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Evaluation of ozonesondes, HALOE, SAGE II and III, Odin-OSIRIS and -SMR, and ENVISAT-GOMOS, -SCIAMACHY and -MIPAS ozone profiles in the tropics from SAOZ long duration balloon measurements in 2003 and 2004

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Abstract. The performances of satellite and sondes ozone measuring instruments available in the tropics between 10 and 26 km during the southern hemisphere summer in 2003 and 2004, have been investigated by comparison with series of profiles obtained by solar occultation in the visible Chappuis bands using a SAOZ UV-Vis spectrometer carried by long duration balloons. When compared to SAOZ, systematic positive or negative altitude shifts are observed in the satellite profiles, varying from <50 m for the GOMOS v6.0b stellar occultation instrument, followed by +100/200 m for solar occultation systems (SAGE II v6.2, HALOE v19 above 22 km), but as large as −900 m for the OSIRIS limb viewing system. The ozone relative biases are generally limited, between −4% and +4%, for measurements in the visible Chappuis bands (SAGE II and SAGE III moon v3, GOMOS above 22 km and OSIRIS), the near IR (HALOE above 22 km) and the ozonesondes, but increase to +5.5% (SCIAMACHY IUP v1.63) though still in the visible, and +7% in the mid-IR (MIPAS NL v4.61) and the submillimetric range (SMR v222). Regarding precision, evaluated statistically from the zonal variability of ozone concentration, the best measurements are found to be those of SAGE II (2%), followed by HALOE above 22 km (3–4%), then the ozonesondes, SAGE III moon, SCIAMACHY and OSIRIS (4–5%), GOMOS above 22 km (∼6%), MIPAS (8.5%) and finally SMR (16%). Overall, all satellite ozone measurements appear to be of little utility in the tropical troposphere except those of SAGE II (and eventually SAGE III), though low biased by 50% and of limited (50%) 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 for a long time from space-borne instruments such as SAGE II since 1984 (Mauldin et al., 1985), HALOE since 1992 (Russell et al., 1993), complemented by the SHADOZ tropical ozonesonde network since 1998 involving now eleven stations (Thompson et al., 2003), the ozone distribution and its variations are still poorly characterised in the 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. For example, although a 8-year comparison between HALOE and SAGE II shows RMS differences not exceeding 4–12% through most of the stratosphere, larger differences (>15%, HALOE low compared to SAGE II) are systematically observed below 22 km in the tropical lower stratosphere which never received satisfactory explanations (Morris et al., 2002). Another example is the 40–60% low bias of SAGE II ozone in the upper tropical troposphere compared to ozonesondes reported by Kar et al. (2002) and Wang et al. (2002), attributed by these authors to the high sensitivity of the retrieved ozone abundance to the background (electronic offset) of the SAGE II channel.

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More recently, the relative performances of SAGE II, HALOE and the SHADOZ ozonesondes in the tropics have been studied by comparison with accurate measurements of a SAOZ UV-Visible spectrometer carried by long duration balloons flown in 2001 and 2003 at 20° S (Borchi et al., 2005). The agreement with SAOZ was found excellent in the stratosphere (2% precision, +2–4% ozone bias, +150 m altitude bias for SAGE II; 4% precision, <1% ozone bias, <100 m altitude bias for HALOE; and 5% precision, −4% bias and +300 m in altitude for the sondes), but degrading rapidly for the two space-borne instruments, below 22 km (−40% at 18 km) for HALOE, and below 19 km for SAGE II, displaying a low bias of 50–60% compared to SAOZ in the upper troposphere. The comparison between SAOZ and the ozonesondes in the troposphere is less conclusive. Indeed, the SAOZ ozone concentrations were found to be comparable with those of the sondes at Reunion Island but systematically high biased by 50–60% at Samoa and Fiji in the Western Pacific. Asides from differences of instrumental arrangements, a possible explanation could be the frequent occurrence of near zero ozone layers in convective clouds over the South Pacific Convergence Zone (SPCZ) because of the ozone destruction at the surface of the ocean (Kley et al., 1996), which could not be seen by SAOZ limited by definition to cloud free areas. A possible systematic overestimation of tropospheric ozone by remote sensing systems was thus suggested over convective areas.

Since 2001 a number of new ozone measuring space instruments have been put into orbit: the Limb viewing visible OSIRIS (Optical Spectrograph and Infrared Imager System) (Warshaw et al., 1996; Llewellyn et al., 1997) and the Limb observing SMR (Sub-Millimetre Radiometer) (Murtagh et al., 2002) both aboard the Swedish-Finish-Canadian-French ODIN satellite launched in February 2001; the NASA SAGE III UV-Vis-near IR spectrometer aboard METEOR III in December 2001 operating by moon occultation in the tropics (McCormick et al., 1991); and finally the ESA ENVISAT satellite in March 2002 carrying the GOMOS (Global Ozone Monitoring by Occultation of Stars) stellar occultation (Bertaux et al., 1991; Bertaux et al., 2001), the limb viewing SCIAMACHY (SCanning Imaging Absorption spectrometer for Atmospheric CHartographY) UV-Vis spectrometers (Burrows and Chance, 1991; Bovensmann et al., 1999) and the Infra-red Limb viewing MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) (Fischer and Oelhaf, 1996; Harris, 2000).

The objective of the present paper is the evaluation of the performances of all above systems with the method applied already by Borchi et al. (2005), with the data of the 9-day SAOZ flight of February 2003 and those of a new 39-day flight carried in February-April 2004 in the frame of a HIBISCUS project of the European Union dedicated to the study of the impact of convection on the stratosphere (Pommereau et al., 2007).

Section 2 provides a brief description of the SAOZ system and the long duration balloon flights performed in 2003 and 2004 and a recall of the technique developed for deriving the relative contributions of horizontal and vertical transport and measurements errors from the observed ozone zonal variability. Section 3 describes the performances of each ozone measuring system, altitude registration, ozone bias and precision, relative to those of SAOZ: HIBISCUS ozonesondes at Bauru and at the three SHADOZ stations overflown by the balloons, and HALOE, SAGE II and III, Odin-OSIRIS and -SMR, and ENVISAT-GOMOS, -SCIAMACHY and -MIPAS instruments into orbit by that time. Section 4 provides a comparison of the relative performances of each of them. Finally, the conclusions of the study are summarised in Sect. 5.

2 SAOZ ozone profiles in the tropics

SAOZ is a balloon borne diode array UV-Visible spectrometer for remote O_3 and NO_2 observations by solar occultation (Pommereau and Piquard, 1994). In the present application it was flown aboard long duration Infra-Red Montgolfier (MIR) balloons operated by the Centre National d'Etudes Spatiales (CNES). The data used here are those of two flights launched from Bauru at 22° S in Brazil during the SH summer season in February-March 2003 and 2004, similar to the 34-day flight already performed in 2001 (Borchi et al., 2005). The first in 2003 lasted for 9 days at about 20°S across the Pacific before being brought down by a hurricane over the Coral Sea, while the second in 2004 flew for 39 days for one and half circumnavigation between 20° S and 9° S.

2.1 MIR SAOZ balloon flight in 2003 and 2004

Ozone profiles are obtained by solar occultation in the visible Chappuis bands in the 450–620 nm spectral range. The spectra are analysed by the differential optical 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. A description of the SAOZ instrument (300–650 nm range, 1.1 nm resolution, 3 pixels over-sampling) is given in Borchi et al. (2005). Although very similar in most aspects, the SAOZ payload flown in 2004 was using a slightly different spectrometer extended to 1000 nm for the measurement of water vapour around 940 nm (400–1000 nm range, 1.2 nm resolution, 1.7 pixels over-sampling). However, the change has no impact on ozone measurements in the broad (30 nm large) Chappuis bands features. The altitude is a geometric altitude above Mean Sea Level (MSL) directly related to that of the GPS 3D location (Larsen LP from Trimble; horizontal accuracy <8 m

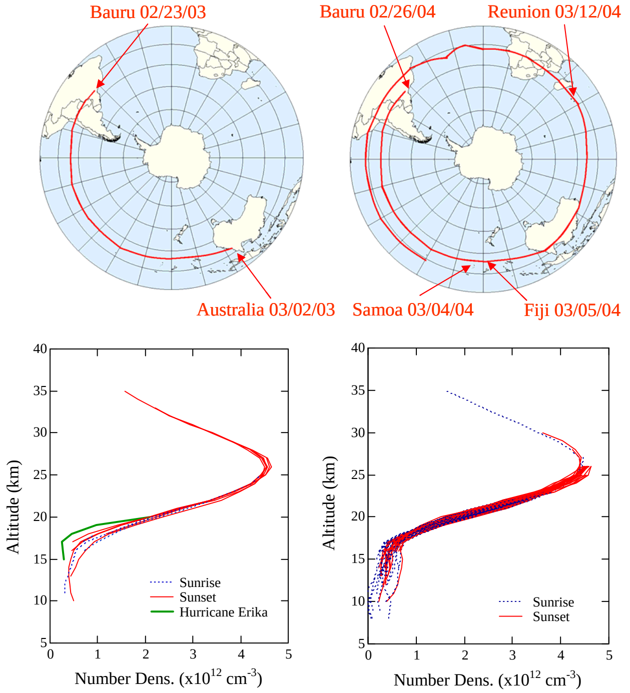

Fig. 1. Balloon trajectories (top) and ozone profiles (bottom) in 2003 (left) and 2004 (right).

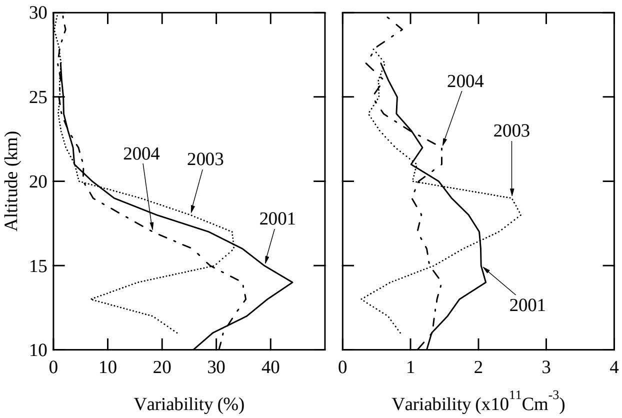

Fig. 2. Zonal variability of ozone concentration in 2001, 2003 (Pacific only) and 2004. Left: relative standard deviation in percent compared to the mean profile; Right: standard deviation in number density.

