



## Interactive comment on "50 years of balloon-borne ozone profile measurements at Uccle, Belgium: short history, scientific relevance and achievements in understanding the vertical ozone distribution" by Roeland Van Malderen et al.

## Anonymous Referee #1

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Review of Van Malderen et al. -50 years of balloon-borne ozone profile measurements at Uccle, Belgium: short history, scientific relevance and achievements in understanding the vertical ozone distribution.

General Comments

In this manuscript, the authors provide an overview of the 50 year history of the ozonesonde program at Uccle.

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The dataset is certainly extremely valuable to the field of atmospheric science, and everyone responsible should be congratulated on maintaining a high quality measurement program over such a long period of time. This is an impressive achievement!

The manuscript describes, in a medium level of detail, previous work that has been done to adjust for inhomogeneities and contamination, after which the authors present trends in total ozone at Uccle measured by ground-based instruments, a height resolved trend analysis of the Uccle ozone record in the stratosphere based on multilinear regression, and separately a trend analysis for the troposphere, a trend analysis of tropospheric folding events and a trend analysis of surface ozone. The analyses also make use of a wide variety of other data sources to compare to the Uccle ozonesonde record. In the final section satellite overpasses are compared for three different instruments (or series of instruments) – GOME2, AURA-MLS and AURA-TES.

In my view, these components in themselves are all very worthwhile and very appropriate subjects for ACP.

Unfortunately I must state however, that I have major concerns with the design of the manuscript in its current form. Because of the very wide scope, none of the subjects are presented in sufficient detail to be of much use to the specific communities who are interested in them. In my view it would be preferable to resubmit the manuscript as three separate pieces of work, each one with a more specific focus allowing greater depth. Alternatively, the number of topics could be reduced to allow a more satisfactory level of detail to be given.

I appreciate that the authors have deliberately chosen to present something more along the lines of a summary of the many applications of the dataset.

Unfortunately, in my opinion, this decision is not optimal for the benefit of the readers of ACP.

(Please be clear I am not intending to be in any way negative about the value of the

dataset or the scientific competence of the authors, merely the structure of the submitted manuscript).

Old work is covered in too much detail, but still not in sufficient detail to enable it to be understood without reference to the original papers anyway. On the other hand, new work is "described" without enough detail provided however for the reader to make a proper assessment of its value. This is most evident in the discussion of tropospheric fold occurrence.

The major contribution of the work I see as being the assessment of long-term trends in the stratosphere and free troposphere (sections 4.2 and 4.3), at very high vertical resolution.

I would prefer these sections to be significantly expanded and sections 1, 2 and 3 shortened. I would also consider removing 4.1 and 4.4 and treating these topics in better detail in separate works. In my view section 4.1 is too superficial and section 4.4 too inconclusive in their current forms. The use of other data sets for comparison (eg DeBilt ozonesondes, Frankfurt IAGOS, surface ozone monitoring) is very interesting but does tend to clutter the analysis and obscure the conclusions.

Specific comments

Line 49: You should also quote WMO 2018 Chapter 3 (and chapter 4 if you want to talk about recovery of Antarctic ozone, seeing you have mentioned it just earlier).

Line 53 A paper about SHADOZ is not appropriate for a statement about the radiative forcing of ozone – the best reference for this would be the IPCC or the major papers they have used.

Lines 50-52 You should be careful about the radiative effect of ozone at different levels in the atmosphere.

Lines 61-64 This is confusing for the reader, because in the first sentence it sounds like "electrochemical ozonesondes" is being used broadly in a way which includes Brewer-

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Mast, but then in the second sentence you distinguish ECC ozonesondes from Brewer-Mast.

Lines 67 Explain to the reader why you say "nowadays".

Lines 68-70 I am sure my Australian colleagues would also like you to mention Aspendale, which has data in WOUDC from as early as 1965 and has continued to the present, but with the launching site having moved somewhat (Aspendale – Laverton – Broadmeadows).

Line 82 I don't like "our" here – this makes it appear the work is intended for an internal audience.

Line 82 This is probably out of scope for the current work, but I would be very curious to know if the high frequency of launches at Uccle can be shown to lead to better results, compared to weekly or fortnightly or monthly launches? (You could easily enough calculate the uncertainties on the trends using only a subset of the data).

Line 91 Change "has been" to "was"

Line 114 – It sounds like the ozone profile is adjusted to agree with the total ozone measurement - You should state this clearly to the reader. (Particularly because it is the less common practice at ozonesonde stations these days).

Lines 119-135 Without more detail, it is not very convincing that this approach has worked very well.

Lines 119-135 I would also be curious to know if the results agree with more recent work, eg Tarasick 2020

Lines 139 - 163 This was all very important work, but the problem is that the reader has to take everything on trust, namely the effect of SO2 on the Dobson, the effect on the ozonesonde, and the assumptions about the SO2 concentration. You don't even show, for example, that when you put all this together the Dobson and the ozonesonde are brought into agreement. Thus, I find this moderate level of detail quite frustrating. I would much prefer you be briefer, and just report the outcomes of the historic work (with full references of course).

