.

ACPD

9, C3194–C3206, 2009

GOME provided the first set of this type of observations and therefore we decided to analyze our method for the 1996-2001 time period.

Comment: Second, it is very difficult to interpret Figures 2 and 4 due to their small size. I also found the labels and captions to be rather confusing, and the description of these figures lacking sufficient detail. Because of the way the information is plotted, it is difficult to see how the addition of the ozone profile information affects the TOC estimates. Based on the information in the tables and figures, it appears that assimilating the ozone profile information does not significantly improve global TOC estimates. Yet in the Discussion (section 5) the authors speculate that future applications of TORA using ozone profile measurements with a smaller footprint and higher horizontal resolution in the chemical transport/assimilation model should improve their results in mid-latitudes. Since the ozone profile information fails to add value to the TOC estimates outside of the tropics, in contrast to expectations, couldn't one also conclude that the assimilation system itself is fundamentally flawed? Based on the evidence presented here, it's not clear that increasing TORA model resolution to try and improve mid-latitude performance is a worthwhile undertaking. The authors need to offer some quantitative calculations in support of their conclusion.

Response: Figures 2 and 4 have been modified to provide a clearer view. The figure captions describe the figures more precisely.

With regard to the added value of the assimilation: we use a similar assimilation method as Stajner et al. [2007]. Furthermore, our results show that our assimilation also does its job as it keeps stratospheric ozone "in check", i.e. transport models have a tendency to accumulate ozone in the stratosphere outside of the tropics due to an "overstrong" Brewer-Dobson circulation, which is related to the use of meteorological reanalysis data to simulate transport. This accumulation results in too large stratospheric ozone amounts, but the assimilation counteracts this accumulation by assuring that modeled stratospheric ozone remains close to the measured values. We show that the GOME ozone profiles are sufficiently accurate for this purpose. However, for a proper deriva-

ACPD

9, C3194–C3206, 2009

tion of tropospheric ozone it is necessary that the observations and model results can resolve ozone features in UTLS ozone. These can be small, smaller than the model resolution and much smaller than the GOME ozone profile spatial footprint of roughly 1000x100 km. Further, for these large footprints tropospheric clouds complicate the situation as for partly clouded pixels it is not known where the tropospheric ozone information originates from (i.e. from which part of the pixel). Smaller footprints will suffer less from this 'smearing' effect. The work by Schoeberl et al. [2007] suggests that residual methods work in case of a spatially finer model grid (1x1.25 degrees) and MLS stratospheric ozone profiles with good vertical resolution. The TM5 model in our study does not have such a fine spatial resolution, and the GOME ozone profiles lack the vertical resolution of the MLS profiles, hence it is likely that either the model resolution or the GOME ozone profile resolution (either vertically or spatially) or a combination of all three issues is the cause for the lack of improvement of tropospheric ozone residuals from the TORA method, as noted in the discussion. However, given the results by both Schoeberl et al. [2007] and Stajner et al. [2008] we expect that improvements to the TORA method can be made, and we have started investigating this. In this paper we present the method and explore the extent to which satisfactory results can be obtained for the GOME time period. Nevertheless, the final result can be that regardless of the spatial model and observation resolution the lack of vertical information in GOME and GOME-like ozone profile observations is the bottleneck and prevents accurate determination of the tropospheric ozone column. This study is only one but an important step along the way to assessing if residual methods using UV/VIS ozone profile assimilation can be used to determine the tropospheric ozone column. It would not be a good idea to reject the presented method based on the use of GOME data, which have known limitations.

We clarified these issues wherever discussed in the document.

Furthermore, we modified the tables and put bold numbers (indicating an improvement) in italics and colored them blue, so that systematic changes in all tables could be more

ACPD

9, C3194–C3206, 2009

easily identified.

Comment: Third, it's not clear to me what the ultimate goal of this work is. Are the author's developing a system to analyze past global TOC behavior or are they interested in producing TOC analyses in near real time for monitoring purposes? If it's the former, the results seem to show that the use of an appropriate ozone climatology with analyzed winds constrained by linearized chemistry is reasonable for estimating global TOC over the 1996-2001 period on monthly time scales. In this case, the meteorological analyses capture the vertical motions that drive tropospheric ozone fluctuations in response to synoptic variability in the upper troposphere. If it's the latter, Figure 5 appears to show that a more sophisticated ozone assimilation approach is needed (assuming that the TM4 results with full chemistry are more representative of the true annual mean TOC).

