

Interactive comment on “Harmonisation and trends of 20-years tropical tropospheric ozone data” by Elpida Leventidou et al.

Anonymous Referee #1

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General Comments:

This study attempts to harmonize a 20-year satellite record including GOME/ERS-2, SCIAMACHY/Envisat, and GOME-2/MetOp-A using SCIAMACHY as a transfer standard. The authors use 6 different schemes in their attempt to harmonize the data and evaluate the relative success of the various approaches via comparisons with in situ measurements (i.e., sondes) when and where possible. The authors suggest that by using 6 different approaches, they are better able to estimate the uncertainty in apparent trends owed to the harmonization itself.

Like prior studies, the authors find few areas of significant tropospheric ozone trends. Their analysis of trends over tropical mega cities seems to produce results not too different from prior studies. Frankly, I am really not sure what to take away from this

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study, and I have several important issues with what is (and is not) presented.

The authors should be aware of a major reprocessing effort of the Southern Hemisphere Additional Ozonesonde (SHADOZ) network data being led by Thompson and Witte. While I realize the papers on their work are just making their way into the literature, their efforts have been ongoing for several years. Since the authors of this study leverage the SHADOZ data, I am surprised that the paper communicates no awareness of the reprocessing effort nor of its potential impact on the results of this study. At the very least, the authors could have contacted SHADOZ PI Thompson to make sure she was aware of this study and had the opportunity to communicate important updates relevant to the authors. While entirely up to the authors, having Thompson as a co-author would have strengthened the credibility of the sonde results presented in this paper.

Finally, while the paper appears to present different approach to tropical tropospheric ozone trend analyses that recent studies, I do not find the results particularly compelling or worthy of publication in this form at this time.

Recommendation:

I recommend this manuscript be declined for publication in ACP at this time pending major revisions.

Detailed Comments:

Page 1 Line 3: What does “good agreement” mean? Quantify. Line 14: “Additionally, over central...” Awkward sentence. Line 19: “...reasons for these decreases are...”

Page 2 Line 2: delete “both”

Page 3 Line 10: “3.8% decade-1 (0.16 ppbv year-1)” What is the difference between these numbers? Unclear. You mention “surface and ozonesonde observations,” so which is it?

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Page 4 Line 4: The reference to Leventidou et al., 2016 may be a recent one for the CCD method, but I think you should be referencing the original paper for this approach, which I believe goes back to Ziemke. . . Line 10ff: This would be a good place to remind the reader of the specific application of the CCD approach you're using. To what altitude is tropospheric column ozone being computed? Does it vary scene-to-scene? On what fraction of pixels can it be applied? Line 14: "overpass time" – is this not a critical element influencing tropospheric ozone, especially in regions near megacities? Line 25: ". . .whole timespan of the operation of the European satellites. . ." Line 26: ". . .since it is the only. . ."

Page 5 Line 2: "Possible reasons for the biases are. . ." The paper is filled with these statements. It would be good to know if this is the problem. Could you test your hypothesis by applying the same cloud algorithm to both retrievals? I realize that requires working with the instrument teams, but even a limited test application could prove useful. The differences that appear between the two panels of Figure 1 are striking. To me, this subject is more interesting than the one that is the current main focus of the paper. Figure 1. It occurs to me in looking at the upper panels that perhaps it would be worthwhile to separate the lower plots into "over land" and "over water" components. Visually, It would also be helpful to the reader if they plots were rotated 90 deg. so that the latitudes ran up and down the page as they do in the top panel. Line 8 – 9: "This behavior may be explained by the short time of common operation. . ." Another undemonstrated hypothesis. How could you test this assumption? And if it's true, is not your transfer standard idea (i.e., reference to SCIAMACHY) compromised?

Page 6 Line 3: ". . .ozonesonde data, it seems reasonable. . ." Figure 2. What is important about this plot is how little of the area is actually statistically significant. Perhaps you should reverse and only mark with "x" those cells that ARE statistically significant. Also, remind the reader, what fraction of the cells are statistically significant? This result is key in your argument that you can use a constant offset to "correct" the GOME 2 data, but you do not seem to make much of a point of that in the text. Line 12: ". . .but

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only a subset of them." Which subset? Which years? Sect. 2.3: This seems to be an important part of the paper, but frankly, I do not find it well motivate. While 6 scenarios? Have you exhausted all possibilities. Does the reader need to know the details or simply your recommendation for the best approach to harmonize the data – with a discussion of the other approaches you tried and how they inform your estimate of the component of the calculated trend uncertainties that arise from the harmonization process itself?

Page 7 Table 1. Not sure what to do with this table. Why are the stations in the order in which they appear? What is the table communicating? Here's where my earlier comment about SHADOZ reprocessing becomes relevant: are you using the reprocessed sonde data? How would these results change if you did? You are integrating the sondes to 200 hPa. How does that compare to the altitude used for the CCD approach?

Page 8 Line 7: "The same occurs for the. . ." delete the comma. Line 9: ". . .the scenarios that can be confidently rejected according to this comparison are. . ." I'm not sure I have confidence that any scenario can be rejected until I know more about the sonde data you used and the altitudes used for the satellite tropospheric column amount.

