

Interactive comment on “A UT/LS ozone climatology of the nineteen seventies deduced from the GASP aircraft measurement program” by C. Schnadt Poberaj et al.

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We thank the anonymous reviewer for his comments and recommendations which have helped us to significantly improve the manuscript. We will first comment on the major changes that we have performed on the manuscript according to the referees' suggestions. Thereafter, we will respond to the minor comments point by point. We have taken into account all suggestions and issues raised by both referees.

Major issues:

1) Both referees strongly recommend to reduce the length of the manuscript. The anonymous referee suggests to significantly shorten the length by at least a factor of two. 2) Due to the questionable quality of the early ozonesonde data, the anonymous

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Interactive Discussion

Discussion Paper

referee suggests to remove the section on the comparison of GASP with ozonesonde data from the manuscript (and to address this issue through validation of sonde by GASP data exclusively in a separate paper). 3) Instead, the anonymous reviewer suggests to focus more strongly on the evaluation of the quality of the GASP ozone data and to implement a thorough uncertainty analysis.

Response to the major issues:

The manuscript text has been shortened by an overall 30%, the number of figures has been reduced to 9. This reduction was achieved by following the anonymous referee's suggestion to remove the section on the comparison with ozonesondes, by rephrasing the remaining parts more concisely, and by removing redundancies. The comparison of GASP with ozonesonde data will be dealt with in a follow-up paper.

However, we were not able to achieve a 50% reduction as we also followed the recommendation of the anonymous referee and the editor to include more detailed information on GASP data quality and QA/QC management. To do this, we have included two more sections on "Quality assurance and quality control (QA/QC) procedures" (Sect. 2.1.2) and the "Internal consistency of GASP ozone measurements" (Sect. 2.1.4). Sect. 2.1.4 discusses two near "simultaneous" flights to gain more quantitative information on the accuracy and precision of the measurements. Three new figures have been added to document the quality of GASP measurements.

Applying the above changes, the first focus of the paper is now on GASP data quality and QA/QC management. The other part of the manuscript deals with discussing the GASP climatology and how it compares to MOZAIC measurements and data in the literature. To enhance the significance of the comparison with MOZAIC data, we now also discuss MOZAIC data in former Sect. 3.1 (now Sect. 4.1).

We have somewhat restructured the manuscript allowing the methodology its own section, and moving the discussion of UT seasonality over the Pacific regions, which was formerly part of the general UT ozone discussion (former Sect. 3.2), to the section on UT ozone over the Pacific Ocean (former Sect. 3.3).

In the following replies to the referee's comments, we have not replied on comments concerning the comparison with ozonesondes since this part has been removed from the manuscript.

Reply to general comments:

“Several rather large corrections of the GASP data have been made, however, the authors failed to quantify the uncertainties inherent to these corrections adequately.”

We applied three significant corrections to the GASP data:

- we had to scale down the ozone data by 9%. This high bias of the ozone data arose from calibrating the GASP instruments against a transfer standard that itself was initially calibrated using the KI method. Later in the program, because the accuracy and reproducibility of this method had been questioned, the transfer standard was calibrated at the Jet Propulsion Laboratory (JPL) against a UV photometry standard. There, it was found that the GASP data had a 9% ±] 2% high bias relative to the JPL standard. The archived data had not been corrected for this discrepancy and therefore had to be corrected by ourselves. All calibration issues and problems were carefully documented throughout the program and can be found in the data reports for the tapes, on which data were stored (e.g., Morris, D.: Tabulations of ambient ozone data obtained by GASP airliners, March 1975 to December 1977, NASA TN-81528; Holdeman et al.: NASA Global Atmospheric Sampling Program (GASP) Data Report for Tapes VL0010 VL0012, NASA TM 79061, 1979). Moreover, a thorough documentation of the GASP ozone instruments including all related calibration and quality issues can be found in Tiefermann (1979), one of the few reports that is freely available at the NASA reports server at <http://ntrs.nasa.gov/search.jsp>. We have changed the text to include more details on this correction and the associated uncertainty (Sect. 2.1.2).

