EVALUATION OF OZONESONDES, HALOE, SAGE II, SAGE III, ODIN-OSIRIS AND -SMR, AND ENVISAT-GOMOS, -SCIAMACHY AND -MIPAS OZONE PROFILES IN THE TROPICS FROM SAOZ LONG DURATION BALLOON MEASUREMENTS

Jean-Pierre Pommereau and Francois Borchi

CNRS, Service d’Aéronomie, Route des Gardes, 91371 Verrières le Buisson, France

ABSTRACT

Within the HIBISCUS European project, long series of ozone profiles between 10 and 25 km have been obtained at almost constant latitude (20°S) in the tropics by remote sensing from circumnavigating IR Montgolfier (MIR) long duration balloons in February and March 2001, 2003 and 2004. The performances of all satellite instruments available during the flight periods have been investigated by comparison with this data set. Systematic altitude shifts could be observed in satellite profiles, varying from <50 m for the GOMOS stellar occultation instrument, followed by +100/200 m for solar occultation systems (SAGE II, HALOE above 22 km), but as large as -900 m or +2000 m for limb viewing systems (OSIRIS, SCIAMACHY). The ozone relative biases are generally limited, between 4 % and +4 %, for measurements in the visible Chappuis bands (SAGE II and III, GOMOS above 22 km and OSIRIS), the near IR (HALOE above 22 km) and the ozonesondes, but increase to 7 % in the UV (SCIAMACHY), and +7 % in the mid-IR (MIPAS) and the submillimetric range (ODIN-SMR). Regarding precision, evaluated statistically from the zonal variability of ozone concentration, best measurements are found to be those of SAGE II (2 %), followed by HALOE above 22 km (3-4 %), ozonesondes, SAGE III and OSIRIS (~5%), GOMOS above 22 km and SCIAMACHY (~6 %), MIPAS (8.5 %) and finally SMR (16 %). Overall, satellite ozone measurements appear little reliable in the tropical troposphere except those of SAGE II (and eventually SAGE III), though low biased by 50 % and of limited precision.

1 INTRODUCTION

Because of the many dynamical and chemical processes involved, i.e. quasi-horizontal transport from mid-latitude, convective uplift from the surface, photochemical production by precursors, and potential destruction by very short lived species such as bromine and iodine compounds, the knowledge of ozone concentration changes in the tropical upper troposphere / lower stratosphere (UT/LS) and particularly in the Tropopause Tropical Layer (TTL) is a prerequisite for understanding possible climatic and long term ozone changes in the future. However and although long series of ozone profile measurements are available from space-borne instruments such as SAGE II since 1984, HALOE since 1992 and more recently OSIRIS and SMR aboard ODIN since 2001 and GOMOS, MIPAS and SCIAMACHY aboard ENVISAT since 2002, the ozone distribution and its variations are still poorly characterised in the Tropical Tropopause Layer (TTL). This is because of the many limitations of remote sensing observations from orbit in tropical areas such as increased Rayleigh atmospheric attenuation, high altitude clouds, low temperature, high humidity and dense aerosols and for ozonesondes, their necessarily limited number. In the frame of a HIBISCUS European project dedicated to the study of transport, microphysics and chemistry in the TTL, a series of ozone profile measurements between 6 and 25 km altitude have been carried by solar occultation in the visible from circumnavigating long duration balloons flown in the southern summer in 2001, 2003 and 2004 from Brazil. The objective of the paper is to report on an evaluation of the altitude registration, possible systematic biases and precision of ozone measurements by all above satellite instruments by comparison with those of the balloons.

2 INSTRUMENTATION AND FLIGHTS

The instrument used is a SAOZ UV-visible diode array UV-Visible spectrometer originally designed for the monitoring of total ozone and nitrogen dioxide by the zenith-sky technique from the ground, later developed as a balloon version for remote profile measurements by solar occultation. The measurements shown here are those performed with a further version designed for long duration flights onboard Infra-Red Montgolfier (MIR) balloons developed by the Centre National d’Etudes Spatiales (CNES).

