**Atmospheric
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Discussions**

Evaluation of tropospheric ozone columns derived from assimilated GOME ozone profile observations

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Abstract

Tropospheric O_3 column estimates are produced and evaluated from spaceborne O_3 observations by the subtraction of assimilated O_3 profile observations from total column observations, the so-called Tropospheric O_3 ReAnalysis or TORA method. Here we apply the TORA method to six years (1996–2001) of ERS-2 GOME/TOMS total O_3 and ERS-2 GOME O_3 profile observations using the TM5 global chemistry-transport model with a linearized O_3 photochemistry parameterization scheme.

Free running TM5 simulations show good agreement with O_3 sonde observations in the upper-tropospheric and lower stratospheric (UTLS). Assimilation of GOME O_3 profile observations improves the comparisons in the tropical UTLS region but slightly degrades the model-to-sonde comparisons in the extra-tropical UTLS for both short day-do-day variability as well as for monthly means. We suggest that this degradation is related to the large ground pixel size of the GOME O_3 measurements (960×100 km) in combination with retrieval and calibration errors. The assimilation of GOME O_3 profile observations does counter the gradual multiyear mid-latitude stratospheric O_3 accumulation caused by the overstrong stratospheric meridional circulation in TM5.

The evaluation of daily and monthly tropospheric O_3 columns obtained from total column observations and using the TORA methodology shows realistic residuals within the tropics but unrealistically large deviations outside of the tropics, although average biases remain small for the monthly means.

The findings of this paper suggest that improvements can be expected by using O_3 observations from present-day instruments like MetOp/GOME-2 and EOS-AURA/OMI.

1 Introduction

Ozone (O_3) is one of the most important atmospheric trace gases. About 90% of atmospheric O_3 is located in the stratosphere, where it shields the lowest 10–15 km of the atmosphere from UV radiation which is harmful for life. The remaining 10% is

located in the troposphere, where it plays a key role in various processes.

Tropospheric O_3 is required for the formation of the hydroxyl radical (OH), which determines the oxidizing or cleansing capacity of the atmosphere. Furthermore, tropospheric O_3 is the harmful atmospheric element in photochemical smog, severely impacting human health and reducing crop yields (Royal Society, 2008). Finally, tropospheric O_3 is an important greenhouse gas contributing about 20% to the current anthropogenic greenhouse gas radiative forcing (IPCC, 2007; SPM, Fig. 2).

The tropospheric O_3 distribution is determined by transport processes in combination with complex chemical production and destruction. Important boundary conditions for tropospheric O_3 are downward transport from the stratosphere and surface deposition. The total tropospheric O_3 column (TTOC) is defined as the sum of the tropospheric ozone concentration from the surface to the tropopause. The TTOC responds to concentration changes as well as to spatio-temporal variability in tropopause height and in surface pressure, the latter largely in relation to variations in surface elevation.

Monitoring of tropospheric O_3 is a challenge because tropospheric O_3 shows large spatio-temporal variability and the number and representativity of observational sites are limited for a complete assessment of long-term tropospheric O_3 trends and tropospheric O_3 distribution changes (IPCC, 2007; WG1, Ch2, pp150).

Satellite observations can potentially contribute in addressing some of the above-mentioned issues, especially the requirement for a spatially dense observational network. Some success has been reported in *direct* observations of tropospheric O_3 using GOME (Global Ozone Monitoring Experiment) UV-VIS nadir observations (Liu et al., 2006). Separating between troposphere and stratosphere depends on determining the exact location of the tropopause. Additional complications stem from the large and rapid tropospheric O_3 variability outside of the tropics, which requires individual observations to be of sufficient accuracy (de Laat et al., 2005). However, due to the limited vertical sensitivity and resolution of direct O_3 profile observations determining the TTOC is at the edge of the satellite observing capability.

Indirect or residual methods – i.e. estimating the stratospheric O_3 column which is

then subtracted from the total O_3 column – have been shown to work in the tropics, where variability of both stratospheric O_3 as well as the tropopause is small compared to tropospheric O_3 variability (NRC, 2008, 44–46). Outside the tropics results have been mostly inconclusive. Recently, results from a new indirect method using strato-
5   spheric O_3 profiles from Microwave Limb Sounder (MLS) observations have been reported (Schoeberl et al., 2007). MLS observations have a relatively high vertical resolution but do not provide daily global coverage. Therefore, trajectory calculations are used to simulate the advection of MLS observed stratospheric ozone and to reconstruct the daily global vertical distribution of stratospheric O_3. Subtracting the MLS-based
10   stratospheric O_3 column from Ozone Monitoring Instrument (OMI) total O_3 column observations yields level-2 tropospheric O_3 residuals. Results suggest that this method works quite well and can deliver realistic tropospheric O_3 variations for relatively short time periods (5 days), also at mid-latitudes.

In this paper we introduce and explore another *indirect* method for estimating tropo-
15   spheric O_3 columns by assimilation of UV-VIS nadir O_3 profile measurements and subtracting the resulting stratospheric column from UV-VIS total column O_3 observations. The rationale behind this method is that upper tropospheric and lower stratospheric (UTLS) O_3 variability and thus the separation between stratosphere and troposphere is dominated by dynamical processes, while photochemical processes are much less
20   relevant (Brasseur, 1999, p. 502). Assimilation of satellite observations of the smooth vertical O_3 profiles in a chemistry-transport model will provide the best estimate of the stratospheric O_3 column. Transport is calculated using weather analyses that provide the best available information about the state of the atmosphere and the separation between stratosphere and troposphere. Subtracting the assimilated stratospheric O_3
25   column from a total O_3 column observation should then deliver a residual product that is the best estimate for the TTOC. A major advantage of this method is that it can be applied globally and possible gaps in the observational system are filled by the assimilated ozone distribution as determined by the linearized O_3 chemistry in the transport model using dynamical information from the weather analyses. This method is referred

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to as the Tropospheric O_3 Re-Analysis or TORA method.

This paper is organized as follows: Sect. 2 briefly describes the setup of the TM5 model and O_3 profile assimilation, Sect. 3 describes the observational datasets used in this study, Sect. 4 shows and evaluates the free running and assimilation model results for the UTLS region as well as the for the modelled TTOC's. Finally the calculated TORA residual product is evaluated. Section 5 contains a discussion of the main results, some preliminary conclusions on the TORA methodology as well as recommendations for future work.

2   TM5 O_3 profile assimilation

The TM5 model has been developed at the Institute of Marine and Atmospheric research Utrecht (IMAU) in cooperation with the Royal Netherlands Meteorological Institute (KNMI) and the Dutch Centre for Mathematics and Computer Science (CWI). The TM5 model has been used to study stratospheric chemistry and transport (Bregman et al., 2003; van den Broek et al., 2003, 2004]) A detailed description of the TM5 model is given in Krol et al. (2005). The TM5 version used here is the same as in de Laat et al. (2007), using meteorological reanalysis data from the European Centre for Medium range Weather Forecast (ECMWF).

Segers et al. (2005) described the assimilation of GOME nadir O_3 profile observations using the TM3 model (TM3 is the predecessor of the TM5 model) in single tracer mode with the linearized O_3 chemistry scheme from Cariolle and Deque (1986), hereafter referred to as the "CARIOLLE chemistry". We use updated parameter values from Cariolle and Teyssèdre (2007), but without the use of a cold tracer to represent heterogeneous O_3 loss. It is anticipated that GOME O_3 profile observations for Antarctic ozone hole conditions are not sufficiently accurate (Meijer et al., 2006). The main focus of this paper is therefore to test the TORA methodology on a day-to-day basis for tropical and mid-latitudes.

