K.-P. Heue (Referee)

klaus-peter.heue@dlr.de

Received and published: 18 February 2016

Referee comment on:

Tropical tropospheric ozone columns from nadir retrievals of GOME-1/ERS-2, SCIAMACHY/Envisat, and GOME-2/MetOp-A (1996-2012) by: Elpida Leventidou, Kai-Uwe Eichmann, Mark Weber, and John P. Burrows

Summary

The paper describes the convective cloud differential algorithm for tropospheric ozone column (TOC) and its application to the WFDOAS level 2 Ozone columns from GOME, SCIAMACHY and GOME-2A. The data are compared to sondes from the SHADOZ network and to TOC from SCIAMACHY's limb nadir matching. General remark The paper is well written and to large parts well structured it includes a good error discussion of the CCD algorithm. Why do the authors stop the timeseries in 2012- while GOME-2 on MetOp-A is still in operation? A detailed comparison between the data sets would be nice. The authors claim to have a 17 years time period, but how good do the data agree in the overlapping periods? The discussion of Figure 6 focuses on the overall TTOC distribution pattern.

As mentioned in the conclusions, the authors are planning to harmonise the three datasets and extend the dataset up to today in a coming paper. The differences in the overlapping periods are investigated in order to define correction factors to be applied.

Detailed Discussion

1 Introduction

L28 The authors explained above that tropospheric ozone has two precursors NOx and VOCs, Lightning does not produce VOCs, only NO or O3 itself.

The phrase "and non-human activities, such as lightning" has been removed and later in the paragraph the following text has been added to comment on natural ozone precursors:

"Considerable amounts of NOx in the tropics are also produced by lightning (upper troposphere), natural savannah burning and by microbial nitrification and denitrification of the soil."

L30: "The precursors interact with convective systems" I am not sure "interact" is the correct word here, because this implies that the convective system is influenced by the ozone precursors. Replace by "are transported by" or "lifted up by"

The following paragraph has been added to explain better the impact of convection on tropospheric O3:

"Tropospheric ozone presents considerable variability, mainly due to its chemical lifetime which coincides with the timescale of weather systems. The globally averaged tropospheric ozone lifetime is 22 ± 2 days (Stevenson et al., 2006). On the other hand, ozone's lifetime in the boundary layer is much shorter (a few hours) because it is more probable to get destroyed by surface deposition and chemical reactions, whereas in the middle and upper troposphere its lifetime is on the order of weeks to months (Cooper et al., 2014). There are several ways that convection impacts on tropospheric ozone and its precursors. First, convection can redistribute tropospheric ozone burden via vertical mixing. Lower tropospheric ozone is lifted up to the upper troposphere (UT) where O_3 lifetime is longer, while due to mass balance UT air rich in ozone mixes and submerges into regions where O_3 lifetime is shorter. As a result, the UT O_3 as well as the overall tropospheric O_3 column decreases (Doherty et al., 2005). Second, convective systems such as tropical cyclones, can transport ozone precursors many kilometers away from their source, resulting in ozone production at remote areas where it builds up (Sauvage et al., 2006)."

L30: the rest of the sentence seems somehow doubled, and confusing

The paragraph has been divided and modified into two paragraphs as suggested by Reviewer 1. The first describes the role of ozone as an oxidizing agent and a greenhouse gas and the second presents the influence of convection and different ozone lifetimes in different tropospheric altitudes to tropospheric ozone abundance.

3 The CCD method

In the description of the CCD method a first step is missing. In the level 2 data total columns are stored, how is the ACCO retrieved? The discussion about the different cloud product for SCIAMACHY (SACURA) and GOME/GOME-2 (ROCINN) is a bit unclear and not well structured. Moreover it is partly mixed with detailed studies on the ACCO columns for different CTH or cf. It may partly be shifted to section 2? Separate between the different algorithms (section 2) and the influence on the ACCO (section 3) and reference the respective sections.