2.1 SAOZ IR Montgolfier instrument

The SAOZ version used here makes use a flat field holographic grating and a 1024 diode-array detector providing spectral measurements of 1.1 nm resolution in the 400-1000 nm spectral range. There is no sun tracker. Sunlight passes into the 50 μm entrance slit after reflection on a conical aluminium mirror (Fig. 1) defining a 360º azimuth field of view.
and −10°, +10° elevation, followed by a pile of three diffusers to homogenise the light beam. Measurements are performed during twilight periods only from 89° to 95° SZA with an exposure duration, controlled by a CPU, varying from 0.5 s to 52 s. Because of limited capacity of the ARGOS satellite collection system used to transfer the data, the measurements are not repeated continuously but at constant tangent height steps of 0.7 km. The spectral data are both analysed onboard and stored in memory in case of recovery. Only the results of spectral retrieval (column density along the line of sight and fitting error of O₃, NO₂, O₅, O₂, H₂O and solar irradiance, both at three wavelengths) are transmitted. Ancillary data also transmitted include 3 D location by GPS, pressure and temperature from Vaïsala radiosonde sensors, spectra information (exposure, wavelength shift) and housekeeping (internal temperatures, voltage). The payload is powered by lithium batteries and additional solar panels for daytime, providing autonomous operation for about one month. The total instrument weight is 28 kg.

Ozone is measured in the visible Chappuis bands between 450 and 620 nm using laboratory absorption cross-sections of 1.5% quoted accuracy. Profiles retrievals are performed by onion peeling from light paths calculated by ray tracing. The vertical resolution is 1.4 km. The error on altitude registration derived form the location and altitude of the balloon measured to within ±10 m by GPS is estimated to ± 50 m. The precision of ozone concentration estimated from the error on the spectral fit is 2% at 20 km, 5% at 17.5 km and 10% at 10 km. Since the cross-sections are independent of temperature in the Chappuis bands, the systematic error is of the order of 1.5%.

2.2 MIR long duration flights during HIBISCUS

The Infra-Red Montgolfier (MIR) is a hot air balloon heated by the Earth’s thermal emission during nighttime and sunlight during the day. Of 45 000 m³ volume it allows carrying about 60 kg at an altitude 28 km (13 hPa) during the day and between 18 and 24 km at night depending on the cloud clover with the exception of the very first 2-3 days of flight when the helium used for the initial ascent not totally escaped, makes the balloon to fly at higher altitude, up to 34 km (4hPa) on the first day. The data used here are those collected during three balloon flights all launched during the southern summer in February, in 2001, 2003 and 2004 from Bauru at 22°S in Brazil. The balloons lasted in flight for respectively 34, 9 and 39 days. In all cases the flights terminated by automatic separation of the payload when the balloon reached 18 km at night over extremely high and cold convective clouds respectively in northern Argentina, over a hurricane in the Coral sea and over the South Pacific Convergence Zone. Their trajectories are shown in Fig. 2. The first and the last provided a complete view on the zonal distribution of ozone while the observations of 2003 are limited to the Pacific. The balloons passed over the SHADOZ stations of Samoa, Fiji and Reunion Island where dedicated ozonesonde ascents could be performed, as well as tropospheric and stratospheric lidar measurements at the last station.

![Fig. 1. SAOZ long duration balloon payload.](image)

![Fig. 2. IR Montglofier trajectories in 2001, 2003 and 2004, all launched in February from Bauru in Brazil.](image)
3 ANALYSIS OF SAOZ MEASUREMENTS

3.1 Ozone profiles

Fig. 3 displays the ozone profiles retrieved along the flights: 54 in 2001, 9 in 2003 and 78 in 2004 (sunrise in blue and sunset in red). With the exception of measurements performed above a hurricane in 2003 (green) showing a lift of the bottom part of the profile, the ozone zonal variability is very limited, 3-4%, above 20 km, in contrast to the upper troposphere and the TTL layer between 12 and 19 km where the ozone concentration varies by ± 70%. The small variability in the lower stratosphere provides an ideal tool for satellite validation.

Fig. 3. Ozone profiles recorded during the three flights at sunrise (blue), sunset (red) and over the hurricane (green).

3.2 Evaluation of SAOZ ozone performances

The existence of collocated lidar and ozonesondes ascents offers a unique opportunity for checking the quality of the SAOZ ozone measurements. Fig. 4 shows on, the left the comparison in 2001 between the profile derived from a combination of the tropospheric and stratospheric ozone lidars at Reunion island, and the closest SAOZ measurements, where the errors bars represent the estimated precision of SAOZ. The difference between the two is shown on the right without (red) and with (blue) an altitude shift of +50 m applied on the lidar. The smallest average difference is reached for a shift of 25 m, meaning the error on the altitude registration of the SAOZ is –25 ± 25 m. The difference in ozone concentration never exceeds ±5% between 16 and 26 km.

