Anonymous Referee #2
Received and published: 14 December 2017

Review of the paper by Leventidou et al.

The authors present in the paper a detailed trend analysis of tropospheric ozone over the tropics, using a long term homogenized data set based on satellite measurements using the Convective Clouds Differential method. This method and its application on individual satellite sensors has already been presented in various studies including a publication from the same group in AMT (Leventidou et al. 2016). In this paper they homogenize the data from three sensors and examine the variability and the trends over regions and mega cities within the tropics. The paper is well written and structured but there are many significant issues that should be considered before being accepted for publication in ACP.
The content of the paper has many similarities with the paper by Heue et al. 2016 in AMT. Although it is clear that the Heue et al., paper uses a different version of total ozone data it is not clear from the current paper what are the differences between these two data sets concerning the application of the CCD method and the resulting tropospheric ozone estimates. The authors should elaborate more here.

Our reply:

Many thanks for the comments.

The discussion on the various merging approaches is shortened and a part of it is moved into a supplement. Now we discuss mainly the lessons learned from looking at trends from differently merged datasets in Subsection 2.4. The estimation of the mean tropical trends is now limited between 15°S and 15°N since the topospheric ozone retrievals are questionable (also for the case of Heue et al. (2016)) due to the fact that the tropical borders are strongly influenced by air masses being transported from the mid-latitudes and stratospheric intrusions (Thompson et al., 2017).

At various places we have expanded on the comparisons to the Heue et al. (2016) results. Although similar instruments have been used, the results from this study and Heue et al. (2016) are different and are discussed in more detail. In page 4, line 6 we added: "The main differences between our CCD algorithm and the one developed by Heue et al. (2016) originate from the corrections that we have applied in the above cloud column calculation of GOME and GOME-2 data and handling of the outlier data (Leventidou et al., 2016)."

Our study shows that despite the fact that the same instruments are used, the trends differ. These differences can be attributed to the different harmonization/merging approaches applied in addition to the different ozone and cloud retrievals used. This paper clearly shows that the merging approach is rather a large source of uncertainty in determining tropospheric ozone trends. This is in our opinion is demonstrated for the first time in this paper.

Detailed comments:

Section 2.2

The authors attribute most of the differences between the TTCO mostly to the different cloud algorithms involved. Why they exclude eventual biases between the sensors also in the initial total columns?

For trend calculations a constant bias (in clouds and ozone) is not really an issue and can be removed using a suitable merging approach as shown here. In the periods of overlaps both total ozone and tropospheric columns agree well after applying a bias correction. In particular the lack of significant drifts in the comparison between GOME-2A and SCIAMACHY over an extended period show that the data records are quite stable. A time-varying bias (drift), however, may add significantly to trend uncertainties if not properly accounted for.

In the text we mention: "Possible reasons for the biases are the different cloud algorithms used for each instrument (SACURA for SCIAMACHY and FRESCO for GOME and GOME-2) and the small biases noticed in the total ozone columns (e.g. ~ -2.5 DU between SCIAMACHY and GOME-2). Differences in spatial resolution and overpass time of the instruments have also minor contributions in the biases."

Is there any explanation for the different behavior of GOME-SCIA differences over 10^{o}N shown in Figure 1?

The larger variation in the bias with latitude in GOME data is most likely due to the short overlap period (10 months, from August 2002 to June 2003, when GOME lost its global coverage). For GOME-2 the overlap with SCIAMACHY was more than 5 years, making the latitude dependence smoother. It should be noted that the shift in the bias at 10°N is within the uncertainty of the observed biases at these latitudes.

In the manuscript (page 5, line 22) it is now mentioned that: "GOME mean biases have stronger latitudinal variability than those of GOME-2. This behavior may be explained by the short time of common operation (Jan. 2002–Jun. 2003) between GOME and SCIAMACHY instruments."

The authors should also provide an explanation for the GOME-2/SCIA drift. Does this originate from a potential drift in the total columns?

There seems to be a positive drift in the GOME-2-SCIAMACHY difference (Fig. 1) which is quite small and statistically not significant. One possible explanation are changes in the instrument response function with time (e.g. De Smedt et al, 2012).

Reference: De Smedt, I., Van Roozendael, M., Stavrakou, T., Müller, J.-F., Lerot, C., Theys, N., Valks, P., Hao, N., and van der A, R.: Improved retrieval of global tropospheric formaldehyde columns from GOME-2/MetOp-A addressing noise reduction

and instrumental degradation issues, Atmos. Meas. Tech., 5, 2933–2949, doi:10.5194/amt-5-2933-2012, 2012.

Section 2.3.
The discussion of six scenarios in the paper is confusing, since they don't differ substantially concerning the outcome. I think the authors should just describe
here the chosen approach of harmonization.

We think that this section is one of the most important results of this paper. In the six scenarios we checked different reasonable assumptions on how to handle the differences between the individual instruments. We show here that the hamonisation procedure (merging) is one of the largest error sources of the trends since the trends derived from the various merged dataset show larger differences than the statistical uncertainty of the trend derived from any of the single dataset (one of the six). This is usually neglected in other studies. We, however, shortened that section a bit and moved some of the figures to the supplementary material. The added uncertainty from the merging approach is also discussed in more detail in the summary section.

Section 3.3.1.
The authors present regional trends in this sections. The choice of the regions to my understanding is based only on the significance of the trends and in a sense this looks like a random choice. Do these regions have some special characteristics that have to do either with prevailing dynamic features or emission sources? In general the discussion here should be improved.

Indeed we studied regional trends focusing on the regions where the trends are statistically significant across many grid points. As we are not trying to speculate too much on the possible causes (would require substantial modelling efforts) we leave it as is. In the introduction and summary we discuss some of the possible causes of trends and make it clear that long-range transport of tropospheric ozone can contribute to trends in rather remote areas.

Section 3.3.2.
The authors attribute the positive trends to South Africa and South America to biomass burning. Is there any indication from another source that there is increased biomass burning over the years that can cause such a trend?

The following text has been added (page 17, line 15): "The burned area in southern tropical Africa increased by 1.8 %/yr during the period 2000 to 2011 (Giglio et al., 2013). Ziemke et al. (2009b) and Wai et al. (2014) estimated that biomass burning can contribute to an increase in tropospheric ozone column by ~20%. Hence, it is very likely that biomass burning could be the origin of the observed ozone increase."

Section 3.3.3.

The authors show trends over mega-cities in the tropics. The authors should provide a comment why they think a grid-point of 2x5degrees can represent the variability of tropospheric ozone caused by a mega city.

This Section (3.3.3) has been removed from the manuscript.

The discussion against NO2 trends as shown in the paper is also not conclusive. Are there studies (modelling or in-situ ones) to support their findings?

The discussion about NO_2 trends has been removed (megacities).

The authors also compare their results with Heue et al and although the approach is pretty similar there are differences. They should elaborate more here to explain this.

The comparison with Heue et al. (2016) trends for 10 mega cities has been removed from the manuscript.