(90%), vertical <16 m (90%)), from which the SZA at the location of the balloon and then the tangent height are derived. However, the GPS MSL altitude refers to an Earth Gravitational Model, which differs from the real MSL by few meters on 90% of the globe but +20/−40 m in extreme cases from recent satellite altimetric measurements. The atmospheric density profile used in the calculation of tangent height after refraction is that of the standard atmosphere at 15° N. Compared to this, the average ECMWF (European Centre for Medium-range Weather Forecast) density at the location of the MIR is larger by 6% with a peak to peak dispersion of ±6% resulting in an average high bias of the SAOZ altitude registration of 24 m at 20 km and 72 m at 15 km. The overall uncertainty of the SAOZ altitude registration is thus +64/−16 m at 20 km and +140/−16 m at 15 km. This is very consistent with the average shift of +30±25 m bias between 17–24 km found between SAOZ and the measurements of an ozone lidar during the overpass of a MIR-SAOZ over the NDSC station of Reunion Island (Borchi et al., 2005). The vertical resolution of the measurements is 1.4 km corresponding to the half-width of the solar disk brightness, while concentrations are retrieved within 1 km thick atmospheric shells. Data contaminated by clouds are removed by looking at the atmospheric extinction at 615 nm.

The precision of the SAOZ ozone retrievals estimated from fitting errors of the spectral analysis is 1.5% at 20 km degrading to 5% at 17.5 km, 10% at 15 km and 23% at 10 km, to which a systematic error of 1.5% has to be added for accuracy corresponding to the uncertainty of the ozone absorption cross-sections used (Borchi et al., 2005).

The data used here are those obtained during two flights at the tropics launched from Bauru, Brazil (22° S, 49° W), the first on 23 February 2003 for the preparation of the HIBISCUS campaign, and the second on 26 February 2004 during the following main campaign (http://www.aerov.jussieu.fr/projet/HIBISCUS/). The first lasted for 9 days only, at almost constant latitude (22°±1° S), providing 11 useful profiles across the Pacific before being lost over a hurricane. The second lasted for 39 days, resulting in 70 useful profiles (roughly half at sunrise, half at sunset) between 9° S and 21° S, before falling over the Pacific Convergence Zone. The trajectories of the balloons in the summer stratospheric Easterlies and the ozone profiles recorded along the flights at sunset and sunrise, are shown in Fig. 1. Only the first thirty-one profiles of 2004 within the 15–21° S latitude range are being used in this study which could be readily compared with those observed within 15–25° S in 2001 and 20–23° S in 2003 (Borchi et al., 2005).

2.2 Ozone variability along a latitudinal circle

The zonal variability of ozone concentration along the flights in 2001, 2003 and 2004 is shown in Fig. 2 (relative standard deviation in percent on the left, standard deviation in number density on the right). In the stratosphere, above 20 km, the figures are very similar on the three years showing little variability (<5%), but increasing rapidly below 20 km, reaching a maximum on the two long circumnavigating flights, of 45% in 2001 and 35% in 2004 at 14–15 km at about the average altitude of the tropopause and then decreasing at lower altitude. The picture is slightly different on the nine days flight in 2003 where the measurements are limited to the Pacific. In this region, the variability displays a maximum around 17–18 km,

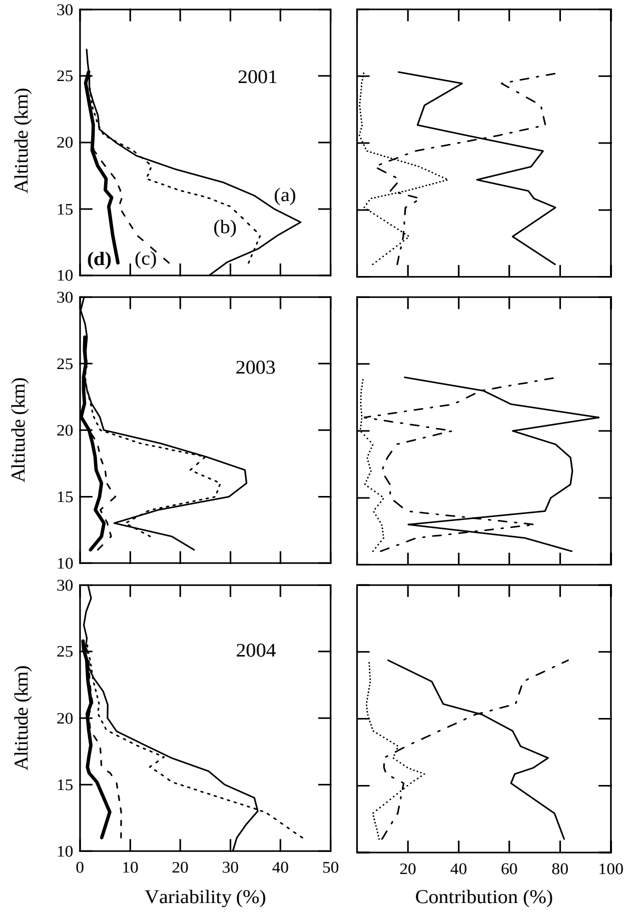

Fig. 3. Analysis of contributions to zonal variability (relative standard deviation) of ozone in 2001 (top), 2003 (middle) and 2004 (bottom). Left: percent variability **(a)** number density at constant altitude **(b)** mixing ratio on isentropic surfaces; **(c)** after removing horizontal transport; **(d)** after removing vertical and horizontal transport. Right: relative contributions of horizontal (solid line) and vertical (dotted) transport and measurement errors (dotted-dashed) to total variability.

higher than the zonal average of long duration flights, largely due to the upward displacement of the ozone profile associated to the hurricane Erika (Fig. 1). The minimum variability below is little significant since it is derived from three profiles only, all over the non-convective eastern Pacific, the western part being covered by high altitude clouds.

Apart from random errors, several mechanisms could contribute to the variability, i.e. horizontal and vertical transport, convective lifting of ozone poor air from oceanic surfaces and photochemical production by precursors. A method for analysing and separating atmospheric and instrumental contributions in the ozone measurements has been proposed by Borchi et al. (2005) based on a multiple regression with proxies of horizontal and vertical transport. The proxy used for horizontal transport is potential vorticity (PV) on isentropic surfaces provided by the MIMOSA high-resolution contour advection model initialised with ECMWF wind fields (Hauchecorne et al., 2002) interpolated at the location of the SAOZ tangent points. For vertical transport, the proxy used is the altitude difference between 370 K and 340 K isentropic levels issued from ECMWF operational analyses at the location of the SAOZ tangent height. It is an indicator of the presence of convection in the troposphere below the balloon and thus of possible overshooting of ozone poor air in the Tropical Tropopause Layer (TTL) or even in the lower stratosphere, currently not captured by the models. The same proxy is used for all altitude levels.

Ozone number densities at geometric altitude as shown in Fig. 1 are first converted in mixing ratios (MR) on isentropic surfaces. The contribution of quasi-horizontal and convective transport is then evaluated by correlating ozone, PV and $\Delta Z_{370-340\,\mathrm{K}}$ changes at each level, following the equation:

$$\Delta O_3(\Theta)=\alpha(\Theta).\Delta PV(\Theta)+\beta(\Theta).\Delta Z_{370-340.\mathrm{K}} \qquad (1)$$

where $\Delta PV(\Theta)$ is the variability of potential vorticity, $\alpha(\Theta)$ the slope of the linear regression between ozone MR and MIMOSA PV, and $\beta(\Theta)$ the slope of the linear regression between ozone MR and $\Delta Z_{370-340\,\mathrm{K}}$, at a Θ isentropic level. The calculations are made on 16 isentropic levels between 335 and 650 K, corresponding to a vertical resolution of ~700–1000 m.

The results of the regression applied to the SAOZ measurements in 2001, 2003 and 2004 are depicted in Fig. 3, where the left panel shows the residual variability after each step of the calculation: (a) number density at geometric levels; (b) mixing ratio on isentropic surface; (c) after removal of horizontal transport (PV) and (d) residual after removing also vertical transport. The right panel shows the relative percent contributions of horizontal and vertical transport as well as of the residual representing the random error of the measurements and possibly also other mechanism, e.g. photochemical production. The results are very similar for the two long circumnavigating flights in 2001 and 2004 displaying a large 60–70% contribution of horizontal transport below 20 km, and a smaller impact of vertical transport of 15–25% limited to the 14–20 km altitude levels. This suggests an overshooting ozone poor air up to 19–20 km in the Tropical Tropopause layer (TTL) and the lower stratosphere. The absence of signature of convective transport below is simply due to the fact that the ozone mixing ratio is constant in the troposphere. A vertical displacement has thus little impact on the profile. If the residual variability, which may still include some contribution from ozone photochemical production, is adopted as an indicator of random error, the precision of the SAOZ ozone measurements would be 2% in the stratosphere, 5–6% at the tropopause and 7–8% at 12 km in the upper troposphere. These figures are very consistent with those estimated from the error of the spectral fitting of 1.5% in the stratosphere and 5–6% at the tropopause, but significantly better in the upper troposphere than the estimation of 10% at

Table 1. Date of SAOZ profiles (sr = sunrise, ss = sunset) and ozonesondes selected ascents in 2004.