- WFDOAS for GOME-2 does not use ROCINN but FRESCO+, it was a mistake that has been corrected in the document.
- The whole Section 3 is focused on the above cloud column ozone (ACCO) calculation and the assumptions and corrections made.
- The second paragraph of Section 3 describes the basic idea of the original CCD technique that the ozone column above deep convective clouds rerpesents the stratospheric ozone column. For the ACCO the basic assumption of longitudinal invariability is made.
- The third paragraph comments on the natural variability of cloud top height and the limitations of the cloud algorithms used and their influence on the ACCO calculation.
- The fourth paragraph discuss the assumption that the top of deep convective clouds defines the tropopause. It explains that this usually does not happen since these clouds are somehow lower and therefore a fixed level of 200 hPa has been selected to adjust the ACCO calculation with the help of an ozone climatology.

L145: "These clouds" refers to those DCCs overshooting the TTL, but this not meant here. I assume the authors mean normal large convective clouds with cloud top heights above 7 or 9 km respectively.

The phrase "these clouds", refers to the DCCs that sometimes overshoot the tropopause so, their retrieved top will be above the tropopause breaking the assumption that their top defines the tropopause height.

L165: "Valks et al. (2014), although it is not documented in detail" I am not sure the authors read the paper by Valks et al. carefully enough: P2517 "To that end, a small correction has been made for the difference between the cloud pressure level and the 200 hPa level (typically 0–2 DU), assuming a constant (small) ozone volume mixing ratio of 5 ppbv (see Sect. 4.2)." Valks et al. (2014) used a constant mixing ratio for the correction and not a climatology as it is used by the authors. Because the correction term is small (2 DU) the difference between the 5 ppbv constant profile and the climatology might be negligible. In Figure 4 a) it seems the correction column might be up to 10 DU. This is much higher than the columns mentioned in Valks et al 2014. Is this related to the cloud product, does it influence the TTOC.

The part referring to the correction term used from Valks et al. (2014) has been rewritten as follows:

" Another correction approach for the difference between the cloud pressure level and the 200 hPa level was also used by Valks et al. (2014) assuming a constant (small) ozone volume mixing ratio of 5 ppbv. Valks et al. (2014) concluded that the correction term is small (less than 2 DU) and, therefore, the difference with the climatology considered to be negligible."

It is true that for GOME and GOME-2 the cloud algorithms detect the effective and not the geometrical cloud tops, resulting generally in lower clouds. We have noticed that the correction term for these instruments may reach 10 DU but these cases are excluded from the ACCO calculation because usually they belong to latitude bands with only few cloudy measurements and therefore no TTCO is retrieved.

L191: "All ACCO resulting in a negative TTOC . . . are screened out" but this depends on the local TCO, So why do screen out the complete latitude band of ACCO if for one TCO a negative TTOC is calculated, instead of removing the local TTOC?

It is not the latitude band that it is screened out, but only the daily ACCO value for that gridbox.

L191 and L194: is "daily binned" correct not "monthly"? According to the following it seems correct.

The daily binning is used only for excluding outliers in ACCO. For the final TTCO result, the TCO and ACCO are monthly binned.

L193 It is a very good idea to screen out the data with higher deviations. However, calculating the daily averages to check the deviation is time consuming. Why not taking the complete ensemble of all data in a grid cell (independent of the day) and remove the outliers (e.g. more than two or three standard deviations)

Since we are making the daily average to screen out the daily ACCO values that result in negative TTCOs we calculate also the daily standard deviations and we remove the daily ACCOs that have standard deviations more than 5 DU which is roughly 1 σ .

Results

L265 "redistribution of ozone from farther north in Africa around Namibia." This may be grammatically correct but can be misunderstood easily "Namibia is not farther North in Africa". Please clarify.

The phrase has been changed as follows:

"redistribution of ozone from north Africa to the area around Namibia"

L278: "collocated ozonesonde measurements. . .($\pm 5 \circ$ in latitude and longitude)". What does it actually mean? The CCD TTOC are gridded on 2.5° x 5° grid. The sonde station is in one grid box, then the data are averaged over the surrounding boxes up to $\pm 5 \circ$?