Lines 163-185 Again, I this was very important work (and I think Uccle should be congratulated for studying the effects of the change away from Brewer-Mast compared to many other stations around the world which were not as careful), however, the method is not actually explained here, you just show the results (and reproduce an old figure). As above, I think you should just mention this work and give the references.

Line 208 For comparison with other work, it would be helpful to give the size of the altitude correction specifically (ie not just the resulting difference in ozone).

Lines 221-223 How confident are you of the homogeneity of this time series? If you are going to show this figure (Figure 3), you need to discuss the calibration history of the Dobson (particularly in the pre-1977 era) and the transition from Dobson to Brewer.

Line 228 How can you be so confident about the attribution to volcanic eruptions? There are other large dips and several peaks as well of comparable size.

Lines 233-241 This section is too superficial. Your trend line has returned to the 1980 level by 2018 which is much faster than expected from ODS emissions. You seem to suggest the increase is therefore due to a large increase in the Brewer-Dobson circulation but then note the seasonality doesn't match. It is hard for the reader to have any confidence that the trend lines are meaningful.

Line 259 What is the justification for treating AOD as constant since 2012? Have there been no volcanic injections into the stratosphere since then that could have affected Uccle?

Line 267 I think this is really interesting work and I would have loved for you to show your full regression model and how well the different proxies can account for ozone variability at different altitudes at a specific site like Uccle.

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Lines 274 I don't like the "etc" – you need to list the specific factors which have been identified.

Lines 276-277 Yes, this brings me to a question. Does it make sense to consider the relative height with respect to the tropopause once heights above 20 km say? Surely from 10 km above the tropopause, the absolute height is more relevant? This point might have been worth some consideration.

Lines 281-295 This is a very important result and deserves more attention.

Line 297 (Figure 4). This is a very important part of the manuscript. The error bars for the recent period are very hard for the reader to assimilate. There does seem to be an offset between DeBilt and Uccle, not just larger uncertainty.

Lines 304-305 What level of "the stratosphere"?

Lines 302-325 Unfortunately I find this discussion very "hand-wavy". There is a general discussion of results from the literature but I find it hard to discern what the specific conclusions are.

Lines 345-350 Frankfurt and Munich seem a long way from Uccle if considering boundary layer ozone – would you expect the trend to be correlated over this distance?

Line 342 (Figure 5) This is a very important figure but the way you've drawn it is unintentionally misleading. The reader's eye sees that the black curve and the red curve agree very well, and agree better than the blue, and it's easy to miss the fact the black curve is for a completely different time period. I would suggest only showing the three 1995-2018 trends on the one plot. If you want to include the 1969-1994 Uccle trend it could be a different panel.

Line 342 (Figure 5) Wouldn't a lot of the height range shown in this plot be above the troposphere at Uccle, particularly in winter?

Lines 399-403 This result seems extraordinary. Could the number of events really

have increased by a factor of eight in only fifty years? It seems very implausible, and if you want to include it you need to show much more convincing evidence. (Otherwise the reader will assume the most likely explanation is that there's a mistake in your algorithm).

Lines 412-465 Unfortunately, I have to say my honest recommendation would be to delete section 4.4 altogether. For one thing, it is not clear that the ozonesonde record adds any benefit compared to the continuous surface monitoring. (Noting that the ozonesonde data before 1985 is of limited value). There is a small offset which you don't seem to explain. Secondly, although you show an increasing trend, after a fairly lengthy discussion you are not able to reach any conclusions about its causes (in terms of changes in the precursors or meteorological conditions or some other factor).

Line 464 (Figure 7) Overall the figure is cluttered and difficult to read. Just from looking at the plot, while there appears to be a good agreement between the ozonesonde and the surface monitor, it is really just the existence of a consistent seasonal cycle. There seems to be a jump around the year 1998?

Line 484 Who is "us" in this context?

Line 503 You should explain why applying the averaging kernels makes a much larger improvement in the lower stratosphere compared to other altitudes.

Lines 505-509 This would be more convincing if you didn't just show the "before" and "after" plots – the reader is left wondering whether the degradation correction has been tuned to match the ozonesondes – perhaps you could show the degradation correction over time?

Line 531 In section 5.2 you don't use the averaging kernels but in section 5.1 you said how important it was to apply them – why is this?

537-538 The figure shows a growing bias at altitudes above 10 hPa – it seems to be a systematic effect and not just that the ozonesondes are "known to be less accurate".

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Lines 545 (Figure 9) Overall I like this figure but the error bars seem strange. If the agreement from one year to the next is so good, it doesn't seem possible that the error bars could really be so large. The description in the caption is too brief and doesn't show what the error bars are really representing.

Line 551 Here, you should also let the reader know that TES was decommissioned in early 2018.

Line 552 You have already described the orbit of Aura in the MLS section.

Line 559 It seems curious that you apply a limit of 300 km in the troposphere but 100 km in the stratosphere – is this a reasonable thing to do?

Line 563 You should explain to the reader what is meant by the term "observation operator". Is it the same as an averaging kernel?

Line 580 In this case, seeing there are only "1-2" degrees of freedom in the troposphere, wouldn't it better to make a plot of the regression described in lines 581-590, rather than figure 10 as it currently stands? Then the reader could see the temporal stability.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-724, 2020.