The assimilation procedure in Segers et al. (2005) is based on a Kalman filter, and

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is an extension of the total O_3 column assimilation methodology presented by Eskes et al. (2003). For this paper the Segers et al. (2005) methodology was adopted and adjusted for use in TM5. The CARIOLLE chemistry has already been used for stand alone TM5 simulations (de Laat et al., 2007). We use the same 44 hybrid-sigma layers between the surface and 0.1 hPa, and a horizontal resolution of 3°×2° (lon-lat) as in Segers et al. (2005). The 44 layers are a subset of the ECMWF operational 60-layer configuration available for the period 1996–2001. The choice of the 44 layers is related to the focus of the assimilation of producing accurate stratospheric O_3 fields and resolving O_3 variability in the UTLS region, and includes all ECMWF layers between 10 and 20 km altitude.

Tropospheric chemistry is not included in the CARIOLLE chemistry. Instead, tropospheric O_3 is relaxed towards an O_3 climatology (Fortuin and Kelder, 1998) between the surface and 500 hPa altitude with a relaxation time of 14 days. Tests with other tropospheric altitude ranges where the relaxation to the climatology is applied showed little sensitivity to the altitude range definition, as long as lower tropospheric O_3 was sufficiently constrained. Note that the TORA methodology only needs the stratospheric O_3 distribution from the assimilation model.

3 Observations

3.1 GOME O_3 profile observations

The GOME O_3 profile observations are retrieved using an updated version of the O_3 ProfilE Retrieval Algorithm (OPERA). A full description of the algorithm can be found in the Algorithm Theoretical Basis Document (van Oss, 2004) produced for the ESA funded project CHEOPS-GOME (Climatology of Height-resolved Earth-Ozone and Profile Systems for GOME). A detailed validation of GOME OPERA O_3 profiles can be found in deClercq (2007). The OPERA version used in this paper is the version R2 as described in deClercq (2007). A detailed description of recent improvements in the

OPERA retrieval algorithm is reported in Mijling et al. (2009).

The analysis of the information content of the GOME OPERA O_3 profiles (deClercq, 2007; chapter 4) reveals that the amount of information – vertical resolution and sensitivity – has decreased over this period due to instrument degradation. Three periods can be discriminated: 1996–1998, when information content is at its maximum, 1999–mid-2001, when tropospheric information content degrades progressively and after mid-2001, when stratospheric information content also starts to degrade and when tropospheric information – in particular between 30° N–30° S – vanishes. Therefore, this paper focuses on the six year period 1996–2001.

Comparison of OPERA O_3 profiles with independent ground-based microwave and O_3 sonde profile measurements shows that in the bulk of stratosphere up to 45 km altitude there is – on average – a good agreement of 10% and better until the end of 2001 (DeClercq, 2007). In the troposphere, on average a good agreement is found both inside and outside of the tropics only during the first years. After 1998 tropical biases were found as well as the effects of the instrument degradation as described above.

Quality checks were applied to the O_3 profiles in order to ensure the selection of accurate O_3 profile observations. Profiles are checked on standard OPERA quality flags for convergence of the retrieval and out of bounds values (including negatives). Non-convergence may for example occur over snow and icy surfaces.

3.2 Total O_3 column observations

The main goal of the TORA methodology presented in this paper is to produce a best estimate of the stratospheric O_3 column and subtract this column from a total column observation. This yields level-2 tropospheric O_3 residuals (or TTOC's). For the evaluations we use two GOME total O_3 column datasets: DLR GDP v4.1 (van Roozendael et al., 2006; Balis et al., 2007) and TOGOMI v1.2 (Valks et al., 2004) as well as TOMS v8 total O_3 column observations (Bhartia, 2004). These total O_3 column measurements have a pixel size of about 320×40 km, which is significantly smaller than the footprint

of the GOME O_3 profile observations (960×100 km). Another advantage of the TORA methodology over direct TTOC estimates from O_3 profile observations is the use of the better spatial resolution of the total O_3 column observations. Furthermore, the assimilation ensures that for every level-2 total O_3 column observation a best estimate for the stratospheric O_3 column is available even in case no co-located O_3 profile observation is available.

3.3 O_3 sonde observations

For evaluation of the model simulations with and without assimilation the model profiles are compared with O_3 sonde observations. These observations are taken from the World Ozone and Ultraviolet radiation Data Center (WOUDC) database (http://www.woudc.org/) wherever sonde observations were available for the period 1996–2001. A comparison was made for all available sonde data. Figure 1 shows the geographical locations of the stations summarized in Table S0 in the supplementary information (http://www.atmos-chem-phys-discuss.net/9/11811/2009/acpd-9-11811-2009-supplement.pdf).

3.4 ECMWF tropopause heights

In order to separate between the stratosphere and troposphere for estimating the stratospheric O_3 column the tropopause height must be known. Although various tropopause definitions exists, the most commonly used definitions are 2 K/km lapse rate within the tropics and the use of the Potential Vorticity (PV) unit outside of the tropics (here we use the PV=3 contour). ECMWF temperature and ERA40 PV data were used to calculate daily tropopause heights on a 3°×2° resolution. Poleward of 40° the PV definition was used, equatorward of 20° the temperature-lapse rate definition was used, while between 20° and 40° latitudes both definitions were linearly combined to ensure a smooth transition between both definitions. The minimum tropopause height allowed was 5 km to account for potential lower tropospheric temperature inversions.

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4 Results

The TORA methodology aims at providing the best estimate for the stratospheric O_3 column and subtracting this column from total O_3 column observations that will yield – in an ideal case – the total tropospheric O_3 column (TTOC) as a residual. First of all, a baseline needs to be defined: how well does a free TM5 run – i.e. without assimilation – compare with sonde observations? Results for UTLS O_3 are discussed in Sect. 4.1 and for the TTOC in 4.2. The next step is to compare the TM5 simulation with assimilation of the GOME O_3 profiles with the same set of sonde observations and establish how the comparisons change (Sects. 4.3 and 4.4, respectively). In addition, the effect of the O_3-profile assimilation on the full 6-year modelled UTLS O_3 is investigated (Sect. 4.5). After that, the TORA methodology is applied to produce tropospheric O_3 residuals and these are again evaluated with the available sonde observations (Sect. 4.6). Statistics of the one-on-one comparisons are presented, as well as annual mean differences in UTLS O_3 for the model simulation with and without assimilation. Finally, we analyse the annual mean spatial distribution of the TORA residuals for the year 2000.

4.1 Evaluation of TM5 UTLS O_3 with sonde observations

The CARIOLLE scheme is particularly suited for the simulation of lower and middle stratospheric O_3, and given that outside of the tropics lower stratospheric and upper tropospheric O_3 are dynamically coupled, also for the simulation of upper tropospheric O_3 outside of the tropics (Cariolle and Deque, 1986; Cariolle and Teyssèdre, 2007; Geer et al., 2007). In de Laat et al. (2007) we showed in a comparison of GOME O_3 profiles with O_3 sonde profiles from Payerne, Switzerland, that O_3 concentrations at 15 km altitude agreed very well on both short synoptic and long seasonal time scales, despite the large ground pixel size (960×100 km) of the GOME O_3 profile. Furthermore, the key issue of the TORA methodology – or any tropospheric O_3 residual method for that matter – is the separation of the stratosphere from the troposphere and thus a realistic representation of the tropopause. We therefore chose to compare modelled

O_3 for the layer between 250 and 50 hPa (approximately the 10–20 km altitude range; hereafter referred to as UTLS column) with sonde observations.