Page 9 Line 1: "where α is the offset. . ." Line 12: ". . .to persist into the next month." Line 16: define "AR(1)" Lines 25ff: "Nevertheless, all scenarios shown in Fig. 3 agree that there is a positive trend. . ." My quick read of Figure 3 is that very little of the map shows statistically significant trends. As to the fact that one appears to exist "over the southern tropical Atlantic Ocean" and a couple of other sites cited by the author, I am not sure what to make of it. The authors provide no explanation for such trends or why they might exist. I would find this more compelling if the authors could simplify the presentation, show the best correction scheme, show the best estimation of uncertainty (including that resulting from the harmonization scheme), and then spent some time in the text discussion what the resulting trend data showed and why. In its current form, I find the presentation more confusing than compelling.

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Page 11 Line 8: “. . .following very well a Gaussian distribution.” You don’t show that in the paper. But I’m not sure what to make of it, either. Are you saying that there is no signal anywhere on the map? What do you mean by “the noise is random?” What is the noise? Line 10ff: “This result is in agreement with Ziemke et al. (2005) and Ebojie et al. (2016). . .” What periods did they examine? What data did they use? Are there any implications from the fact that it does not appear to have changed from their analyses to your analysis? What new have we learned from your analysis?

Page 12 Line 10: “Figure 5 summarizes the tropical tropospheric ozone trends. . .” Line 25: “. . .may still be an artifact of the data-set.” Is there a way to know? Line 31ff: Just to be clear, you selected your regions based on where you found statistical significance? That led to larger regions that then had statistically significant trends? Your Table 2 shows some impressive trend results. You then follow that with a list of possible factors that led to the trends (anthropogenic NO_x, population, energy consumption, biomass burning, changes in meteorology, dynamical oscillations, stratospheric intrusions) and you cite some prior works that have made these suggestions, but you provide no evidence within this paper for the proximal cause (or causes) in each of the regions you list in Table 2, nor is there really any justification for the selection of the boundaries of those regions other than they produce significant trends. If, as the title of this section suggests, mega-cities are responsible, it seems the regions might have been more narrowly defined. It would have been nicer to select regions based on a hypothesis and then identify the existence of significant trends (or not) to accept or reject that hypothesis.

Page 16 Line 10: “Despite the fact that might appear to be. . .” What might appear? Lines 22ff: “. . .cloudiness and humidity which contribute to photochemical O₃ loss. . .” I think the missing factor identified in the Morris et al., 2010 paper was significant lightning production, which they hypothesized led to NO_x production and O₃ loss in the absence of sunlight. The presentation here is a bit oversimplified. Deep convection alone can loft relatively low O₃ concentrations from near the surface (especially over

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the sea) to the upper troposphere. Those decreases are not “loss” but reductions resulting from transport.

Page 17 Line 2: “winter” – what does “winter” mean in the tropics? Perhaps it’s better to identify seasons by months rather than such ambiguous names. Line 3: “as NO₂ over North America and Europe may have affected the O₃ trends. . .” moved the comma. Line 8ff: “Possible reasons. . .” There’s a whole list of possibilities here with no conclusions or evidence to support any one (or combination) of them. Line 10: “. . .water vapor in the troposphere accounts for one of the most important. . .” Lines 10 – 12: “An increase in vertical convective patterns over the tropical oceans may result in lower ozone mixing ratios in the upper troposphere. . .” True if lofting low ozone from the surface. If lightning is present in the convection, however, you might see enhancements. Thus, the influence is unclear.

Page 18 Lines 23ff: I would replace all of this text with a table. Non need to write it all out.

Page 19 Table 3. Like previous tables, what is the logic of the order of cities in this table? How do the periods of study for Heue, Ebojie, Schneider, Hillboll, and this work compare? What impacts do differences in study periods have on interpretation of the results? The data in this table appear to have been compiled using 2.5 X 5 deg. boxes. That’s roughly an area 250 km X 500 km in the tropics. Can you actually see signatures from megacities spread out over such a large area? For a control, should you also compute trends around cities that have not grown (perhaps ones that have shrunk) or that have reduced emissions just to see if they behave any differently than the ones you list here? Line 3: “The derived tropospheric trends clearly show that the tropospheric ozone increase is not proportional to. . .” If you’re going to make this claim, I think you need to show the population data, perhaps in Table 3, and the proxy you’re using for industrial activity as well. Lines 8 – 9: “The degree of tropospheric ozone change strongly depends on the NO₂ amount. . .” As the second half of this sentence correctly relates, it depend on the relative NO₂ and VOC concentrations. I think I would

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get rid of the word “strongly” in this sentence.

Page 20 Line 21: “. . .since the uncertainties in the trends are larger. . .” Line 24: Cite the uncertainties associated with the trends published in Ebojie et al. Line 31 – 32: “They might be linked to. . .” You list a whole bunch of possibilities. Has anyone shown the specific relevant link for your study? If not, how can you test these hypothetical influences?

Page 21: Line 4: “. . .tropical latitudes (> 18 0N and S). . .” What range? Line 10: “It has been shown that tropospheric ozone increase is not linearly related. . .” I’m not sure this study has shown that result conclusively or persuasively. Line 18: “. . .the fact that their retrieval reaches up to the tropopause. . .” That seems like an important factor. How different are the retrievals? What impact do the differences have on your results/homogenization scheme? Line 28: “. . .(expected lifetime of the Sentinel 5 precursor satellite).” No need to introduce an abbreviation in the last paragraph.

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