

Reply to comments of reviewer #2 on the manuscript “Tropical tropospheric ozone column retrieval for GOME-2” by P. Valks et al.

We would like to thank the reviewer for his helpful comments and suggestions. In the following, we will reply to them point by point, including the reviewer’s text in italic.

1. Introduction - The discussed method works for the tropics where a zonally invariant stratospheric column can be assumed. Could you please hint on more details (or give a reference to literature) regarding the situation in the extra tropics and your expectancies e.g. due to rapid variation of the tropopause and larger spatial variability.

We have added a short comment in the introduction about the important limitation of the CCD method that it can only be applied in the tropics (because of the assumption of a zonally invariant stratospheric column).

In section 4.2, we have extended the discussion on the validity of the CCD method for the extra-tropics. Using tropopause information from the EMAC model, we analysed the functionality of the CCD method for the sub-tropical area around 20°N and 20°S. During the winter half-year, there is a significant longitudinal variation in the tropopause height within these latitude bands, resulting in a much larger st. dev. in the above-cloud ozone and in unrealistic (erroneous) variations in the derived TOC. The large errors in the derived TOC in these situations are also visible in the ozonesonde comparisons for Hilo (20°N, 155°W) and La Reunion (21°S, 55°E) (note that these measurements are rejected for the ozonesonde comparisons described in Section 6). The criteria for the validity of the CCD method is mainly based on the number of measurements and the st. dev. in the above-cloud ozone columns (see comment below).

The authors list literature references for TOC. I recommend adding [A trajectory-based estimate of the tropospheric ozone column using the residual method, M. R. Schoeberl et. al.]
This reference has been added.

3.1 GOME-2 total ozone - The citations for GDP 4.7 refer to Loyola, 2011 (and Hao 2014), which discusses GDP 4.4. Please clarify the different algorithm versions and/or give a quick overview of changes for v4.4. to v4.7. or otherwise where to find them.

The only important change in the GDP 4.7 algorithm (compared to the GDP 4.4) is the use of Brion-Maliget-Daumont (BMD) ozone cross-sections in the DOAS slant column retrieval, which results in ~2% larger ozone columns than the ones retrieved using GOME FM98 ozone cross sections (GDP 4.4). Since for this study only GDP 4.7 ozone column have been used, we changed the corresponding sentence in the manuscript to avoid confusion: “The retrieval is performed with the GOME Data Processor (GDP) version 4.7 (Hao et al., 2014), the latest incarnation of the GDP 4 algorithm (Van Roozendaal et al., 2006; Loyola et al., 2011)”.

3.2 GOME-2 tropospheric NO₂ column - Where does the stratospheric NO₂ profile climatology come from?

This harmonic climatology (Lambert et al., 2000) has been derived from satellite measurements by UARS/HALOE (Gordley et al., 1996) and SPOT-4/POAM-III (Randall et al., 1998) and complementary information from ground-based measurements from the Network for the Detection of Atmospheric Composition Change (NDACC). The stratospheric NO₂ profiles are time dependent and given for 16 latitude bands. We have added the reference Lambert et al., (2000) in the updated manuscript.

3.3 Cloud properties - Fig 2: Please provide a small statement on observed differences in the comparison of ROCINN vs. ISCCP-D2 cloud pressure distribution.

We have added the following sentence to the caption of Fig 2: ” ... ROCINN and ISCCP-D2 cloud pressure distributions are in good agreement (see Sect 3.3 for an explanation of the plots).”

The information in Fig 2 is sparse, maybe the authors could merge the January and July plots, thus showing combined plots in only two panels.

In the updated paper, we have merged the cloud pressure and cloud albedo plots into two panels (keeping separate plots for January and July).

4.1 GOME-2 CCD method - The authors indicate that the selection criteria for deep convective clouds is $pc \leq 300$ hPa. Later (~monthly number of pixel occurrence 150 - 800) $pc \leq 350$ hPa is used. Please explain.

This is a typo. The corrected sentence is: “The number of cloudy GOME-2 pixels with $f \geq 0.8$ and $pc \leq 300$ hPa varies between 150–800 per month for each (1.25°) latitude band.”

Is there a lower limit in the number of pixels to be used? Could that be systematically handled by a dynamic criteria on the selected longitude (e.g. 70 E - 170 W). The authors state, that sub-tropical air is sometimes present in the outer latitude bands (15–20 N or 15–20 S), resulting in a small number of deep-convective cloud tops etc. The boundary for the GOME-2/CCD analysis will be limited to lower latitudes. What is the criteria (e.g. number of deep convective clouds) to determine such a situation and which is the updated latitude band.