- Although the ozone data were supposed to have a temporal resolution of five to ten minutes, sometimes up to 16 samples per minute were recorded. For the purpose

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of building a climatology, the high temporal resolution data were averaged to one minute values. For consistency with the normal operating procedure, only a single one-minute average out of five within any given five minute period was considered. This pre-processing of the data is merely technical and therefore does not require any further quantification of inherent uncertainties.

- Another problem when using GASP ozone data is that some data were obviously not correctly flagged such that spuriously low data were found within the data set. These low values are found over all seasons and over all latitudes. We have used probability density functions to identify the problematic data range to be able to eliminate these data from the data set. Within the 5-minute data set, this reduces the number of measurements by little more than 8%. The remaining data can appear to be reliable as is documented by the overall excellent agreement with MOZAIC described in the results' sections.

“A thorough uncertainty analysis and assessment of GASP data quality should be provided.”

As stated in our reply to the raised major issues this has been done by a) more thoroughly describing the quality assurance and quality control procedures of the GASP program in the new Sect. 2.1.2. Among other things, the probably most critical problem of ozone measurements, the ozone destruction upstream the ozone monitor, has been documented in much more detail and been displayed for the five aircraft as a function of time over the GASP period; b) a new section that focuses on the comparison of two near “simultaneous” flights and demonstrates the general reliability of the system under flight conditions.

“The presentation and investigations of the large-scale ozone distributions of GASP UT/LS and comparison with MOZAIC is far too long and too detailed.”

We have tried to present Sect. 4.2 more concisely and have shortened it by 28%. This includes shifting of the paragraphs on UT ozone over the Pacific to Sect. 4.3

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and moving the discussion on UT ozone over California from Sect. 4.3 to Sect. 4.2. Additionally, a few lines on the comparison of GASP UT ozone distribution over North America with respective surface observations have been added.

Reply to specific comments

Abstract and Introduction have been shortened by 21% and 17%, respectively.

- S1802, 2. Data and Methodology, P3456/17:

The aim of the GASP program was to monitor the temporal and spatial distribution of particulate and gaseous constituents related to aircraft engine emissions in the upper troposphere and lower stratosphere. For this reason and to prevent contamination in the airport environment, the instruments were only switched on between 6 and 13.7 km. The text of Sect. 2.1 has been updated accordingly.

- S1802, Fig. 1: “what scale (lat, lon, time) the measurements are determined?”

The measurements were counted over a 5°x5° grid from 1975-1979 (Fig. 1a) and 1994-2001 (Fig. 1b). The figure caption has been updated accordingly.

- S1802, Section 2.1.1:

The subsection has been reduced in length by almost 50%. This was achieved by omitting the details on photometry, leaving out the technical details of the instrument, and shifting the parts on calibration and uncertainty of the measurements to Sect. 2.1.2.

- S1803, “How accurate is 16Since ozone destruction upstream of the ozone monitor most probably constitutes the most critical problem of GASP measurements, we have now included a much more detailed discussion on it in Sect. 2.1.3. To document ozone destruction over time and for all GASP aircraft, we have included a new figure showing the evolution of ozone destruction with time as determined in regular tests (Fig. 2). This figure shows that ozone destruction was as high as 16% in 1975 and 1976, with a random error in determining the amount of destruction as high as $\pm 8\%$. The amount

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Discussion Paper

of destruction was reduced in 1977 to less than 6% (and the associated error to $\pm 2\%$) when the Buna-N rubber diaphragms were changed to silicone rubber. This information has been added to the text (Sect. 2.1.3).

- S1803, “What type of materials were used for inner walls of inlet tubing, pump, 3-way valve, and absorption cell?”