A subset of four O_3 sonde locations representative for different tropospheric and stratospheric circumstances is used to illustrate the comparisons between model results and sonde observations. Figure 2 shows the comparison between UTLS columns from the TM5 assimilation run and sonde observations for the four location subset: Payerne, Switzerland (stn156); Broad Meadows, Australia (stn394), Samoa (stn191) and Ascension Island (stn328). Payerne and Broad Meadows represent mid-latitude Northern and Southern Hemisphere locations and are characterized by strong seasonal cycles with the largest UTLS columns during local winter, associated with a lower tropopause and thus a larger part of the stratosphere located between 250 and 50 hPa. Superimposed on the seasonal cycle are rapid UTLS column variations associated with synoptical scale weather phenomena driven by Rossby wave activity. Figure 2 shows that there is a good agreement between UTLS O_3 from the TM5 simulation and sonde observations, which is also reflected in the monthly means. For individual days considerable differences do occur. These differences are likely related to model resolution and representativity of the sonde data as well as to the exact timing of UTLS events by for example rapidly travelling frontal zones and the corresponding time differences of the O_3 sonde measurements (mainly around local noon) and the time of model output (always at 12:00 UTC). The amplitude of the seasonal cycle and day-to-day synoptic scale variability are larger for the Northern Hemisphere location (top left panels) compared to the Southern Hemisphere location (top right panels), related to the stronger Northern Hemisphere Rossby wave activity.

The bottom four panels show two tropical locations: Ascension Island for the equatorial Atlantic Ocean and Samoa for the equatorial Pacific Ocean. The seasonal cycles for both locations are much smaller in amplitude than those at the mid-latitude locations. This is partly related to the smaller seasonal cycle of the tropical tropopause and tropical lower stratospheric O_3, and partly due to the higher tropopause in the tropics so that a smaller part of the stratosphere is located between 250 and 50 hPa. The free-

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running TM5 simulation also agrees rather well with tropical observations and captures the synoptic-scale, seasonal and interannual variability in the tropical UTLS. A small positive model bias appears for Samoa (bottom left panel). This bias is related to the zonal-mean tropospheric ozone climatology used in the simulations which because of the zonal wave 1 pattern of tropical tropospheric O_3 with a minimum over the Pacific leads to an overestimation of the modelled UTLS O_3 for the equatorial Pacific.

Table 1 summarizes the comparison between modelled and sonde observed UTLS variability for a selection of 25 sonde locations over the full range of latitudes. The statistics show that in general at mid-latitudes model results and observations agree well, with correlations varying between 0.55 and 0.92. The correlations improve considerably for the monthly means, and are even better for the 6-year climatological values, which in many cases are near perfect. This proves that the day-to-day differences caused by synoptical scale variability can be considered random variations, i.e. weather noise which averages out. This variability is large outside of the tropics as reflected in the root-mean-square (rms) differences. When looking in detail at the biases, we find that at high northern latitudes (>60° N) the model slightly underestimates UTLS O_3. For the region between 20° N and 50° N it slightly overestimates UTLS O_3. This also occurs in the Southern Hemisphere mid-latitudes. At Southern Hemisphere high latitudes (>60° S) modelled UTLS O_3 is too large, caused by the absence of heterogeneous O_3 loss in the TM5 model simulation.

In the tropics between 20° N and 20° S, correlations are smaller although also here the correlations improve for monthly and climatological means, while biases and rms differences are small. This different behaviour is caused by the smaller seasonal cycle and the relatively larger influence of tropospheric O_3 variability. Spatio-temporal variations in upper tropospheric O_3 relate via convection processes to lower tropospheric O_3 which is not well reproduced by the TM5 simulations because of the relaxation to the zonal-mean tropospheric climatology and the lack of tropospheric chemistry. Overall, given the use of a linearized O_3 chemistry model, the applied model resolution and differences in collocations, the agreement between the free-running TM5 simulation

and observed UTLS O_3 from sondes is considered rather good.

4.2 Evaluation of TM5 tropospheric O_3 columns with sonde observations

In the previous section we compared modelled and observed UTLS variability. Since the end goal of this paper is to derive total tropospheric O_3 columns (TTOC's), we also briefly investigate the agreement between the free-running TM5 simulation and observed TTOC's as derived from ozone sondes (Table 2). Similar to the UTLS comparison the correlations outside of the tropics are larger than within the tropics, and correlations improve for monthly and climatological means, although the correlations are slightly worse than for the UTLS columns. The agreement between modelled and observed TTOC variability outside of the tropics is closely related to the influence of upper tropospheric O_3 and the coupling between upper tropospheric O_3 and atmospheric dynamics. The model is well suited to reproduce O_3 variations related to such processes. However, the variations are superimposed on a tropospheric O_3 climatology which does not result in realistic day-to-day and interannual TTOC variability; hence correlations for TTOC are worse than those for the UTLS. In absolute terms the TTOC bias is smaller, but in relative terms the biases – compared to the mean – are similar for the UTLS column and the TTOC (not shown). The same applies for the rms differences, which are comparable for the UTLS and TTOC relative to the rms variability or mean of the UTLS and TTOC.

4.3 Evaluation of TM5 assimilation UTLS O_3 with sondes

The next question to address is how the comparison and statistics change when GOME O_3 profiles are assimilated in TM5. Table 1 shows that correlations with or without assimilation are comparable for the individual, monthly mean and climatological values. Outside of the tropics the correlations are generally a bit worse for the assimilated UTLS columns. The differences in correlations are small and the change should be considered insignificant for most cases, but nevertheless this occurs for nearly all

sonde locations. On the other hand, within the tropics the correlations improve, albeit also slightly. The rms differences follow suite, i.e. the rms differences decrease or increase with decreasing or increasing correlations. The rms values are large for the UTLS columns outside of the tropics, reflecting the influence of synoptic scale day-to-
⁵ day UTLS variability associated (2005).

Earlier studies have shown that the spatial correlation length of UTLS O_3 at mid-latitude locations is about 300–400 km for the LS and less than 100 km for the UT (Sparling et al., 2006). These spatial scales are considerably smaller than the size of the GOME pixel (960×100 km). The question rises whether the GOME O_3 profiles on
¹⁰ average contain sufficient and accurate information to improve the already quite good TM5 UTLS O_3 profiles from the stand-alone simulation. Furthermore, because of the vertical smoothing of the GOME O_3 profile, it becomes very important to know exactly where information of the tropospheric part of the GOME O_3 profile originates from (e.g. from climatology in case of obscuring cloudiness). These retrieval uncertainties,
¹⁵ together with errors due to measurement biases and uncertainties may degrade the accuracy of the GOME O_3 profiles in the UTLS region.