After the comments and from other reviewers, the comparison of CCD TTCOs with ozonesondes using a $5^{\circ}x5^{\circ}$ box around the sonde station has been replaced by the comparison with the closest gridbox that the sonde station belongs to.

In L290 ". . .from CCD covers a larger area (grid box of 2.5° by 5°)", it seems just the grid box of the sonde station is considered.

We concluded that either using the fixed grid and comparing with the box that contains the ozonesonde station or using a grid around the ozonesonde, the statistics do not change dramatically.

L299: How is the "bias" defined? Averaged difference? Difference of the averages? Intercept of the correlation? Figure 7-9 I am not sure a correlation between two data sets that both show no variation (two flat time series e.g. Kuala Lumpur) makes much sense. It might be useful for stations like Ascencion that show a pronounced annual cycle.

Bias is defined here as the difference between the estimated value (CCD) and the true value (ozonesondes). Correlation is not the only parameter used for the comparison. The relative difference and bias also indicate levels of agreement between ozonesondes and CCD.

5.2 SCIAMACHY limb/nadir matching

L359: Is the tropopause height in the SCIAMACHY data stable enough to subtract 10%, instead of calculating it correctly, under the assumption of a constant mixing ratio? The big advantage of satellite observation is the global coverage. Here I suggest not focusing on the sonde stations again but using a more global approach. Both datasets have been compared to the sondes independently and the results were satisfying. So for the comparison regional or season averages might be used. Alternatively the differences might be studied and compared to the errors or standard deviations of the two dataset.

This part has been changed and the comparison with the sondes has been removed. The two datasets are now compared directly with each other in Fig. 15 and 16.

In order to make the LNM columns comparable to the CCD TTCOs, we adjust the LNM columns to the 200 hPa level using climatological values from the Fortuin et al. (1998) climatology. Therefore, we subtract the ozone between the tropopause and the 200 hPa. The LMN data have been gridded with the same grid as used for CCD.

Acknowledgment

Please acknowledge also the work of the SHADOZ network, and your colleagues providing the level2 raw data, unless they are co-authors.

The following sentence has been added to the acknowledgment section:

"This work was supported in parts by the DLR S5P project (50EE1247) and the federal state of Bremen.The authors would like to thank the principal investigators of the Southern Hemisphere ADditional OZonesondes (SHADOZ) network, K.P. Heue and the two anonymous reviewers for their helpful comments and suggestions."

Tables and Figures

Figure 4a: Compared to the text (L 196 ff) the content of the figure is not clear: The red line is the ACCO used for August 2008. The crosses indicate the means for the first second and last ten days. But why it the difference up to 10 DU? Is it the removal of the outliers or correction to 200 hPa, or both? If it is both it might be worth to separate the effects. To include the stations Kuala Lumpur and Hilo is well justified, since they are in the reference region, or close by. But Ascencion Island is far away in the Atlantic Ocean and influenced by the African outflow (figure 4b), how about Fiji? (178°E 18.13°S) or American Samoa?

The figure has been replaced. It now shows the above cloud column of ozone (ACCO) for 2.5° latitude bands in the Indian and western Pacific Ocean from GOME-2 (cf<0.8 and cth<7km) and SCIAMACHY (cf<0.8 and cth<9km) in August 2008. Blue and yellow dashed lines show the zonal ACCO values before applying any corrections to GOME-2 and SCIAMACHY, respectively. Red and green lines are the zonal ACCO values, after corrections applied for adjusting to the 200 hPa level and screening out outlier data. The black boxes show the stratospheric ozone columns from ozonesondes. The ozonesonde stations of Natal, Nairobi, Paramaribo, and Hilo have been added. The Atlantic stations are kept on the plot because we want to see to which degree the ACCO represents the stratospheric column. For Fiji and Samoa we had no available ozonesonde launches in the selected month.

