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REMOTE SENSING OZONE MEASUREMENTS FROM STRATOSPHERIC BALLOON DURING THE MAP/GLOBUS CAMPAIGN 1983*

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Abstract—Remote sensing measurements of ozone in ultraviolet, visible and infrared domains performed in September 1983 by means of stratospheric balloons during the MAP/GLOBUS campaign in France are presented and discussed. Ozone concentrations deduced from planet and solar occultation measurements in the Huggins and Chappuis absorption bands are given with an uncertainty of ±10%. The results obtained from absorption measurements in the Chappuis bands give systematically higher concentration values than those deduced from *in situ* techniques. Discrepancies of the order of 20% are found between 24 and 34 km altitude. They are larger below the ozone maximum and above 35 km. The analysis of the data shows an aerosol contribution in the measured optical depth in the Chappuis bands for altitude below 28 km, giving overestimated inferred ozone concentrations. Discrepancies at higher altitudes with *in situ* soundings are difficult to explain.

INTRODUCTION

Quantitative measurement of ozone concentration in the stratosphere is of fundamental importance in order to understand the aeronomic processes controlling the ozone budget and to distinguish between possible changes due to natural variations (e.g. solar variability response, see Brasseur and Simon, 1981; Garcia *et al.*, 1984) and those due to anthropogenic influences.

Several methods are presently used to derive the ozone vertical profile from balloon observations. Among these, remote sensing methods based on solar occultation in the ultraviolet and in the visible ranges, similar to satellite observations on infrared emission and on planet occultation in the visible range, have been used during the MAP/GLOBUS campaign in 1983 in order to compare results obtained from different techniques. Previous campaigns (Aimedieu *et al.*, 1983; Hilsenrath *et al.*, 1986; Robbins *et al.*, 1985) have already pointed out systematic differences among various methods of observations.

The purpose of this paper is to present the intercomparison of remote sensing measurements of ozone performed in September 1983 by means of balloon observations in the frame of the MAP/GLOBUS project (Offerman, 1983). Ground-based remote measurements made during the same epoch are reported by de La Noë *et al.* (1987).

PRINCIPLES OF MEASUREMENTS AND INSTRUMENTATION

The data reported and analysed in this work were obtained from three different instruments separately integrated in three gondolas carried by stratospheric balloons. Table 1 summarises the information concerning these three flights.

The first flight carried a stabilized platform of the

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TABLE 1

Flight number	Gondola	Date	Time (U.T.)	Observation method	Institution
29	G5	13–14 Sept.	Night	Planet occultation (Chappuis bands)	LPCE, Orléans
34	G1	23 Sept.	4:30	i.r. Emission (9.6 μm)	Univ. of Wuppertal
37	G2b	28 Sept.	Sunset	Solar occultation (Huggins and Chappuis bands)	IASB, Bruxelles

Geneva Observatory (Huguenin and Magnan, 1978) in which a double grating spectrometer JY DH 10 was integrated. This instrumentation has been extensively described by Rigaud *et al.* (1983). Ozone is measured by its absorption in the Chappuis bands, from 647 to 672 nm, during the night, by observing Venus occultation by the Earth.

The second flight measured the thermal infrared emission of atmospheric trace gases by means of an Ebert–Fastie grating spectrometer working in the wavelength interval 5–19 μm. Ozone emission at 9.6 μm were measured by vertical limb scanning performed by a mirror, covering $\pm 2.4°$ with respect to the balloon horizon. The whole instrument is cooled by liquid helium. An extensive description of instrumentation and calibration methods is given by Rippel *et al.* (1987).

The third flight measured the ozone content by absorption in the ultraviolet and visible domains, using the Sun as a source. The instrument is a double grating spectrometer with holographic gratings of 10 cm focal length made by Jobin–Yvon. A silicon photodiode EG & G type UV444-B is used as detector. The wavelength range extends from 250 to 700 nm and the band-pass of the spectrometer (FWHM) is 1 nm, with a triangular shape. A high-pass filter with a cut-off at 300 nm has been set in front of the detector. Wavelength scans below this limit provide accurate measurement of the instrument "optical zero", including the electronic and telemetry offset and possible residual stray-light. Two wavelength intervals, namely 320–340 and 540–660 nm, corresponding to the Huggins and Chappuis absorption bands of ozone, are used to measure the solar absorption during sunset. In addition, differential absorption method as developed for nitric dioxide measurements between 430 and 450 nm can be applied in the Huggins bands for comparison purposes with Bouguer's plot. A more detailed description of the instrumentation is given elsewhere (Simon *et al.*, 1987).