Fig. 4. Left: lidar (red solid line) and closest SAOZ ozone profiles at La Reunion in March 2001. Right: difference in red and after applying a vertical shift of +50 m to the lidar. Black solid lines: random errors of the lidars.
Similarly, Fig. 5 shows an example of comparison between SAOZ and ozone sondes at Bauru and at Samoa. In the stratosphere above 20 km, the smallest difference is observed after shifting the sondes down by 150-250 m. This result applies to all individual comparisons with ozonesondes as well to zonal mean profiles. It just corresponds to the time constant of 50-60 s of the Electro Chemical Cell (ECC) on a balloon ascending at 5-6 m/s. The situation is different in the troposphere where the sondes and SAOZ are found in good agreement at Bauru and Reunion Island, but not at Samoa and Fiji where the sondes are found systematically low biased by 30-50% compared to SAOZ. Besides instrument differences (solution buffering or not, dark current measurements, sonde analysis software), a possible problem in the comparison with Samoa and Fiji located in the South Pacific Convergence, is the presence of thick clouds which makes collocated measurements difficult (the closest SAOZ measurements available are 1400 km away). Another possible explanation could be the frequent presence of “zero ozone layers” in the ascents in this convective region because of the uplift of air having experienced ozone destruction at the surface of the ocean. SAOZ as well as all remote sensors could not perform within clouds. There is a possibility that this could introduce a systematic bias in these conditions.

3.3 Ozone zonal variability

The reason for ozone variability in the SAOZ data (standard deviation compared to mean zonal profile) was further explored by a multi-regression analysis using two proxies: potential vorticity (PV) provided by an isentropic CTM from ECMWF analysis for horizontal transport, and height between the 340 K and 370 K surfaces used as an indicator of convection. The impact of each of horizontal and vertical transport was thus removed using the slope of the correlation. Fig. 7 shows the result of this analysis in 2004. On the left is the observed variability in blue, the residual variability after removing the contribution of horizontal transport in green, and after removing the influence of both horizontal and vertical transport in red. The plot on the right shows the percent relative contribution of each of them. According to this analysis most of the ozone variability in the tropical UTLS could be attributed to horizontal transport that is quasi-isentropic exchange with the mid-latitude stratosphere. The contribution of vertical exchange is relatively small and significant only between 13 and 18 km. After removing those contributions, the residual is of the order or smaller than the precision estimated for SAOZ measurements, leaving little room for additional contributors such as photochemical production. But more important for the comparison with satellite measurements, the residual variability including SAOZ errors does not exceed 2 % above 20 km, between 4 and 7% in the TTL and 7-8% in the UT.

4 COMPARISON WITH SATELLITE DATA

The procedure for comparing satellites to SAOZ data is based on these findings. Fig. 8 illustrates the procedure on the example of ODIN-OSIRIS in 2004. The plot on the left is the comparison of mean zonal profiles of both instruments (SAOZ in solid line, OSIRIS dotted) and on the middle the difference. A systematic vertical shift could be observed which is adjusted to provide the minimum average difference between 20 and 30 km (dotted line). The altitude shift is the altitude registration error (-900 m in present case), while the average difference above 20 km is a measure of the relative bias (+2.0%). The plot on the right shows the comparison of zonal variability before (black) and after (red) subtraction of the contribution of horizontal transport. The last is a measure of precision. In the OSIRIS case it is ±5% in the stratosphere above 18 km, but increases below reaching about 20% at the altitude of the tropopause. However, the question here, as in all retrievals of limb observations, is the significance of concentrations in the lower layer strongly weighted by the a priori profile used in the retrieval process.
couple of instruments derived here are very consistent with those of previous evaluations.

The same procedure has been applied to all ozone profiles available during the balloon flights: SHADOZ, SAGE II and HALOE in 2001; the same plus ODIN and ENVISAT instruments in 2004. The results for each of them, for 2004 and (2001) into brackets, are displayed in Table 1. Also indicated is the version of the algorithm used, the last available at the time of this study. A remarkable feature is the frequent errors in altitude registration varying from +100/+200 m for solar occultation systems (SAGE II, HALOE) to shifts as large as +2000/-900 m for limb viewing instruments (SCIAMACHY, OSIRIS). As expected, the most precise altitude is that of the GOMOS star occultation system whose observing direction is very accurately known since the light source is punctual. As expected also the ozonesondes are shifted up by 200-300 m because of their time constant. Curiously, the SAGE III moon occultation instrument, using a moon edge detection algorithm similar to that of the SAGE II solar system, is showing a shift down by 550 m, which may come from errors in the satellite location. Another feature, which did not receive any explanation yet, is the increasing shift up or ozone low bias of HALOE profiles below 22 km, as large as 1500 m (or 40% in ozone) at 18 km, present at both sunrise and sunset, in 2001, 2003 and 2004 compared to SAOZ, identical to that already reported between SAGE II and HALOE. Since no such difference was found between SAOZ, SAGE II and all other systems, it can be safely attributed to HALOE.