For tropical locations the above mentioned problems may be less of an issue because UTLS variability is smaller, tropospheric O_3 variability occurs more on longer temporal time scales compared to mid and high latitudes (de Laat et al., 2005) and
²⁰ the sensitivity of GOME to tropospheric O_3 is larger in the tropics (Liu et al., 2005, 2006). Furthermore, the comparison between modelled and observed tropical UTLS O_3 showed that the comparison was not as good as outside of the tropics. Hence, there is room for improvement at tropical latitudes, where the O_3 profile assimilation leads to improvements in the modelled UTLS columns.

²⁵ **4.4 Evaluation of TM5 assimilated TOC with sonde observations**

We only very briefly discuss the differences in TTOC's for the TM5 simulations with and without assimilation of GOME O_3 profiles. Here, a consistent picture arises from Table 2. Correlations are nearly always better for the simulation without assimilation com-

pared to the simulation with assimilation. This is reflected in the rms differences, which are better for nearly all locations. On the other hand, the bias improves somewhat for nearly all locations when GOME O_3 profiles are assimilated. Apparently the effect of assimilation of GOME O_3 profiles is the increasing variability in modelled TTOC's, while at the same time the average TTOC's improve.

4.5 Effect of O_3-profile assimilation on the 6-year free-running UTLS O_3

Chemistry-transport models show a general tendency to overestimate the residual stratospheric circulation, i.e. too fast transport from the tropics to middle and high latitudes (Douglass et al., 2003), also called the overstrong meridional circulation (Stajner et al., 2008). The effect of this overly strong circulation is a gradual accumulation of stratospheric O_3 outside of the tropics, as stratospheric O_3 is produced in the tropics and slowly transport to higher latitudes where destruction occurs. This effect has also been reported for TM5 (de Laat et al., 2007), but can be countered by assimilation of stratospheric O_3 profiles (Stajner et al., 2008).

Figure 3 shows the annual mean differences for all years from 1996 to 2001 for TM5 UTLS ozone without and with assimilation. Differences in extra-tropical UTLS O_3 gradually increase year by year, while tropical UTLS O_3 differences are close to zero. This spatial pattern is typical for the overstrong meridional stratospheric circulation in chemistry-transport models (de Laat et al., 2007). It also shows that the assimilation – on average – counters this O_3 build up. It explains why biases in the model-sonde UTLS O_3 improve when O_3 profiles are assimilated despite worse rms differences (Table 1). The overstrong circulation is a long-term process affecting the average ozone whereas the assimilation affects individual days and profiles, therefore short synoptic-time variability and thus rms differences. Summarizing, we find that the effect of assimilating GOME O_3 profiles compensates for a known model bias and thus improves the analysis with regard to the long term variations in UTLS O_3.

4.6 Evaluation of TORA residuals with O_3 sondes

The comparison of sonde measurements and modelled O_3 – either without or with assimilation of GOME observations – showed that there are still large differences on a day-to-day basis in tropospheric and lower stratospheric O_3. Furthermore, the assimilation of the GOME O_3 profiles gave only limited improvements in day-to-day UTLS O_3 at tropical latitudes and mainly degraded UTLS O_3 at mid-latitudes. Since it is crucial to be able to separate stratosphere from troposphere, the presence of biases and the limited improvement – if any – due to the assimilation very well may not have lead to an improvement in estimating the stratospheric O_3 column.

Figure 4 shows the TORA residuals from the GDP v4.1 total O_3 columns minus the assimilated SOCs and the corresponding O_3 sonde TTOC's (when available) for two locations also shown in Fig. 2 (Payerne and Ascension). The comparison for Payerne shows that variations in the individual TORA residuals are unrealistically large compared to the sonde observations. This type of behaviour is representative for all mid-latitude locations. For the tropical location the comparison looks better although there are only a limited number of collocations with sondes available for validation.

The monthly means for the TORA residuals and O_3 sonde TTOC's show similar seasonal cycles, but the seasonal cycles in the residuals are too large. Note that using total column observations from a different algorithm (TOGOMI instead of GDP v4.1) yielded similar results, as did the use of the model simulation without assimilation, which is unsurprising given earlier findings about the quality of the free-running TM5 simulation. Furthermore, it should be noted that many earlier studies have shown that within the tropics other – simpler – residual methods gave satisfactory results (see Tarasick and Slater (2008) for an overview). In addition, Liu et al. (2006) have shown that directly measured tropical TTOC's from GOME O_3 profiles agreed very well with model simulations, a result that we could reproduce in great detail with the GOME O_3 profiles used in this study (not shown). It is reassuring that the TORA residual method does not appear to degrade the residual tropical tropospheric O_3 columns.

Table 3 shows the statistical evaluation of the TORA residuals. Note that the number of collocations with sonde observations was reduced compared to previous model-sonde comparisons due to fewer GOME GDP total column observations. The overall picture is that, with only a few exceptions, correlations and rms differences degrade
5 when using the assimilated stratospheric columns, often significantly. Nevertheless, for quite a few locations the bias still improves, showing that the assimilation increases the "noise" in the residual but not the average differences. At high southern latitudes very large biases occur due to the fact that neither the model simulation nor the GOME O_3 profiles could provide realistic ozone hole O_3 profiles. In addition, opposite to the
10 comparisons between modelled and measured TTOC and UTLS columns, the correlations do not improve for the TORA residuals going from individual columns to monthly mean columns (Rm, Table 3).

Figure 5 shows a comparison of the mean TORA residuals using respectively GOME GDP and TOMS v8 total columns for the year 2000. For comparison we added the
15 TTOC from a TM4 chemistry-transport model simulation using a full tropospheric chemistry scheme and realistic emission estimates (RETRO, 2007). The TORA residuals using the model simulations with and without assimilation are larger than the full-chemistry TM4 modelled TTOC's and show some realistic features, for example the biomass burning O_3 maximum over the equatorial Atlantic, high TTOC values around
20 the subtropical jets (30° N/S) and low columns over the equatorial Pacific. However, small and even negative residuals are found around Antarctica due to the lack of an ozone hole in the TM5 model simulations as well as in the GOME O_3 profiles. Although results might appear realistic at mid and high latitudes, given the large amount of noise for individual TORA residuals it remains questionable whether the spatial variations of
25 the residuals contains much useful information. The spatial coherence of residuals from either GOME or TOMS total columns is an indication that the lack of useful information is related to the (assimilated) stratospheric O_3 column – in particular to the quality of the GOME O_3 profile observations – and not a problem with the total column observations. Other years yielded similar results in terms of representativeness of the

TORA residuals and coherence by using either GOME or TOMS total O_3 columns.

5 Discussion and conclusions

This paper presents an evaluation of level-2 residual tropospheric O_3 columns derived from subtracting free-run and assimilated model stratospheric O_3 columns from total column observations, the so-called TORA method. TORA residuals have been calculated using GOME O_3 profile and GOME total O_3 column observations for the six year period 1996–2001.

The comparison of model simulations without assimilation for the UTLS region (O_3 column between 250 and 50 hPa) with independent O_3 sonde observations shows that the TM5 model with the CARIOLLE chemistry scheme produces realistic O_3 variations in the UTLS, both on short day-to-day as well as longer seasonal timescales, although for the tropics some tropospheric biases occur because of the use of the zonal-mean tropospheric O_3 climatology. The agreement improves for monthly and climatological means. The results also show that the rms differences are rather good for middle and high latitudes.