For the calculation of the stratospheric ozone column, we use both a lower limit for the number of cloudy GOME-2 pixels per month for each latitude band (in the eastern Indian Ocean/ Western Pacific), and an upper limit for the st. dev. in the above-cloud ozone columns. Between 12.5°N and 12.5°S, the number of cloudy GOME-2 pixels (with $f \geq 0.8$ and $pc \leq 300$ hPa) is > 300 for each (1.25°) latitude band, and the st. dev. < 5 DU. The CCD method can normally be applied for this equatorial region without any restrictions. In the latitude bands pole wards of 12.5°N and 12.5°S, the number of cloudy GOME-2 pixels can be significantly smaller (< 300), and the st. dev. in the above-cloud ozone columns can become larger, especially in the winter month. We use a lower limit of 50 for the number of cloudy GOME-2 pixels and an upper limit of 5 DU for the st. dev. (note that these criteria depend on the spatial resolution of the satellite instrument and the latitude-longitude grid size). Only when these criteria are met, the CCD method is applied for the corresponding latitude band. We have clarified this in the updated manuscript.

5 Tropical tropospheric ozone and NO distributions - Fig 5. It would be interesting to see as well GOME-2A data here. Please indicate why this data has not been considered (e.g. signal degradation, horizontal resolution).

We agree with the reviewer that it is interesting to add the TOC data from GOME-2A for Oct. 2013 to Fig. 5 of the paper. Fig. 1 below shows the TOC for Oct. 2013 for both GOME-2A and GOME-2B. There is a good agreement between the TOC of GOME-2A and GOME-2B, but the GOME-2B TOC data generally show a small negative off-set of ~1-2 DU. Analysis of the GOME-2 (level-2) total ozone column data for 2012 and 2013 indicate that this off-set is related to a small negative bias in the GOME-2B total ozone column for cloud-free measurements. The reason for this small cloud fraction dependent offset in the GOME-2B total ozone data is not known, and is under investigation. The increased spatial resolution of GOME-2A (40x40 km²) does not seem to have a large impact on the monthly averaged TOC distribution (the number of GOME-2 measurements in each 1.25°x2.5° grid cell within one month is generally similar for GOME-2A and GOME-2B, and the different (daily) coverage and revisiting times of the two instruments do not have a large impact on the monthly averaged TOC field).

The instrument degradation of the GOME-2 instruments only has a small effect on the level-2 total ozone columns retrieved with the GDP v4, as described in Hao et al., 2014. We did not notice any significant degradation effects on the GOME-2 TOC (e.g. in the comparisons with the ozone sondes in Fig. 7). This is to be expected, since the TOC is derived from the difference between cloud-free and cloudy GOME-2 ozone column measurements. In the updated manuscript, we have replaced Fig. 5 by a similar figure using both the GOME-2A and GOME-2B data for Oct. 2013. We have also added a short discussion in Sect. 6 on the insensitivity of the CCD method for the GOME-2 instrument degradation.

