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Evaluation of SHADOZ sondes, HALOE and SAGE II ozone profiles at the tropics from SAOZ UV-Vis remote measurements onboard long duration balloons

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Abstract. Ozone profiles from 10 to 26 km have been obtained at almost constant latitude (20±5°S) in the tropics using SAOZ UV-vis spectrometers flown onboard long duration balloons in 2001 and 2003. The precision of the measurements is estimate to be better than 2% in the stratosphere (3.5% accuracy) and 5–6% in the troposphere (12% and 25% accuracy at 15 km and 10 km respectively) with an altitude uncertainty of -30 ± 25 m. The variability of ozone concentration along a latitudinal circle at 20°S in the SH summer is found smaller than 3–4% above 20 km, but increasing rapidly below in the Tropical Tropopause Layer (TTL). The high correlation between PV and ozone suggests that most of this variability can be attributed to quasi-horizontal exchange with the mid-latitude stratosphere.

The performances of the SHADOZ ozonesonde network, HALOE and SAGE II in the tropics have been studied by comparison with SAOZ measurements. In the stratosphere, the main discrepancies arise from differences in altitude registration, particularly sensitive between 20 and 26 km in the tropics because of the strong gradient of ozone concentration. In the upper troposphere, the SAOZ measurements are consistent with those of the sondes and the lidar in cloud free conditions, but biased high by 60% on average compared to ozonesondes over the Western Pacific, at American Samoa and Fiji. The likely explanation is the frequent occurrence of near zero ozone layers in the convective clouds of the South Pacific Convergence Zone which cannot be seen by SAOZ as well as all ground-based and space borne remote sensing instruments. Compared to SAOZ, SAGE II displays a 50–60% low bias similar to that already known with the ozonesondes, and a larger zonal variability. However, the significant correlation with PV suggests that useful information on tropospheric ozone could be derived from SAGE II. Finally, the unrealistic large offsets and variability in the HALOE data compared to all others, indicates that the measurements of this instrument are of limited use below 17 km.

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1 Introduction

The ozone distribution in the tropical upper troposphere and lower stratosphere (UT/LS) is the result of a combination of transport and chemical mechanisms. Among them are: quasi-horizontal exchange between equatorial and mid-latitude regions (Dessler et al., 1995); deep convection in the troposphere (Dessler, 2002) and slow upward Brewer-Dobson circulation above; photochemical production from precursors lifted by convection from surface levels or NO_x production by lightning (Jeker et al., 2000); and heterogeneous reactions on thin cirrus (Solomon et al., 1997). However, because of the limited data available, particularly in the upper troposphere, the relative contribution of each of the processes is still poorly understood in the tropics. Although remote ozone profile measurements from space platforms have been available for some years, their performances near or below the high tropical tropopause are still debated (e.g. since 1984 from the Stratospheric Aerosols and Gas Experiment II (SAGE II) (Mauldin et al., 1985) and since 1991 from the HALogen Occultation Experiment (HALOE) (Russell et al., 1993)). To fill-in the gap, an ozonesonde network was initiated in 1998, the Southern Hemisphere ADditional OZonesondes (SHADOZ) project, now involving eleven stations distributed throughout the tropics and subtropics (Thompson et al., 2003a).

Here we report on remote sensing ozone measurements performed with a SAOZ UV-Visible spectrometer flown on board circum-navigating long duration balloons in 2001 and 2003 at the Southern Tropics during the SH summer. This

Fig. 1. SAOZ long duration balloon payload. The cone at the top of the instrument defines a 360° azimuth, −10°, +10° elevation FOV within which the full sun could be observed at sunrise and sunset without additional tracking system.

new independent data set is used for studying the relative performances of both space-borne instruments and sondes.

The following Sect. 2 provides a description of the SAOZ measurements, the retrieval process, an estimation of the accuracy and precision, and a summary of the data available from the flights. Section 3 is devoted to the validation of the data from a comparison with lidar measurements at Reunion Island during a balloon overpass in 2001, and from the residual ozone variability along a latitudinal circle after removing the contribution of horizontal transport by correlation with PV (Potential Vorticity). Section 4 describes the results of comparisons between SAOZ and SHADOZ sondes, HALOE and SAGE II ozone measurements. Finally, the relative performances of all the systems in the tropical UTLS are summarised in Sect. 5.

2 SAOZ ozone profiles in the tropics

SAOZ is a diode array UV-Visible spectrometer originally designed for the monitoring of total ozone and nitrogen dioxide by the zenith-sky technique from the ground (Pommereau and Goutail, 1988), later developed as a balloon version for remote profile measurements by solar occultation (Pommereau and Piquard, 1994). 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) (Malaterre et al., 1996), after an idea of Pommereau and Hauchecorne (1979). The data are those of two flights at the tropics launched from Bauru at 22°S in Brazil during the SH summer season in February-March. The first in 2001 circumnavigated twice in 34 days, while the second in 2003 lasted for only 9 days across the Pacific (brought down by a hurricane over the Coral Sea).

2.1 SAOZ ozone measurements

Ozone is measured in the visible Chappuis bands in the 450–620 nm spectral range. The spectra are analysed by the differential absorption technique (DOAS). The profiles are retrieved by onion peeling, after calculating the light path by ray tracing. Compared to a satellite, the advantage of a balloon moving slowly is to allow the exposure to vary from 0.5 s to 52 s for compensating the strong increase of attenuation at low tangent height, allowing the measurements to be continued down to cloud top or to about 6 km in clear conditions.

2.1.1 SAOZ long duration balloon payload

This version of the SAOZ instrument makes use a flat field holographic grating and a 1024 diode-array detector providing spectral measurements of 1.1 nm resolution in the 300–650 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 360° azimuth field of view and −10°, +10° elevation, followed by a pile of three diffusers to homogenise the light beam (Pundt et al., 2002). Measurements are performed during twilight periods only from 89° to 95° SZA with 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 O_3, NO_2, O_4, O_2, tropospheric H_2O and solar flux 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, which provide autonomous operation for about one month. The total instrument weight is 28 kg.

2.1.2 Spectral analysis

The onboard spectral retrieval, identical to that in use for post processing in the laboratory, is based on the DOAS technique (least squares fitting with laboratory cross-sections) after rationing the spectra to a reference one recorded at high altitude (30 km) and high sun (40° SZA) with the same instrument during a previous test flight. Ozone is measured in the 450–620 nm Chappuis bands using laboratory absorption cross-sections of Brion et al. (1998) of 1.5% quoted accuracy. Within the 410–690 nm spectral range, those data have been shown to be consistent within 2% with all other recently available laboratory measurements (Orphal, 2003). They all agree in showing very small or no temperature changes of the peak cross-sections between 550–650 nm, but some irregular results at wavelengths shorter than 500 nm, however of small impact in the SAOZ retrieval because of the very small cross-sections in this region. No significant difference in ozone could be found between the results of the onboard retrieval and those of the post flight re-analysis of the data stored in memory in the 2001 payload (recovered in Argentina). In both cases, the fitting error on the slant column is $5e^{17}$ mol/cm^2, almost constant during the occultation down to 15 km, increasing to $2e^{18}$ mol/cm^2 at the bottom of the profile at 6 km.

2.1.3 Profile retrieval

Profiles of ozone concentration are retrieved by the onion peeling technique within 1 km thick shells after calculating light paths including refraction effects by ray tracing (SAOZ algorithm version 3.4). The ozone amount in the reference spectrum is determined from the reading at 89° SZA on the first morning of flight when the MIR, still inflated with helium, is floating at 36 km. The random uncertainty (precision) on ozone concentration is derived from the fitting error of the column density in the retrieval process. On average, it varies from 1.2% at 25 km at the altitude of the ozone maximum, to 2% at 20 km, 5% at 17.5 km, 10% at 15 km, 23% at 10 km and 33% at 7 km.

The altitude is a geometric altitude. Its accuracy is directly related to that of the GPS 3D location and clock (±100 m and 0.1 s in 2001, improved to ±10 m and 0.1 s in 2003 after removal of scramble in the GPS system) from which the SZA at the location of the balloon and then the tangent height, are derived. The vertical resolution of 1.4 km is limited by the size of solar disk. Pressure and temperature are those of the ECMWF operational analyses (6h, T106, 2°×2°, 60 levels) at the location of the tangent point, very consistent (0.5±0.4 hPa, −1.2°±1.9°C) with that measured onboard the balloon in the stratosphere. The average difference in altitude between ECMWF and MIR density levels is 100 m at 25 km, 50 m at 20 km, and 25 m at and below 18 km. Finally, data contaminated by clouds are removed by looking at the atmospheric extinction at 615 nm. The profile is cut when the optical thickness along the line of site exceeds that of pure Rayleigh attenuation by 0.2 (cloud extinction of 1×10^{-3} km^{-1}, that of a thin cirrus). In the absence of clouds, profiles could be retrieved down to 6 km. Otherwise the profiles are cut immediately above the cloud layer, observed occasionally up to 19 km.

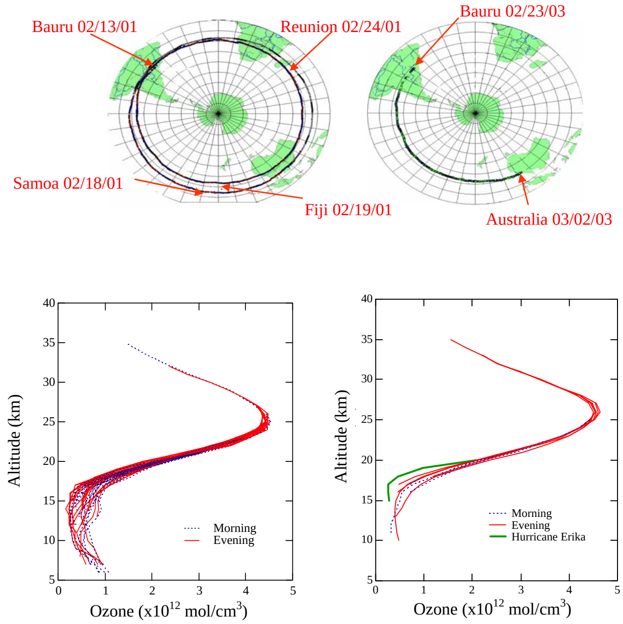

Fig. 2. Balloon trajectories and ozone profiles in 2001 (left) and 2003 (right). Blue: sunrise; Red: sunset; Green: hurricane.