The differences before and after screening the outliers and adjusting to 200 hPa may reach 10 DU for latitudes where less cloudy ozone measurements are available (in this case at Southern tropics, since the ITCZ moves to northern latitudes in summer, see Fig. 3a on the right). The assumption made by Valks et al. (2014) that the correction term is small (less than 2 DU) and therefore the difference with the climatology are negligible can be true only for latitude bands that have enough cloudy data and the standard deviation of the monthly mean is small. After the corrections applied to GOME-2 and SCIAMACHY ACCO we can see that the agreement between them improves.

Figure 5a: The Quasi Biannual Oscillation has a strong influence on the stratospheric column. So for a more detailed study of the stratospheric column (or ACCO) I suggest to split the average according to positive or negative QBO index. How about El Niño?

This is beyond of the scope of this paper.

Figure 5b: From my point of view this figure does not add much information, so it might be skipped.

The figure has been removed

Technical corrections

L86 Later on in chapter 4 (error discussion) the abbreviation for the total ozone column is "TCO" instead of "TOC". Change in either of the two occurrences.

Done

L53: "usng" -> "using"

Done

L85 "Above Cloud Columnar Ozone" is the correct term for ACCO

Done

L166 vertical column (VC) is usually used in a different context - it often describes the total vertical column, so a more telling name for the correction term might be useful.

It is changed to Cor_{ACCO}

L198 remove blank after the bracket " (in this case.." -> "(in this case.." L200 "Kulala Lumpur" -> "Kuala Lumpur"

Done

L205f: "The agreement . . . is less than 2 DU" -> "The difference . . . is less than 2DU"

Done

L275: NOx is already explained in the introduction (L27) Skip the part in the brackets, the sentence gets too difficult to read, especially with the full stop inside.

It is shifted to the Introduction.

Conclusion

MetopB compare to introduction MetOpA and title MetOp-A

Is planned for the future.

References

In some cases a lowercase "a" is written instead of the capital "A", please recheck the references and correct. I found this error at:

All Done

L408 Bracher

L439 Ladstätter-Wießenmayer and Rozanov

L520-525 Folberth, G. MacKenzie I. Plummer, D. and Strode, S.

L436 Diab et al. Please update to ACP version

L479 PHD thesis, Author is missing

L485 Loschnigg and Webster, remove the * after Ocean

L487 Loyola et al. From -> from

L496: Rex et al. remove blank in front of the coma between title and journal: . . .Composition, Atmospheric . . .

L507: Sierk et al. Savigny, C. Von -> Savigny C. von no capital letter

L530: Valks et al. 2003. Mention the Co Authors: Valks, P. J. M., R. B. A. Koelemeijer, M. van Weele, P. van Velthoven, J. P. F. Fortuin, and H. Kelder, Variability in tropical tropospheric ozone: Analysis with Global Ozone Monitoring Experiment observations and a global model, J. Geophys. Res., 108(D11), 4328, doi:10.1029/2002JD002894, 2003.

All Done

The list of figures and list of tables are not necessary

They have been removed

Table 5: This table is quite big and the last line over writes the page number. Therefore the mean TTOC at Fiji for 2008 -2012 is not readable. It might help if you skip the unit for the TTOC for GOME-2 and SCIA? Is the bias the same as mentioned in L299?

Table 5 is now Table 2 and its size has been decreased.

Figure 7: c) replace Kuala by Kuala Lumpur Figure 7 and Figure 11: The CCD data and the sondes in these figures are the same for Paramibo and Kuala Lumpur. I failed to discover the same features in the two figures, e.g. linear increase in CCD data mid 2007 is shown in figure 7 but not in figure 11.

Kuala is replaced by Kuala Lumpur. The Paramaribo figure has been corrected. Figure 11 has been removed and replaced by Figures 15 and 16 presenting the comparisons with the LNM tropospheric columns.