

***Interactive comment on “Long-term changes in
UT/LS ozone between the late 1970s and the 1990s
deduced from the GASP and MOZAIC aircraft
programs and from ozonesondes” by
C. Schnadt Poberaj et al.***

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Reply to comments by Jennifer Logan:

We have updated the manuscript according to the recommendations by all reviewers and the editor. Overall, the paper has significantly been reduced in length. This has been accomplished by creating appendixes (Sects. 2.2 and 3.4), and by shortening the rest.

Responding to the referee’s and the editor’s concern about the used cutoff values for tropospheric ozone, one side effect was that we found out that the assignment of

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tropopause information to the MOZAIC data went wrong in some places (when there were more than one aircraft measuring in one minute). To mend the problem, we have recalculated all MOZAIC averages. While there was no major quantitative effect on the MOZAIC averages, still some results have changed marginally and some numbers given in several Figs. have slightly changed (e.g., Fig. 5).

Significant changes that we have carried out are listed below in a point-to-point reply.

Overall comments:

We have added a sentence to the abstract expressing that the comparison of aircraft with ozonesondes was included to determine whether the aircraft changes are consistent with those from ozonesondes, which are the only other data set covering the 1970s in the UT/LS.

1. We have tested whether there is a fair weather bias in the Wallops Island data using NCEP reanalysis 1 water vapour data. Further details see author comment for the editor.

2. We have put most emphasis on the regional results in the updated version of the paper, but retained Fig. 1. We have also followed the reviewer's recommendation to only give values in case of significant trends. We agree with the reviewer to retain the indication of significant changes in Fig. 1 (see also author comment to third comment by D. Parrish). Section 4 has been rewritten following the recommendation of the reviewer to either focus on significant changes or no change in ozone.

3. We have kept the cutoff values as in the ACPD version of the paper for the following reasons: In the case of small GASP regional or 10x10 degree samples, individual flights measuring anomalously high UT ozone ("aged stratospheric air") strongly biased and significantly distorted the typical UT frequency distribution shifting the mean to higher values. Therefore, it was necessary to remove anomalously high ozone from the GASP data set. Since it can be assumed that the MOZAIC samples are sufficiently

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large to provide representative UT ozone probability density functions, they were used to define seasonally dependent upper limits. The cutoff values at 80 ppbv, 120 ppbv, 120 ppbv, and 90 ppbv in DJF, MAM, JJA, and SON, respectively, resulted in 97% (95%), 98% (97%), and 99% (99%) of the MOZAIC (GASP) samples at middle, sub-tropical, and tropical latitudes considered tropospheric, respectively. We acknowledge that by removing aged stratospheric air from the tropospheric samples, we may potentially miss a certain contribution to long-term UT ozone changes by stratosphere-troposphere (STE) exchange processes suggesting the possibility of a) a changed frequency of STE and/or b) changed ozone concentrations entering the troposphere. However, not applying the cutoff values for MOZAIC does not significantly alter UT ozone mixing ratios (not shown). Thus, it can be assumed that the effect of the cutoff values is minor in the case of a well-defined frequency distribution and it may help to restrict GASP UT ozone to more typical mixing ratios. We have adapted the text of Sect. 2.3 accordingly. 3. Boxes surrounding the regions for averaging have been added to Fig. 1a. A sentence explaining how the averages were formed has been added to Sect. 2.3.

4. We have greatly shortened the comparison with surface measurements. However, since there may be a connection with UT changes (as discussed for East USA), we have not removed the discussion completely, but confined it to a qualitative discussion.

6. For the purpose of comparing the aircraft with sonde data, we have refined the EUR region reducing its size to 42°N–57°N and 0°E–20°E (EUR SONDE). While the associated uncertainty of the changes increases somewhat, the results are not changed by the size reduction of the area. The definition of the region has been included in Sect. 2.3.

Figures:

Figures 2, 7, and 9: These figures have been designed to be printed over the width of two columns and preferably on a whole page. When printed on a whole page, it should be possible to read all numbers.

Other comments:

1. Whereas David Parrish recommends the authors to clarify in the introduction that parts of the regional variability in ozone trends may be due to imperfect measurements and thus difficulties to adequately quantify trends, the reviewer does not have a problem with different trends in different regions, as they are not necessarily unphysical. In fact, in our opinion, both views may be right. On one side, it makes sense that trends differ in different regions because underlying causes for trends differ, such as e.g. regional differences in industrial development and related emissions of pollutants. On the other side, quantification of trends may actually be difficult in some cases due to imperfections of the ozone sensors or their improvements over time, as stated by David Parrish and discussed in the text. Therefore, we have changed the according part in the introduction including a sentence on potential difficulties in quantifying trends. Furthermore, we have removed the above-cited sentence on differences of long-term ozone changes in different regions of the world. Instead, we introduce the very same paragraph with the following sentence: "The current state of knowledge of tropospheric ozone trends based on ozonesonde measurements can be summarised as follows: ...". The subsequent sentences clearly indicate themselves that trends are different in different regions of the world.

Comments 2., 7., and 9.: adopted as recommended.

3. Whole Sect. 2.2 has been moved to an appendix. Nevertheless, while the original text on the Uccle homogenisation has been somewhat shortened to become more concise, parts have been added to motivate the paragraph. In our opinion, the information on the homogenisation should be kept for several reasons:

a) a description of the homogenisation of the Uccle data can neither be found elsewhere in peer-reviewed literature, nor is there any meta information of the data given in the WOUDC archive. Hence, it actually makes sense to once provide this information to the interested reader, and to give a link to the homogenisation report by De Backer,

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which contains a detailed description of the data processing.

More specifically and more importantly, a basic understanding of the homogenisation procedure is relevant for understanding b) why the 1970s Uccle data have been corrected by a different altitude shift than MOHp and Payerne (same section further below), and more importantly,

c) the quantitative characteristics of the Uccle data, specifically relevant in the comparison with the other 1970s BM ozonesonde data (Sect. 3.3). Here, the homogenisation covers one essential problem of the 1970s sonde data: underestimation of tropospheric and UT/LS ozone, the same problem also encountered at Payerne and at the Canadian stations (then BM devices).

4. Apparently, the reviewer is not aware that the paper she is referring to (Schnadt Poberaj et al., ACPD, 2007) is not the finally revised paper. While the ACPD version contains a comparison of the GASP with ozonesonde data and thus, a discussion of the altitude shift of the ozonesonde data, in the ACP final version of the paper, this comparison was removed and with it, the description of the altitude shift. For this reason, the paragraph on the altitude correction of ozonesonde data is new and should remain in this paper (now in Appendix A).

5. According to the CO emissions time series from the RETRO inventory that we got from Martin Schultz to evaluate emissions in SE Asia and Indonesia separately, CO emissions have developed very similarly over Indonesia and continental SE Asia. This is mainly because one of the two parameters to calculate emissions, burned area, was constructed on the basis of the very same decadal deforestation trends and the ENSO index (as indeed in these regions burned area and thus, as the reviewer states, biomass burning, is significantly correlated with ENSO). The text of Sect. 3.1 has been changed to additionally include a statement on the CO emissions increases in SE Asia alone, as well as a statement on the uncertainty of the emissions estimates in these regions due to data sparseness in the fundamental parameters required in the fire

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emissions equation used in Schultz et al. (2008).

8. Sections B, first paragraph, and section C. have largely been shortened.

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