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Atmospheric Chemistry and Physics Discussions

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 the editor:

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 J. Logan'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

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 to the editor's comments.

1) Concern about the cutoff values for UT ozone analysis (O. Cooper, J. Logan): 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 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, subtropical, 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 (this condition also includes those cases where anthropogenic ozone in warm conveyor belts gets mixed into stratospheric Intrusions and re-enters the troposphere in these, editor's comment). 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

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restrict GASP UT ozone to more typical mixing ratios. We have adapted the text of Sect. 2.3 accordingly.

Do the 95 percentiles change from the 1970s to the 1970s? We calculated the 95% percentiles for all regions of Fig. 2. There are no consistent differences between the percentiles in the 1970s and 1990s, possibly also connected to GASP data limitations in some regions and seasons.

2) "The paper should focus on regional results more than the results for the individual boxes, but retaining Fig. 1". We have changed the text everywhere accordingly. Fig. 1 is now only discussed qualitatively, whereas the main focus and the quantitative discussion are laid on Fig. 2. We now put less emphasis on changes that are statistically not significant. For the discussion how to understand confidence intervals that include zero, please see author comment for D. Parrish.

3) Potential dry bias of sonde data: There should not be a dry bias of the European BM data, since there are regularly launched two to three times a week, as also J. Logan states. For the Wallops Island station, to identify whether there actually is a dry bias in the 1970s and/or 1990s tropospheric sonde data, we used daily NCEP reanalysis 1 water vapour mixing ratios at 400 hPa, interpolated onto the Wallops Island coordinates, to calculate multi-annual seasonal means a) using all available daily data and b) only those dates where launching occurred. Comparing the a) and b) averages for the 1970s and 1990s, slightly dryer conditions by about 10% were identified in the sonde launching means in most seasons, but not as pronounced as in April/May 2000-2003 and except for JJA 1994-2001, where water vapour mixing ratios were the same in a) and b). Comparing the 1990s ratios b/a (91%, 91%, 100%, and 92% in DJF, MAM, JJA, and SON, respectively) with differences between MOZAIC and Wallops Island UT ozone profiles in Fig. 9, no clear dependency of the seasonally varying magnitude of the water vapour ratio on differences could be identified. Thus, the effect on long-term averages may be assumed to be minor. We have changed the text of Sect. 2.2 accordingly.

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We have chosen NCEP reanalysis data to carry out the analysis, because the data are available at no charge on the internet, and in addition using them is computationally inexpensive. In contrast, re-processing the ERA40 data extracting water vapour mixing ratios for both the 1970s and 1990s would have meant a large effort (two to three weeks computational time).

We also tried to reach Frank Schmidlin by email to get more information on the launching policy and a potential fair weather bias of Wallops Island station. However, until now, we have got no answer from him. If he knows anything about the issue, we will update the manuscript again in the final correction phase before publication.

Figure 1: As recommended, two more panels showing actual mean mixing ratios in each 10x10 degree box for the 1970s and 1990s have been added to Fig. 1. Additionally, outlines around the regions addressed in Fig. 2 have been added to Fig. 1a (1970s mean, DJF).

Figure 2: The fact that there are differences plotted for the N IND region (60-90°E, 20-30°N) in SON, but differences in the 10°x10° boxes are not displayed over the same region and season, is not inconsistent. This is because stricter rules apply for calculating long-term differences over the 10°x10° boxes than over the greater N IND region: Within every box, at least 10 daily means need to be available to calculate long-term averages for the 1970s and 1990s. Over the N IND region, 19 daily means (referring to app. 19 flights) are available for computing the average in the 1970s (see number in Fig. 2). However, when distributing these 19 flights over subregions, it is easily understood that there must be less flights in the individual boxes. In SON in the 1970s, none of the subregions of N IND contains equal or more than 10 daily means. A corresponding explanation has been added to the text in Sect. 2.3.

Figure 4: The labelling around the graphs has been enlarged, and to enlarge the graphs, they have been arranged more favourably reducing the bounding boxes, as well as using the whole width of the page.

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Figure 7: We agree that the figure is far too small the way it is printed in ACPD. The figure was designed to be printed on a whole A4 page, not on half a page as customary in ACPD. When printed on a whole A4 page, the figure improves significantly. We hope that then it will be sufficiently readable. The figure caption has been reworded to make the meaning of the grey triangles more easily understandable: ", ... and grey triangles to indicate biases of the average to one year: if the sample contains data from greater equal (less than) three years and more than 50% (70%) of data are from one year.".

Figures 8 and 10: the legend has been corrected.

Figure 9: A statement has been included that Wallops Island is red.

Figures 11, 12, and 13: All triangles in Fig. 1, Fig. 6 and Figs. 11-13 have been enlarged. Additionally, the triangle colour green has been substituted by pink, because it is more noticeable than green. .

Minor comments:

All references concerning spelling, wording and grammar have been considered.

Page 2437 line 16, page 2458, lines 3-4: references have been added.

Page 2456, line 4: "10% increase above N JP in MAM does not appear to be significant ..." The editor is right. Therefore, we have reworded the whole paragraph and put more emphasis on the fact the estimated changes over N JP have to be considered with caution due to data sparseness of the GASP data set in this region.

Page 2457 line 7: The wording must be "wildland fires" instead of wildfires. The authors of the cited study (Schultz et al., 2008) define wildland fires as fires in open vegetation. These fires both include uncontrolled wildfires, as well as those ignited by humans either on purpose or inadvertently. The text of Sect. 3.1, paragraph on Southeast Asia, has been changed accordingly.

Page 2457 lines 12-14: Fig. 6 shows data from all available longitudes. A sentence has

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been added to Sect. 2.3 pointing to two publications in which the equivalent latitude method is described (SP2007 and Hegglin et al., 2006).

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 2435, 2009.

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