Response: The ultimate goal here is to investigate if it is possible to derive valuable information on tropospheric ozone from UV-VIS nadir ozone profiles, both in retrospect and for near-real time. Given the availability of ozone profile measurements from UV-VIS instruments like GOME, OMI and GOME-2 and the prospect of continuation of such observations for at least another decade provides an incentive to further explore the possibility of using UV-VIS ozone profiles for determining tropospheric ozone columns. Because of the limited vertical resolution of UV-VIS ozone profiles other information is required to constrain the vertical distribution of tropospheric and stratospheric ozone. As outlined in the paper, it can be motivated that assimilation of UV-VIS ozone profiles in a transport model should – at least in theory – give you a realistic vertical distribution of ozone, which then can be used in combination with total ozone column measurements to provide a tropospheric ozone column. In this paper we introduce and explore this method, which has not been done before using UV-VIS nadir ozone profile measurements.

Our results show that the assimilation improves the modeled ozone distribution and uses the information content of the UV-VIS nadir ozone profiles. For reasons outlined

ACPD

9, C3194–C3206, 2009

in the paper, the use of GOME UV-VIS nadir ozone profiles in combination with spatial resolution of the model does not result in satisfactory residual tropospheric ozone columns.

We have put more emphasis on the main objectives of the paper in the introduction.

Specific Comments:

Comment: Abstract: In the last sentence, the authors state that the present results indicate that TORA residual should improve using MetOp/GOME-2 and EOS-AURA/OMI observations. Why exactly would this be so? Couldn't the present system be used to demonstrate this assertion using synthetic data?

Response: This paper presents results from using GOME with known limitation as explained above. We are working on the implementation of GOME-2 and OMI ozone profiles, but this still in the development phase, as the quality of both GOME-2 and OMI ozone profiles still needs to be evaluated. Performing a synthetic study to demonstrate that assimilation of GOME-2 or OMI ozone profiles results in realistic tropospheric ozone columns is beyond the scope of this paper. For this paper we want to present the method, use GOME data, analyze the results, and investigate if improvements can and should be made.

Comment: Section 2: The paper would benefit from some general descriptive remarks about chemistry-transport models in general and the TM5 in particular. Why does TM5 employ a linearized ozone photochemistry parameterization rather than full photochemistry? Can the authors describe in more detail the assimilation system? What are the main differences between the TM5 here and in Segers et al.? It was not clear at first what is being assimilated. I was under the impression that both total ozone measurements and profile measurements are being assimilated, but this does not appear to be the case. What are the relevant spatial scales for error propagation of the assimilated ozone profiles? Is the Fortuin and Kelder climatology used for the troposphere different than the climatology used in the Cariolle and Deque scheme? If so, are the

ACPD

9, C3194–C3206, 2009

climatologies merged in some way?

Response: Employing a linearized ozone chemistry parameterization rather than a full photochemical scheme is simply a matter of focus and computational power: a linearized model is very fast and uses only one single tracer. Furthermore, it has been proven to produce realistic ozone variations for a variety of situations [Teyssedre and Cariolle, 2007]. A full stratosphere-troposphere photochemical scheme involves many dozens of tracers and requires detailed chemistry and photolysis calculations, which is very time consuming. It also introduces many uncertainties – in particular in stratospheric ozone chemistry – and potentially unwanted effects. Combined with the computationally expensive assimilation method this is for our purposes completely unpractical. Furthermore, the focus of this paper is on ozone. This explains why most ozone assimilation efforts so far prefer to use the linearized ozone chemistry [Lahoz et al., 2007]. The linearized ozone chemistry has proven its value for many years.

Apart from the use of a newer TM version (TM5 vs. TM3) the only difference between the TM5 model used here and the TM3 model in Segers et al. [2003] is how the growth of the 2-D standard deviations due to uncertainties in chemistry and meteorological input is vertically distributed. Segers et al. [2003] distributed this by applying one constant factor for all altitudes. However, on long time scales of months to years this leads to unwanted behavior (infinitely small standard deviations). Vertical distribution of the standard deviation following the vertical distribution of ozone rather than using a constant factor is more appropriate, which we therefore implemented after discussion with Segers and colleagues.