OBSERVATIONS AND DATA ANALYSIS

The balloons were successfully launched by the Centre National d'Etudes Spatiales (CNES) from the French balloon launching facility situated at Aire sur l'Adour (France, 43°42′N, 0°15′W). Table 1 gives the dates, time of observation, type of measurements and wavelength ranges used for ozone profile determinations.

(a) In the case of planet occultations, the reference spectrum (called the extra-atmospheric spectrum of Venus) is recorded at zenith angles smaller than 80°. For the float altitude of this flight, namely 39 km, the optical depth due to absorption of atmospheric constituents is negligible for such zenith angles in the wavelength range of the observations (647–672 nm), and cannot be detected.

The wavelength scale of each spectrum is adjusted to the Hα line at 656.3 nm. A synthetic attenuated spectrum is fitted to each measured transmission during occultation, by using the least-squares method. The ozone absorption cross-sections are those compiled by Ackerman (1971). Attenuation due to Rayleigh scattering is calculated using the extinction cross-section formula published by Nicolet (1984) and a density profile taken from the U.S. Standard Atmosphere, 1976. For the lower altitudes of the line of sight (tangent height below 20 km), the residual optical depth is attributed to stratospheric aerosol assuming no wavelength dependence in the observed wavelength range (647–672 nm). A more complete description of the experimental procedure and data analysis has been published by Rigaud *et al.* (1983).

(b) For solar absorption measurements in the Huggins and the Chappuis bands of ozone, the Bouguer law was applied to determine the total content of ozone along the line of sight. The so-called reference spectrum has been recorded for a solar zenith angle of 82° at about 29 km altitude. Consequently, this measured spectrum has been corrected for residual ozone absorption in the wavelength ranges of interest and also for Rayleigh scattering extinction according to the formula published by Nicolet (1984). The representative ozone vertical profile defined for the MAP/GLOBUS campaign (de La Noë *et al.*, 1987) given in Table 2 and model published by Krueger and Minzner (1976) have been taken for altitudes above the balloon. The absorption cross-sections measured by Bass (1984, private communication) have been

TABLE 2. MAP/GLOBUS REPRESENTATIVE ATMOSPHERE. Average 9–28 September 1983

Altitude (km)	Pressure (mb)	Temperature (K)	Ozone conc. (cm^{-3})
14	150	208	1.11×10^{12}
16	107	211	1.72
18	77	215	2.70
20	56	218	3.86
22	41.5	220	4.61
24	30.2	222	4.67
26	22.5	224	4.43
28	16.3	228	3.50
30	12.0	233	2.65
32	8.7	238	2.03†
34	6.5	243*	1.58†

*Extrapolated values.
†Krueger and Minzner (1976).

used for absorption correction computation up to 340 nm and those published by Griggs (1968) for the Chappuis bands. Ratios between the corrected reference spectrum and spectra recorded for zenith angles larger than 90° have been calculated. Their natural logarithms have been plotted vs absorption cross-section of ozone. Intensity values have been corrected for Rayleigh scattering extinction along the optical path corresponding to each recorded spectrum. The integrated content of ozone is given by the slope of the linear regression in equation:

$$\ln \frac{I_0(\lambda)}{I(\lambda)} - \tau_{RS}(\lambda) = N_{0_3}\sigma_{0_3}(\lambda) \qquad (1)$$

where

λ is the wavelength;
$I_0(\lambda)$ is the solar irradiance above the atmosphere;
$I(\lambda)$ is the solar irradiance after its absorption in the atmosphere;
τ_{RS} is the optical depth due to Rayleigh scattering extinction;
N_{0_3} is the total content of ozone along the line of sight (cm^{-2});
σ_{0_3} is the absorption cross-section of ozone.

On the other hand, the three-wavelength differential absorption method has been used for analysing the same data in the Huggins bands, according to publications of Brewer *et al.* (1973) and Gillis *et al.* (1982). Table 3 defines the maxima and minima in the absorption cross-sections selected between 320 and 340 nm.