When assimilating GOME O_3 profiles in the TM5 model simulation the UTLS biases are reduced in comparison to O_3 sonde data. Within the tropics correlations and rms differences also improve slightly. On the other hand, correlations and rms differences outside of the tropics degrade albeit not very significantly. This result can be explained as follows: mid-latitude stratospheric O_3 in TM5 has a tendency to slowly increase over years, an effect that is related to the overstrong stratospheric meridional circulation generally present in chemistry-transport models driven by off-line analysed meteorological fields (Douglass et al., 2003). The overstrong circulation pumps too much O_3 from the tropical stratosphere (O_3 production) to the stratosphere at higher latitudes where O_3 accumulates as destruction remains similar but transport increases. The assimilation of GOME O_3 profiles counters this effect, reducing the bias in average UTLS O_3.

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At the same time the increase in rms differences is likely related to the very large footprint of the GOME pixel (960×80 km), which is larger than the typical correlation length of upper tropospheric (∼100 km) and lower stratospheric (∼300–400 km) O_3 variations. Hence, the assimilation introduces errors in single model O_3 profiles as much as it improves the average representation of UTLS O_3. Additional errors may be introduced by retrieval uncertainties, including the vertical smoothing of the GOME O_3 profiles and instrumental errors. The latter are not analyzed here, but are subject of another paper (Mijling et al., 2009).

Within the tropics the TORA residuals appear realistic and the assimilation helped to slightly reduce the bias in modelled UTLS O_3 compared to ozone sondes. Outside of the tropics the day-to-day variations in the individual residuals are much larger than the observed variability – even yielding negative values. Using both GOME and TOMS total column observations results in very similar spatial patterns of the average residuals, showing that the too large day-to-day variations are not related to the total column observations but to the (assimilated) stratospheric O_3 columns. The tropical stratospheric column shows much less variability than mid-latitude stratospheric columns, providing an additional indication that the GOME O_3 profile observations are too coarse to fully capture UTLS O_3 variability outside of the tropics. Given the results from the validation with sonde observations it is not very likely that the spatial variations in the annual means and the interannual variability contain valuable information outside of the tropics.

Nevertheless, the results also clearly point in the direction where to improve the TORA methodology to obtain realistic residuals outside of the tropics. The spatial resolution of the model can be improved to at least 1°×1°, and preferably better, so that mid-latitude UTLS O_3 variations can be more realistically simulated. Realistically means that the model resolution should be smaller than the empirically established correlation length of UTLS variations, which is about 100 km or 1° according for the upper troposphere according to Sparling et al. (2006). In addition, the observations that are assimilated should preferably have a footprint that is at least of similar size as

the model simulation on $1°×1°$, and also preferably smaller.

Schoeberl et al. (2007) presented results for a similar-but-different methodology, similar in a sense that the method also assimilated observed O_3 profiles, but different in sense that a different assimilation approach and different observations were used (O_3 profiles from the Microwave Limb Sounder). Nevertheless, with a better spatial resolution and using MLS stratospheric O_3 profiles – which have a much better vertical resolution – realistic residuals were derived and they found realistic O_3 variations on relatively small spatio-temporal scales (roughly $1°×1°$ resolution for a five-day average).

Important for the TORA methodology is that considerable improvement in O_3 profile quality and accuracy has been reported for GOME-2 O_3 profiles compared to GOME O_3 profiles (van Peet et al., 2009), while at the same time GOME-2 O_3 profiles can be retrieved with the necessary smaller footprints of $80×40$ km. Future application of the TORA methodology may also use OMI O_3 profiles having an even smaller footprint ($60×30$ km) than GOME-2.

A possible drawback for the use of for example GOME-2 is that the number of observations that need to be assimilated increases considerably. The assimilation step is by far the time limiting step in the TORA methodology, hence increasing the number of observations to be assimilated increases the computational time accordingly. Realistic results might be achieved by using only a subset of all available high-resolution O_3 profile observations.

Summarizing we conclude that the TORA methodology to get level-2 tropospheric O_3 columns by subtracting best-estimate stratospheric O_3 columns from total O_3 column observations is valuable for past and present-day satellite observations, and using GOME total O_3 column and O_3 profile observations it provides realistic tropospheric O_3 columns for tropical latitudes. Outside of the tropics, accurately estimating the tropospheric O_3 column is hampered by the too large ground pixel of the GOME O_3 profile observations and possible insufficient accuracy of the O_3 profile observations as well as a too large horizontal resolution of the TM5 assimilation model. Recommendations for future application of the TORA methodology are to use O_3 profile observations with

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a smaller spatial footprint – preferably 1°×1° degrees or smaller – and a better horizontal resolution of the assimilation model – preferably 1°×1° degrees or smaller. UV-VIS O_3 profile observations with the required spatial footprint are becoming available from the GOME-2 and OMI instruments, which in the near future will be used for applying the TORA methodology.

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Table 1. TM5 evaluation of UTLS O_3 columns (250–50 hPa) for a subset of available WOUDC sonde observations. Locations are ordered according to their latitude. The statistics shown here are the standard Pearsons correlation coefficient (R) for the six year period 1996–2001, the correlation for monthly means (Rm), the correlation for the 6-year climatological monthly means (Rcl), the average bias (Δ; sonde-model) and the root-mean-square differences (σ). Biases and differences are in Dobson Units (DU; 2.69×10^{18} molecules/cm^2). The bold numbers indicate the better statistical values for the comparison of sonde measurements and model results with and without assimilation. "better" refers to larger correlations, smaller biases and smaller rms differences. Stations shown in Fig. 2 are indicated in grey. Statistics for all available sonde stations can be found in supplementary information Table S1 (http://www.atmos-chem-phys-discuss.net/9/11811/2009/acpd-9-11811-2009-supplement.pdf).