The ozone bias relative to SAOZ is generally small, between-2 % and +4 % for most of the instruments measuring ozone in the visible Chappuis bands and near IR (SAGE II and III, GOMOS, OSIRIS and HALOE), within their ±5% known accuracy for ozonesondes, but it increases for SCIAMACHY in the UV (-6.6 %), MIPAS in the mid-IR (+7 %) and SMR in the sub-millimetre region (+7 %). Most of above biases are not new. On average, differences between a couple of instruments derived here are very consistent with those of previous evaluations.
New here is the evaluation of the precision of each by a statistical method based on the limited ozone variability in the tropical stratosphere, moreover shown to be mainly due to horizontal transport efficiently removed by a multi-regression analysis using potential vorticity (PV) as proxy. Most precise are found to be SAGE II and SAOZ (~2 %), followed by HALOE (3-4 % above 22 km), the sondes, SAGE III moon and OSIRIS (4.5 %), GOMOS above 22 km and SCIAMACHY (~6 %), MIPAS (8.5 %) and finally SMR (16 %). All these figures are more or less consistent with previous precision estimates by other methods, but more importantly they fully agree with known or suspected limitations of instrumental and retrieval techniques.

Table 1. Summary of performances relative to SAOZ

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Alt. Bias (m)</th>
<th>O$_3$ Bias (%)</th>
<th>Precision (%)</th>
<th>Version / Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td>SAOZ</td>
<td>-</td>
<td>-</td>
<td>2 (2)</td>
<td></td>
</tr>
<tr>
<td>Ozonesondes</td>
<td>+200 (300)</td>
<td>+1.5 (~4)</td>
<td>4 (5)</td>
<td></td>
</tr>
<tr>
<td>HALOE Z&gt;22km</td>
<td>+200 (&lt;+100)</td>
<td>+1.8 (&lt;1)</td>
<td>3.4 (4)</td>
<td>V19 Bias or altitude, and precision degrading below 22 km</td>
</tr>
<tr>
<td>SAGE II</td>
<td>+150 (+150)</td>
<td>+2.5 (+4.5)</td>
<td>2.5 (2)</td>
<td>V6.2</td>
</tr>
<tr>
<td>SAGE III moon</td>
<td>-550</td>
<td>+2.5</td>
<td>4.5*</td>
<td>V3, 4 profiles only. * H. transport not removed</td>
</tr>
<tr>
<td>Odin-OSIRIS</td>
<td>-900</td>
<td>+2</td>
<td>5</td>
<td>V2.3</td>
</tr>
<tr>
<td>Odin-SMR</td>
<td>+300</td>
<td>+7</td>
<td>16</td>
<td>V222</td>
</tr>
<tr>
<td>GOMOS Z&gt;22 km</td>
<td>-50</td>
<td>-1.5</td>
<td>6</td>
<td>V6.0 Bias and precision degrading below 22 km</td>
</tr>
<tr>
<td>SCIAMACHY</td>
<td>+2000</td>
<td>-6.6*</td>
<td>6</td>
<td>V1.6, *Z shape profile</td>
</tr>
<tr>
<td>MIPAS</td>
<td>-300</td>
<td>+7</td>
<td>8.5</td>
<td>V4.61</td>
</tr>
</tbody>
</table>

Finally, tropospheric ozone measurements by remote sensing from space remain a difficult task. There are several reasons for that: the strong signal attenuation by Rayleigh scattering, the masking by clouds, the weakening of IR emission at low temperature, or the increased scintillation because of turbulence, etc. Another concern for limb observing systems using optimisation retrievals is the information effectively carried by the measurements compared to the weight of the a priori climatic profile. Finally, since remote observations are not possible within clouds, there is a risk of systematic overestimation of tropospheric ozone if the concentration is reduced inside the clouds particularly in convective regions. The best instrument for measuring ozone in the troposphere remains ozonesondes. The performances of most satellite instruments degrade rapidly below 20 km and sometimes 22 km (HALOE, GOMOS). The only satellite, which may provide some meaningful information in the upper troposphere, is SAGE II (may be SAGE III also) although low biased by 50 % and on limited (50 %) precision.

6 ACKNOWLEDGMENTS

The authors thank the engineers and technicians of the Service d’Aéronomie, A. Garnier the project manager, P. François and E. d’Almeida who built the SAOZ instrument and M. Nunes-Pinharanda for the data processing, and the personnel of the Institute of Meteorology (IPMet) of the State University of Sao Paulo (UNESP) and of the Centre National d’Etudes Spatiales (CNES) for their support in balloon operations. The project was supported by the ENVISAT validation programme ESABC, project 713, the Programme National de Chimie de l’Atmosphère (PNCA) in France and the European Commission within the HIBISCUS project (contract EVK2-2001-000111). They are all gratefully acknowledged.

7 REFERENCES