(c) The infrared emission measurements at the limb were performed only at 36.5 km altitude because of the failure of the spectrometer cryogenic system. Consequently, the observing time was reduced to 10 min. A more extensive description of the experimental procedure and data evaluation is published by Rippel *et al.* (1987).

TABLE 3. WAVELENGTH TRIPLETS

Triplet	Wavelengths (nm) Min	Max	Min
1	321.250	322.250	323.850
2	323.850	324.950	326.700
3	326.700	327.900	329.475
4	329.475	330.925	332.575
5	332.575	333.750	335.600

DISCUSSION OF RESULTS

Figure 1 shows the integrated concentration along the line of sight for flights 29 and 37 performed respectively on 14 September and 28 September 1983. The comparison between these two observations is not straightforward because, first, of the differences in balloon altitudes during the occultation measurements and, second, observations were made eastward by pointing the planet Venus and, obviously, westward for sunset. The lowest tangent altitude is respectively situated at about 500 km away from the balloon, North of the Observatoire de Haute Provence (OHP), and at about 200 km above Bordeaux. Differences between the two curves on Fig. 1 are explained by a longer optical depth for flight 29 for which the balloon altitude during occultation is 38 km instead of 29 km for flight 37. When optical depths become comparable, the two curves merge.

It should be mentioned that the Bouguer law as described in the previous section is not verified over the wavelength interval 540–660 nm used for solar occultation measurements on 28 September 1983. Deviation from this law increases for lower tangent altitudes of the line of sight and cannot be explained either by errors on ozone cross-sections or Rayleigh scattering corrections. According to the 2 y of observations made after the eruption of El Chichon in April 1982, by means of an airborne lidar at various latitudes and reported by McCormick and Swissler (1987), the aerosol loading in the stratosphere is still very important in September 1983, at latitudes corresponding to the balloon flights (44°N). This fact is also supported by the *Solar Mesosphere Explorer* (*SME*) satellite observations showing an important increase of limb radiances at various wavelengths after April 1982, between 20 and 38 km altitude (Thomas *et al.*, 1986). The residual optical thicknesses have

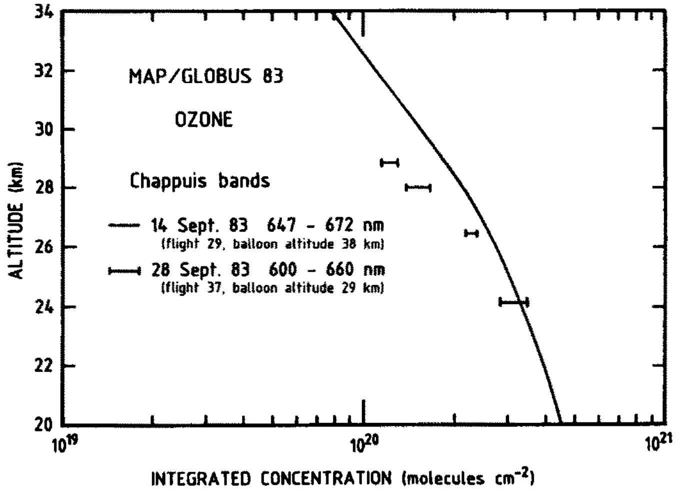

FIG. 1. COMPARISON BETWEEN INTEGRATED CONCENTRATION OF OZONE ALONG THE LINE OF SIGHT DEDUCED FROM ABSORPTION MEASUREMENTS IN THE CHAPPUIS BANDS PERFORMED ON 14 SEPTEMBER 1983 (VENUS OCCULTATION AT 38 km ALTITUDE) AND ON 28 SEPTEMBER 1983 (SOLAR OCCULTATION AT 29 km ALTITUDE).

been evaluated for both balloon observations in the Chappuis bands. The data inversion yields to a peak value of 8×10^{-3} km^{-1} at 16 km altitude. The vertical profiles are systematically higher than those deduced from previous balloon flights performed in 1980 and 1981 (Naudet, 1983). In addition, for solar occultation measurements, the residual optical thickness strongly increases for wavelengths lower than 620 nm (Simon *et al.*, 1986). This fact could explain the higher values for the ozone integrated concentrations deduced from the data around 620 nm and below. A more detailed analysis is presented in Simon *et al.*, (1987).

Figure 2 gives detailed results deduced from the data measured during flight 37 on 28 September 1983, by using the Bouguer plots in the Huggins and Chappuis bands and also the differential absorption method applied to the Huggins bands.