Station	lat	UTLS (250–50 hPa) assimilated GOME					UTLS (250–50 hPa) no assimilation				
		R	Rm	Rcl	Δ	σ	R	Rm	Rcl	Δ	σ
stn315	80.0	0.60	0.74	0.92	25.4	32.7	**0.63**	**0.77**	0.92	**18.2**	**31.6**
stn089	78.9	0.56	0.73	**0.96**	9.6	**32.1**	**0.57**	**0.75**	0.94	**3.2**	32.3
stn024	74.7	0.76	**0.85**	0.95	20.0	27.9	**0.78**	0.84	0.95	**12.9**	**26.6**
stn262	67.4	0.72	0.82	0.97	**1.6**	25.1	**0.76**	**0.85**	0.97	–2.0	**23.5**
stn043	60.1	0.79	0.89	0.98	6.5	23.8	**0.85**	**0.93**	**0.99**	**3.1**	**20.8**
stn077	58.7	0.82	0.89	0.98	17.8	26.5	**0.84**	**0.91**	0.98	**9.6**	**24.6**
stn316	52.1	0.73	0.85	0.97	**–1.3**	23.1	**0.80**	**0.89**	**0.98**	–6.2	**20.3**
stn156	46.5	0.76	0.88	0.98	**1.4**	19.2	**0.84**	**0.93**	**0.99**	–4.0	**15.9**
stn012	43.1	0.88	0.95	**1.00**	**–0.9**	20.2	**0.92**	**0.96**	0.99	–10.3	**16.6**
stn107	37.8	0.72	0.81	0.97	5.8	20.8	**0.74**	**0.82**	**0.98**	**0.8**	**20.0**
stn190	26.2	0.45	0.48	0.60	**–11.7**	**7.7**	**0.47**	**0.54**	**0.69**	–14.2	8.1
stn109	19.4	**0.68**	**0.79**	**0.88**	**–1.9**	**6.8**	0.66	0.75	0.87	–3.8	7.2
stn435	5.8	**0.43**	**0.58**	**0.80**	2.3	**5.3**	0.34	0.55	0.78	**1.0**	6.1
stn434	–0.9	**0.33**	**0.43**	**0.69**	**–4.0**	**5.3**	0.32	0.42	0.67	–4.5	5.5
stn175	–1.3	**0.46**	**0.62**	**0.85**	**0.2**	**4.9**	0.45	0.61	0.84	–0.9	5.1
stn219	–5.9	**0.37**	**0.53**	**0.87**	**–3.1**	**5.3**	0.29	0.49	0.82	–4.1	5.8
stn328	–8.0	**0.49**	**0.68**	**0.83**	**–0.6**	**5.1**	0.44	0.64	0.77	–1.6	5.5
stn191	–14.2	**0.71**	0.81	0.91	**–4.3**	**3.9**	0.70	0.81	**0.92**	–5.0	4.2
stn438	–18.1	0.57	0.67	0.93	**–4.7**	8.7	**0.62**	**0.70**	**0.95**	–6.3	**8.5**
stn436	–21.1	**0.65**	**0.73**	**0.77**	**–1.5**	**5.9**	0.63	0.66	0.68	–3.2	7.0
stn394	–37.7	0.81	0.91	0.96	**–1.5**	15.5	**0.82**	**0.92**	**0.97**	–9.2	**15.1**
stn256	–45.0	0.83	0.94	0.97	**0.5**	15.8	**0.88**	**0.96**	**0.98**	–6.4	**13.3**
stn029	–54.5	0.71	0.80	0.95	**1.0**	20.5	**0.79**	**0.84**	**0.97**	–5.8	**18.1**
stn233	–64.2	0.48	0.39	**0.59**	–10.3	29.1	**0.55**	**0.49**	0.56	**–14.4**	**27.7**
stn101	–69.0	0.53	0.65	0.75	**–8.7**	35.3	**0.66**	**0.72**	**0.78**	–12.5	**32.8**

Table 2. As Table 1 but for the total tropospheric O_3 columns (TTOC's), based on ECMWF tropopause heights. Locations are ordered according to their latitude. The statistics shown here are the standard Pearsons correlation coefficient (R) for the six year period 1996–2001, the correlation for monthly means (Rm), the correlation for the 6-year climatological monthly means (Rcl), the average bias (Δ; sonde-model) and the root-mean-square differences (σ). Biases and differences are in Dobson Units (DU; 2.69×10^{18} molecules/cm^2). The bold numbers indicate the better statistical values for the comparison of sonde measurements and model results with and without assimilation. "better" refers to larger correlations, smaller biases and smaller rms differences. Stations shown in Fig. 2 are indicated in grey. Statistics for all available sonde stations can be found in supplementary information Table S1 (http://www.atmos-chem-phys-discuss.net/9/11811/2009/acpd-9-11811-2009-supplement.pdf).

Station	lat	TTOC (surf-tropP) assimilated GOME					TTOC (surf-tropP) no assimilation				
		R	Rm	Rcl	Δ	σ	R	Rm	Rcl	Δ	σ
stn315	80.0	0.48	0.43	**0.47**	**−3.8**	6.7	**0.57**	**0.49**	0.40	−3.9	**5.3**
stn089	78.9	0.43	0.58	0.86	**−4.0**	7.7	**0.53**	**0.73**	**0.94**	−4.7	**6.6**
stn024	74.7	0.41	0.38	0.67	−6.8	7.8	**0.46**	**0.50**	**0.72**	**−6.0**	**6.6**
stn262	67.4	0.48	0.67	0.69	**−5.7**	8.0	**0.74**	**0.81**	**0.90**	−8.1	**4.8**
stn043	60.1	0.47	0.64	0.86	**−3.6**	8.1	**0.71**	**0.82**	**0.94**	−6.5	**4.6**
stn077	58.7	0.45	0.61	**0.93**	**−3.9**	8.3	**0.54**	**0.71**	0.90	−5.1	**6.6**
stn316	52.1	0.29	0.36	0.70	**−7.0**	10.7	**0.57**	**0.64**	**0.83**	−10.7	**6.6**
stn156	46.5	0.51	0.79	0.86	**−1.5**	8.2	**0.65**	**0.83**	**0.90**	−4.2	**6.1**
stn012	43.1	0.65	0.76	0.92	**−7.9**	7.6	**0.80**	**0.89**	**0.98**	−7.7	**5.4**
stn107	37.8	0.67	0.83	0.93	**−3.3**	8.0	**0.74**	**0.87**	**0.95**	−4.1	**6.4**
stn190	26.2	0.60	**0.66**	**0.72**	**−9.6**	8.0	0.60	0.65	0.68	−11.5	**7.7**
stn109	19.4	0.75	0.86	0.93	**−10.9**	6.8	**0.77**	0.86	0.93	−11.6	**6.4**
stn435	5.8	0.19	0.22	0.22	**−2.7**	6.5	**0.33**	**0.55**	**0.74**	−4.0	**5.7**
stn434	−0.9	**0.57**	**0.68**	**0.85**	**−11.3**	3.9	0.51	0.65	0.82	−13.4	4.0
stn175	−1.3	0.34	0.24	**0.35**	**−1.8**	5.5	**0.43**	**0.34**	0.27	−2.9	5.0
stn219	−5.9	0.61	0.74	0.89	**−3.1**	6.5	**0.67**	**0.76**	**0.90**	−4.3	**6.2**
stn328	−8.0	0.57	0.66	0.94	2.8	7.8	**0.64**	**0.74**	**0.97**	**1.3**	**7.4**
stn191	−14.2	0.62	0.64	0.74	**−13.3**	5.4	**0.68**	**0.70**	**0.81**	−15.0	**4.8**
stn438	−18.1	0.69	0.77	0.86	**−12.3**	6.1	**0.70**	0.77	0.86	−15.4	**5.7**
stn436	−21.1	0.54	0.66	0.78	**−1.2**	7.0	**0.69**	**0.76**	**0.84**	−6.5	**5.7**
stn394	−37.7	0.33	0.36	0.59	0.3	7.9	**0.54**	**0.71**	**0.79**	**−2.1**	**5.8**
stn256	−45.0	0.43	0.45	0.62	**−4.6**	6.4	**0.64**	**0.72**	**0.73**	6.3	**3.8**
stn029	−54.5	0.36	0.53	**0.75**	**−1.7**	7.7	**0.63**	**0.70**	0.69	−2.7	**5.0**
stn233	−64.2	0.33	0.66	0.75	−2.7	9.2	**0.52**	**0.69**	**0.79**	**−2.1**	**7.0**
stn101	−69.0	0.26	0.41	**0.67**	−1.4	9.1	**0.42**	**0.46**	0.63	**−0.9**	**6.3**