2.2 Summary of measurements in 2001 and 2003

The data used here are those obtained during two flights at the tropics launched two years apart from Bauru, Brazil (22° S, 49° W), on 13 February 2001 and 23 February 2003 in preparation for the HIBISCUS project held in 2004. The first flight lasted for 34 days and two circumnavigations, resulting in 54 useful profiles (roughly half at sunrise, half at sunset) at almost constant latitude (22°±5° S), before falling in northern Argentina where the payload and the onboard recorded data were recovered. The second lasted for 9 days only, also at almost constant latitude (22°±2° S), providing 11 useful profiles across the Pacific before being lost.

The balloons (45 000 m^3 volume) were carrying a service payload controlling the flight (balloon technical parameters, safety systems, cut down below 16 km, GPS, pressure, temperature, and global IR radiometer) and the SAOZ gondola 50 m below was independent of the service system. The total weight of the system was about 60 kg.

Since MIRs are hot air balloons only heated by the Earth radiation at night and by the sun during daytime, their altitude varies from 26–27 km at noon to 18–24 km at night depending on the cloud cover. However, since they are inflated with helium for their first ascent, they fly at higher altitude during the first three days, 34 km on the first morning, the duration required to lose excess helium.

The trajectories of the balloons in the summer stratospheric Easterlies and the ozone profiles recorded along their flights at sunset (red) and sunrise (blue), are shown in Fig. 2. The few profiles starting at high altitude are those from the

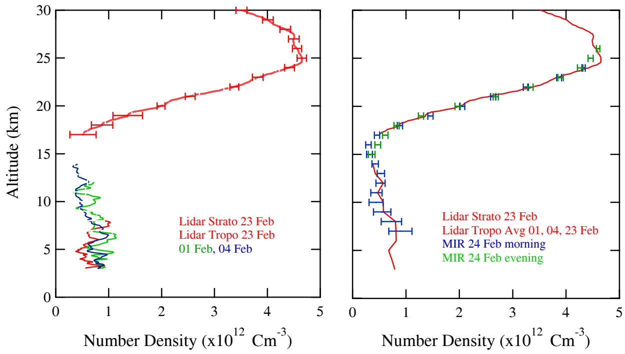

Fig. 3. Left: Stratospheric and tropospheric ozone lidar measurements at Reunion Island. The error bars indicate shot noise. Note the drop at 20 km between lower and higher channels. Right: comparison between average lidar on 1, 4 and 23 February and SAOZ measurements on 24 February in the morning and the evening. The error bars indicate SAOZ random errors derived from the spectral fit.

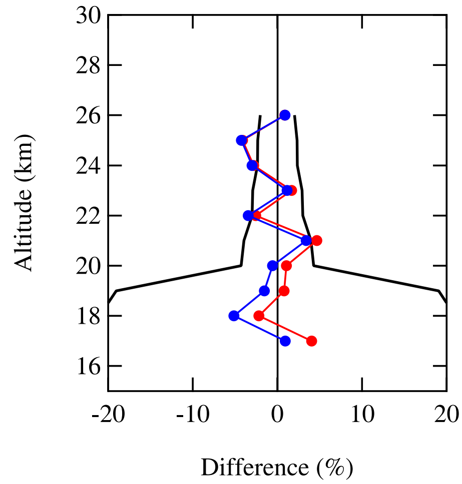

Fig. 4. Percent difference between SAOZ and lidar ozone (red), and after shifting SAOZ up by 50 m (blue). The black solid line indicates the sum of SAOZ and lidar random errors.

first days of flight. The ozone concentration along the flight is almost constant in the lower stratosphere, but highly variable in the upper troposphere between 12 and 19 km. The profile departing most from the mean is that observed near the hurricane in 2003 (green line in Fig. 2) showing a remarkable upward shift up to 19 km.

3 Validation of SAOZ altitude registration and precision

In complement to the error analysis already provided, the quality of the SAOZ measurements has been evaluated by two independent methods. The first is a comparison with stratospheric and tropospheric lidar observations at Reunion Island in the Indian Ocean, which could be performed during an overpass of the balloon close to the station. The event provided a unique opportunity for checking the altitude registration as well the accuracy of the SAOZ retrieval. The second method proposed is to evaluate the precision of the ozone measurements from their variability, after subtracting the contribution of horizontal and vertical transport by the use of adequate proxies derived from ECMWF analyses.

3.1 Validation of altitude registration from lidar at Reunion Island

The 2001 flight passed close to the NDSC (Network for Detection of Stratospheric Change) and SHADOZ station of Reunion Island, where three instruments could be activated for simultaneous measurements: a tropospheric ozone lidar (Baray et al., 1999), a dial stratospheric ozone lidar (Portafaix et al., 2003) and a dedicated ozonesonde ascent. Since other ascents of the SHADOZ network are available along the balloon flights, the comparison with the ozonesonde will not be considered here, but together with the others in the next section. Only the lidar measurements will be compared, whose unique characteristic is to provide a precise (±25 m) check of the altitude registration.

The MIR passed close to Reunion Island on the afternoon of 24 February. The closest SAOZ measurements are those of the morning and the evening of the same day when the tangent points at 20 km were located respectively at 22° S, 70.7° E (1400 km East of the station) and 21.2° S, 51° E (400 km West). The first was performed in clear sky down to 7 km, while the second was stopped at 16 km because of the presence of high-level clouds. The stratospheric ozone lidar could be activated for 168 min during the evening of February 23 prior to the MIR pass, while the troposphere lidar performed during the same night but from 3 to 8 km only.

Figure 3 shows all lidar measurements made on the evening of 23 February as well as two other tropospheric profiles performed earlier in the month, on 1 and 4 February, reaching higher altitudes. Also shown by error bars is the shot noise of the stratospheric lidar (only every kilometre for clarity). A drop could be seen in the error bars at 20 km at the transition between lidar lower and higher channels. The uncertainty of the tropospheric ozone lidar (Baray et al., 1999) is estimated as 10 ppb (7 mol/cm^3 at 10 km, about 10%). The comparison between lidar and SAOZ ozone profiles is displayed on the right side of Fig. 3, the SAOZ data being represented by error bars. The measurements are consistent within their error bars with the exception of a single point at 25 km which is likely due to the relatively poor (1.4 km vertical and 200 km horizontal) resolution of SAOZ compared to that of the lidar.

A possible bias in the SAOZ altitude registration has been investigated by looking at the change of average standard deviation between the two measurements when shifting the

Table 1. Variation of SAOZ-LIDAR difference between 17–24 km after shifting SAOZ by DZ meters.

DZ (m)	Bias (%)	Standard Dev. %
+200	−7.1	4.6
+100	−3.1	3.1
+50	−1.1	2.8
+30	−0.05	2.7
+25	−0.05	2.8
0	1.0	3.0
−50	3.0	3.4
−100	5.0	4.1

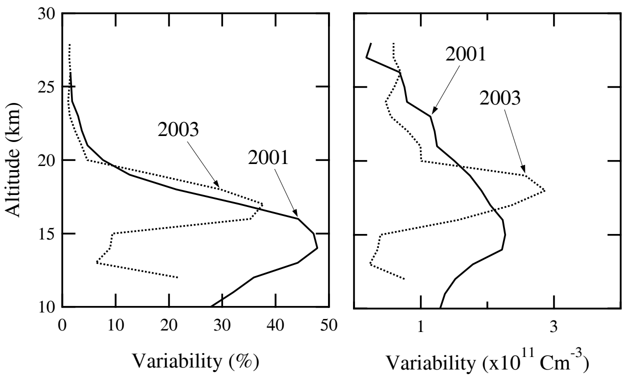

Fig. 5. Zonal variability of ozone concentration in 2001 and 2003 (Pacific only). Left: percent; Right: number density.

SAOZ altitude in the 17–24 km altitude range where the ozone gradient is the strongest. The results of the calculations are shown in Table 1. The impact of a vertical shift of 50 m on the profile of the SAOZ-lidar difference can be seen in Fig. 4. A minimum standard deviation corresponding to a minimum slope of the difference is reached for a SAOZ shift up by about 30 m. Since this is the precision of the lidar altitude, it is barely significant, as well as the corresponding bias. The SAOZ and lidar profiles are thus very consistent in both altitude registration (−30±25 m) and ozone concentration (0.05±2.7%).

3.2 Evaluation of SAOZ precision from ozone zonal variability

The variability (standard deviation compared to the mean) of ozone concentration along the flight track in 2001 and 2003 is shown in Fig. 5 (percent on the left, concentration on the right). In the stratosphere, above 20 km, the figures are very similar in both years showing low variability (<5%). Below 20 km the variability increases, reaching a maximum of 40% (in 2001) at 14 km in the TTL and then decreases at lower altitude. The picture is a little different in 2003 over the Pacific where the ozone is less variable below the tropopause

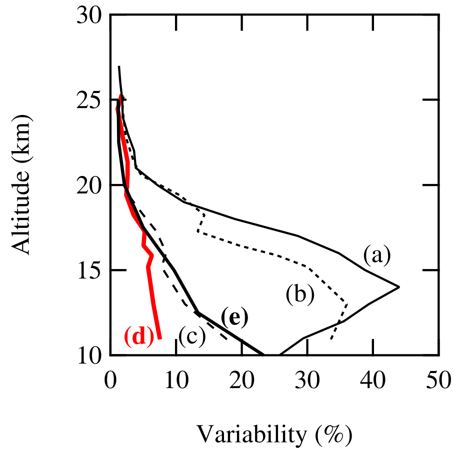

Fig. 6. Analysis of contributions to ozone variability in 2001: **(a)** number density; **(b)** MR on isentropic surfaces; **(c)** after removing horizontal transport; **(d)** red, after removing vertical and horizontal transport; **(e)** bold, SAOZ random error estimate from spectral fit.

at about 16 km, but larger immediately above between 17–19 km due to the influence of the hurricane.