Station	SAOZ profiles	Sondes ascents	Time (day)	Lat (deg)	Lon (deg)	Distance (km)
Bauru	27 Feb (sr, ss)	21, 23, 24 Feb	3–6	2.1–2.6	11.6–30.6	1235–3210
Samoa	4 March (sr, ss)	4, 5 March	0–1	4.2–4.8	2.8–5.4	611–734
Fiji	5 March (sr, ss)	4 March	1	1.5–2.4	5.0–12.7	548–1353
Reunion	11 March (sr), 12 (sr)	10, 11 March	0–2	3.1–4.8	3.1–15.6	475–1717

15 km and 23% at 10 km. The identified reason for this is the presence of water vapour lines introducing systematic errors in the spectral analysis but of small impact on the precision of ozone measurements.

3 Evaluation of relative performances of other systems

A number of ozone profiles within the latitude range and during the period of the MIR flights in 2003 and 2004 are available from the SHADOZ ozonesondes and those performed at Bauru during HIBISCUS, from three NASA space instruments, HALOE, SAGE II and III respectively aboard UARS, ERBS and METEOR 3M, from the two OSIRIS and SMR instruments aboard ODIN, and from GOMOS, SCIAMACHY and MIPAS aboard ENVISAT. The performances of each have been studied by comparison with SAOZ after adaptation of vertical resolution by smoothing. Higher resolution measurements are smoothed to the resolution of the other measurement to which they are compared, i.e. ozonesondes of 50 m sampling adapted to the 1.4 km SAOZ resolution or of SAOZ adapted to the broad 2–3 km satellite limb viewing resolution. For better evaluation of the precision of each instrument the contribution of horizontal transport is removed following the technique applied to SAOZ as described above. But since the contribution of convective transport is relatively limited, it will be ignored. The mathematical background used in this section is given in Appendix A.

3.1 Ozonesondes

A first statistical comparison between SHADOZ ozonesondes (Borchi et al., 2005) at Samoa, Fiji and Reunion Island and SAOZ-MIR flights carried in February and March 2001 and 2003 concluded at (i) a small systematic altitude shift up of the sondes by 300 m consistent with the known 40–50 s time constant of the Electrochemical Concentration Cell (ECC) on a balloon ascending at 5–6 m/s (SPARC, 1998, Johnson et al., 2002); (ii) an average relative ozone low bias of 4±1% in the stratosphere between 20 and 26 km, and (iii) a 5% precision. As already explained, at lower altitude in the upper troposphere the SAOZ measurements were found comparable with those of the sondes at Reunion Island but systematically high biased by 50–60 % compared to the ascents of Samoa and Fiji in the high clouds area of the South Pacific Convergence Zone (SPCZ) suggesting a systematic high bias in remote sensing measurements in oceanic convective clouds areas. This problem has been revisited in 2004 by dedicated ozonesondes ascents during the pass of the balloon over the stations.

3.1.1 Ozonesondes at Bauru and at three SHADOZ stations in 2004

The data used here are those of thirteen ozonesondes ascents performed by IPMet and the Danish Meteorological Institute (Pommereau et al., 2007) from Bauru during the HIBISCUS campaign in February and March 2004 and those of special ascents during the overpass of the balloon over the SHADOZ stations of Samoa (14.2° S, 171° W) and Fiji (18.1° S, 178° E) in the Western Pacific (S. Oltmans, personal communication) and Reunion Island (21° S, 55.5° E) in the Indian Ocean (F. Posny, personal communication). For statistical studies several other ascents at the three stations between 23 February and 31 March 2004 available from the SHADOZ database (http://croc.gsfc.nasa.gov/shadoz/), have been added. All ozonesondes are ECC (Electrochemical Concentration Cell) but of various types, i.e. SPC 6A, 2% unbuffered KI solution at Fiji and Samoa, ENSCI Z 0.5% buffered at Reunion, and partly SPC 6A partly ENSCI both 1% buffered at Bauru, and in addition using different correcting factors for pump efficiency. In the following, ozone partial pressure is converted into number density using the radiosonde pressure and temperature. The altitude is a geopotential height derived from the pressure sensor of the sonde. Since the sonde sampling of 50 m is higher than that of SAOZ, the sonde data have been smoothed with a binomial filter to match the SAOZ resolution of 1.4 km, and then linearly interpolated to 1 km steps.

The circumstances of the ozonesondes ascents and the SAOZ measurements are displayed in Table 1. Though the MIR passed close to the stations, because of the time difference with the ascents, the observations are always separated by more than 475 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 clouds. Even more difficult to interpret are the SAOZ measurements of Reunion Island close to the powerful Gafilo cyclone present over the area on

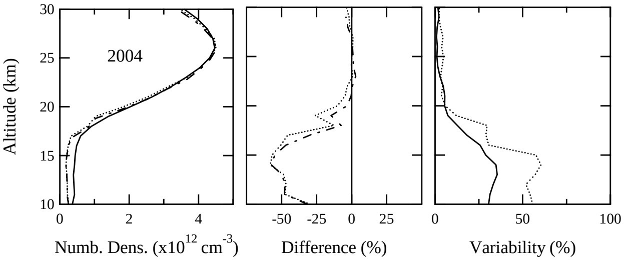

Fig. 4. Comparison between SAOZ and SHADOZ ozonesondes in 2004. From left to right: mean zonal profile, relative percent difference compared to the mean, and relative standard deviation. Solid line: SAOZ; dotted: SHADOZ; dotted-dashed: SHADOZ altitude adjusted.

2–17 March which prevented lidar observations during the period. Indeed, the temperature and ozone profiles of March 10 and March 11 ascents display a large upwelling and cooling of the tropopause compared to the monthly average, and at the opposite, an increase of ozone in the lower stratosphere highly correlated with a PV increase indicative of strong advection from mid-latitude.

3.1.2 SHADOZ statistical comparison in the stratosphere

Twelve ascents are available between 23 February and 31 March 2004 at Samoa (14.2° S, 171° W), Fiji (18.1° S, 178° W) and Reunion Island (21° S, 55.5° E), ignoring those of February 10 and 11 at the latter station because of the cyclone.

The sondes and SAOZ mean profiles, percent difference and relative variability are displayed in Fig. 4. The analysis confirms the results of 2001 and 2003 displaying a shift up by some 200 m of the sondes compared to SAOZ, and a 4% precision. But, after adjustment for the altitude shift, the average relative O_3 bias of +1.5% in 2004 is different from the −4% of 2001. The possible significance of such difference derived from comparisons with 12 ascents in 2004 and 19 in 2001 of different instrumental arrangement and data processing software, is difficult to access. All which can be safely concluded is that the differences remain within the ±5% accuracy quoted for the SHADOZ sondes (Thompson et al., 2003).

3.1.3 Individual overpass comparisons in the troposphere

On average as shown in Fig. 4, the SAOZ profiles are high biased by some 50% in the troposphere compared to the sondes. But given the high variability of tropospheric ozone concentration, this is of little meaning. A better approach is to look at collocated profiles above individual stations. The comparison between sondes and SAOZ profiles over each station is displayed in Fig. 5 and the results of the analysis are summarized in Table 2. In the troposphere, the results confirm the analysis of 2001 and 2003 displaying for the two Pacific stations, a high bias of some 30% of SAOZ compared to the sondes, even for the closest MIR overpass 611/734 km over Samoa. However, a similar high bias of 26% is found at Reunion Island in 2004 in contrast to 1% in 2001. In that case, the difference might be partly attributed to a similar lift of low ozone air by the cyclone, which was not be seen by SAOZ more than 500 km west. Low or even zero ozone layers as frequently reported by the sondes in convective regions have never been observed with the SAOZ whose measurements are limited to cloud free areas. Although differences of technical arrangements could certainly contribute to differences between ozonesonde stations (e.g. the data processing software of Reunion Island was changed in 2003 for the NOAA/CMDL system), the results of 2004 support the idea of a possible systematic overestimation of average tropospheric ozone in the tropics by remote sensing instruments because of the absence of measurements in the presence of clouds.

Table 2. Ozonesondes-SAOZ percent difference in the stratosphere and the troposphere after altitude adjustment (NA = Not Applicable).

Station	Stratosphere	Troposphere
Bauru	−0.9±5.3	NA
Samoa	+2.3±1.5	−32.7±16.6
Fiji	+2.6±2.5	−34.0±13.4
Reunion	+3.3±3.4	−26.0±24.8

3.2 HALOE

The HALogen Occultation Experiment (HALOE) aboard the Upper Atmosphere Research Satellite (UARS) launched in September 1991 and still in operation, performs solar occultation measurements to infer mixing ratio profiles of trace gases (Russell et al., 1993). Ozone measurements are carried out in a broadband channel centred at 9.6 μm. The geometry of the UARS orbit (57° inclination, circular at 585 km with orbit period of 96 min) results in about 15 sunrises and 15 sunsets daily, with tangent point locations during a day confined to small latitude bands, different for the two events, which cycle over varying extremes, roughly every 36 days. Tangent heights are derived from a solar edge detection system. 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. They vary from about 5–10% in the middle and upper atmosphere, to about 30% at 100 hPa (Bhatt et al., 1999). The vertical resolution of 2 km of HALOE profiles is close to that of SAOZ, and the sampling 0.3 km.