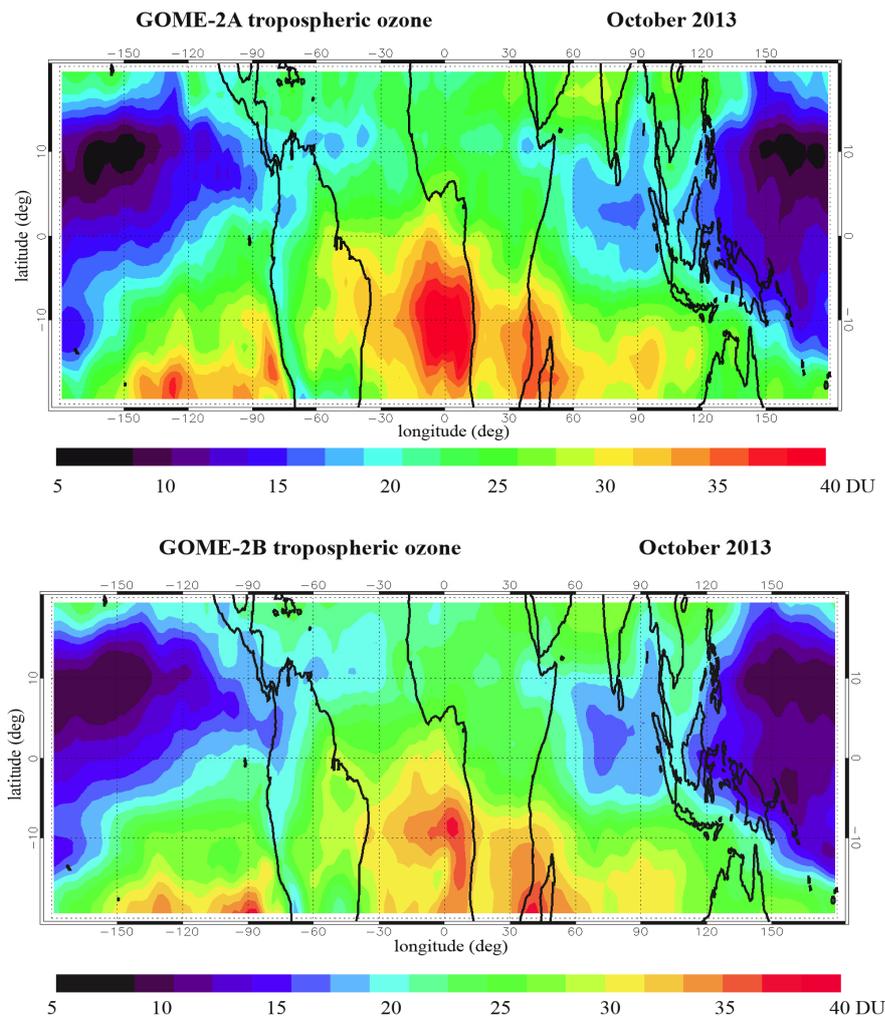


Fig 1. Tropospheric ozone columns for October 2013 as measured by GOME-2/MetOp-A (top) and GOME-2/MetOp-B (bottom).

page 741: Replace 'plate' with 'panel'
Done.

6 Comparisons with ozonesonde measurements - page 743, line 16: Samoa -> American Samoa - page 753, line 29: You mention the sparse nature of monthly GOME-2 TOC measurements. Please quantify in text and/or if possible add e.g. some histogram on number of contributing measurements to the statistics in Fig 7.

The authors state that the validation with ozone sondes is sometimes difficult because of horizontal coverage of sondes vs. satellite instruments. Still I believe that showing plots (not only information in Table 1) for all relevant SHADOZ stations adds value to the validation part of the paper.

We included the number of ozonesonde measurements in the ozone sonde comparisons in Fig. 7 of the updated paper, and added plots of the mean differences (GOME-2 minus sonde, EMAC minus sonde) (see also comment Reviewer #1).

Furthermore, the ozone sonde comparisons for the Equatorial Eastern African site Nairobi (1°S,37°E) has been added to Fig. 7, because of its position with respect to high pollution sources by countries bordering the Indian Ocean (see Fig. 2 below).

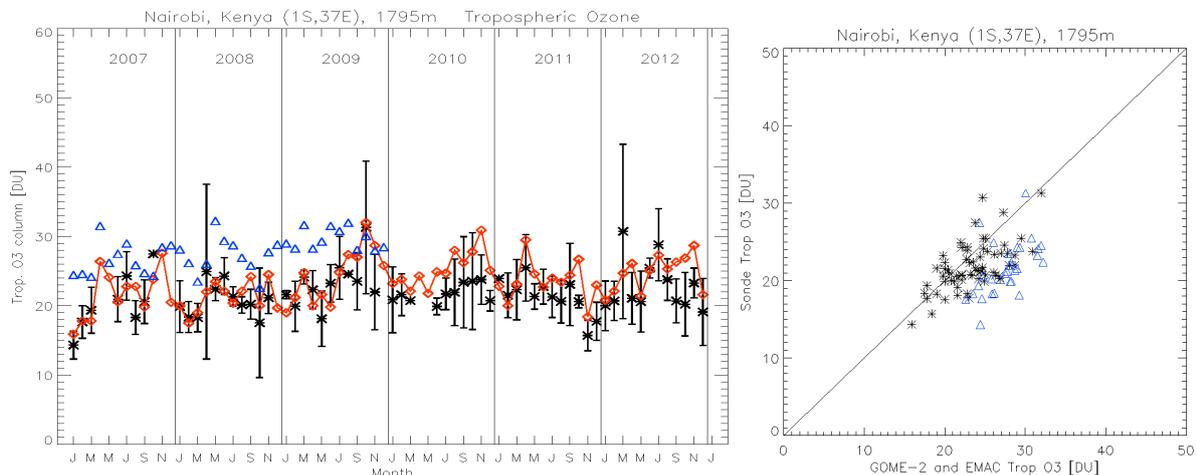


Fig. 2. Tropospheric ozone columns for Nairobi (1°S,37°E) for the period January 2007 – December 2012. The asterisks denote the integrated ozonesonde measurements (monthly ensemble average) with 1σ error bars. The red diamonds denote the tropospheric ozone columns derived with the GOME-2/CCD method; the blue triangles are tropospheric ozone columns from the EMAC model. A scatter plot of ozonesonde versus GOME-2 (black) and EMAC (blue) monthly mean TOCs is shown on the right.

7.2 Tropospheric ozone variations - For Fig. 7 the author state that the model has a larger offset for higher tropospheric ozone columns. Please explain the generally high offset for Natal station in the second half of 2007. Here the model doesn't seem to reflect the seasonal variation very well, while for Ascension Island and American Samoa the situation is better. Is this only tied to issues with the emission datasets as suggested?