Apart from random errors, several mechanisms could contribute to the variability, e.g. horizontal and vertical transport, convective lifting of ozone poor air from oceanic surfaces and photochemical production by precursors. A well-known feature of upper tropospheric ozone in the tropics is the wave number one distribution displaying a maximum over the Atlantic and Africa and a minimum over Western Pacific (Thompson et al., 2003b).

Although the study of mechanisms controlling the ozone distribution in the tropics is beyond the scope of this paper, we propose a method for removing the impact of horizontal and vertical transport, resulting in a residual variability indicative of the measurement precision. The method is based on a multiple regression with several proxies all provided by ECMWF operational analyses interpolated at the location of the SAOZ tangent points. The impact of the longitudinal change of isentropic surface altitude is first removed by converting ozone number density at constant altitude into mixing ratios on potential temperature surfaces. The quasi-horizontal transport is evaluated by correlating the ozone changes to that of potential vorticity (PV) on isentropic surfaces provided by the MIMOSA high-resolution contour advection model (Hauchecorne et al., 2002). The contribution of vertical convective transport is removed by correlation with the altitude difference between the 370 K and 340 K isentropic levels.

The results of the calculations applied to the SAOZ measurements of 2001 are shown in Fig. 6 where: (a) is the initial standard deviation of concentration at constant altitude levels; (b) is that of mixing ratios on isentropic surfaces; (c) is the residual after removing horizontal transport by correlation with PV; (d) is the remaining variability after removing

Table 2. SHADOZ selected ascents.

Station	February	March	Total
2001			
Samoa	16, 23	none	2
Fiji	2, 9, 16, 23	2, 9, 19, 26	8
Reunion Isl.	2, 15, 21, 24, 28	7, 15, 21, 28	9
2003			
Samoa	7, 21, 28	7, 14, 21, 28	7
Fiji	21	14	2

vertical convective transport, and (e) is the average SAOZ random error derived from the profile retrieval process. At all levels above 12 km the largest contribution is that of horizontal exchange. Convection becomes more important only below that level, its impact drops above the tropopause around 15.5–16 km, but is still present up to 19 km (the top of the TTL). Curiously, the final residual is smaller than SAOZ errors below about 18 km suggesting some overestimation of the error in the SAOZ spectral fit. The very likely explanation for that is the presence of a systematic error due to an interference with water vapour absorption bands in the visible missing in the spectroscopic HITRAN data base as shown recently by Coheur et al. (2002). If the residual variability, which may still include some photochemical signal, is adopted as an indicator of the random error, the precision of SAOZ ozone measurements would be 2% in the stratosphere, 5.7% at the tropopause and 6.5% at 12 km in the upper troposphere at 12 km. Measurement accuracy, after adding the uncertainty on ozone cross-sections and the water vapour systematic contribution, would be of about 3% ($1.2e^{11}$ mol/cm^3) at 25 km in the stratosphere, 7% ($3.5e^{11}$ mol/cm^3) at the tropopause, and 15% ($7.5e^{11}$ mol/cm^3) at 12.5 km.

4 Comparison with SHADOZ, HALOE and SAGE II

A number of ozone profiles close to the latitude and during the period of the MIR flights in 2001 and 2003 are available from the SHADOZ ozonesonde network as well as from two NASA space instruments, HALOE onboard UARS and SAGE II onboard ERBS. The comparison with those of SAOZ and the use of the PV correlation method described above allow studying the relative performances of each. The mathematical background used in this section is given in appendix A.

4.1 SHADOZ ozonesonde network

The SHADOZ network was initiated by NASA in 1998 to develop a coordinated ozonesonde network at the tropics, putting together some 10–12 stations supported by in-country agencies or universities (Thompson et al., 2003a). Relevant stations at the latitude of the MIR flight are Reunion Island (21° S, 55.5° E) in the Indian Ocean and American Samoa (14.2° S, 171° W) and Fiji (18.1° S, 178° W) in the Western Pacific.

Ozone measurement at SHADOZ sites are made with balloon-borne ECC (Electrochemical Concentration Cell) ozonesondes coupled with a standard Vaïsala radiosonde and a sensor for relative humidity. However, as various stations prepare their sondes and process the raw data differently, and as each sonde is a new instrument, the evaluation of the precision and accuracy of the SHADOZ data is not straightforward. According to Thompson et al. (2003a), (a) the sonde precision is 5%; (b) integrated total ozone column amounts from the sonde profiles are usually to within 5% of independent measurements from ground-based instruments and overpass measurements from the TOMS satellite; (c) systematic variations in TOMS-sonde offsets and in ground-based-sonde offsets from station to station reflect biases in sonde technique as well as satellite retrieval; and finally (d) there is evidence for a zonal wave-one pattern in total and tropospheric ozone, but not in stratospheric ozone.

4.1.1 SHADOZ Data selection

The data used in this study are taken from the SHADOZ database (http://croc.gsfc.nasa.gov/shadoz/). Ozone partial pressure is converted to number density using the radiosonde pressure and temperature. The altitude is a geopotential height.

The ascents selected are those of the weekly flights available in the database in February and March at each of the three stations in 2001, and at Samoa and Fiji only in 2003, since the MIR flight was limited to the Pacific on that year. The dates of the ascents are displayed in Table 2. In total, 19 events are available in 2001 and 9 in 2003.

4.1.2 Statistical comparison

The comparisons between ozonesonde and SAOZ mean profiles, difference and variability are displayed in Fig. 7 and the results are summarised in Tables 3 and 4 for the stratosphere and the troposphere respectively.

In the stratosphere above 20 km, the two mean profiles are very similar but shifted in altitude. The relative difference decreases with altitude. But the comparison improves when shifting down the sonde in altitude. In 2001, an average minimum deviation between 18 and 26 km is reached by shifting the sonde down by 300±25 m (dashed-dotted line). After applying this correction the average bias drops from −14.3±6.6% to −3.7±1.0% (sonde low biased compared to SAOZ). In 2003 over the Pacific, the optimum shift is even larger, 500 m, and the difference drops from −18.2±7.9% to −5.9±1.3%. A systematic and constant 300 m altitude shift of the sonde is not a surprise. It corresponds exactly to the lag expected from the known 50–60 s response time of the

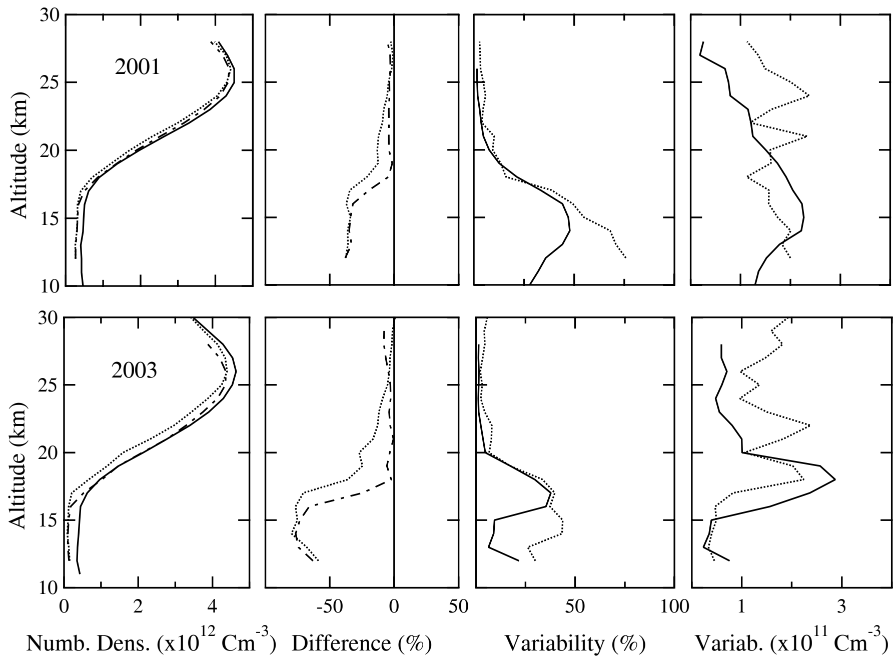

Fig. 7. Comparison between SHADOZ and SAOZ mean profiles. Top: 2001. Bottom: 2003. From left to right: mean profiles, percent difference, percent and absolute standard deviation. Solid line: SAOZ; dotted: SHADOZ; dotted-dashed: SHADOZ altitude adjusted.

Table 3. SHADOZ-SAOZ difference and variability in the stratosphere.

	Diff %	Diff % Alt adjusted	Var SAOZ %	Var Shadoz %	Var SAOZ e^{10} mol/cm^3	Var Shadoz e^{10} mol/cm^3
2001	−14.3±6.6	−3.7±1.0	2.8±2.3	5.3±2.7	8.7±4.5	16.8±4.6
2003	−18.2±7.9	−5.9±1.3	2.1±1.3	4.8±2.0	7.0±1.9	15.3±4.4

Table 4. SHADOZ-SAOZ difference and zonal variability in the troposphere.