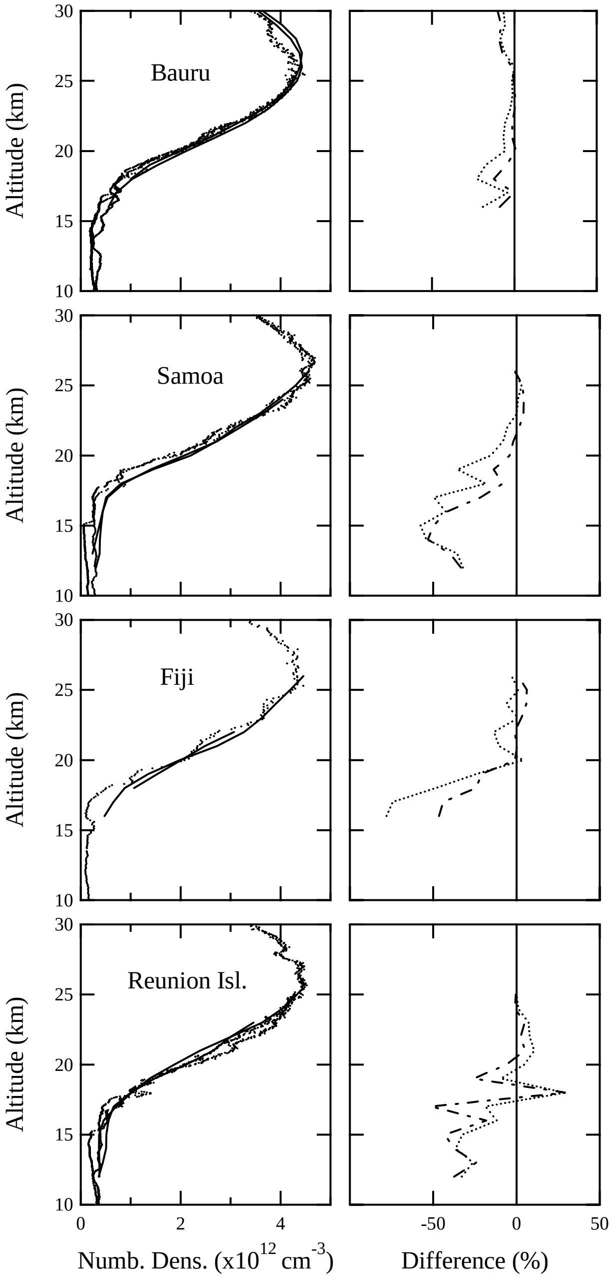

Fig. 5. Comparison of best collocated SAOZ and sondes ozone profiles in 2004. Left: profile; right: percent difference. solid line: SAOZ; dotted: sondes; dotted-dashed: sondes altitude adjusted. From top to bottom: Bauru, Samoa, Fiji and Reunion Island.

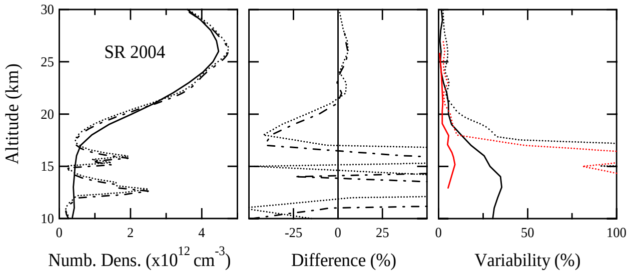

Fig. 6. Comparison between SAOZ and HALOE sunrise ozone in 2004. From left to right: mean zonal profile, relative percent difference, and relative percent standard deviation. Solid line: SAOZ; dotted: HALOE; dotted-dashed: HALOE altitude adjusted for bias; Red: variability after removal of horizontal transport.

A first comparison between HALOE v19 and SAOZ in 2001 and 2003 at the Southern Tropics at 20°±5° S during the SH summer (Borchi et al., 2005) showed an insignificant ozone bias in the stratosphere between 22 and 26 km of +1% to −0.7% depending on the year and the event, a small altitude shift up by 100 m and a precision of 4%. However, the agreement was observed to degrade rapidly below 22 km, where increasing differences with SAOZ, as well as decreasing precision, were reported.

3.2.1 HALOE data selection

The data in use here are those of version 19 of the retrieval algorithm available on the HALOE website http://haloedata.larc.nasa.gov/. The data selected are a series of measurements on 15 consecutive orbits during or as close as possible to the balloon flight period within the latitudinal range 15–21° S. The closest available in 2004 are 15 sunrise profiles within 14.9–20.9° S on 11–12 February before the launch of the balloon on 26 February. There is no sunset measurement available during the flight period nor sunrise profiles in March within 15–21° S. The HALOE products available in the data files are ozone mixing ratios on altitude levels. Since the SAOZ measured quantity is a number density, the HALOE mixing ratios data have been converted in concentrations using NCEP (National Center for Environmental Prediction) pressure and temperature also provided in the data files. Finally, since the HALOE vertical resolution is close to that of SAOZ and provided every 300 m, the data have been just linearly interpolated to the SAOZ 1 km altitude grid.

3.2.2 Comparison with SAOZ

The comparisons between HALOE and SAOZ ozone mean profiles, difference, and relative variability, are displayed in Fig. 6 and the results of the analysis for the stratosphere between 22 and 26 km are summarised in Table 3. Above 22 km, the HALOE mean profile shows a small altitude shift up by 200 m and a limited ozone high bias of 1.8% after adjustment for the altitude shift. After removing the contribution of horizontal transport by correlation with PV, the residual variability or precision (in red in the plots) is 3.4% between 22 and 26 km, which is slightly larger than the 1.5% of SAOZ. However, as in 2001 and 2003, the difference between HALOE and SAOZ increases rapidly below 22 km

Table 3. Difference between HALOE and SAOZ in 2004 between 22 and 26 km before and after altitude adjustment, and relative variability before / after removal of horizontal transport.

	Diff %	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var Haloe %
2004 SR	3.7±1.9	+200	1.8±1.6	2.2/1.5	4.7/3.4

Table 4. Difference in ozone number density or altitude registration between HALOE and SAOZ below 22 km in 2004, after adjustment for the HALOE altitude shift up by 200 m above 22 km.

Alt. (km)	ΔO_3 (%)	ΔAlt. (m)
22	–0.5	0
21	–11	450
20	–25	800
19	–38	1100
18	–41	1550

reaching 1550 m in altitude or 41% in number density at 18 km (Table 4), below which the HALOE data appear not to be reliable.

The comparison with SAOZ in 2004 fully confirms the results of 2001 and 2003. The altitude shift up by 100–200 m in the lower stratosphere is a well-known feature of HALOE (SPARC, 1998), thought to be due to a bias in the detection of the top edge of the sun in the presence of heavy aerosol or cirrus clouds, particularly in the tropics. The 1.8±1.6% ozone high bias between 22 and 26 km is comparable to the $+1/-0.7\%$ difference observed in 2001/2003 at both sunrise and sunset. Finally, the increasing difference between the two systems below 22 km is very similar on the three years. These results are very consistent with the small bias ($<4\%$) between HALOE v19 and SAGE II v6.00 through most of the stratosphere, but increasing systematically (HALOE low compared to SAGE II) below 22 km as reported by Morris et al. (2002), but for which no satisfactory explanation was provided. The only piece of information we could add here is that it that the increasing difference below 22 km is to be attribute to HALOE since nothing similar could be seen between SAOZ and all other systems, including SAGE II v6.0, as described in the following sections. However, the still open question is to know if it is resulting from an underestimation of the ozone mixing ratio or a progressively growing error in the HALOE altitude registration.

3.3 SAGE II

The Stratospheric Aerosols and Gas Experiment II (SAGE II) aboard the Earth Radiation Budget Satellite (ERBS) launched in October 1984 and still operational in 2004, utilizes the solar occultation method to retrieve ozone, water vapour, aerosols and NO_2 (Mauldin et al., 1985). ERBS orbits the Earth along a circular path at an altitude of 610 km with an inclination of 56° allowing the measurement of 15 sunset or sunrise profiles every month at a given tropical latitude. Ozone is retrieved in a channel centred at 600 nm in the Chappuis bands (Cunnold et al., 1989) using cross-sections of Shettle and Anderson (1994). The vertical resolution is $\sim$1 km and the altitude sampling 0.5 km. Tangent heights are calculated by a solar edge detection algorithm. The precision of the ozone measurements is estimated to 1% in the middle stratosphere (2.5% accuracy), degrading to about 2% near the stratopause and the tropopause and to higher values in regions of very low ozone (Manney et al., 2001).

A first comparison between SAGE II v6.2 and SAOZ in 2001 and 2003 at the Southern Tropics at 20°±5° S during the SH summer (Borchi et al., 2005) showed a relative ozone high bias of 2–4% in the stratosphere between 20 and 26 km, a small altitude shift up by 100–200 m and a 2% precision. In the troposphere, the comparison showed a systematic 50–60% low bias of SAGE II with a precision of about 40–60%, very consistent with that found between SAGE II and ozonesondes by Kar et al. (2002) and Wang et al. (2002), attributed by these authors to the high sensitivity of the retrieved ozone abundance to the background (electronic offset) of the SAGE II channel.

3.3.1 SAGE II data selection

The data in use here are those of version 6.2 of the retrieval algorithm available on the SAGE website www-sage2.larc.nasa.gov. Eleven SAGE II sunset profiles within 15.4–19.7° S on 20–21 March 2004 are available for comparison with SAOZ. There is no sunrise measurement during the balloon flight period. Since the SAGE II products are number densities at geometric altitude on a vertical resolution closed to that of SAOZ and a 500 m altitude sampling, they have been compared directly to SAOZ by just selecting the data at the 1 km SAOZ grid.

3.3.2 Comparison with SAOZ

The comparison between SAGE II and SAOZ in 2004 is displayed in Fig. 7 in the same format as for HALOE and the results are summarised in Table 5. The SAGE II mean profile shows a small altitude shift up by 150 m and an ozone high bias of 4.5% in the stratosphere after adjustment for the

Table 5. Same as Table 3 for SAGE II and SAOZ, between 20 and 26 km.

	Diff %	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var SAGE %
2004 SS	5.3±4.3	+150	4.5±2.3	3.2/1.6	4.7/2.5

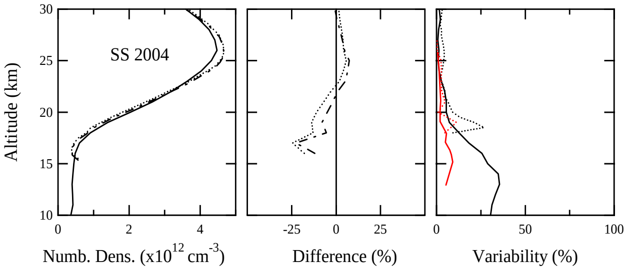

Fig. 7. Same as Fig. 6 but for SAGE II in 2004 at sunset (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid).

altitude shift. Above 20 km, the 2.5% precision of SAGE II estimated from the variability after subtracting the contribution of transport is of the same order of the 1.6% found for SAOZ. Unfortunately, only one profile is available between 15.5 and 18 km, thus nothing could be said in 2004 on SAGE II performance in the troposphere.