The tropospheric ozone column in EMAC show a generally high bias in all months. This could in principle be related to: 1) shortcomings in the chemical kinetics (uncertain reaction coefficients, missing reactions etc.), 2) biased emissions of ozone precursors, and 3) deficiencies in tracer transport. It is difficult to judge the quantitative effects of all this, however, we can almost safely rule out point 1), and point 2) is known to give the largest uncertainty in chemistry-climate model simulations. For the transport (point 3), large scale advection can also be ruled out, since the model in the presented simulation has been nudged by Newtonian relaxation towards ECMWF operational analysis data.

The situation for Natal in the model does not seem very specific for the year 2007, the maximum here is rather flat in the other years as well (this is also visible in Fig. 7). Thus the issue is somehow persistent. It is likely that the deviation over Natal is a systematic, yet combined effect of the timing of the precursor emissions (mainly biomass burning activity in South America and Africa) and transport. Despite the nudging, smaller scale transport, in particular by convection, could under-/overestimate the vertical redistribution of emitted ozone precursors, which will alter their lifetimes and therefore also their long-range transport. This effect is non-linear and hard to quantify (we think that an in-depth model analyses is not within the scope of this paper). Yet, it is consistent with the fact that the horizontal gradient in the tropospheric column is large at this time of the year (see for instance Fig. 8), i.e., even small deviations of transported air mass characteristics could cause a time dependent high bias.

We have extended the discussion on the EMAC model results in Sect. 7 of the updated paper.

For Fig. 8 the high EMAC tropospheric ozone values in the area east of India is shortly mentioned. A difference map on a common grid between GOME-2 CCD and EMAC for the discussed month October 2009 would support this. Again, it would be useful to show more data from other months of EMAC data coverage.

We have added the EMAC trop. ozone map for Oct. 2008 to Fig. 8 of the updated paper, and the corresponding GOME-2 - EMAC difference maps. In contrast to the more general high bias of the EMAC trop. ozone columns, especially over the Near East and India (as explained in the paper), the GOME-2 vs. EMAC difference plot for Oct. 2009 also shows a negative bias in EMAC for west tropical Africa (see Fig. 3. below). This EMAC trop. ozone “minimum” could be explained by an underestimation of precursor emissions (from biomass burning and lightning), since the trop. ozone column in this area is very sensitive to changes in precursor emissions (e.g. see Fig 3. in Sauvage et al., 2007). In addition, dynamical processes could play a role as well (i.e. too strong convective uplift and subsequent large scale transport). However, we think that an in-depth model analyses is not within the scope of this paper.

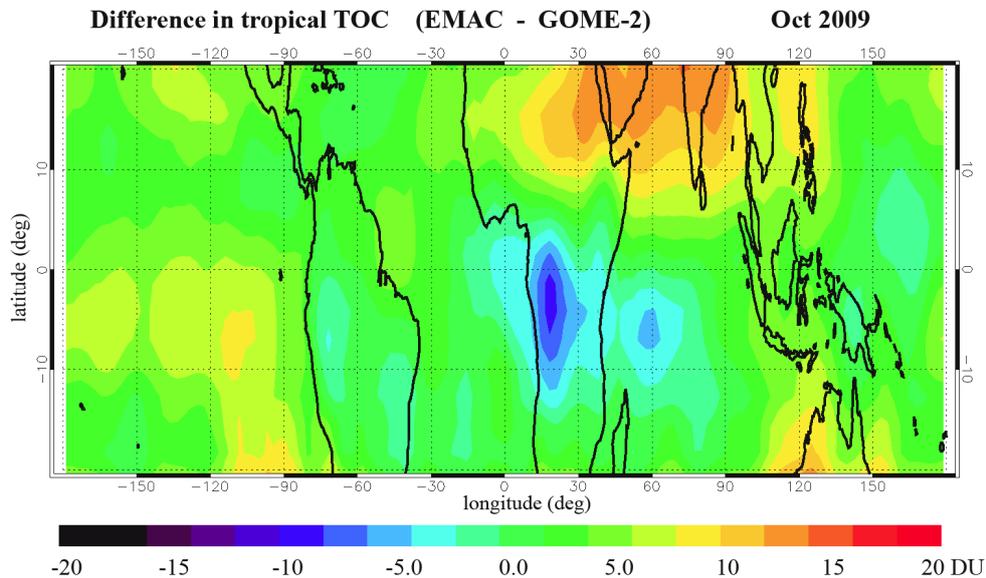


Fig. 3. Difference between the tropical TOC calculated with the EMAC model and the TOC derived with the GOME-2/CCD method for Oct. 2009.

*Figures - At least Fig 2, 4, 6, 7 need larger labels.
The labels of Fig 2, 4, 6 and 7 have been enlarged.*