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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.

O. Cooper (Editor)

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This review is by Owen Cooper, editor of this manuscript. I have carefully read the manuscript and at the present time I find it to be highly relevant to ACPD/ACP, thorough and well written. However, I have not yet had the benefit of reading the reviews of both anonymous referees and their opinions and comments will have a major influence on my decision regarding acceptance or rejection of the manuscript. Below please find my suggestions for improving the manuscript.

Regarding the comparison of the GASP and MOZAIC data I find the analysis to be quite thorough and only have a few suggestions. 1) I'm a little concerned about the cutoff values used to throw out samples strongly influenced by the stratosphere as described on page 2446. Were these cutoff values derived from GASP and MOZAIC data, or just one of the data sets? Do the 95th percentiles change from the 1970s to the 1990s? I found in Cooper et al. [2004] that pollution in the upper troposphere within WCBs can travel close to stratospheric intrusions and the two eventually mix. So aged intrusions can contain ozone of anthropogenic origin and if anthropogenic ozone is increasing it might cause a trend in the ozone of aged intrusions. Cooper, O., et al. (2004), On the life cycle of a stratospheric intrusion and its dispersion into polluted warm conveyor belts, J. Geophys. Res., 109, D23S09, doi:10.1029/2003JD004006.

2) When discussing the significance of ozone changes there are two approaches in the paper, discussing either the changes in the 5x5 degree boxes or within the larger regions. Switching back and forth between these two scales makes it difficult for the reader to grasp the results. To make the major results more clear, most emphasis should be placed on the regional results in Figure 2 as these have larger sample sizes and are generally easier to interpret, and then just focus on those that are statistically significant which should be those in which the 95% confidence bars do not contain zero. Once the significant changes in the regions have been highlighted you should then take the more detailed approach of looking at the 5x5 degree boxes. Also, in the conclusions you should focus on the regional scale. As it stands you appear to focus on the 5x5 degree boxes which don't always agree with the regional analysis. For example on page 2472 line 12 you state that changes over the NE USA in summer and autumn are statistically not or only marginally significant. But Figure 2 would argue that on a regional basis there is no significant change in either season. And on page 2473 line 10 it says mostly significant changes are found, but Figure 2 shows that changes in spring are not significant over NJP. These are mainly minor discrepancies but it would still help to clarify the results if the focus is placed on the regional analysis.

ACPD

9, S656–S662, 2009

Interactive Comment

Regarding the comparison between the sondes and the aircraft data, one important aspect that needs to be explored is the strong possibility that the ozonesondes are biased towards clear sky conditions which tend to have greater ozone mixing ratios in the upper troposphere than moist conditions. This potential sampling bias could explain why the ozonesondes consistently show more ozone in the upper troposphere than the aircraft measurements. My reasoning is as follows:

In 2005 I published a manuscript in JGR [Cooper, et al., J. Geophys. Res., 110, D05S90, doi:10.1029/2004JD005183, 2005] that compared ozone on the east coast (MOZAIC profiles at Boston, NYC and Philadelphia, ozonesondes at Wallops Island and Huntsville) of the USA with ozone on the west coast (ozonesondes at Trinidad Head) during springtime. We found that there was little difference in ozone between the two coasts, with the exception of Wallops Island which showed 5-10 ppbv more ozone in the mid-upper troposphere than Trinidad Head. Wallops Island also showed more ozone in the mid and upper troposphere than the east coast MOZAIC profiles, in agreement with your study and that of Thouret. I wondered if there was a sampling bias in the Wallops Island ozonesodnes and compared the ozone profiles to daily weather maps. I noticed that the sondes were rarely launched during warm conveyor belt (WCB) conditions which bring warm, moist and cloudy air from the south. This seems reasonable seeing as the ozonesonde measurements are compared to the Dobson spectrophotometer, which cannot give a column ozone measurement under cloudy conditions. But if the sondes are not measuring air in warm conveyor belts they are missing the periods when depleted ozone from the marine boundary layer rises into the upper troposphere through the WCB. This sampling bias should then result in a high bias of the average ozone values.

Following is an excerpt from Cooper et al 2005 (page 17) describing an analysis that proved that Wallops Island ozonesondes during April-May 2000-2003 were biased towards dry conditions:

"We compared water vapor mixing ratio (WVMR) profiles extracted from the NCEP FNL

ACPD

9, S656–S662, 2009

Interactive Comment

analyses (1x1 degree, 26 vertical levels) at the times of the Wallops Island ozonesondes to a climatology of NCEP WVMR profiles at 00 and 12 UTC for each day in April and May from 2000 to 2003. The median WVMR profile at the times of the sondes was less than the corresponding climatology profile at all levels. For example, at 3 km (6 km) median WVMR for the ozonesondes was 85% (75%) of the climatology; and at 3 km (6 km) the 90th percentile WVMR for the ozonesondes was 81% (56%) of the climatology. We compared measured ozone mixing ratios in the lower to mid troposphere associated with WVMR measurements above and below the climatological median and found no large difference. However, between 6-9 km median ozone above Wallops Island is 5-13 ppbv greater when WVMR is less than the climatological median compared to times when WVMR is greater than the median. These ozone differences have a similar magnitude to those between Wallops Island and Trinidad Head. We conclude that the launching of the Wallops Island ozonesondes is biased toward dry conditions and that this dry bias accounts for the greater ozone values at Wallops Island in the 6-9 km range."