	Diff %	Diff % Alt Adjusted	Var SAOZ %	Var Shadoz %	Var SAOZ e^{10} mol/cm^3	Var Shadoz e^{10} mol/cm^3
2001	−36.1±1.5	−32±6.3	35.8±12.9	48.3±24	19.6±2.7	16.8±4.6
2003	−68.2±7.0	−61.5±16.2	20.1±12.3	33.9±9.2	13.9±10.9	9.0±7.7

ECC cell on a balloon ascending at 5–6 m/s (SPARC, 1998; Johnson et al., 2002). The 200 m additional shift in 2003 could be largely explained by the large latitude difference. Indeed on that year (Fig. 2) and in contrast to 2001 when the MIR passed close to the three stations, the balloon travelled across the Pacific at an average latitude of 22.0±0.6° S, that is 4° south of Fiji and 8° south of SAMOA where 7 of the 9 available ascents were performed.

For the best comparison with 19 sondes in 2001, the average bias with SAOZ in the stratosphere between 18 and 26 km after provision for the time lag is −3.7±1.0%. Finally, the standard deviation of the ozone profiles (right panels of Fig. 7 and Table 3) provides also an estimation of the relative precision of the sondes (5% or about 16e^{10} mol/cm^3), twice larger than that of SAOZ in the stratosphere.

In the upper troposphere below 18 km (Table 4), the sondes are low biased compared to SAOZ, by a larger amount over the Pacific in 2003 than over the whole latitudinal circle in 2001. But since tropospheric ozone is known to vary by more than a factor 2 between Africa/Atlantic and the West

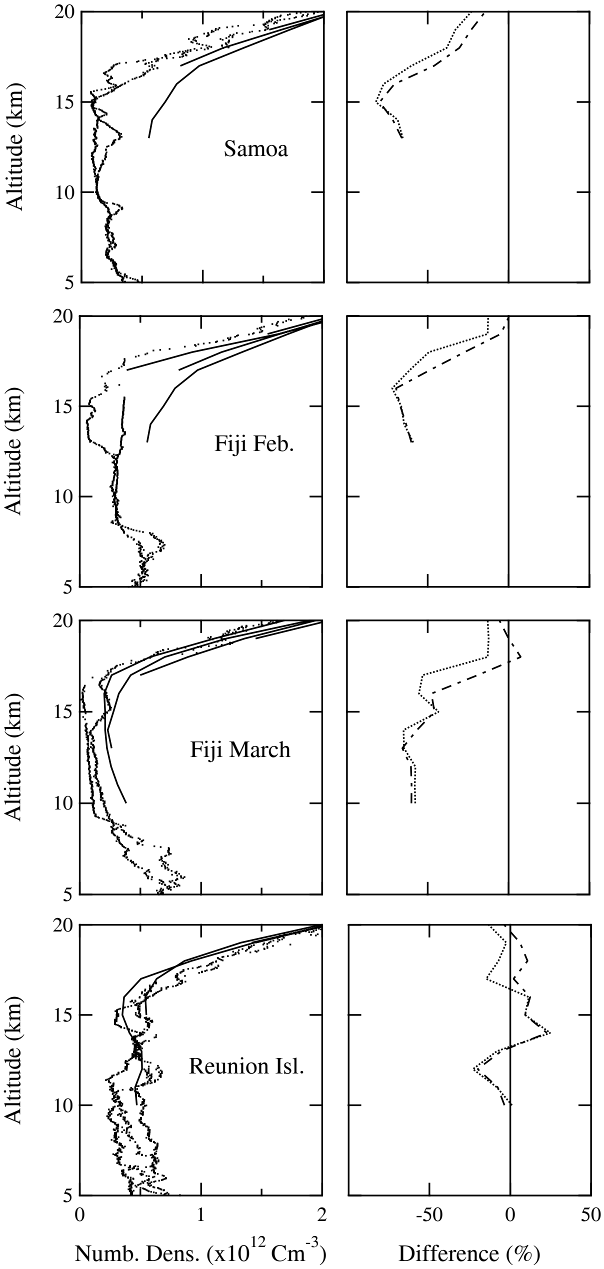

Fig. 8. SAOZ and ozonesondes below 20 km at the three SHADOZ stations in 2001. Left: profiles, right: percent difference. Solid line: SAOZ; Dotted: SHADOZ; Dotted-dashed: SHADOZ altitude corrected. From top to bottom: Samoa, Fiji February, Fiji March, and Reunion Island.

Pacific (Thompson et al., 2003b), the statistical comparison is not valid. Individual collocated comparisons only could be conclusive. However, it is worth noting the very similar amplitude of variability of the sondes and SAOZ measurements in the troposphere (Fig. 7, Table 4). This suggests that (i) their precision is very comparable and (ii) transport induced ozone variations at local scale in the upper troposphere are of the same origin of that observed at zonal scale.

4.1.3 Tropospheric ozone comparisons during MIR overpasses close to the SHADOZ stations

On average, the sondes of the three stations report less ozone in the troposphere than the MIR. However, this has little meaning since the tropospheric ozone is highly variable on a zonal scale. For example ozone concentrations are systematically larger at Reunion Island than over the Western Pacific, and also there are local episodic zero ozone layers over convective oceanic areas such as the South Pacific Convergence Zone (SPCZ). Collocated measurements only could be thus meaningful. Though no dedicated ascent is available, except at Reunion Island, several sondes are available at the three stations, less than a week apart from the MIR overpasses. The dates and differences in time, latitude and longitude between the closest ascents and the SAOZ measurements in 2001 are displayed in Table 5.

Though the MIR passed close to the stations, the observations are always separated by more than 400 km in distance. In addition, at several occasions the SAOZ measurements at the closest distance were limited to altitude levels above the tropopause at 16–17 km because of the presence of high convective clouds especially over the Western Pacific. Unfortunately and largely for these reasons, no ascent closer than 8 days could be found in 2003. Therefore, no reliable individual comparison is available for that year.

SAOZ and sonde profiles during the four overpasses and their average percent difference below 20 km are displayed in Fig. 8. The vertical shift of 300 m identified in the stratosphere has been applied to all sondes, but this has almost no impact on the comparison since the ozone vertical gradient is small in the troposphere. The factor two difference in ozone concentration between the Reunion and Western Pacific stations is consistent with that reported by Thompson et al. (2003b) for the tropospheric column. The three ascents of Reunion Island are similar within ±20%. In contrast, those of Samoa and Fiji in the South Pacific Convergence Zone are highly variable, often displaying zero ozone layers in the vicinity of deep convective cells resulting from the lifting of ozone depleted air at the surface of the ocean (Kley et al., 1996; Oltmans et al., 2001).

The average difference between SAOZ and the sondes between 10 km or the lowest altitude of SAOZ measurements and 16 km (below the tropopause) is shown for each station in Table 6. No systematic difference is observed at Reunion Island, as between SAOZ and the tropospheric ozone lidar shown in section 3. In contrast, the sondes are low biased by 60–70% over the Pacific stations, even on the closest MIR overpass 2.5/3.8° south of Fiji in March. A possible explanation for that could be likely the frequent presence of near zero ozone layers associated with convective clouds in the sondes. Indeed, the GOES West satellite pictures (not shown) indicate that all ascents at the two Pacific stations, except on 9 March at Fiji where the SAOZ and sondes measurements around 15 km are better consistent, were carried within the

Table 5. Date of SAOZ overpass and SHADOZ selected ascents in 2001.

Station	SAOZ overpass	SHADOZ ascent	ΔTime (day)	ΔLat (deg)	ΔLon (deg)	Distance (km)
Samoa	18–19 Feb.	16, 23 Feb.	2–5	9.8–10.0	2.7–15.8	1150–2000
Fiji	18–19 Feb.	16, 23 Feb.	2–5	5.3–6.1	4.4–17.9	650–2000
Fiji	8–9 Mar.	2, 9 Mar.	0–7	−2.5/−3.8	3.3–17.9	500–1900
Reunion	24–25 Feb.	21, 24, 28 Feb.	0–4	0.1–1.0	4.0–15.0	400–1600

Table 6. SHADOZ-SAOZ difference in the troposphere.

Station	Difference %
Samoa	−72.0±5.7
Fiji Feb.	−65.0±4.2
Fiji Mar.	−56.7±7.2
Reunion	1.0±14.4

Table 7. HALOE selected profiles.

Date	Event	Number	Lat ° S
2001			
17–19 Feb.	SS	13	15.2/24.8
4–6 Mar.	SR	25	15.1/24.9
2003			
4–6 Feb.	SS	14	15.8/23.0
19–20 Feb.	SR	14	17.2/24.6

convective belt of the SPCZ. In contrast the three sondes of Reunion Island were performed in cloud free conditions far south of the Intertropical Convergence Zone (ITCZ).

Since the remote sensing SAOZ measurements are restricted to cloud free areas and zero ozone layers have never been reported with this instrument, it is possible that the average SAOZ tropospheric ozone could be significantly high biased over oceanic convective areas. This remark applies also to all ground or space-based remote sensing measurements. It could explain at least part of the larger high bias between TOMS, SBUV and Dobson, and the SHADOZ sondes reported over oceanic areas and especially over the West Pacific (Thompson et al., 2003a).

In summary, when corrected for the known 50–60 s time constant of the ECC cell, the SHADOZ ozonesondes and SAOZ measurements are very consistent in the tropical stratosphere and in the cloud free troposphere. They show very little relative bias (−4%), insignificant compared to the cumulative uncertainty of both instruments, and a precision of 5% for the sondes very consistent with that claimed by previous evaluations. Finally a possible cause of systematic high bias in the remote measurements in the troposphere around oceanic convective regions has been identified, related to the low or near zero ozone layers frequently reported by the sondes within clouds, but which could not be observed remotely.

