Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2016-139-AC1, 2016

Atmospheric Measurement Techniques Discussions
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Interactive comment on "Trends of tropical tropospheric ozone from twenty years of European satellite measurements and perspectives for Sentinel-5 Precursor" *by* Klaus-Peter Heue et al.

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Received and published: 26 August 2016

Author's reply to the interactive comment #1 on

"Trends of tropical tropospheric ozone from twenty years of European satellite measurements and perspectives for Sentinel-5 Precursor" by Klaus-Peter Heue et al.
Anonymous Referee #1
Received and published: 1 July 2016

We thank the referee for the comment and the helpful advice for a more robust verification.

This paper is the first attempt, as far as I am aware, to merge several remotely sensed

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products to produce a long-term trend of tropical tropospheric column ozone. This is an excellent endeavour and the only way to understand how tropospheric column ozone has changed across the entire tropical region over the past 20 years. I hope that the authors succeed in their effort but before I can recommend this paper for publication the authors need to conduct a more robust evaluation of the product against available in situ observations.

Major comments: 1. My main concern lies with the evaluation of the satellite product with the limited ozonesonde observations across the tropics. The sampling rate at these tropical sites is far too low to provide an accurate quantification of the monthly mean ozone profile given the high day-to-day variability in tropospheric ozone. For example, Saunois et al. found that 12 profiles are needed per month at mid-latitudes to give a monthly mean profile that is accurate to within plus or minus 5%. When you have just 4 profiles per month (weekly sampling) the accuracy drops to $\pm$ 10%. The accuracy was even worse for the tropical site they considered, with 4 profiles per months being accurate to only $\pm$ 12%. Logan (1999) concluded that an even higher sampling rate is required, arguing that 20 profiles are needed per month to ensure that the monthly mean is accurate to within $\pm$ 15%. While the NASA SHADOZ program has been extremely valuable in expanding our understanding of the tropical ozone distribution, the sampling frequency of roughly 2 profiles per month is entirely insufficient to allow us to accurately quantify the monthly mean ozone column. Unfortunately this means that your satellite/ozonesonde comparison in its present form is meaningless.

I recommend the following evaluation:

Divide the year into quarters or 4 seasons. For a given ozonesonde site, gather all ozone profiles in this period for at least 10 years. So if a site has 2 profiles per month then it will have a total of 60 profiles (3 months x 2 profiles x 10 years), which is a good sample size. Calculate the 10th, 50th and 90th percentiles for the tropospheric ozone column corresponding to each profile. Extract your satellite product at the location of the sondes on the days the sondes were launched (5x5 degree grid cell is fine) and

also calculate the 10th, 50th and 90th percentiles. Now you have adequate sampling at all sites and you can conduct a robust comparison between in situ and satellite observations.

The referee is absolutely right that one or two soundings per month do not represent the monthly mean adequately. However, the CCD method always results in mean values; usually monthly means are used to have a good representation of the data and the variability.

To overcome this principle difficulty we developed a near daily data product. We still assume the stratospheric column to be monthly and longitudinal invariant and we averaged the tropospheric data for three days and an area of $5° \times 5°$ *around the sounding station. We also tried a daily dataset, but the data coverage was too low (GOME has a three day global coverage). With respect to the finding of Saunois et al. (2012) that 12 profiles result in a good representation, we assumed that one sounding every other day is enough, or a sounding is representative for a two or three days period. We made three comparisons between the sondes and the harmonized satellite data:*

1) We sorted the data per season, and calculated the average, and the 10th and 90th percentile for running 5-years periods. For the station with 4 soundings per month (e.g. Nairobi) this resulted in up to 58 soundings per samples. The comparison showed there is a bias between the soundings and the CCD TTOC at least for Nairobi and Natal, but within the 10th to 90th percentile margin the data agree. Moreover for the selected stations no trend in the bias is visible.

2) A histogram of the deviation showed a bias of 1.7 DU and a width of the distribution of 5.8 DU. The satellites underestimate the sondes as already realised for the stations.

3) We plotted the time series of the daily differences; here the position of the station was neglected. Neither in the monthly mean differences nor in the daily data the fit resulted in a robust trend. The fit coefficients were smaller than the fit error. A similar bias is observed.

The respective section in the manuscript was changed. The major finding of the manuscript remained unchanged, from the comparison.

2. The Introduction is not very well written and needs to be thoroughly revised. a. The opening paragraph needs to provide a more thorough summary of global ozone trends. Saying that ozone "has at least locally doubled in the last 50 years" is very vague. Cooper et al 2014 could only show that ozone doubled over Europe as this is the only region with 50 years of data. Increases elsewhere are more modest due to shorter time series. A broader perspective is as follows: Using a multi-model ensemble, Young et al. estimate that approximately 30% of the present-day tropospheric ozone burden is attributable to human activity.

Changed. The uncertain finding that ozone has at least doubled in Europe since 1850 was clarified. The model results by Young et al. 2013 that 30% percent of the current ozone is caused by anthropogenic emissions of ozone precursors are now included in the manuscript. The introduction was reorganised and insitu trends and satellite based trends are clearly separated.

b. You need to provide references for the impact of the stratospheric intrusions. Good ones are Tang et al. 2011 and Stohl et al. 2003. c. Lines 8-25 on the specifics of ozone formation chemistry should be deleted as this is far too much detail for a paper on satellite retrievals. Instead just refer the reader to review articles such as Monks et al 2009, 2015.