These findings fully confirm previous SAGE II performances estimations. The altitude shift up by 100–150 m 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). The small SAGE ozone high bias is consistent with the 2% smaller absorption cross-sections in use in the SAGE retrieval compared to that of SAOZ (Bazureau, 2001). Finally, the comparable precision of the SAGE broadband channels and the SAOZ spectral measurements suggests that aerosol and Rayleigh attenuations are efficiently removed in the SAGE v6.2 retrieval algorithm.

3.4 SAGE III moon occultation

The Stratospheric Aerosol and Gas Experiment III (SAGE III) was launched into a sun-synchronous orbit aboard the Russian Meteor 3M platform in December 2001 (McCormick et al., 1991). SAGE III adds solar and lunar occultation and limb modes. Lunar occultation, the only one available in the tropics, provides profiles of ozone, nitrogen dioxide, nitrogen trioxide, and chlorine dioxide. The hardware configuration of the SAGE III instrument is significantly different from either SAGE I or SAGE II. SAGE III, a grating spectrometer measuring ultraviolet, visible and near infrared radiances from 280 to 1040 nm, builds on and improves on the previous experiments by including additional wavelength, allowing better O_3 measurements at lower altitude (Thomason et al., 1999). Because of the spatial and spectral non-uniformity of the lunar albedo, the variation in lunar illumination as a function of lunar phase, and the moon's relatively low brightness, the lunar altitude registration is much more difficult than that of the sun (SAGE III ATBD, 2002). Tangent heights are derived using a radiometric model of the moon for predicting the radiometric center different from the geometric center in the case of the moon. Below 50 km, ozone is retrieved in the Chappuis bands using cross-sections of Shettle and Anderson (1994). The precision of O_3 profiles in lunar occultation mode is about 10% between 16 and 35 km. The vertical resolution is 500 m. More details could be found in the SAGE III Algorithm Theoretical Basis Document: Solar and Lunar Algorithm (2002).

Table 6. SAGE III moon occultation selected profiles.

Date	Total	Latitude ° S	Longitude ° E
13 March 2004	2	19.0 and 24.4	110.6 and 56.5
11 April 2004	2	18.5 and 24.1	99.3 and 45.2

3.4.1 SAGE III moon data selection

The data in use here are those of version 3 of the retrieval algorithm available on the SAGE III website www-sage3.larc.nasa.gov. Four profiles could be found in 2004 close to the latitude and time of the SAOZ flights (Table 6). No collocated data were available for 2003. Since the SAGE III products are ozone number densities at geometric height of 500 m resolution every 500 m, they have been smoothed and linearly interpolated to the SAOZ 1 km grid.

3.4.2 Comparison with SAOZ

The comparison between SAGE III and SAOZ is displayed in Fig. 8 and the results in Table 7. The SAGE III mean profile shows a large shift down by 550 m, but after adjustment for this altitude shift the ozone bias in the stratosphere drops to 2.5%. Above 20 km and without removing the contribution of horizontal transport (only four selected profiles), the variability of the concentration is 4.5% suggesting a precision of the same order of that of SAGE II. But remarkably and in contrast to SAGE II, the bias and precision hold down to the lowest altitude of the measurements at 15 km.

Table 7. Same as Table 3 for SAGE III moon occultation and SAOZ between 20 and 26 km.

	Diff%	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var SAGE %
2004	16.3±5.3	–550	2.3±2.0	3.2	4.4

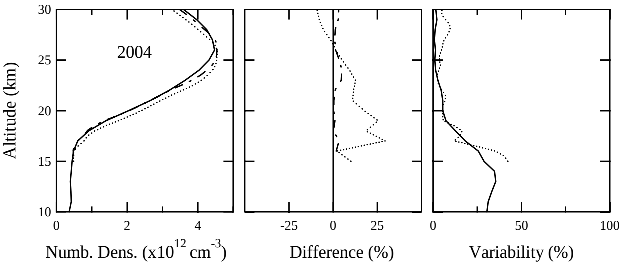

Fig. 8. Same as Fig. 6 but for SAGE III moon occultation (only four collocated profiles) in 2004 (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid). The limited number of profiles available does not allow reliable removal of horizontal transport.

Likely because of the larger uncertainty of the lunar altitude registration compared to the sun, the SAGE III altitude bias is larger than that of SAGE II. However, the small ozone high bias is similar to that observed between SAGE II and SAOZ, consistent with the 2% smaller absorption cross-sections in use in both retrievals compared to that of SAOZ (Bazureau, 2001). Finally, the variability of 4.5% derived from this study, close to that of SAOZ, is slightly better than the 10% estimated by the SAGE III ATBD (2002), although the small number of profiles available for comparison do not allow definite conclusions on this aspect.

3.5 Odin-OSIRIS

The Swedish/Canadian/French/Finish Odin satellite was launched on February 20, 2001 into a circular quasi-polar orbit at 600 km. Detailed information on Odin and its mission can be found in Murtagh et al. (2002) and Llewellyn et al. (2003). Odin carries two co-aligned instruments: a Sub-Millimeter Radiometer (SMR) and an Optical Spectrograph and Infrared Imager System (OSIRIS), both enable to scan the Earth's limb with tangent heights ranging from about 10 to 70 km. The OSIRIS spectrograph measures scattered sunlight within a field of view of 1.288 arc min in the vertical direction, equivalent to ∼1 km height at the tangent point. Ozone number density profiles are retrieved from the OSIRIS limb radiance spectra in the Chappuis absorption bands between 10 and 50 km on a 2 km grid, using the inversion technique suggested by Flittner et al. (2000) and McPeter et al. (2000) and adapted to OSIRIS by von Savigny et al. (2003). The vertical resolution is 2 km. The absorption cross-sections are those of Burrows et al. (1999). Tangent heights are calculated from the viewing direction provided by a star tracker and the orbitography of the satellite. The overall accuracy of an individual profile is estimated to be better than 10% within the 15 to 35 km altitude range (von Savigny et al., 2003).

Comparison with 1220 POAM III profiles and 205 ozonesondes (with a much broader latitudinal range in the both Hemisphere) during the 12 month period from November 2001 to October 2002 showed an ozone high bias ranging from 5–7% for altitudes between 15 and 32 km and a strong altitude low bias of about 1 km during the April–July period for which no clear explanation has been provided yet (Petelina et al., 2004). The random error of 100–300 m at the tangent point was found too small for accounting alone for the offset. The cause was suggested to be presumably due to other altitude registration issues such as misalignment or shift of the optical axis of the instrument in respect to the star-tracker, although it is also possible that systematic errors within the current operational inversion algorithm, such as aerosol climatology, are adding to the problem (Petelina et al., 2004).

3.5.1 Odin-OSIRIS data selection

The data in use here are those of version 2.3 of the retrieval algorithm available on the OSIRIS website http://osirus.usask.ca. 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. Forty-six OSIRIS profiles within 19.6–24.5° S between 15 February and 6 March are available for comparison with SAOZ in 2003 and 42 within 15.0–20.9° S between 22 February and 16 March 2004. Since OSIRIS products are number densities every km geometric height with a resolution close to that of SAOZ, the data have been just interpolated at the SAOZ 1 km grid.

3.5.2 Comparison with SAOZ

The comparison between OSIRIS and SAOZ in 2003 and 2004 is shown in Fig. 9 and the results summarised in Table 8. The OSIRIS mean profiles display an average altitude shift down by 300 m in 2003 over the Pacific and by 900 m in 2004 over the entire latitude band. After adjustment for this shift, OSIRIS is found insignificantly high biased compared to SAOZ by 1–2% in the whole stratosphere down to 19 km in 2003 and 15 km in 2004. The precision above 20 km estimated from the variability after correction for horizontal transport is 7.4% in 2003 and 4.7% in 2004, about 3 times

Table 8. Same as Table 3 for OSIRIS- SAOZ between 20 and 26 km.

	Diff. %	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var Osiris %
2003	4.5±3.3	–300	1.0±0.7	4.2/2.5	11.3/7.4
2004	15.9±13.1	–900	2.0±1.8	3.2/1.6	7.2/4.7

worse than that SAOZ. But it degrades progressively below reaching 35% and 20% at 15 km respectively in 2003 and 2004. An important question is the weight in the results of the a priori profile used in the inversion at low altitude. According to von Savigny et al. (2003), the retrieved concentrations should be quite insensitive to the a priori down to 18 km at low latitude. Overall, the results of the comparison with SAOZ are very consistent with previous evaluations of OSIRIS performances showing a relatively large low bias in altitude registration of unknown origin, but after adjustment for that, a very small 1–2 % ozone bias compared to SAOZ, corresponding exactly to the difference of absorption cross sections (Orphal et al., 2003). The 5% OSIRIS-ozonesondes mean difference in the 20–26 km altitude range derived from this study is very consistent with the 5–7% bias found by Petelina et al. (2004), both after OSIRIS altitude adjustment. The sum of the 1–2% systematic error and the 5–7% precision derived from the variability corrected for transport is even better than the 10% accuracy on individual profiles quoted by von Savigny et al. (2003). The only limitation is the increasing systematic errors in the upper troposphere due to the sensitivity of the retrievals to the a priori profiles. The information provided by the OSIRIS data degrades rapidly below 18 km in the tropics.