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Table 3. As Table 2 but for the TORA residuals, i.e. total O_3 column observations minus strato-spheric O_3 columns from the TM5 CARIOLLE assimilation of GOME OPERA O_3 profiles. For the total O_3 columns the GOME GDP v4.1 product was used. Climatological monthly mean statistics were not calculated due to insufficient sonde collocations for most sonde stations. Locations are ordered according to their latitude. The statistics shown here are the standard Pearsons correlation coefficient (R) for the six year period 1996–2001, the correlation for monthly means (Rm), the average bias (Δ; sonde-model) and the root-mean-square differences (σ). Biases and differences are in Dobson Units (DU; 2.69×10^{18} molecules/cm^2). The bold numbers indicate the better statistical values for the comparison of sonde measurements and model results with and without assimilation. "better" refers to larger correlations, smaller biases and smaller rms differences. Stations shown in Fig. 3 are indicated in grey. Statistics for all available sonde stations can be found in supplementary information Table S1 (http://www.atmos-chem-phys-discuss.net/9/11811/2009/acpd-9-11811-2009-supplement.pdf).

| | | TORA residual | | | |
| | | GDP v4.1 minus TM5/GOME | | | |
Station	lat	R	Rm	Δ	σ
stn315	80.0	−0.03	−0.05	**0.1**	38.7
stn089	78.9	0.03	0.08	8.4	38.9
stn024	74.7	−0.10	0.06	−10.9	31.9
stn262	67.4	−0.38	−0.64	**−0.2**	20.4
stn043	60.1	0.02	0.06	−7.8	35.8
stn077	58.7	−0.31	−0.34	−17.2	25.9
stn316	52.1	0.06	0.08	−9.4	24.3
stn156	46.5	0.25	0.29	**−1.4**	25.2
stn012	43.1	0.27	0.23	−15.1	25.6
stn107	37.8	0.24	0.39	**−1.4**	21.3
stn190	26.2	0.34	0.24	**−2.7**	15.2
stn109	19.4	0.19	0.30	**9.6**	14.9
stn435	5.8	−0.21	−0.18	−13.6	7.4
stn434	−0.9	0.47	0.44	**−4.5**	14.3
stn175	−1.3	0.24	0.24	3.0	12.4
stn219	−5.9	0.26	0.24	−12.0	9.6
stn437	−7.5	0.47	0.47	**−3.7**	12.3
stn328	−8.0	0.37	0.41	−11.4	12.1
stn191	−14.2	0.40	0.44	**−8.9**	13.0
stn438	−18.1	0.62	0.61	−17.9	18.7
stn436	−21.1	0.42	0.41	−12.3	13.5
stn394	−37.7	0.25	0.25	**3.0**	20.6
stn256	−45.0	0.18	0.30	**−0.7**	28.6
stn029	−54.5	−0.28	−0.48	56.8	46.5
stn233	−64.2	−0.21	−0.39	53.1	46.7
stn101	−69.0	−0.30	−0.69	46.1	44.3

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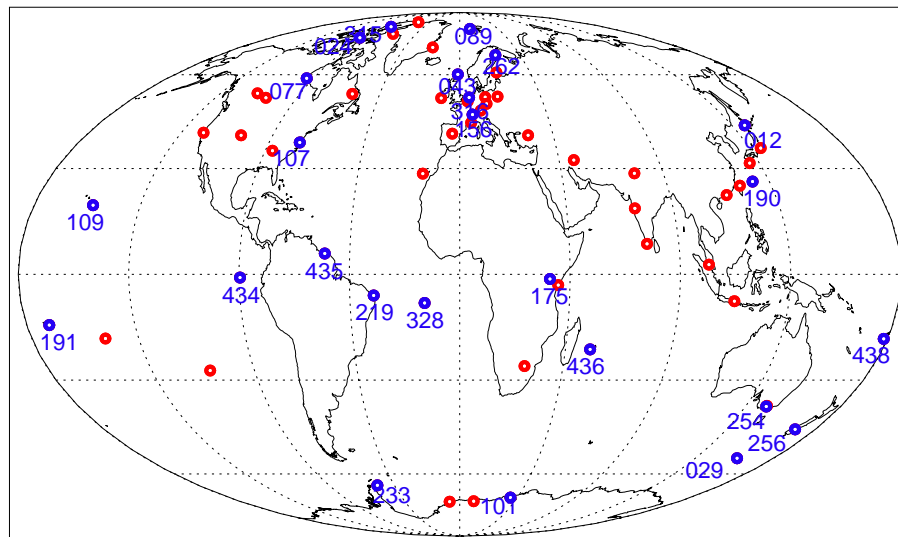
Fig. 1. Locations of WOUDC stations used in this paper. The stations used and discussed in the paper are indicated in blue, including the WOUDC station identifier (see also Table S0: http://www.atmos-chem-phys-discuss.net/9/11811/2009/acpd-9-11811-2009-supplement.pdf). Additional sondes that have been compared but not discussed in the paper are in red, the results of their comparison can be found in the supplementary information.
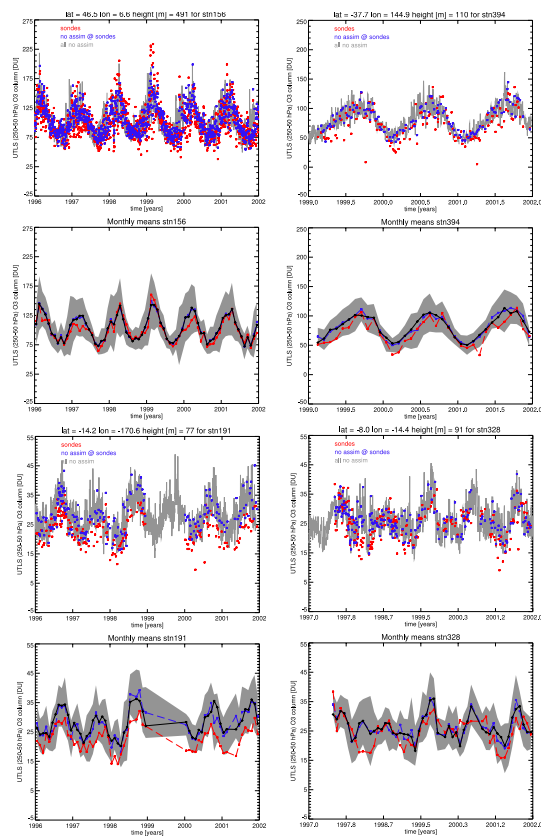
Fig. 2. Comparison of assimilated UTLS (250–50 hPa) columns and WOUDC O$_3$ sondes for the period 1996–2001 for four locations: Payerne, Switzerland (stn156); Broad Meadows, Australia (stn394), Samoa (stn191) and Ascension Island (stn328). See Fig. 1 and Table S0 for location details. The upper panels show the observed (red) and assimilated daily columns (blue markers: collocated with sondes; grey lines: all modelled columns). The lower two panels show the corresponding monthly means including the variability envelope (2σ) of all modelled daily columns within that particular month.