The section about the sources of tropospheric ozone is reduced to a very short summary of the anthropogenic sources and reference to Monks et al 2015. Therefore also the reference to the stratospheric intrusions is no longer necessary.

d. The discussion of broad tropospheric ozone trends is not well done and should instead rely on the conclusions of three recent and authoritative papers on ozone trends: IPCC, 2013, see section 2.2.2.3 on Tropospheric Ozone Oltmans et al., 2013 Cooper et al., 2014.

The discussion was rewritten. All the above mentioned references are review papers and give an excellent summary on the state of knowledge. Therefore they are used as basis of the new discussion. The literature cited therein may however still be used.

e. Page 2 line 7. Here you talk about stratosphere-troposphere exchange occurring at the subtropical jetstream. What about the polar jetstream? What reason do you have to believe that STE is more important at the subtropical jet? References?

We originally focussed on the subtropical jetstream, because our data product is available in the tropics only. Hence our data are not affected by any intrusion due to the polar jetstream. In the general context of the sources on a global scale this was indeed incorrect. The respective section on sources was reduced.

f. page 2 line 2: make it clear that Jack Fishman was the first to produce a satellite retrieval of tropospheric ozone by saying something like: Fishman and Larsen (1987) produced the first satellite retrieval of tropospheric ozone. . .

Clarified. I tried to cite the title of the paper by Fishman and Larsen directly: "...: Implications for the distribution of tropospheric ozone"

g. page 3 line 1: what do you mean by "high reaching"? Typically we say deep convective clouds in reference to those that reach the upper troposphere. Don't some (i.e. Ziemke) methods assume that the cloud tops reach the tropopause and therefore any ozone measured above the cloud is entirely in the stratosphere?

Clarified. "High reaching" is replaced by "deep convective". The cloud top height is uncertain, with respect to the light path inside the cloud top. In our algorithm we rely on the cloud height retrieval for the different instruments. There are only very few clouds reaching the upper tropical troposphere (15 km). So to get a reasonable number of ACCOs we decided to use the clouds between 8.5 and 14.9 km. The average was about 10 km. (see 4.)

h. page 3 lines 6-7: Smog is not a scientific expression (I have measured plenty of

ozone, but never smog) so please use a better term. Oltmans et al. 2013 review long-term rural and remote ozone changes but don't really discuss the impacts of emissions reductions. Better papers would be Cooper et al 2012 or Cooper et al 2014

The word smog is deleted. It is clear that you can not measure smog, but only ozone, formaldehyde, nitrogen oxides, (secondary organic) aerosols or other parameters that are typical for high pollution episode in the summer. Summarised in http://nationalgeographic.org/encyclopedia/smog/

i. Page 3 lines 4-7 are out of place and belong somewhere else

Deleted

3. The English in the manuscript is not up to the standards of AMT and there are far too many grammatical errors for me to take the time to correct. The authors need to either find a colleague with excellent English skills to edit the entire manuscript, or they need to work with the journal to secure the services of a copy editor.

4. page line 11 If I understand this part correctly, your tropospheric ozone column is only from the surface to 10 km? Does this differ from the Ziemke method which is for the full tropospheric column?

The altitude of the TTOC retrieved with convective cloud differential method is not as certain as it may look like. Here we used the cloud top altitude as given in the cloud data for collocated ozone column observations. The cloud altitude retrieval is based on oxygen absorption structures (O2 or O4) and is typically lower than physical cloud top. Therefore the ozone inside the cloud top may be observed as well. For the reference region over Pacific it is generally believed that the influence is small (Ziemke et al. 2009). However it can not easily be distinguished down to which altitude the above cloud column ozone really extends. In an earlier publication by Ziemke and Chandra (1999) no altitude information is given for this first step. A small section on this altitude uncertainty was added to the section 2.2. Convective Cloud Differential Method for

TROPOMI

5. page 18 line 1: here you compare your tropospheric column ozone trend to observations from Samoa as reported by Oltmans et al. 2013. You say that the trend at Samoa is 0.1 +/- 1.7 DU/decade for 1991-2010. But I could find no such value in Oltmans et al. All I could find was the surface ozone trend of 0.02 +/- 0.34 ppb/year. Comparing a column value to a marine boundary layer surface observation is not valid. If you want to compare your results to surface observations then the only site that might be relevant is Mauna Loa because at 3.4 km it samples the free troposphere. While it cannot report the trend of the tropospheric column ozone it is still probably a good indicator of how tropospheric ozone has changed in this part of the world.

It would certainly be better to compare our TTOC trends to sonde based trend data or similar integrating measurements. However, such data are rare, and if we want to compare regional trend data, we have to use the available measurements. It was clarified that the original data in Oltmans et al. (2013) are marine boundary layer measurements and that we have to be aware of the different air masses observed.

Minor Comments: if no explanation is given for a comment, please insert the suggested text into the appropriate place in the manuscript Page 4 line 12 It would be best to give more explanation of the cloud slice method up front as the reader has to venture a long way into the paper before it becomes clear how the method generally works. A short summary of CCD method and the harmonisation is now given in the introduction (see also comment #2).

The title would sound better as: Trends of tropical tropospheric ozone from twenty years of European satellite measurements and perspectives for *THE* Sentinel-5 Precursor
corrected

Abstract line 3 ". . .of the satellite instruments GOME. . .." It's important to distinguish the instruments from the satellite platforms.
corrected

Page 17 line 1 and elsewhere: your use of the word “global” makes it sound like the product covers the whole world when it really only covers the tropics, please correct.
corrected

Page 18 line 13 Change “grand” to grant.
corrected

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