3.6 Odin-SMR

The Sub-Millimetre Radiometer (SMR) also aboard Odin employs 4 tuneable single-sideband Schottky-diode heterodyne receivers in the 485–580 GHz spectral range. In aeronomy mode, various target bands are dedicated to observations of trace constituents relevant to stratospheric/mesospheric chemistry and dynamics such as O_3, ClO, N_2O, HNO_3, H_2O, CO, and isotopes of H_2O and O_3 (Murtagh et al., 2002). Profile information is retrieved from the spectral measurements of a limb-scan by inverting the radiative transfer equation for a non-scattering atmosphere. A retrieval algorithm based on the Optimal Estimation Method (Rodgers, 1976) has been adopted (Urban et al., 2004). In stratospheric mode, the instrument scans the limb between roughly 7 and 70 km in about 90 s. Tangent-heights are calculated from the satellite's orbitography and the information of the star tracker. The altitude range of the ozone measurements in the 501.8 GHz band used here is 17–45 km with vertical resolution of about 2.5 km. The data are provided every 2 km. The precision for a single-scan is of the order of 20–25% (0.25–1.5 ppmv). The upper limit of systematic errors for O_3 in the lower stratosphere is estimated to ~1 ppmv (Urban et al., 2005).

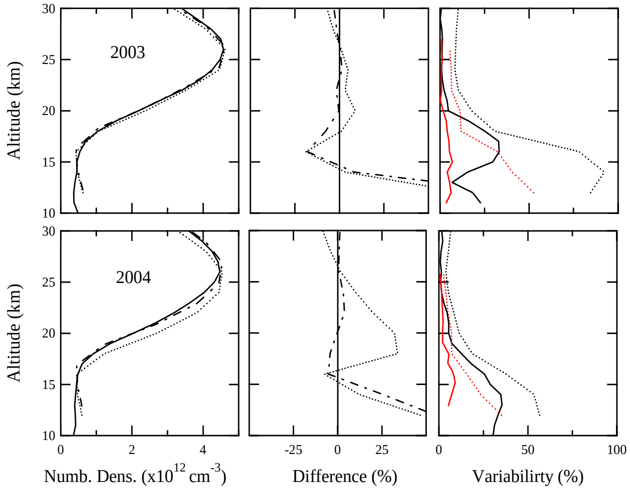

Fig. 9. Same as Fig. 6 but for OSIRIS (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid).

The two comparisons available between SMR and independent data such as ozonesondes and MIPAS at different periods and locations showed a high O_3 bias smaller than 10% between 20 and 55 km (Murtagh et al., 2002; Kerridge et al., 2004).

3.6.1 Odin-SMR data selection

The data in use here are those of version 222 of the retrieval algorithm available on the SMR website http://ether.ipsl.jussieu.fr. Thirteen SMR profiles within 15.2–26.4° S and 45° W–150° E (over the Pacific) are available on 18 February 2003 which could be compared to SAOZ and 189 at all longitudes within 15.3–20.7° S between 22 February and 20 March 2004. The SMR products available in the data files are ozone-mixing ratios at altitude levels. Since the SAOZ measured quantity is a number density, the SMR data have been converted using ECMWF (European Centre for Medium-Range Weather Forecasts) average pressure and temperature at the SAOZ tangent locations. As the SMR vertical resolution is of 2.5 km the SAOZ data have been smoothed and then compared to that of SMR on a 2 km grid.

Table 9. Same as Table 3 for SMR-SAOZ between 20 and 26 km.

	Diff %	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var SMR %
2003	6.3±6.3	–350	4.3±4.5	4.2/2.5	24.8/18.2
2004	8.0±1.9	+300	6.7±5.7	3.2/1.6	21.9/15.5

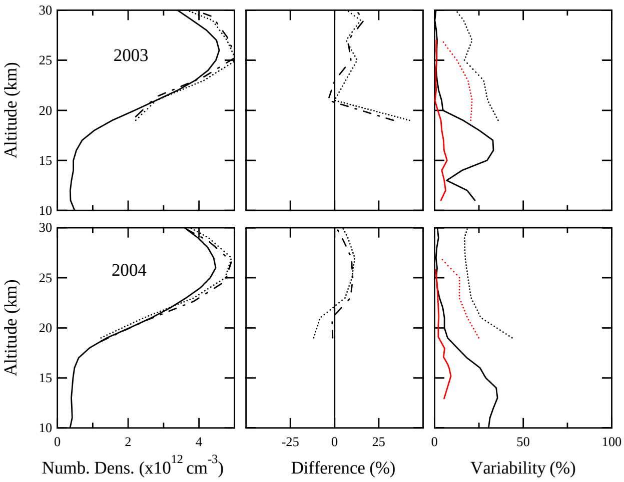

Fig. 10. Same as Fig. 6 but for SMR (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid).

3.6.2 Comparison with SAOZ

The comparisons between SMR and SAOZ in 2003 and 2004 are shown in Fig. 10 and the results summarised in Table 9. The SMR mean profiles display an average altitude shift down by 350 m in 2003 over the Pacific and up by 300 m in 2004 over the entire latitude band. After adjustment for this shift, the SMR ozone is found high biased compared to SAOZ by 4.3% in 2003 (with only 13 profiles) and 6.7% in 2004 (with 189 profiles) between 20 and 26 km. The precision above 20 km estimated from the variability is 18.2% in 2003 and 15.5% in 2004, about 8 times worse than that of SAOZ.

According to Urban et al. (2005), the altitude shift could be resulting from a scan-bias assuming an uncertainty of 500 m. The 9% SMR high bias compared to the sondes is consistent with the 10% reported by Murtagh et al. (2002). The precision of 16–17% derived from this study is slightly better than the 20–25% estimated by Urban et al. (2005). An important question here also is the weight of the a priori profiles used in the inversion at low altitude. According to Urban et al. (2004) and Urban et al. (2005) retrieved concentrations should be quite insensitive to the a priori down to 19 km at low latitude. The SMR measurements should have thus little meaning below 19 km in the tropics.

3.7 ENVISAT-GOMOS

The Global Ozone Monitoring by Occultation of Stars (GOMOS) instrument aboard the ENVIronmental SATellite (ENVISAT) has been launched by the European Space Agency on 1 March 2002. ENVISAT is a sun-synchronous polar-orbiting satellite at about 800 km altitude. GOMOS is a star occultation UV-visible-near IR spectrometer providing O_3, NO_2, NO_3, H_2O, aerosols and stratospheric clouds measurements, from 15 to 100 km for O_3 and from 20 to 50–60 km for other constituents (Bertaux et al., 1991; Bertaux et al., 2001). Slant columns of all species are retrieved simultaneously by global minimization of the mean squares difference between measured and modelled transmissions. Below 40 km, ozone is retrieved in the Chappuis bands using cross-sections of Burrows et al. (1999). Tangent heights are calculated from the orbitography of the satellite. Since the light source is punctual and the location of the satellite and thus the direction of the star are very accurately known, the altitude registration of the measurements is better than ± 50 m. Vertical profiles are derived using an onion peeling inversion technique (Connor and Rodgers, 1988) assuming a linear variation of concentration between 2 layers. A Tikhonov regularization is further applied during the profile inversion for smoothing out unphysical oscillations that can appear on the vertical profiles due to atmospheric scintillation. The smoothing results in a degradation of the vertical resolution to 3 km. The vertical sampling is about 400–500 m. The data used in the following are those produced by a research retrieval algorithm (version v6.0b). Only nighttime measurements are considered.

Comparisons with balloon and ground-based measurements in the 19–64 km altitude range carried with the version v5.4b of the algorithm have suggested a small negative bias of 2.5–7.5%, within a relative standard deviation of 11–16% (Meijer et al., 2004). Compared to SAGE II, the products of v6.0 have shown to display also a small negative bias of 0–5% between 21 and 52 km (Bracher et al., 2004).

3.7.1 ENVISAT-GOMOS data selection

Fifteen GOMOS profiles within 18.4–25.1° S and 45° W–150° E (over the Pacific) between 26 February and 4 March in 2003 are available for comparison with SAOZ and 35 within 14.9–21.0° S between 22 February and 20 March 2004. Since the GOMOS products are number densities at

Table 10. Same as Table 3 for GOMOS-SAOZ between 22 and 26 km.

	Diff %	Alt bias (m)	Diff% Alt. adj.	Var SAOZ %	Var GOMOS %
2003	–2.5±0.5	0	NA	2.1/1.1	4.5/3.1
2004	–1.1±0.8	-50	–1.0±0.6	2.2/1.5	8.9/6.1

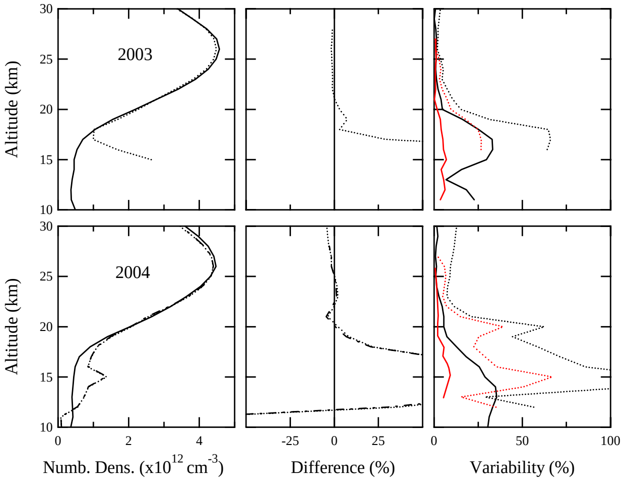

Fig. 11. Same as Fig. 6 but for GOMOS (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid).

geometric height, they can be compared directly to SAOZ. As the GOMOS vertical resolution is 3 km and the sampling 400–500 m, the SAOZ data have been smoothed and those of GOMOS linearly interpolated to the SAOZ altitude grid of 1 km.

3.7.2 Comparison with SAOZ

The comparisons between GOMOS and SAOZ in 2003 and 2004 are displayed in Fig. 11 and the results summarised in Table 10. On both years, the mean profiles of the two instruments are very consistent above 22 km in the stratosphere showing no altitude shift within ±50 m and a small, little significant, low ozone bias of GOMOS of 1–2.5%. At these altitude levels, the only difference is in the worse precision of GOMOS of 3.1% over the Pacific in 2003 and 6.1% over the full latitude band in 2004 above 22 km, but degrading very rapidly below that level.