4.2 HALOE V19

The HALogen Occultation Experiment (HALOE) on board the Upper Atmosphere Research Satellite (UARS), launched in September 1991 and still in operation, performs solar occultation measurements to infer high resolution mixing ratio profiles of trace gases (Russell et al., 1993). Ozone measurements are carried out in a channel centered at 9.6 μm. The geometry of the UARS orbit (57° inclination, circular and 585 km with orbit period of 96 min) results in 15 sunrises and 15 sunsets daily, with tangent point locations during a day generally confined to small latitude bands which cycle over varying extremes, roughly every 36 days. This measurement strategy provides good longitudinal coverage on two latitude circles each day, one corresponding to the sunrise locations, and the other corresponding to the sunset locations. Latitude coverage of HALOE varies continuously over each UARS "month". The error estimates provided in the HALOE data files include random components due to noise and altitude dependent quasi-systematic errors due to uncertainties in aerosol corrections. Estimates of total uncertainty, including these systematic effects, range from about 5–10% in the middle and upper atmosphere, up to around 30% at 100 hPa (Bhatt et al., 1999).

4.2.1 HALOE data selection

The data in use here are those of version 19 available at http://haloedata.larc.nasa.gov/. The data selected for this study are a series of measurements on 13–15 consecutive orbits during or as close as possible to the balloon flight period, within the latitudinal range (20°±5° S). Since the MIR travelled only over the Pacific in 2003, a sub-set of data has been built from orbits between 45° W and 150° E. The dates, the events and the latitude range of the HALOE measurements are displayed in Table 7.

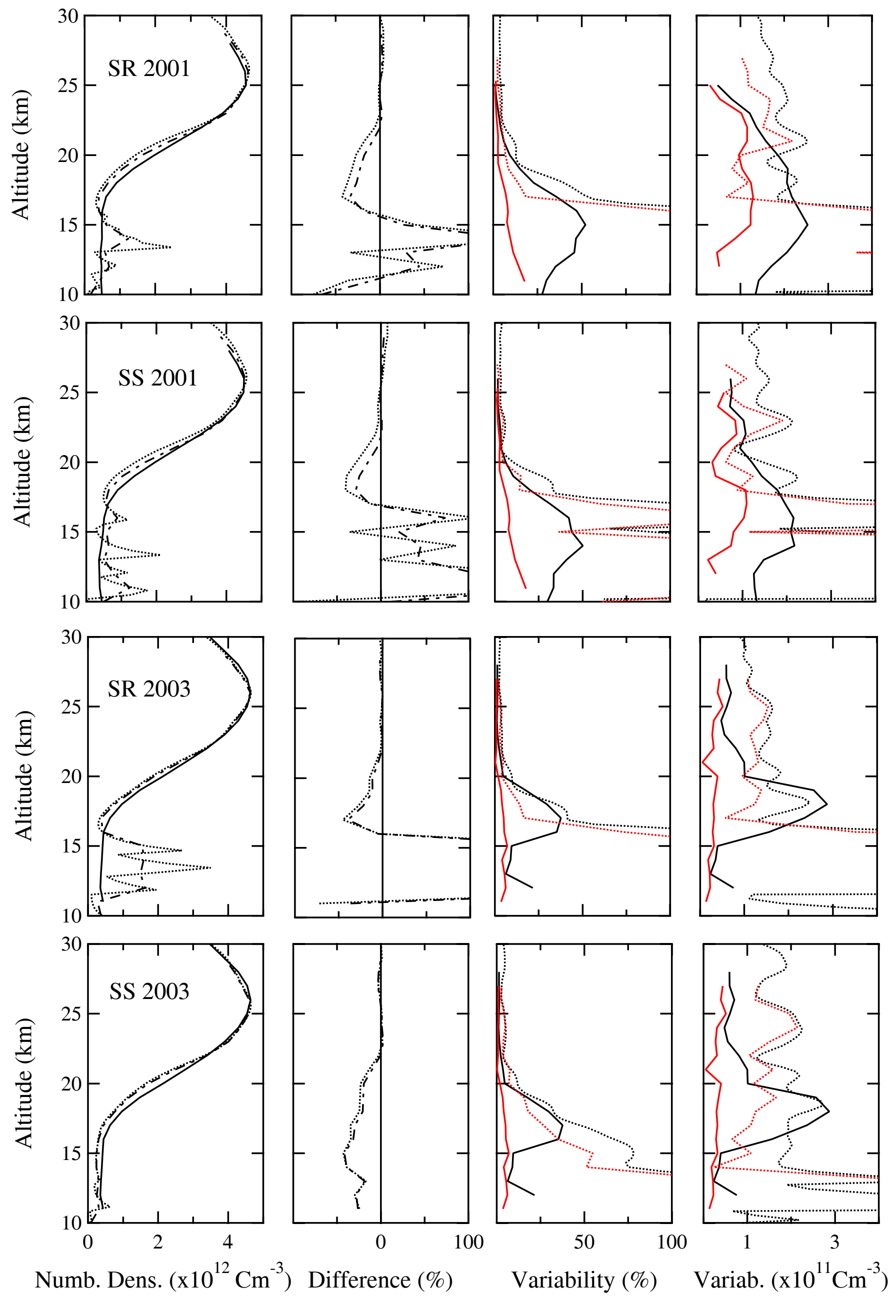

Fig. 9. Same as Fig. 7 for HALOE (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid line) in 2001. The red lines show the variability after removal of horizontal transport. From top to bottom: 2001 SR, 2001 SS, 2003 SR and 2003 SS.

4.2.2 Comparison with SAOZ

The HALOE data available in the data files are ozone-mixing ratios. Since the SAOZ measured quantity is a number density, the HALOE data have been converted using NCEP (National Center for Environmental Prediction) pressure and temperature also provided in the data files.

The comparison between HALOE and SAOZ ozone profiles is displayed in Fig. 9 in the same format as in the previous section. Overall, the agreement is good in the stratosphere (no significant bias, comparable zonal variability though a little larger for HALOE). However, the agreement degrades rapidly below 22 km, where large differences with SAOZ of either altitude or ozone concentration, as well as variability are seen. Although the average difference in the stratosphere at altitudes above 22 km (Table 9) is insignificant at SR and SS (SunRise and SunSet) for the two years, the standard deviation improves by a factor 2 by shifting

Table 8. Altitude difference between SAOZ, SHADOZ, ECMWF and HALOE/NCEP air density profiles in 2001.

SAOZ Altitude (km)	ΔZ SHADOZ (m)	ΔZ ECMWF (m)	ΔZ HALOE SR (m)	ΔZ HALOE SS (m)
10	30	60	160	140
15	0	60	−30	−60
20	−40	20	140	140
25	−40	100	230	250
30	0	160	300	280

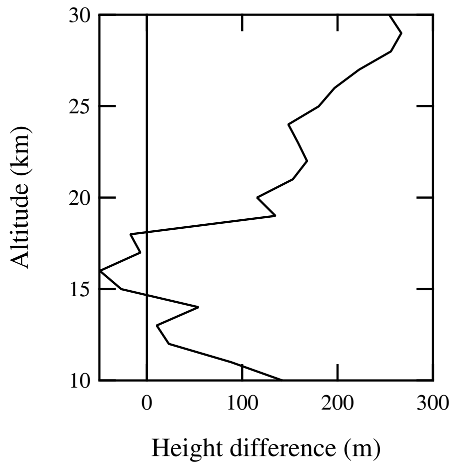

Fig. 10. Altitude difference between NCEP and SAOZ-MIR average density profiles at sunrise in 2001. Other events and year are very similar.

down the HALOE profiles by 250–300 m in 2001 and 100–150 m in 2003 (less significant because of the small number of SAOZ profiles).

The reason identified for this shift is a systematic difference in atmospheric density (or pressure) profiles between NCEP with which the HALOE VMR/pressure profiles are registered in altitude, and the SAOZ zonal average derived from actual MIR GPS, pressure and temperature measurements. The differences in altitude between the HALOE and SAOZ density profiles, as well as with those of ECMWF and the average geopotential height derived from the SHADOZ sondes at Reunion, Fiji and Samoa, are displayed in Table 8. An example of altitude dependence of the HALOE-SAOZ difference for SR in 2001 is shown in Fig. 10, the results for SS and 2003 (not shown) being very similar. SAOZ and SHADOZ altitudes are consistent between 10 and 30 km within ±50 m. ECMWF is a shifted upwards by 20 m at 20 km to 160 m at 30 km, while the NCEP profiles at SR and SS are displaced by 140 m at 20 km and 300 m at 30 km. The shift could be due either to model analyses or systematic errors in pressure measurements (a difference of 300 m at 30 km altitude corresponds to a pressure change of only 0.5 hPa). However, the use of the single density profiles improves the ozone profile comparison. On average in 2001 the bias in ozone concentration between HALOE and SAOZ reduces to +0.5±1.2%. However a difference exists in the stratosphere between standard deviations of ozone concentration compared to zonal averages. After removing changes due to horizontal transport by the PV correlation technique (Fig. 9), the 4% residual variability of the HALOE data is slightly larger than the 2–2.5% of SAOZ, suggesting a precision about twice that of SAOZ.

The agreement between HALOE and SAOZ degrades rapidly below 22 km where the difference increases systematically on all events (Fig. 9). Below 22 km (but still above the tropopause) HALOE displays an increasing systematic bias compared to SAOZ. This behaviour is very similar to that found at the tropics when compared to SAGE II (Morris et al., 2002). The HALOE percent variability also increases in this altitude range (but not the absolute variability), comparable to that of SAOZ. This indicates that if the bias was not due to a loss of ozone sensitivity but instead to altitude registration, useful information could be derived down to about 17 km. This is confirmed by the significant reduction of variability after applying the PV correlation method. However, nothing can be said about the origin of the bias, which could results either from a loss of ozone sensitivity (e.g. temperature dependence since the largest bias is observed at 17 km at the altitude of the cold point tropopause) or a progressively growing error in the HALOE altitude registration. The magnitude of the difference expressed in ozone concentration or altitude shift is shown in Table 10. It could be as large as 40% in ozone or 600–800 m at 18 km. Below that altitude, the variability of the HALOE profiles increases rapidly in both relative (>100%) and absolute (>4e^{11} mol/cm^3) units. The HALOE data appear little unreliable in the tropical troposphere.

