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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.

Anonymous Referee #1

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

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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 +/- 10%. The accuracy was even worse for the tropical site they considered, with 4 profiles per months being accurate to only +/- 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 +/- 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.

Logan JA. 1999. An analysis of ozonesonde data for the troposphere: Recommendations for testing 3-D models and development of a gridded climatology for tropospheric ozone J. Geophys. Res 104: 16,115–16,150.

Saunois M, et al. 2012. Impact of sampling frequency in the analysis of tropospheric ozone observations. Atmos. Chem. Phys 12: 6757–6773. doi:10.5194/acp-12-6757-

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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. 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. 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 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? 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... 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? 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

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Cooper et al 2014 i) Page 3 lines 4-7 are out of place and belong somewhere else

Cooper OR, Gao R-S, Tarasick D, Leblanc T, Sweeney C. 2012. Long-term ozone trends at rural ozone monitoring sites across the United States, 1990–2010. J. Geophys. Res 117: D22307. doi:10.1029/2012JD018261

Cooper, O. R., D. D. Parrish, J. Ziemke, N. V. Balashov, M. Cupeiro, I. E. Galbally, S. Gilge, L. Horowitz, N. R. Jensen, J.-F. Lamarque, V. Naik, S. J. Oltmans, J. Schwab, D. T. Shindell, A. M. Thompson, V. Thouret, Y. Wang, R. M. Zbinden (2014), Global distribution and trends of tropospheric ozone: An observation-based review, Elementa: Science of the Anthropocene, 2, 000029, doi: 10.12952/journal.elementa.000029

IPCC (2013), Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the IntergovernÂňmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp.

Monks, P. S., et al. (2009), Atmospheric Composition Change – Global and Regional Air Quality, Atmos. Environ., 43, 5268-5350.

Monks, P. S., et al. (2015), Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer, Atmos. Chem. Phys., 15, 8889-8973, doi:10.5194/acp-15-8889-2015.

Oltmans SJ, Lefohn AS, Shadwick D, Harris JM, Scheel HE, et al. 2013. Recent tropospheric ozone changes âĂŤ A pattern dominated by slow or no growth. Atmos. Environ 67: 331–351.

Stohl A, Bonasoni P, Cristofanelli P, Collins W, Feichter J, et al. 2003. Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO. J. Geophys. Res 108(D12): 8516. doi:10.1029/2002JD002490

Tang, Q., M.J. Prather, J. Hsu (2011), StratosphereâĂŘtroposphere exchange

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ozone flux related to deep convection, Geophys. Res. Lett., 38: L03806, doi:10.1029/2010GL046039.

Young PJ, et al. (2013), Pre-industrial to end 21st century projections of tropospheric ozone from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP). Atmos. Chem. Phys 13: 2063–2090. doi:10.5194/acp-13-2063-2013

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?

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 clumn 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.

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.

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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

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

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.

Page 18 line 13 Change "grand" to grant

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