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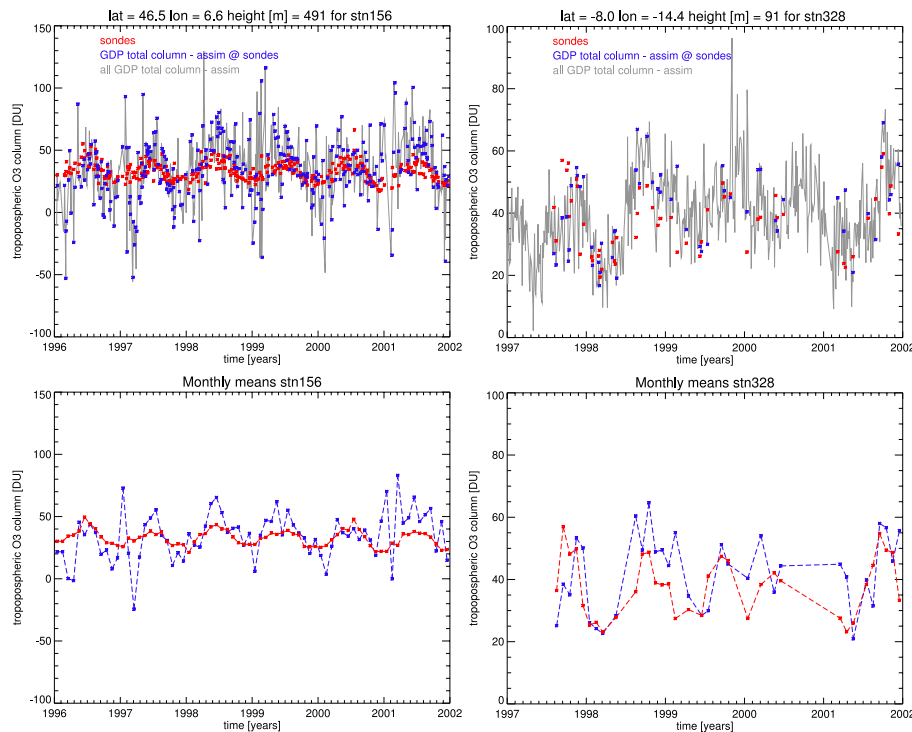Printer-friendly Version

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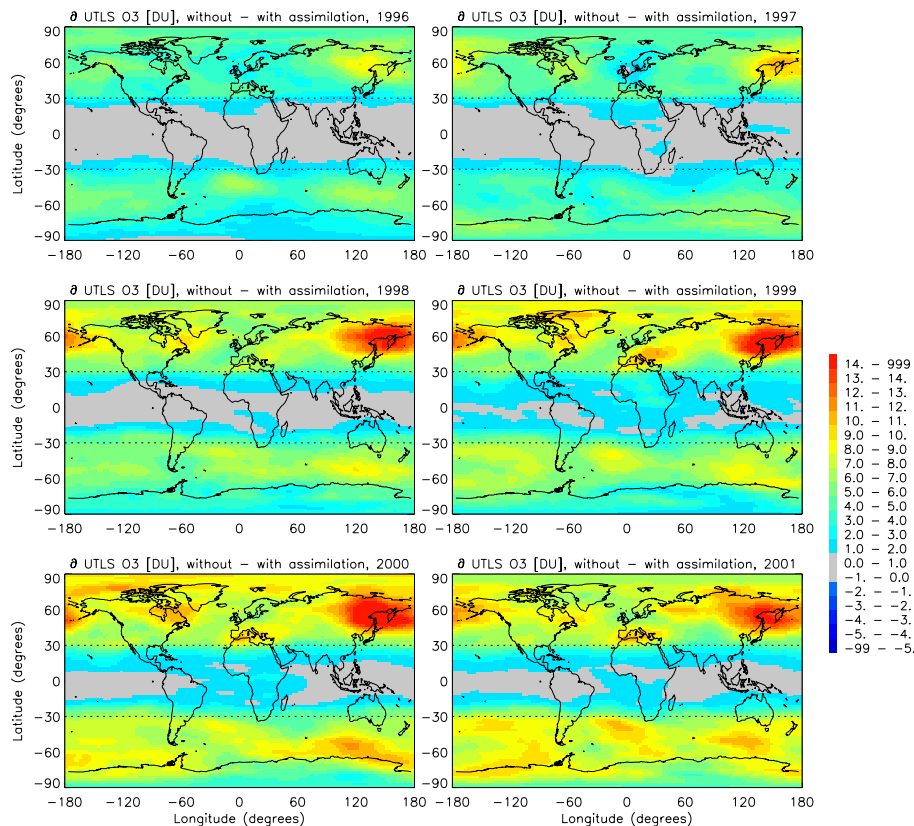
Fig. 3. Annual mean UTLS (250–50 hPa) O_3 column differences [DU] of the free running TM5 model simulation and the TM5 GOME O_3 profile assimilation run for every single year from 1996 (upper right) to 2001 (lower left). Values between −1 and +1 DU are indicated in grey.

Fig. 4. Comparison of GDP v4.1 GOME total O$_3$ columns minus the TM5 assimilated strato-spheric O$_3$ columns and WOUDC O$_3$ sondes for Payerne, Switzerland (stn156) and Ascension Island (stn328) (see Fig. 1 and Table S0 for location details). The upper two panels show the individual observed (red) and total-minus-assimilation columns (blue: GOME GDP collocated with sondes; grey: all GOME GDP columns). The lower two panels show the corresponding monthly means.

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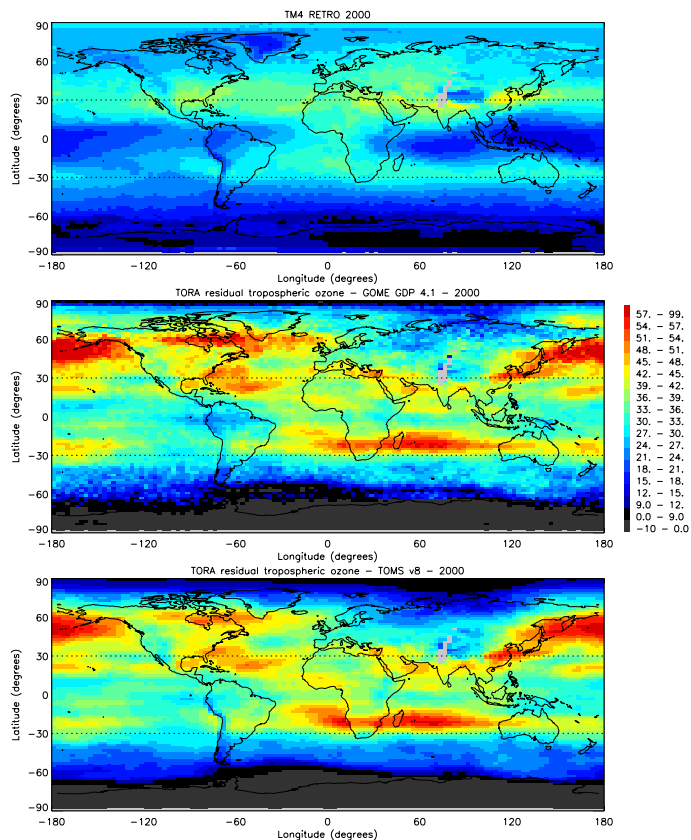
Fig. 5. Global mean total tropospheric O_3 columns (TTOC's) for the year 2000 in DU. Upper panel: TM4 simulated TTOC from the RETRO-project reanalysis simulations (RETRO, 2007). Middle panel: TORA O_3 residuals from GDP v4.1 total O_3 columns minus TM5 GOME assimilated stratospheric O_3 columns. Lower panel: as middle panel but using TOMS v8 instead of GOME GDP v4.1 total O_3 columns.