The absence of altitude shift between GOMOS and SAOZ is very consistent with the expectation of a star occultation system. The small low ozone bias is fully corresponding to the difference of 2.5% between ozone absorption cross-sections used respectively by the two algorithms in the visible Chappuis bands (Orphal et al., 2003). The 1% GOMOS low bias compared to SAGE II is also consistent with the 0–5% reported by Bracher el al. (2004), both after altitude adjustment. The main limitation of GOMOS ozone measurements demonstrated by the comparison is the relatively large random error on individual profiles of ∼6% (one standard deviation) in the stratosphere above 22 km related to chromatic scintillation due to small-scale atmospheric turbulence structures. Finally, because of the fast increase of noise below 22 km, current GOMOS ozone products are of little use in the Tropical Tropopause Layer (TTL) and in the troposphere.

3.8 ENVISAT-SCIAMACHY

SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY) is a satellite-borne instrument also aboard ENVISAT capable of performing spectroscopic measurements of the chemical composition of the Earth's atmosphere in three different observation geometries: nadir, solar/lunar occultation and limb scattering (Burrows and Chance, 1991; Bovensmann et al., 1999). The O_3 limb profiles used here are derived by the Institute of Environmental Physics at University of Bremen (IUP). The retrieval algorithm employed is similar to that developed by Flittner et al. (2000) and McPeters et al. (2000) for the Shuttle Ozone Limb Sounding Experiment (SOLSE) also used for OSIRIS (von Savigny et al., 2003). The retrieval exploits the differential structure of the O_3 absorption cross sections, measured in the laboratory with the SCIAMACHY Flight Model, between the centre (600 nm) and the wings (525 nm and 675 nm) of the Chappuis absorption bands. A linearised version of optimal estimation is used together with the radiative transfer model SCIARAYS (Kaiser, 2001) to iteratively retrieve stratospheric O_3 concentration profiles. The altitude range from about 15 km up to 40 km with vertical a resolution of about 3.5–4.0 km can be covered with this technique as long as there are no clouds or aerosols in the instrument's field of view. The data are provided every 1 km in the altitude range 10–50 km. Tangent heights are calculated from the viewing direction provided by a star tracker and the orbitography of the satellite. The instrumental error is smaller than 1%, but retrieval errors due to errors in the assumptions of albedo, aerosol profile and background atmosphere are generally larger. Sensitivity studies have shown that these are the major systematic error sources and vary between 6% and 10% at 15–40 km altitude and around 6–7% between 20 and 25 km (Rozanov et al., 2003).

The SCIAMACHY v1.6 ozone profiles have been shown to be low biased by 10–15% for an unknown reason in the

Table 11. Same as Table 3 for SCIAMACHY-SAOZ between 20 and 26 km.

	Diff %	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var SCIA %
2003	–1.4±4.5	+250	2.1±1.5	4.2/2.5	7.7/5.2
2004	7.2±3.7	–300	5.5±2.5	3.2/1.6	5.4/3.7

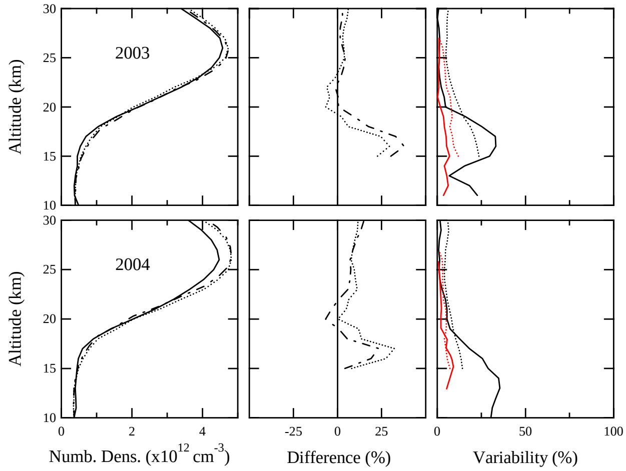

Fig. 12. Same as Fig. 6 but for SCIAMACHY (dotted), altitude adjusted (dotted-dashed) and SAOZ (solid).

altitude range 20–35 km compared to HALOE, SAGE II and III, GOMOS, MIPAS, lidars and sondes, as well as shifted up by 1 to 2 km compare to all others (Bracher et al., 2005; Brinksma et al., 2005; Rozanov et al., 2003; Segers et al., 2005). The altitude shift was identified to mainly result from pointing errors caused by an incorrect knowledge of the satellite orientation and/or position (Rozanov et al., 2003). A monthly mean tangent height offset of 800 m amplitude was identified before the improvement of the ENVISAT orbit propagator model in December 2003, but a constant offset component of about 1 km is still presents after (von Savigny et al., 2005).

3.8.1 ENVISAT-SCIAMACHY data selection

The data in use here are those of a new version 1.63 of the retrieval algorithm provided by IUP. Version 1.63 includes the following main changes and improvements compared to version v1.6. First, the retrievals are performed on a 1 km altitude grid compared to the previously used 3.3 km altitude grid – corresponding to the tangent height grid – in v1.6. Second, the v1.63 retrievals are corrected for pointing errors using the version 1.7 of the TRUE (Tangent height Retrieval by UV-B Exploitation) algorithm (Kaiser et al., 2004; von Savigny et al., 2005), which employs ECMWF background atmosphere and ozone profiles taken from the dynamic ozone climatology of Lamsal et al. (2004). The ozone profiles are selected from the climatology following the TOMS v8 total ozone column for a given day and location. Forty-three SCIAMACHY profiles within 15.9–24.8° S and 45° W–150° E (over the Pacific) are available between 18 February and 4 March 2003 for comparison with SAOZ and 65 within 16.2–20.9° S on 15 to 29 February 2004. The SCIAMACHY products are number densities at geometric height, which can be therefore compared directly to SAOZ. As the SCIAMACHY vertical resolution is of 3.5–4.0 km the SAOZ data have been smoothed and then compared to that of SCIAMACHY on a 1 km grid.

3.8.2 Comparison with SAOZ

SCIAMACHY and SAOZ ozone profiles in 2003 and 2004 are shown in Fig. 12 and the results of the analysis summarised in Table 11. The SCIAMACHY mean profiles display an altitude shift up by 250 m over the Pacific in 2003 and down by 300 m in 2004 on average over the entire latitude band. After adjustment for this shift, the ozone concentration is found high biased compared to SAOZ by 5–6% in the stratosphere between 20 and 26 km, that is 3% high biased compared to SAGE II. Still above 20 km, the precision estimated from the variability is 5.2% in 2003 and 3.7% in 2004, about 2 times worse than that of SAOZ.

The new v1.63 version of the retrieval is encouraging showing significant improvements compared to v1.6. The altitude shift of ±300 m is significantly reduced compared to the constant offset of about 1 km found by von Savigny (2005) with the v1.6. The 3% SCIAMACHY high bias compared to SAGE II is also reduced compared to the 6% low bias reported by Brinksma et al. (2005). Finally, the precision of 4% is also better than the error estimate of 6–7% in the 20–25 km altitude range of Rozanov et al. (2003).

However, little change is observed below 20 km in the upper troposphere, where the difference with SAOZ increases rapidly (30% and 25% at 17 km respectively in 2003 and 2004). As concluded by Brinksma et al. (2005) and confirmed by the small atmospheric variability compared to that observed by SAOZ, the profiles are strongly weighted by the a priori used in the retrieval. The information provided by SCIAMACHY is thus of no use in the tropical troposphere.

Table 12. Same as Table 3 for MIPAS-SAOZ between 20 and 26 km.

	Diff %	Alt. bias (m)	Diff% Alt. adj.	Var SAOZ %	Var MIPAS %
2003	3.7±3.3	–350	0.6±0.8	2.1/1.0	8.2/5.3
2004	7.8±9.0	–300	7.1±3.1	3.2/1.6	11.8/8.5

3.9 ENVISAT-MIPAS

MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) aboard ENVISAT is a limb-viewing Fourier transform infrared (FTIR) emission spectrometer covering the mid-infrared region in five spectral bands (Fischer and Oelhaf, 1996; Harris, 2000). The data used here (ESA level-2 near real time operational products) are taken in the standard observation mode, which consists in rearward limb scans covering the altitude range from 6 to 68 km within 17 steps. The vertical resolution is 3 km for the 13 lowermost tangent altitudes and increases to 8 km at the upper end of the limb scan. The ozone profiles are derived using an optimized retrieval model (ORM) (Carli et al., 2004; Ridolfi et al., 2000). The vertical sampling is of about 2.3 km. At low latitude, the ozone total error is estimated around 8–10% at 20–50 km, rapidly increasing below 20 km to 50–60% (Dudhia et al., 2002). Between 20 and 25 km, HALOE-MIPAS and SAGE II-MIPAS ozone mean difference shows a low bias of 7–8% and 4–5% respectively (Kerridge et al., 2004). MIPAS measurements in 2002 and 2003 indicate a slow drift in instrumental pointing towards lower tangent heights (von Clarmann et al., 2003). Since December 2003, a correction scheme based on both engineering and orbit model updates has been provided by ESA that leads to improved limb pointing. Nevertheless, some residual pointing errors still appear to be present in the data.

3.9.1 ENVISAT-MIPAS data selection

The data used here are those of MIPAS NL (the Level 2 near real time operational products) version 4.61 of the retrieval algorithm available on ftp-ops.de.envisat.esa.int. Twelve MIPAS profiles within 17.7–23.3° S between 18 February and 7 March 2003 are available for comparison with SAOZ and 441 within 16.9–20.1° S between 27 February and 15 March 2004. The data used are volume mixing ratios in pressure coordinate, where the pressure is retrieved from the MIPAS spectra. They have been converted in number density and altitude coordinate using ECMWF pressure and temperature. Indeed, the altitude coordinate also available in the MIPAS data files is not being used since appearing to show significant errors. Although the geometric altitudes are derived from retrieved tangent-pressures, the need to fix the geometric altitude of a reference pressure level introduces the possibility for an offset in absolute heights assigned (Kerridge et al., 2004). Since the MIPAS vertical resolution is 3 km the SAOZ data have been smoothed and linearly interpolated to the MIPAS altitude grid of ∼2.3 km altitude grid.