4.3 SAGE II v6.1 and 6.2

The Stratospheric Aerosols and Gas Experiment II (SAGE II) aboard the Earth Radiation Budget Satellite (ERBS), launched in October 1984 and still operational, utilizes the solar occultation method to retrieve ozone, water vapour, aerosols and NO_2 (Mauldin et al., 1985). Ozone measurements are carried out in a channel centered at 600 nm (Cunnold et al., 1989). ERBS orbits the Earth along a circular path at a distance of 610 km above the surface of the Earth with an inclination of 56°. Over the course of roughly 1 month, SAGE II records observations at latitudes between 70° S and 70° N. Thus, about 15 profiles for each event, sunrise and sunset, are available each month at given tropical latitude.

Table 9. Difference between HALOE and SAOZ above 22 km before and after altitude adjustment, and variability before/after removal of horizontal transport.

	Diff %	Diff % Alt. adj.	Var SAOZ %	Var Haloe %	Var SAOZ e^{10} mol/cm^3	Var HALOE e^{10} mol/cm^3
2001 SR	0.1±1.9	1.0±1.3	4.2/2.5	4.1/3.8	12.1/8.5	19.2/14.6
2001 SS	−1.5±2.1	0.1±1.0	3.2/1.9	5.4/4.1	8.1/5.9	14.5/9.8
2003 SR	−1.3±1.5	−0.7±0.8	2.1/1.0	4.6/4.1	7.0/3.3	14.0/12.3
2003 SS	−0.4±2.2	0.8±1.4	2.1/1.0	3.5/3.1	7.0/3.3	17.4/15.1

Table 10. Difference in ozone number density or altitude registration between HALOE and SAOZ below 22 km in 2001.

Alt. (km)	Diff % SR	Diff % SS	Δ Alt. m SR	Δ Alt. m SS
22	−6	−6	−15	−50
21	−18	−17	230	135
20	−27	−28	475	370
19	−32	−39	630	585
18	−37	−40	635	835
17	−43	−12	925	

Table 11. SAGE II selected profiles.

Date	Event	Total	Latitude ° S
2001			
3–4 Feb.	SS	30	15.2/24.8
21–22 Mar.	SR	16	15.1/24.7
2003			
7–8 Mar.	SR	8	18.8/24.6

In older versions of the algorithm, the precision of ozone measurements was usually less than 1% in the middle stratosphere, increasing to around 2% near the stratopause and the tropopause and to higher percentage in regions of very low ozone (Maney et al., 2001). A rough estimate, including uncertainty in cross sections and knowledge of spectral response, suggests uncertainties around 2.5% from all systematic sources (Maney et al., 2001). The main changes in version 6.1 are a better aerosol clearing and the inclusion of the absorption of the oxygen dimmer. Version 6.1 improved algorithms has led to reduce errors in altitude registration that resulted in a vertical resolution of 1 km, gridded at 0.5 km and the effects of clouds on ozone retrievals (Wang et al., 2002). Compared to previous versions, the change in ozone is 0.5%. The primary change of version 6.2 is an improvement of water vapour products and has no impact on ozone. The only change, which could have an impact, is the correction of an error in the interpolation of the NCEP meteorological data used to remove Rayleigh attenuation.

4.3.1 SAGE II data selection

The data in use here are those available on the SAGE II website www.sage2.larc.nasa.gov. For this study, they have been selected using the same collocation criteria as for HALOE. Selected profiles are displayed in Table 11. Both v6.1 and v6.2 are available for 2001, but only v6.2 for 2003. In addition, because of the short duration of the MIR flight in 2003, only sunrise data are available during the flight period for this year.

4.3.2 Change between v6.1 and v6.2 in 2001

The SAGE II data available are ozone number densities, which could be thus directly compared to that of SAOZ. The results of the comparison between v6.1, v6.2 and SAOZ are displayed in Fig. 11. As could be seen on mean profiles, the ozone maxima are a little larger at SR than SS, which could simply result from slight ozone increase between 3–4 February and 21–22 March. The v6.1 and v6.2 mean profiles are very similar at sunrise displaying a small high bias of 4.5% and 5.3% respectively compared to SAOZ in the stratosphere and a low bias of 60% in the troposphere. However since the MIR started on 19 February the bias in the stratosphere is little significant. The altitude difference with SAOZ is of the order of +60/+80 m for both versions. The main change is in the variability, which drops significantly between 18 and 25 km from v6.1 to v6.2, close to that of SAOZ for v6.2 above 18 km. A little different are the results of the comparison at sunset where the v6.1 mean profile is shifted up by 300 m compared to v6.2 and 450 m to SAOZ. After correction for this altitude shift, the bias relative to SAOZ is 0.4±1.8% for v6.1 and 2.4±2.9% for v6.2 that is little significant in the stratosphere, but again low by 50–60% in the troposphere. The variability is unchanged between the two versions.

In summary, v6.2 compares better than v6.1 with SAOZ, the main improvements being a smaller variability at SR and a better altitude registration at SS.

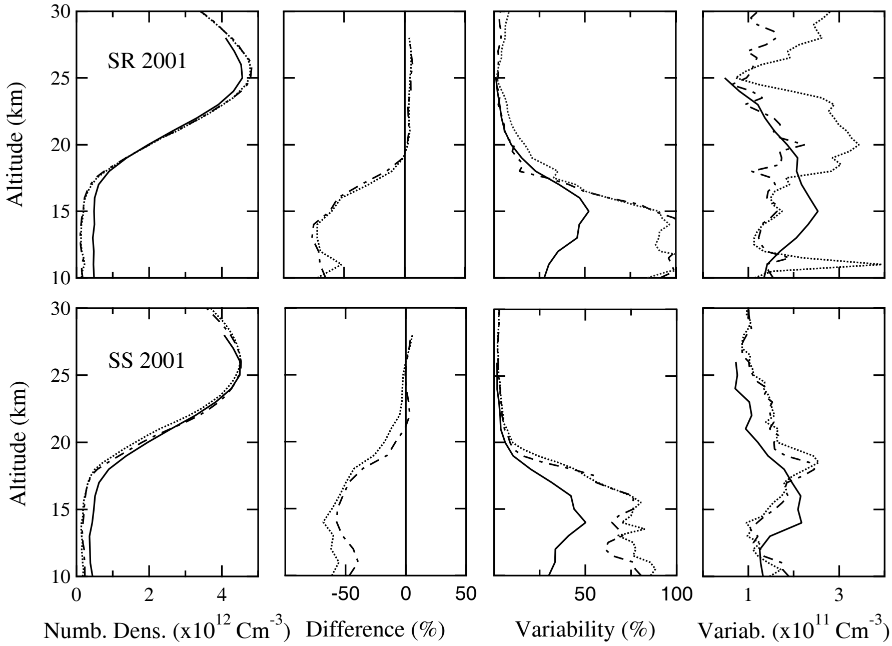

Fig. 11. Same as Fig. 7 for SAGE v6.1 (dotted), SAGE v6.2 (dotted-dashed) and SAOZ (solid) in 2001. Top SR, bottom SS.

4.3.3 Comparison between SAGE v6.2 and SAOZ in 2001 and 2003

The SAGE v6.2 and SAOZ profiles of 2001 and 2003 are compared in Fig. 12. Though the number of profiles is more limited, the results of the comparison between v6.2 and SAOZ at sunrise over the Pacific in 2003 confirm the conclusions of 2001: small SAGE altitude shift up by 100–250 m and small high ozone bias of 2–5% in the stratosphere, large low bias of 60% in the troposphere. Also shown in the right panels of Fig. 12 is the percent and number density variability for each event before (black) and after (red) removal of horizontal transport by correlation with PV. Above 20 km in the stratosphere, the variability of SAGE and SAOZ is very similar (Table 12), suggesting that the precision of the measurements of both instruments is of comparable magnitude, about 2–2.5%.

The altitude high bias in the lower stratosphere is a well-known feature of SAGE II, thought to be due to a bias in the solar edge detection algorithm in the presence of a strong gradient in the 1020 nm extinction profile (SPARC, 1998). However, the bias between SAOZ and SAGE II v6.2 in the tropics is smaller than the 300–500 m found with the 5.96 version. The small high ozone bias relative to SAOZ of 2.4% at SS for the 3–4 February 2001 and 5.3% at SR for 21–22 March, is also consistent to that found between SAGE II and HALOE around 20–25 km by Randal et al. (2003) and the sondes (5%) over Hilo in the Northern tropics (Wang et al., 2002). The small SAGE high bias could be explained by the 2% smaller absorption cross-sections of Shettle and Anderson (1994) in use in the SAGE retrieval compared to that of Brion et al. (1985) used by SAOZ (Bazureau, 2001).

Finally, the comparable precision of the SAGE broadband channels and the SAOZ spectral measurements suggests that aerosol and Rayleigh attenuation are efficiently removed in the SAGE v6.2 retrieval algorithm.

The results of the comparison between SAGE v6.2 and SAOZ in the stratosphere therefore fully confirm previous estimations, but the difference dramatically increases below 19 km, SAGE displaying a systematic 50–60% low bias, as well as 40–60% variability after removal of horizontal transport compared to the 5–10% of SAOZ. The underestimation by 50% of tropospheric ozone by SAGE, is similar to that already reported between SAGE and ozonesondes at the tropics (Kar et al., 2002; Wang et al., 2002), attributed to the high sensitivity of the retrieved ozone abundance to the background (electronic offset) of the SAGE II 525 nm channel. Indirectly, the similarity of the SAGE-sondes and SAGE-SAOZ systematic differences, confirms the agreement previously found between SAOZ and sondes. However, although the SAGE precision in the troposphere is certainly worse than that of the sondes and SAOZ, the significant correlation with PV, suggests, as concluded by Kar et al. (2002), that useful information on tropospheric ozone could be derived from SAGE II.