The relevant spatial scales for error propagation are discussed in Segers et al. [2003]

From the adjustments to the introduction it should be clear now that we assimilate UV-VIS ozone profile measurements.

The CARIOLLE scheme uses the ozone climatology of Fortuin and Langmatz [Proc. SPIE Vol. 2311, p. 207-216, Atmospheric Sensing and Modelling, 1994], which is

ACPD

9, C3194–C3206, 2009

a predecessor of the Fortuin and Kelder [1998] climatology. Both climatologies have been developed using largely observations from the 1980s.

Comment: Section 3: I could not find Mijling et al 2009 in the reference list. Is this a published article? If not, it should be listed as a footnote following ACP style guidelines.

Response: We could not determine from the ACP guidelines how a manuscript in preparation should be referred to. Footnotes are not recommended. We therefore follow the guidelines of the AGU, in which such publications are not allowed in the reference list but should be indicated in the document as "manuscript in preparation".

Comment: Section 4: I found the discussion in Section 4.I to be very confusing. According to the text, the UT/LS ozone columns from the free model simulation (i.e., no assimilation of ozone profile) are compared with selected sonde data in Figure 2. Yet the Figure 2 caption states this is a comparison of assimilated UT/LS columns with sondes. What ozone information is being assimilated in Figure 2? According to the figure legend, there is no assimilation. Figure 2 should be expanded into two four panel figures, one for the mid-latitudes and one for the tropics. It is unreadable in its present form. Also, I could not locate station 394 in Figure 1. It would help the reader to place boxes around the selected 4 stations or otherwise highlight their locations in Figure 1.

Response: The confusion arose because the figure 2 caption was incorrect and it has been adjusted. It should read the free running simulation. Figure 2 was modified according to both referees' suggestions. Boxes have been placed around the four stations highlighted by adding grey filled boxes and using a different color. We also noted that table 3 only highlights two of the four locations from figure 2 and highlighted in tables 1+2, which we adjusted. Finally, table 3 contained one stations (437) not present in tables 1+2, so that one was removed.

Comment: The apparent good agreement between the free model run and the monthly mean sonde data seems to reflect that the model chemistry relaxes the ozone back to climatology with 2 week time scale while the analyzed winds capture the local fluctu-

ACPD

9, C3194–C3206, 2009

ations in tropopause height. Isn't the Fortuin and Kelder climatology is based on the same network of sondes that you are comparing against?

Response: The Fortuin and Kelder climatology is based not only ozone sonde observations but also SBUV satellite observations, and uses data only up to 1993 (with one exception up to 1995), so it is actually different than the sonde data used in this study. The agreement between the sondes and free model run should occur as the scheme is specifically designed to simulate a realistic ozone distribution and compared to other observations generally does a good job [Teyssedre and Cariolle, 2007]. In the long run, ozone variability is strongly determined by seasonal variations in circulation patterns and photochemistry, which do not change much from year to year. Hence, it is expected that the agreement between the model simulation and observations improves for monthly and climatological means. In both cases the role of short-term synoptical variations is mostly removed by the averaging. With regard to tropopause height variations: de Laat et al. [2005] show only part of the variations in UTLS ozone can be attributed to tropopause height and thereby dynamical variations. The other part is caused by variations in tropospheric and stratospheric ozone. The chemical and dynamical ozone variations in the UTLS are captured by the linearized chemistry. So, given the good performance of the model one could actually wonder where the added value of the GOME ozone profile observations lies, if based on these statistics the free run of the linearized chemistry already does a proper job (which is what the linearized chemistry is supposed to do anyhow). As we show, in our case the added value lies in countering the overstrong stratospheric circulation, which in combination with the linearized chemistry leads to accumulation of ozone in the stratosphere at higher latitudes. Differences are now shown in the new Figure 3.

Comment: Section 4.2 could probably be merged with section 4.1 since it also deals with the free model run and finds essentially the same result.