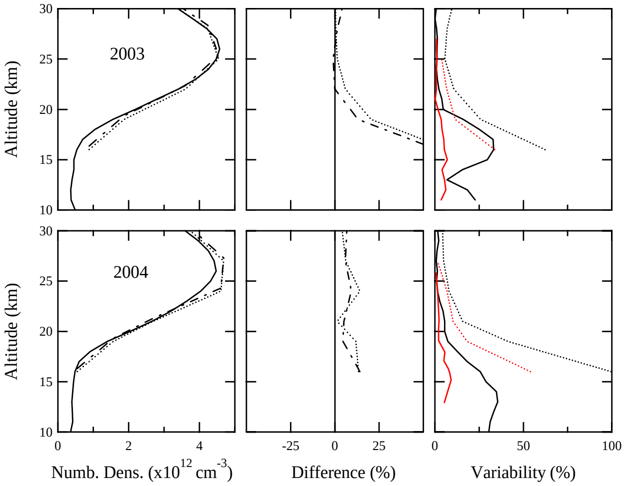

Fig. 13. Same as Fig. 6 but for MIPAS (dotted) in 2003, altitude adjusted (dotted-dashed) and SAOZ (solid).

3.9.2 Comparison with SAOZ

The MIPAS and SAOZ profiles in 2003 and 2004 are compared in Fig. 13 and the results summarised in Table 12. The MIPAS mean profiles display an altitude shift down by 350 m in 2003 over the Pacific and by 300 m in 2004 over the entire latitude band. After adjustment for this shift, MIPAS is found high biased in the stratosphere between 20 and 26 km compared to SAOZ by 0.6% in 2003 and 7.1% in 2004, the last established on 441 profiles being more significant than the first based on only 12 profiles. The difference increases below 20 km reaching 14% at 16 km in 2004. The precision above 20 km estimated from the variability is 5.3% in 2003 and 8.5% in 2004, but decreases below reaching 34% and 54% at 16 km respectively in 2003 and 2004.

Overall the results of the comparison with SAOZ are very consistent with previous evaluations of MIPAS performances. However, the present work does show a systematic altitude shift down by ∼300 m in the tropics in both 2003 and 2004 presumably due to instrumental pointing errors. The −5% HALOE-MIPAS and −7% SAGE II-MIPAS differences derived from their respective differences with SAOZ

Table 13. Summary of performances of all instruments relative to SAOZ in the stratosphere above 20 km, unless stated otherwise, in 2004 *(2001)*.

Instrument	Alt. bias (m)	O$_3$ Bias (%)	Precision (%)	Remark
SAOZ	–	–	2 *(2)*	
Ozonesondes	+200 *(300)*	+1.5 *(−4)*	4 (5)	
HALOE $Z>22$ km	+200 *(<+100)*	+1.8 *(<1)*	3.4 *(4)*	Bias or altitude, and precision degrading below 22 km
SAGE II	+150 *(+150)*	+2.5 *(+4.5)*	2.5 *(2)*	
SAGE III moon	–550	+2.5	4.5*	4 profiles only. * H. transport not removed
Odin-OSIRIS	–900	+2	5	
Odin-SMR	+300	+7	16	
GOMOS $Z>22$ km	–50	–1.5	6	Bias and precision degrading below 22 km
SCIAMACHY	–300	+5.5	4	
MIPAS	–300	+7	8.5	

Table 14. Mean differences between instruments in the 20–26 km altitude range derived from the present study in 2004 and available from the literature.

	Diff %	Diff in literature%	Altitude adjusted
OSIRIS-Sondes	+5	+5/+7	Yes
SMR-Sondes	+9	+10	Yes
GOMOS-SAGE II	–1	–5/0	No
SCIAMACHY-SAGE II	+3	–6	Yes
HALOE-MIPAS	–5	–7/–8	No
SAGE II-MIPAS	–7	–4/–5	No

are of the same order of magnitude of the −7/−8% and −4/−5% noted by Kerridge et al. (2004). The precision of 8.5% above 20 km in 2004 reported here degrading rapidly below, is comparable with the 8–10% reported by Dudhia et al. (2002). According to Carli et al. (2004), the retrieved concentrations should be insensitive to the a priori profile used in the inversion down to 16 km at low latitude. However, the increased high bias and the degradation of precision suggest that the MIPAS ozone profiles are of little use in the Tropical Troposphere Layer.

4 Summary of performances of ozone measuring instruments in tropics

The performances of all available ozone-measuring instruments in the tropical stratosphere relative to SAOZ in 2003 and 2004 are summarized in Table 13. For ozonesondes, HALOE and SAGE II, they fully confirm the conclusions of 2001 (Borchi et al., 2005). 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 −900 m for the limb viewing 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. Another feature, which did not received any explanation yet, is the increasing shift up or ozone low bias of the 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 performing in the visible Chappuis bands and near IR (SAGE II and III, GOMOS, OSIRIS and HALOE), within their ±5% accuracy for the ozonesondes, but it increases for SCIAMACHY still in the visible (+5.5%), MIPAS in the mid-IR (+7%) and SMR in the sub-millimeter region (+7%). Most of above biases are not new. On average, as shown in Table 14, the differences between a couple of instruments derived here are very consistent with those of previous evaluations.

But 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 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, OSIRIS and SCIAMACHY (4–5%), GOMOS above 22 km (∼6%), MIPAS (8.5%) and finally SMR (16%). 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.

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 of limited (50%) precision.

5 Conclusions

A large number of high accuracy ozone profiles has been obtained from 10 to 26 km in 2001, 2003 and 2004 in the tropical UTLS by solar occultation spectroscopic measurements from long duration balloons carrying a SAOZ UV-Vis spectrometer. As shown by a statistical study, the ozone variability along a latitudinal circle at ∼20° S, is smaller than 5% in the stratosphere above 19 km but increases rapidly below in the TTL reaching a maximum of 40% at 14 km immediately below the tropopause. In addition, the high correlation between Potential Vorticity and ozone at all levels suggests that most of the variability (60–70%) could be attributed to horizontal transport and could therefore be efficiently removed from the observed variability by a regression technique. Assuming that other contributions, i.e. photochemistry or convective transport, are negligible, the residual variability after removing horizontal transport provides a rigorous estimation of the precision of the measurements. In the case of SAOZ it is shown that it could vary from ∼2% in the stratosphere, 5–6% at the tropopause and 7–8% at 12 km in the upper troposphere.

Using SAOZ as reference, the relative performances of all ozone-measuring instruments available in the tropics during the period of the balloon flights have been evaluated. It is shown that frequent positive or negative altitude shifts could be observed in the ozone profiles, varying from zero for the GOMOS stellar occultation instrument, followed by +100/200 m for solar occultation systems (SAGE II, HALOE), but as large as −900 m for the limb viewing system OSIRIS. In the case of HALOE, a fast increasing degradation of altitude registration is observed below 22 km. The ozone biases are generally limited, between −4% and +4%, for measurements in the visible Chappuis bands (SAGE II and III, GOMOS and OSIRIS), the near IR (HALOE above 22 km) and the ozonesondes, but increases to +5.5% (SCIAMACHY) although still in the visible, and 7% in the mid-IR (MIPAS) and the submillimetric range (ODIN-SMR). Regarding precision, the best measurements are found to be those of SAGE II (2%), followed by HALOE (3–4% above 22 km), the sondes, SAGE III moon, SCIAMACHY and OSIRIS (4–5%), GOMOS but only above 22 km (∼6%), MIPAS (8.5%) and finally SMR (16%). All satellite measurements appear very little reliable in the tropical troposphere, except those of SAGE II, though low biased by 50% and of limited (50%) precision. Finally, it must be recognised that the retrieval algorithms associated to the various systems investigated here are not at same stage of maturity. For most recent instruments, e.g. onboard ENVISAT, the data shown here are frequently those of research versions of the algorithms expected to receive improvements in the near future. It is hoped that this analysis could contribute. The SAOZ-MIR data remain available for that.

Appendix A

Mathematical background

The statistical method is to compare mean zonal profiles indicative of potential relative biases in concentration or altitude registration, mean differences, and relative variability along a latitudinal circle, indicative of measurements precision. The bias in altitude is given by minimizing the mean value of the relative differences between 20 and 26 km.

$$\mathrm{Diff}_i = 100\,\frac{\overline{x_i} - \overline{\mathrm{SAOZ}_i}}{\overline{\mathrm{SAOZ}_i}}$$

All variables are implicitly altitude dependent. The i index refers to the ith altitude. $\overline{x_i}$ is the mean ozone number density of the correlative data and $\overline{\mathrm{SAOZ}_i}$ that of SAOZ. Diff_i is the relative percent difference.

$$\mathrm{Std\,Dev}_i = \mathrm{Abs.\ Var}_i = \sqrt{\frac{1}{n_i} \sum (x_i - \overline{x_i})^2}$$

Std Dev_i is the standard deviation, equivalent to the absolute ozone variability Abs. Var_i.

$$\text{Rel. Std Dev}_i = \text{Rel. Var}_i = 100\,\frac{\text{Abs. Var}_i}{\overline{\text{SAOZ}_i}}$$

Rel. Std Dev_i is the relative standard deviation, equivalent to the relative variability Rel. Var_i.

$$\text{Diff Satellites}_i = 100\left(\frac{\overline{\text{Instrument A}_i} - \overline{\text{Instrument B}_i}}{\overline{\text{Instrument B}_i}}\right)$$

Diff Satellites_i is the difference between two instruments, equivalent to the ozone bias.

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