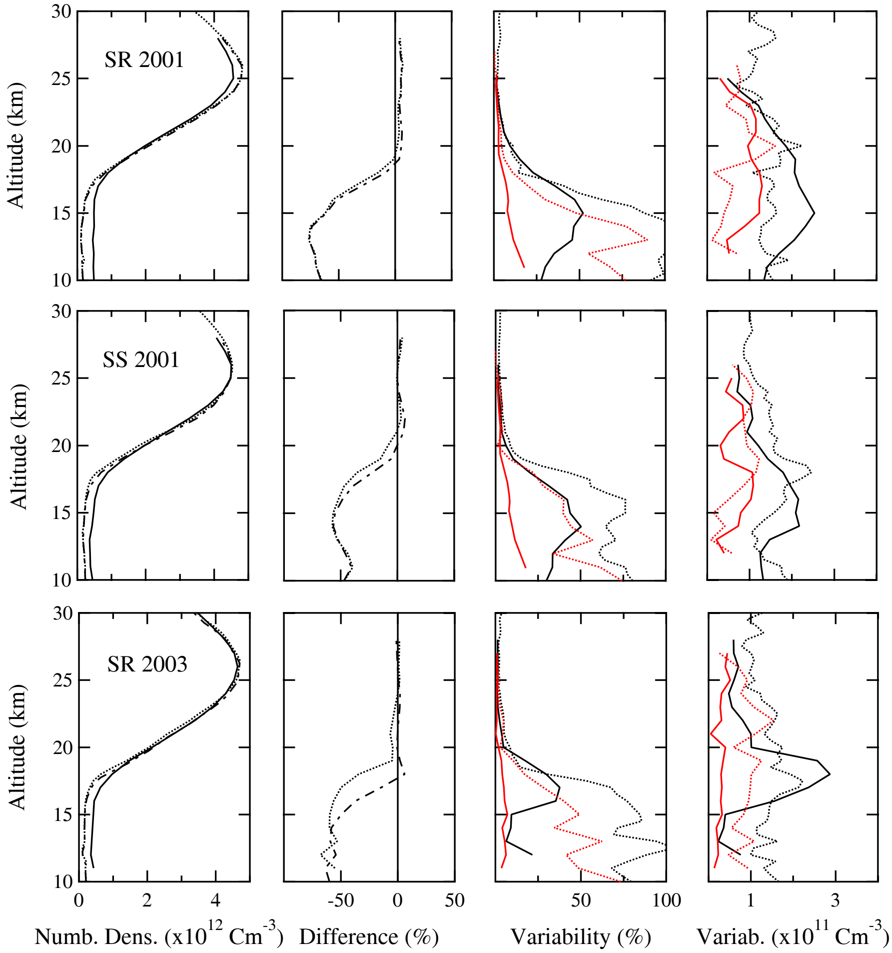

Fig. 12. Same as Fig. 7 for SAGE v6.2 (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid). The red lines show the variability after removal of horizontal transport. From top to bottom: 2001 SR, 2001 SS and 2003 SR.

Table 12. Difference between SAGE v6.2 and SAOZ above 20 km before and after altitude adjustment and variability before/after removal of horizontal transport.

	Diff %	Diff % Alt. adj.	Var SAOZ %	Var SAGE %	Var SAOZ e^{10} mol/cm^3	Var SAGE e^{10} mol/cm^3
2001 SR v6.2	3.9±1.6	5.3±0.7	4.2/2.5	3.8/2.3	12.1/8.5	13.1/8.8
2001 SS v6.2	0.4±3.8	2.4±2.9	3.2/1.9	4.1/2.1	8.1/5.9	12.6/9.0
2003 SR v6.2	−2.2±3.1	0.9±1.0	2.1/1.0	3.5/2.6	7.0/3.3	12.2/8.7

5 Summary

Ozone profiles from 10 to 26 km have been obtained at almost constant latitude (20±5° S) in the tropics using SAOZ UV-Vis spectrometers flown onboard long duration balloons in 2001 and 2003. The error analysis, confirmed by comparisons with tropospheric and stratospheric ozone lidars, indicates that the precision of the measurements is better than 2% in the stratosphere (3.5% accuracy) and 5–6% in the troposphere (12% and 25% accuracy at 15 km and 10 km respectively). The degradation of accuracy at decreasing altitude might result from spectral interference with water vapour absorption bands recently identified but not introduced yet in the retrieval algorithm. Between 17 and 24 km where the ozone gradient is the largest, SAOZ profiles agree with that of the lidar within −30±25 m in altitude and 0.05±2.7% in ozone concentration.

Table 13. Summary of performances in the stratosphere.

Instrument	Precision	Bias	Alt Registr.	Remark
SAOZ	2 %	–	−30±25 mm	
Ozonesondes	5%	−4%	+300 m	
HALOE Z>22 km	4%	< 1%	<100 m	Bias or Alt degrading rapidly below
SAGE II	2%	+2–4%	+150±50 m	

Table 14. Summary of performances in the upper troposphere.

Instrument	Precision	Bias	Remark
SAOZ	5–10 %	–	
Ozonesondes	5%	0%	Cloud free conditions
HALOE	>100%	NA	
SAGE II	50 %	−50%	

According to the SAOZ measurements, the variability of the ozone concentration along a latitudinal circle at 20° S in the SH summer would be smaller than 3–4% above 20 km. But it increases rapidly below in the Tropical Tropopause Layer (TTL) up to a maximum of 50% at 14–15 km immediately below the tropopause. The high correlation between PV and ozone suggests that most of this variability can be attributed to horizontal exchange, while, as shown by the positive correlation with a convection index, vertical transport also contributes in the troposphere. The residual variability of the SAOZ ozone profiles after removal of the impact of horizontal and vertical transport that is the sum of the precision of the measurements and other processes such as ozone chemical or lightning production, is 2% in the stratosphere, 5.7% at the tropopause and 6.5% at 12 km in the upper troposphere.

The performances of the SHADOZ ozonesondes network, HALOE and SAGE II in the tropics have been studied by comparison with SAOZ measurements. The findings are summarised in Table 13 for the stratosphere and Table 14 for the upper troposphere. In the stratosphere, the main discrepancies arise from differences in altitude, particularly sensitive between 20 and 26 km because of the strong gradient of ozone concentration in the tropics. In respect to SAOZ, the ozonesondes are found shifted up by 300 m, consistent with the known 50–60 s time constant of the electrochemical cell. HALOE is found shifted up by about the same. But in that case the reason identified is a systematic bias between the NCEP pressure vertical profile with which the HALOE altitude is registered, compared to that of SAOZ as well as ECMWF. A shift up by 100–200 m compared to SAOZ is observed in the SAGE profiles thought to be due to a bias in the solar edge detection algorithm, but of smaller amplitude than previously thought. After taking into account those vertical displacements, the relative ozone bias compared to SAOZ is insignificant (<1%) for HALOE, a little low (−4%) for the sondes and a little high (2–4%) for SAGE II. Finally, the respective precisions of the instruments have been derived from the zonal variability of ozone concentration after removing the impact oh horizontal transport by correlation with PV. According to this analysis, the best precision (2%), similar to that of SAOZ, is reached by SAGE II, while the HALOE (4%) and the sondes (5%) measurements could be little less precise. All the above figures are generally very consistent with the results of previous comparisons between the instruments, as well as known causes of errors of each of them.

The only feature not explained is the increasing systematic low bias of HALOE compared to others at altitudes below 22 km, which could be caused either by a bias in altitude registration or in ozone concentration. It is similar to that reported between SAGE and HALOE in the tropics by Morris et al. (2002), but for which no explanation could be proposed. The only new piece of information we can provide here is that since it exists also with SAOZ, it must be attributed to HALOE.

At lower altitude in the upper troposphere, the SAOZ measurements are found consistent with those of the sondes and the lidar at Reunion Island, and biased high by 60% on average compared to ozonesondes over the Western Pacific, at American Samoa and Fiji. The likely explanation is the frequent occurrence of near zero ozone layers in the convective clouds of the South Pacific Convergence Zone because of the ozone destruction at the surface of the ocean. Since these layers cannot be seen by SAOZ as well as by all ground-based and space borne remote sensing instruments, it is very likely that the average tropospheric ozone concentration could be overestimated by instruments other than ozonesondes over these areas. The ozone variability of the soundings is found comparable to that of SAOZ, suggesting that the precision of both instruments is comparable, of the order of 5–10%. The high correlation of both ozone profiles with PV changes suggests that most of the ozone variability in the upper troposphere would be due to quasi-horizontal exchange with the mid-latitude lower stratosphere. Compared to SAOZ, SAGE II displays a 50–60% low bias similar to that already identified with the ozonesondes, and a larger zonal variability, suggesting a precision of the order of 50%. Nevertheless, the significant correlation with PV indicates that useful information on tropospheric ozone could be derived from the SAGE II profiles. Finally, the unrealistic large offsets and variability in the HALOE data compared to all others, suggests that the measurements of this instrument are of limited use below 17 km.

Appendix: Mathematical background

The statistical method is to compare mean zonal profiles indicative of possible relative bias in concentration or altitude registration, mean differences (equation 1), as well as absolute and relative variability along a latitudinal circle (respectively Eq. 2 and 3), indicative of measurements precision.

$$Diff_i = 100\frac{\overline{x_i} - \overline{SAOZ_i}}{\overline{SAOZ_i}}, \quad (1)$$

$$StdDev_i = Abs.Var_i = \sqrt{\frac{1}{n_i}\sum(x_i - \overline{x_i})^2} \quad (2)$$

$$Rel.Var_i = 100\frac{Abs.Var_i}{\overline{SAOZ_i}} \quad (3)$$

All variables are implicitly altitude dependent. The i-index refers to the variable at ith altitude. $\overline{x_i}$ and $\overline{SAOZ_i}$ refer to mean ozone number density of correlative data (SHADOZ, SAGE II and HALOE) and SAOZ. $Diff_i$ is the relative percent difference. $StdDev_i$, the standard deviation, is equivalent to the absolute ozone variability, Abs. Var_i. The relative variability, $Rel.Var_i$, is expressed in percent. SHADOZ and HALOE data having a vertical sampling different from that of SAOZ, they are linearly interpolated to the SAOZ altitude grid of 1 km steps. SAGE profiles available on a 0.5 km vertical altitude grid are used directly without further interpolation.