My strong suspicion is that the dry air bias affects the average ozone mixing ratio above Wallops Island for the entire data record (30 years?), or at least as long as the sondes have been launched at the times of spectrophotometer measurements. I also hypothesize that this sampling bias also applies to the European sondes that are compared to spectrophotometers. Seeing as you have the ECMWF reanalysis data you can easily conduct a similar analysis of water vapor mixing ratio above Wallops Island in the 1970s and 1990s and see if same bias applies. You can then do the same for the European stations.

If my hypothesis is correct then there will be no point in comparing the sondes to the aircraft because the aircraft take off and land in all weather conditions and are not biased towards dry conditions. The only way to compare the sonde and aircraft data would be to select aircraft measurements with the same range of WVMR values as the sondes. But this might reduce your sample size by a factor of two leaving too little data

ACPD

9, S656–S662, 2009

for your analysis. Another important result of this analysis would be to demonstrate that any study of long term trends based on ozonesondes biased towards dry conditions will have to be evaluated only in the context of dry air conditions. Please give this analysis a try and see if it helps to explain why the sondes show more ozone than the aircraft.

Figures:

Figure 1 It would be very helpful to include two additional panels, showing the actual mean ppbv values in each 5x5 degree box for the 1970s and 1990s. Also please draw outlines around the regions that are addressed in Figure 2.

Figure 2 Results are shown for NIND during SON but there does not appear to be a 5x5 degree box above NIND in Figure 1 during this time period.

Figure 4 is too small and needs to be enlarged

Figure 7 Far too small and almost impossible to read the number or identify the triangles. Also, I don't follow the discussion of the trianglse in the figure caption. Please re-word.

Figures 8 and 10 Shouldn't panel b show MOZAIC-GASP instead of GASP-MOZAIC?

Figure 9 Please state that Wallops Island is red.

Figure 10 change caption to say: only displayed if 10 or more daily means

Figures 11, 12 and 13 Too small and hard to see the triangles. Maybe use a black dot in the center of the rectangles instead.

Minor comments (if no reason is given for my comment then it means that text in the manuscript should be replaced with what is listed)

page 2437 lines 8-10 break into two sentences

page 2437 line 16 need a reference for transport to the UT/LS via convection

page 2347 line 17 the warm conveyor belts of mid-latitude cyclones

ACPD

9, S656–S662, 2009

page 2438 line 4-5 North America

page 2438 line 11 have leveled off, or have become

page 2438 line 16-17 At which level is this trend?

page 2438 line 25 period up to the mid-1990s

page 2439 line 25 the analysis distinguishes between

page 2441 line 1 Airbus aircraft

page 2441 line 8 14558 flights occurred consisting of

page 2445 line 2 we introduce a correction

page 2446 line 8 Data were classified as tropospheric or stratospheric

page 2450 line 15 differences are not statistically significant or only marginally significant

page 2454 line 4 When you say "vertically integrated UT ozone" it implies that you summed up the UT ozone column in Dobson units. Do you mean to say "vertically averaged UT ozone"?

page 2454 line 7 Do you mean to say: "while MOZAIC gathered data closer to the tropopause" ? I think the point you are trying to make is that if MOZAIC flies closer to the tropopause than GASP, then the higher ozone values that are found closer to the tropopause will be represented in the MOZAIC data. Is this correct?

page 2454 line 25 have been attributed to producing unusually high

page 2456 line 4 the increase of 10% above NJP in MAM does not appear to be significant in figure 2 as the 95% confidence bars encompass zero. Please clarify.

page 2457 line 7 Do you know for certain that the increase in biomass burning is just due to uncontrolled wildfires, or is it also due to controlled burning of forests/cropland

ACPD

9, S656–S662, 2009

for land clearing purposes.

page 2457 line 12-14 Does Figure 6 show data form all longitudes or just one longitudinal band? Please provide a brief description of the EL method.

page 2458 line3-4 A reference is needed that demonstrates a decrease in N. Hemispheric stratospheric ozone and its link to halocarbons

page 2463 line 6 device at the end of 1997

page 2472 line 25 Since CTM modeling of these events was not carried out specifically for this analysis it would be better to say "summer increases may be related to"

page 2474 line 5 the US station Wallops Island

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