Response: We decided to keep sections 4.1 and 4.2 because of the relevance of UTLS and TTOC ozone for the TORA method. Maybe the confusion mentioned above led to

ACPD

9, C3194–C3206, 2009

this recommendation. UTLS ozone is crucial for the ozone climatology, and deserves a thorough evaluation. TTOC variability is not relevant for the TORA method although the results can provide some additional information about the effect of the assimilation. Hence, we prefer to separate both (also for sections 4.3 and 4.4).

Comment: The discussion in Section 4.3 focuses largely on temporal correlations. Is this the only relevant metric? Can you say whether or not spatial patterns improve with the use of the profile data? For example, does the bias arising from the use of a zonal mean climatology in the free model run in Figure 2 (lower left panel) improve when ozone profile measurements are assimilated?

Response: The validation with ozone sondes shows that overall the bias improves everywhere as a result of the assimilation, which indicates that the spatial patterns of the bias do improve from the assimilation. Furthermore, we also show that in the tropics the TTOC bias improves. Since the ozone climatology used in the CARIOLLE scheme is a zonal mean, it cannot capture the wave-1 pattern in tropical tropospheric ozone. The assimilation improves the correlation and bias in the tropics. This was already noted in the discussion.

Comment: Section 4.4 could also be merged with 4.3.

Response: See answer to merging sections 4.2 and 4.1.

Comment: What is lacking in Section 4.5 is a direct comparison of results between the free run and the run with assimilation. It is very difficult to ask the reader to examine two large sets of tables to determine the effect. Can you plot a sonde record along with results with and without profile assimilation (perhaps one for mid-latitudes and one for tropics)? Alternatively, the mean difference between the results with and without assimilation for each case, tropics and mid-latitudes, could be plotted.

Response: We added figures for the same locations as in figure 2 showing UTLS ozone variations and the differences. The results indicate that the assimilation had a

ACPD
9, C3194–C3206, 2009

strong effect on mid-latitude UTLS ozone, but not much on tropical ozone. This is an indication that tropical ozone is already well captured by the CARIOLLE chemistry and that variability is small, so the ozone profiles cannot add much to the tropical ozone profiles. Outside of the tropics, most variability of UTLS ozone is related to dynamical movement (tropopause faults, changes in tropopause heights etcetera). The GOME ozone profiles capture these variations and thus have a strong effect on UTLS ozone. We also added table S3 with the correlations and rms differences for all sonde locations for the free run and assimilation run.

Comment: I don't understand Figure 5. Is the top panel supposed to be the "true" TOC distribution? Both the GDP and TOMS results show the high ozone over the equatorial Atlantic mentioned in the text. I don't see that in the TM4 result. Is this an improvement?

Response: We added the TM4 model results purely as a reference. The model distribution is what we would expect based on a model that is similar to many other transport models and produced realistic tropospheric ozone variations [Stevenson et al., JGR, 2006]. Without such a reference – and lack of validation – one could easily mistake the spatial patterns in the annual mean residual ozone as true variations. This has happened in papers before. The comparison with two different total column products and the coherence between both residual fields shows that the spatial variations in the average residual columns is not related to the total ozone column measurements.

Comment: Section 5: Unless the authors provide additional evidence based on their model calculations, I don't see any support for the claim that increasing the resolution should improve the assimilation.

Response: Our results in themselves do not provide direct support for the claim that increasing the model resolution should improve the assimilation. However, the results from Schoeberl et al. [2008] show that with a better resolution realistic residuals are derived. Furthermore, the study by Sparling et al. [2006] show that the correlation length

ACPD

9, C3194–C3206, 2009

of mid-latitude UTLS ozone variations is less than 400 km in the LS and less than 100 km in the UT. It thus can be expected that a model resolution of 300x200 km is insufficient to resolve the typical mid-latitude variations and that GOME observations with a spatial footprint of about 1000x100 km are limiting for tropospheric ozone variability. In order to determine realistic tropospheric ozone columns either observations or model results and preferably both should be able to capture ozone variations on those scales. Whether or not assimilation of nadir UV/VIS profiles in a 3D chemistry-transport model does result in more realistic tropospheric ozone variations remains to be seen, but that is the least that should be done.

Minor corrections:

Page 11815, line 24: Meijer et al. 2006 missing from reference list. Page 11833, line 23: Tarasick and Slater reference is out of order.

Response: This has been corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 11811, 2009.