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References

Baray, J. L., Leveau, J., Porteneuve, J., Ancelet, G., Kehkut, P., Posny, F., and Baldy, S.: Description and evaluation of tropospheric ozone lidar implemented on an existing lidar in the southern tropics, Applied Optics, 38, 6808–6817, 1999.

Bazureau, A.: Preparation de l'inversion des mesures de transmission de l'experience spatiale SAGE III: mise en place et etude des algorithmes d'inversion, PhD thesis, Université des Sciences et Technologies de Lille, 2001.

Bhatt, P. P., Remsberg, E. E., Gordley, L. L., McInerney, J. M., Bracket, V. G., and Russel III, J. M.: An evaluation of the quality of HALogen Occultation Experiment ozone profiles in the lower stratosphere, J. Geophys. Res., 104, 9261–9275, 1999.

Brion, J., Chakir, A., Charbonnier, J., Daumont, D., Parisse, C., and Malicet, J.: Absorption spectra measurements for the ozone molecule in the 350–830 nm region, J. Atmos. Chem., 30, 291–299, 1998.

Coheur, P.-F., Fally, S., Carleer, M., Clerbaux, C., Colin, R., Jenouvrier, A., Mérienne, M. -F., Hermans, C., and Vandaele, A. C.: New water vapour line parameters in the 26 000–13 000 cm^{-1} region, J. Quant. Spectrosc. Radiat. Transfer, 74, 493–510, 2002.

Cunnold, D. M., Chu, W. P., Barnes, R. A., McCormick, M. P., and Veiga, R. E.: Validation of SAGE II ozone measurements, J. Geophys. Res., 94, 8447–8460, 1989.

Dessler, A. E., Hintsa, E. J., Weinstock, E. M., Anderson, J. G., and Chan, K. R.: Mechanism controlling water vapor in the lower stratosphere, "A tale of two stratospheres", J. Geophys. Res., 100, D11, 23 167–23 172, 1995.

Dessler, A. E.: The effect of deep tropical convection on the tropical tropopause layer, J. Geophys. Res., 107, doi:101029/2001JD000511, 2002.

Hauchecorne, A., Godin, S., Marchand, M., Heese, B., and Souprayen, C.: Quantification of the transport of chemical constituents from the polar vortex to midlatitudes in the lower stratosphere using the high-resolution advection model MIMOSA and effective diffusivity, J. Geophys. Res., 107, doi:10.1029/2001JD000491, 2002.

Jeker, D. P., Pfister, L., Thompson, A. M., Brunner, D., Boccippio, D. J., Pickering, K. E., Wernli, H., Kondo, Y., and Staehelin, J.: Measurements of nitrogen oxides at the tropopause, attribution to convection and correlation with lightning, J. Geophys. Res., 105, 3679–3700, 2000.

Johnson, B. J., Oltmans, S. J., Vömel, H., Smit, H. G. J., Dessler, T., and Kröger, C.: Electrochemical concentration cell (ECC) ozonesonde pump efficiency measurements and tests on the sensitivity to ozone of buffered and unbuffered ECC sensor cathode solutions, J. Geophys. Res., 107, doi:10.1029/2001JD000557, 2002.

Kar, J., Trepte, C. R., Thomason L. W., Zawodny, J. M., Cunnold, D. M., and Wang, H. J.: On the tropospheric measurements of ozone by the Stratospheric Aerosol and Gas Experiment II (SAGE II, version 6.1) in the tropics, Geophys. Res. Lett., 29, doi:10.1029/2002GL016241, 2002.

Kley, D., Crutzen, P. J., Smit, H. G. J., Vömel, H., Oltmans, S. J., Grassl H., and Ramanathan, V.: Observations of near-zero ozone concentrations over the convective Pacific, effects on air chemistry, Science, 274, 230–233, 1996.

Malaterre, P., Dubourg, V., Letrenne, G., and Durand, M.: The long duration MIR balloon, in: Proc. 12th ESA symp. on Rocket and balloon programs, ESA SP-370, 329–334, 1996.

Manney, G. L., Michelsen, H. A., Bevilacqua, R. M., Gunson M. R., Irion, F. W., Livesey, N. J., Oberheide, J., Riese, M., Russell III, J. M., Toon, G. C., and Zawodny, J. M.: Comparison of satellite ozone observations in coincident air masses in early November 1994, J. Geophys. Res., 106, D9, 9923–9943, 2001.

Mauldin, L. E., Zaun, N. H., McCormick, M. P., Guy, J. H., and Vaugh, W. R.: Stratospheric Aerosol and Gas Experiment II instrument: A functional description, Opt. Eng., 24, 307–312, 1985.

Morris, G. A., Gleason, J. F., Russell III, J. M., Schoeberl, M. R., and McCormick, M. P.: A comparison of HALOE V19 with SAGE II V6.00 ozone observations using trajectory mapping, J. Geophys. Res., 10, doi:10.1029/2001JD000847, 2002.

Oltmans, S. J., Johnson, B. J., Harris, J. M., Vömel, H., Thompson, A. M., Koshy, K., Simon, Patrick, Bendura, R., J., Logan, J. A., Hasebe, F., Shiotani, M., Kirchhoff, V. W. J. H., Maata, M., Sami, G., Samad, A., Tabuadravu, J., Enriquez, H., Agama, M., Cornejo, J., and Parades, F.: Ozone in the Pacific tropical troposphere from ozonesonde observations, J. Geophys. Res, 106, D23, 32 503–32 525, 2001.

Orphal, J.: A critical review of the absorption cross-sections of O_3 and NO_2 in the ultraviolet and visible, J. Photochem. Photobiol., A Chem., 157, doi:10.1016/S1010-6030(03)00061-3, 2003.

Pommereau, J. P. and Hauchecorne, A.: A new Atmospheric vehicle: The Infra-Red Montgolfier, Adv. Space Res., 20, 55–58, 1979.

Pommereau, J. P. and Goutail, F.: O_3 and NO_2 Ground-Based Measurements by Visible Spectrometry during Arctic Winter and Spring 1988, Geophys. Res. Lett., 15, 891–894, 1988.

Pommereau, J. P. and Piquard, J.: Ozone, Nitrogen dioxide and Aerosol vertical distributions by UV-visible solar occultation from balloons, Geophys. Res. Lett, 21, 1227–1230, 1994.

Portafaix, T., Morel, B., Bencherif, H., Baldy, S., Godin-Beekmann, S., and Hauchecorne, A.: Fine-scale study of a thick stratospheric ozone lamina at the edge of the southern subtropical barrier, J. Geophys. Res., 108, doi:10.129/2002JD002741, 2003.

Pundt, I., Pommereau, J.-P., Chipperfield, M. P., Van Roozendael, M., and Goutail, F.: Climatology of the stratospheric BrO vertical distribution by balloon-borne UV–visible spectrometry, J. Geophys. Res., 107, doi:10.1029/2002JD002230, 2002.

Randall, C. E., Bevilacqua, R. M., Lumpe, J. D., and Hoppel, K. W.: Validation of POAM III aerosols: Comparison to SAGE II and HALOE, J. Geophys. Res., 106, D21, 27 525–27 436, 2001.

Russel III, J. M., Gordley, L. L., Park, J. H., Drayson, S. R., Hesketh, W. D., Cicerone, R. J., Tuck, A. F., Frederick, J. E., Harries, J. E., and Crutzen P. J.: The HALogen Occultation Experiment, J. Geophys. Res., 98, D6, 10 777–10 797, 1993.

Shettle, E. P. and Anderson, S. M.: New visible and near IR ozone cross-sections for MODTRAN, Proc. 17th Annual Review. Conf. on Atmospheric Transmission Models, Gail, P. (Ed.), Vol. AD-A295591, 1994.

Solomon, S., Borrmann, S., Garcia, R. R., Portmann, R., Thomason, L., Poole, L. R., Winker, D., and McCormick, M. P.: Heterogeneous chlorine chemistry in the tropopause region, J. Geophys. Res., 102, 21 411–21 429, 1997.

Stratospheric Processes and their Role in Climate (SPARC), Assessment of trends in the vertical distribution of ozone, Report 1, edited by: Harris, N., Hudson, R., and Philips, C., report No. 1, World Meteorological Organisation, Rep# 43, 1998.

Thompson, A. M., Witte, J. C., McPeters, R. D., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Johnson, B. J., Vömel, H., and Labow, G.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998-2000 tropical ozone climatology 1. Comparison with TOMS and Ground-based Measurements, J. Geophys. Res., 108, doi:10.1029/2001JD000967, 2003a.

Thompson, A. M., Witte, J. C., McPeters, R. D., Oltmans, S. J., Schmidlin, F. J., Logan, J. A., Fujiwara, M., Kirchhoff, V. W. J. H., Posny, F., Coetzee, G. J. R., Hoegger, B., Kawakami, S., Ogawa, T., Fortuin, J. P. F., and Kelder, H. M.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998-2000 tropical ozone climatology 2. Tropospheric variability and the zonal wave-one, J. Geophys. Res., 108, doi:10.1029/2002JD002241, 2003b.

Wang, H. J., Cunnold, D. M., Thomason, L. W., Zawodny, J. M., and Bodeker, G. E.: Assessment of SAGE version 6.1 ozone data quality, J. Geophys. Res., 107, doi:10.1029/2002JD002418, 2002.