The differential absorption method gives systematically higher values with an average divergence of 20%. This difference is difficult to explain except that when using the Bouguer technique, the integrated ozone concentration is deduced by a linear regression through about 80 data points scattered in the Huggins bands instead of five triplets, namely only 11 different wavelengths, for the differential absorption (see Table 3). This could introduce an additional but not systematic error.

Vertical profiles of ozone were derived by using a non-linear iterative inversion method (Naudet, 1983) for flight 29 and a layer-by-layer linear inversion method for flight 37. Results are summarized in Fig. 3 with the data obtained at 36.5 km from infrared limb emission measurements performed on 23 September 1983. Corresponding numerical values are given in Tables 4, 5 and 6. The uncertainties on inferred ozone concentrations from planet and solar occultation measurements are ±10%.

The error sources for these observations come from the signal-to-noise ratio, absorption cross-section values, ozone concentration above the balloon, air masses along the line of sight, altitude determination and resolution and refraction for altitudes below 20 km. According to Rigaud *et al.* (1985), uncertainties due to the signal-to-noise ratio is less than ±2%. The absorption cross-section values are generally accurate to within ±5%. Possible errors due to their temperature dependence could be excluded (Humphrey and Badger, 1947). The balloon altitude and the geometry of the line of sight are based on pressure measurements performed by sensors integrated in the payload. An altitude accuracy of the order of 500 m can only be estimated and affects the vertical profiles by approximately the same amount. The altitude resolution is very good in the case of planet occultations but decreases from 230 m to 1.4 km during solar occultation for flight 37. The ozone concentration profile above the balloon altitude is based on the aforementioned models, introducing an error of about ±6% mainly for the first ozone concentration value nearest to the balloon altitude. Calculated air mass values for atmospheric layers below the balloon altitude could introduce errors of ±3%. The errors bars on Fig. 3 illustrate the total uncertainty of ±10% except for the solar absorption data in the Chappuis bands which also include the uncertainties coming from the Bouguer law departures.

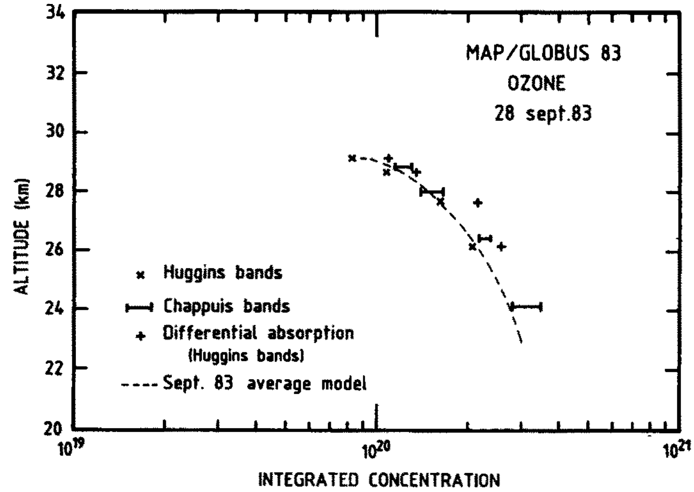

Fig. 2. Comparison between integrated concentration of ozone along the line of sight deduced from absorption measurement in the Huggins and Chappuis bands performed on 28 September 1983 (solar occultation at 29 km altitude).

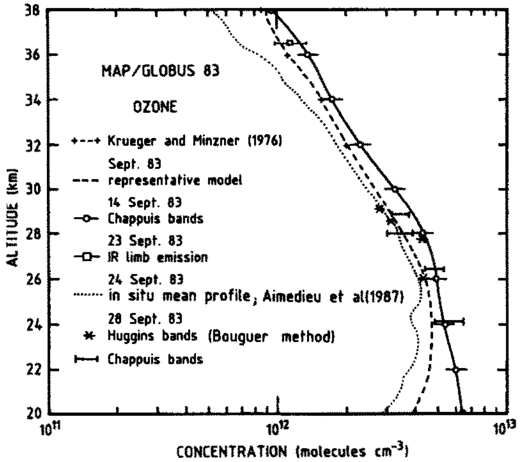

Fig. 3. Comparison between ozone profiles deduced from remote sensing measurements performed during the MAP/GLOBUS campaign in September 1983.
In situ average profile deduced from measurements performed on 24 September 1983 is also shown for discussion purposes. The flight dates refer to the following institutions: 14 Sept. 83, LPCE, Orléans (P. Rigaud); 23 Sept. 83, U. of Wuppertal (D. Offermann); 24 Sept. 83, *in situ* average profile (Aimedieu *et al.*, 1987); 28 Sept. 83, IASB, Bruxelles (P. C. Simon).