All lines were made of Teflon and the pump diaphragm was covered with a Teflon-impregnated fibreglass cloth material. The absorption chamber was a folded-tube arrangement with an effective length of 71 centimetres. The tubes were aluminium internally coated with Kynar, and had quartz windows and mirrors. This information has been added to the text (Sect. 2.1.1).

- S1803, “How often the flown photometers were calibrated in the laboratory against the reference Dasibi and when was the change to the JPL-ozone photometer as reference?”

Ozone monitors were frequently (about two to four times per year) exchanged for calibration and functional tests in the laboratory. The secondary (transfer) standard used in GASP for calibrating all ten ozone monitors has been re-calibrated against the JPL-ozone photometer in February 1977. However, the archived values have never been changed to this new calibration. They all refer to the old calibration of the secondary standard which was based on the KI method. This information has been added to the text (Sect. 2.1.3).

- S1803, “The 9% high bias correction: how accurate? How large are the differences and their variability between pre- and post flight calibration???”

The uncertainty of the correction of the 9% high bias was estimated by Tiefermann (1979) to $\pm 2\%$. Calibrations of the monitors against the secondary standard before and after installation on the airplane showed excellent stability and changed by than not more than 1% over the course of a whole year (Tiefermann, 1979). This information is now given in the text.

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- S1803, “Are these corrections for wall losses and bias and total random error constant over entire period between 1975 and 1979?”

Instrument precision was constant over the whole period and the 9% high-bias correction affects all measurements in the same way. However, ozone losses in the sampling system and hence the overall uncertainty of the measurement was dependent on the material used for the pump diaphragm. Initially, Buna-N rubber was used, but in 1977 they changed to silicone. Ozone destruction was reduced from about $16\% \pm 8\%$ to $6\% \pm 2\%$, and the overall uncertainty was reduced from $\pm 8.4\%$ to $\pm 3.3\%$. This information has been added to the text (Sect. 2.1.3).

- S1803, Section 2.1.2:

Respective passages P3459/21-28 and P3460/1-19 have been significantly shortened.

- S1804, Section 2.3, P3463/10-11:

The text has been changed following the recommendation of the referee.

- S1804, “First paragraph too detailed”:

The paragraph has been significantly shortened.

- S1804, P3463/23-26:

The updated text reads as follows: The ozone analyser is a dual beam UV absorption instrument (Thermo-Electron, model 49-103) with an absolute accuracy of 1% and instrument precision of 2 ppbv. The characteristics of ozone measurements on board the MOZAIC aircraft are a detection limit of 2 ppbv and uncertainties for individual measurements of $\pm [2 \text{ ppbv} + 2\%]$. According to Philippe Nédélec from MOZAIC, the accuracy and precision was constant over the entire period of 1994-2001.

- S1804, Section 2.4, paragraphs 2 and 3:

See reply to comments by J. Logan, “Reply to Other comments”, S1673, p3456

- S1804, Section 2.4, P3466/3:

Thanks to the reviewer for pointing to this mistake in the text. Aircraft temperature

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and pressure were used to calculate potential temperature at the measurement coordinates. Using the additional information of ERA40 potential temperature at the 2 PVU tropopause, potential temperature differences from the local tropopause could be calculated. The text has been corrected.

- S1804, Section 3.1:

A motivation for Section 3 has been added. For purpose of highlighting the reliability of GASP ozone, climatological mean profiles of MOZAIC data have been included into former Fig. 3 (now Fig. 5). The text has been changed to include a discussion on the comparison between GASP and MOZAIC seasonal cycle, vertical gradients, and absolute mixing ratios. In addition, a short statement saying that the indicated GASP features are also in agreement with ozonesonde and satellite data has been added (following recommendation of J. Logan).

- S1804, Section 3.2: See reply to general comments.

- S1805, chapters 4 and 5:

The authors agree that the present paper should focus more on validation and evaluation of the GASP data themselves. Sections 4 and 5 have thus been removed.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 3451, 2007.

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