At 36.5 km altitude, the only ozone concentration value deduced from infrared remote sensing is in agreement with the ozone profile obtained by planet occultation measurements. This latter method gives systematically higher values than the representative model of ozone based on *in situ* soundings made from OHP and Biscarosse (de La Noë *et al.*, 1987). Differences are between 10 and 20%, except below 24 km altitude where day-to-day variations due to dynamical changes in the lower stratosphere should be taken into account. In addition, aerosol absorption becomes significantly more important, also leading to larger discrepancies. Solar occultation measurements in the Huggins bands give ozone concentrations in agreement over 3 km in altitude with *in situ* soundings except at one altitude, namely 27.7 km. The same observations in the Chappuis bands at 25.5 and 24.2 km altitude yield values very close to those obtained from planet occultation. This fact supports the assumption of an important contribution of aerosol in September 1983, even around 660 nm, to the optical depth measured below 28 km of altitude.

Systematic discrepancies with *in situ* measurements at higher altitudes reported by Aimedieu *et al.* (1987) are difficult to explain in terms of aerosol loading of the stratosphere. On the other hand, direct influence of transport is weak in such a region (above 30 km and at 44°N) where the ozone budget is chemically controlled according to the 2-D model of Garcia and Solomon (1985). However, the comparison of other trace species observed during the same campaign shows similar discrepancies. These have been tentatively interpreted in terms of wavelike dynamical disturbances (Offermann *et al.*, 1987).

In conclusion, ozone measurements by occultation techniques are very precise but the final accuracy depends upon a good knowledge of all atmospheric absorptions along the line of sight. Such a knowledge can be reached by extending the wavelength range of observation out of the absorption bands of ozone in

TABLE 4. GONDOLA G5; 14 SEPTEMBER 1983; NIGHT

Pressure (mb)	Altitude (km)	O_3 conc. ($cm^{-3} \times 10^{-12}$)	ΔO_3 ($\times 10^{-12}$)	O_3 mixing ratio (ppmV)
75.7	18	4.94	±0.50	1.95
55.3	20	6.33	±0.42	3.43
40.5	22	5.97	±0.33	4.45
29.7	24	5.34	±0.27	5.47
21.9	26	4.92	±0.22	6.91
16.2	28	4.28	±0.14	8.21
12.0	30	3.25	±0.11	8.50
8.9	32	2.31	±0.08	8.20
6.6	34	1.75	±0.07	8.50
5.0	36	1.37	±0.06	9.10
3.8	38	0.95	±0.09	8.56

Errors related to altitude uncertainties are not included.

TABLE 5. GONDOLA G1; 23 SEPTEMBER 1983; 4:30 U.T.

Pressure (mb)	Altitude (km)	O_3 conc. ($cm^3 \times 10^{-12}$)	Accuracy	O_3 mixing ratio (ppmV)
4.85	36.5	1.25	±15%	8.50

TABLE 6. GONDOLA G2b; 28 SEPTEMBER 1983; SUNSET

Pressure (mb)	Altitude (km)		O_3 conc. ($cm^{-3} \times 10^{-12}$)		Mixing ratio (ppmV)	
	Huggins	Chappuis	Huggins	Chappuis	Huggins	Chappuis
13.8	29.1		2.73 ± 10%		6.31	
14.2		28.9		3.17–3.87		7.10–8.67
14.7	28.7		3.13 ± 10%		6.76	
16.1		28.1		3.00–3.96		5.88–7.76
17.2	27.7		4.28 ± 10%		7.81	
20.7		26.5		4.42–5.40		6.63–8.10
22.2	26.1		4.27 ± 10%		5.96	
29.5		24.2		4.82–6.50		5.01–6.76

order to accurately determine optical depths of the stratosphere at various wavelengths and to reveal atmospheric extinction by other species like aerosols. A good altitude resolution is needed in order to resolve possible layer structure in aerosol profiles.

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