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STRATOSPHERIC OZONE MEASUREMENTS BY SOLAR ULTRAVIOLET ABSORPTION

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Abstract—Vertical distributions of ozone in the stratosphere have been determined by means of u.v. differential absorption technique, using the Sun as a source. The filter radiometer and the data processing are described. Results obtained between altitudes of 22 and 32 km during the Intercomparison Ozone Campaign held in France in June 1981 are presented and discussed.

INTRODUCTION

Many experimental techniques have been developed for several years to determine ozone vertical distribution between 20 and 60 km altitude. Among these, u.v. absorption measurements by means of filter radiometers have been extensively performed by balloon and rockets, using the Sun as a source (see for instance, Kulcke and Paetzold, 1957; Krueger, 1973).

The principle of measurement is based on the difference in the absorption by ozone of solar radiation, at several wavelengths. They are selected according to the optical depth to be measured in the chosen altitude range.

Using the Beer–Lambert's law, ratios between irradiances measured at two wavelengths yield the ozone column content above the sensor. Vertical profiles are deduced by differentiation. This method does not require the knowledge of the absolute value of solar extraterrestrial irradiance but only the spectral bandpass of all of the optical components, the relative quantum yield of the detectors, the relative distribution of solar irradiance and the absorption cross-section of ozone within the filter band passes.

Extinction by Rayleigh and aerosol scattering also has to be taken into account.

A description of such a measurement principle has been published in Kobayashi *et al.* (1966).

This u.v. differential absorption technique can be applied between altitudes of 20 and 40 km using large stratospheric balloons. Observations can take place during the ascent and/or during descent slowed down either by parachute or by a controlled leak at the balloon apex.

The purpose of this paper is to present and to discuss ozone profiles obtained with u.v. radiometer developed and built at the "Institut d'Aéronomie Spatiale de Belgique", and launched during the intercomparison ozone campaign held in France in June 1981 (Chanin, 1983).

INSTRUMENTATION AND CALIBRATION

The optical part of the u.v. filter radiometer is shown in Fig. 1. Two channels are selected by means of interference filters for the differential absorption measurement. The first wavelength, called the active channel, is chosen with respect to the ozone content to be measured in a given altitude range. The second, called the reference channel, is set at 340 nm where ozone absorption is negligible above 20 km altitude (optical depth less than 0.01). The sunlight is diffused into the instrument by a ground quartz window. A beam-splitter is inserted to reflect selectively about 90% of the sunlight in the wavelength range of the active channel and to transmit about 85% in the reference wavelength range. A Schott filter UG 11 is added only in the active channel in order to improve the blocking (better than 10^{-4}) of the interference filter in the visible and the near i.r. Two silicon photodiodes from EG & G, type UV 444B are used as detectors. The spectral characteristics of the filters (transmittivity) and of the beam-splitter (transmittivity and reflectivity) are measured by means of a Perkin–Elmer 330 double beam spectrophotometer with an accuracy of the order of 1% for transmissions greater than 0.1 and 7% for transmissions lower than 0.001. The detectability limit is 10^{-4}. The responsitivity of the detectors is taken from a typical curve given by the manufacturer. Only relative values are needed for all these parameters. Figure 2 gives one typical example of the convolution of the filter and beam-splitter transmissions with the detector sensitivity. Table 1 summarizes the filter characteristics used for the flight of 19 June 1981.

Paper relating to the Intercomparison Ozone Campaign, France, 1981.

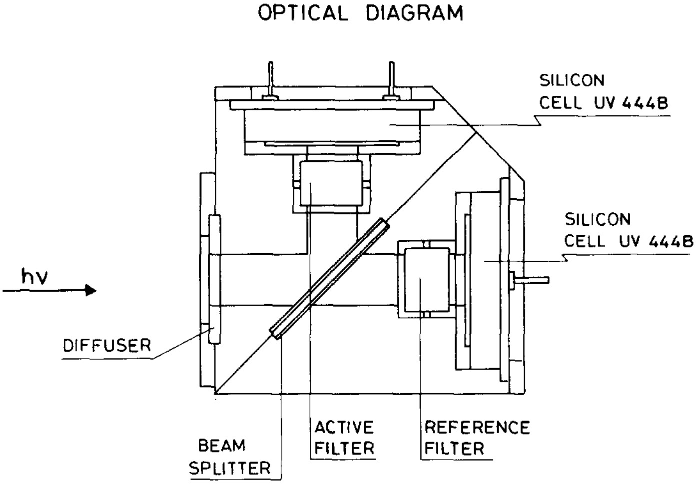

FIG. 1. OPTICAL DIAGRAM OF THE U.V. RADIOMETER.

The angular response of the instrument has been measured carefully in two perpendicular planes, corresponding to rotation in the horizontal and vertical planes of the gondola. The field of view of the ozone sensors is ±25° at FWHM. An asymmetry in the sensitivity of the two channels was detected and corrections to the measured signals have been applied on the basis of laboratory tests.

The output current of the detectors is converted by means of electrometers into a voltage. A heating system is included in order to maintain the detectors and the filters at a constant temperature during the flight, namely 25°C, close to the laboratory temperature at which calibrations and tests were performed. A block diagram of the electronics is presented in Fig. 3.

Absolute calibration has been performed before and after flight. Two sets of transfer standard sources, namely deuterium and quartz halogen lamps, have been used for that purpose. The spectral irradiance scale is traceable to the National Bureau of Standards radiometric scale. The uncertainty of the quartz halogen lamps giving the best signal to noise ratio, is less than 3% in the near u.v.

Two similar sondes were launched from Gap on 19

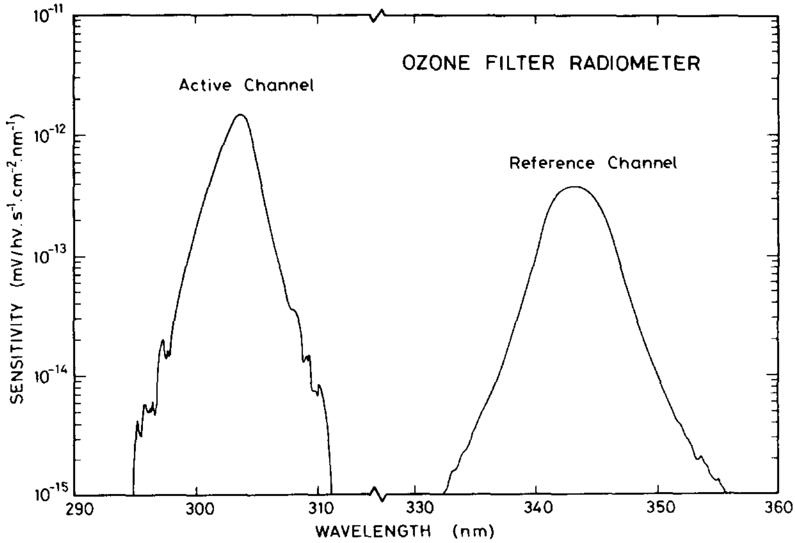

FIG. 2. TYPICAL SENSITIVITY OF THE ACTIVE AND REFERENCE CHANNELS OF THE U.V. RADIOMETER VS WAVELENGTH.

TABLE 1. SPECTRAL DATA OF THE FILTERS USED IN THE OPTICAL OZONESONDES LAUNCHED ON 19 JUNE 1982

	Peak wavelength (nm)	FWHM (nm)
Sonde 1		
active channel	303.7	2.7
reference channel	343.1	5.0
Sonde 2		
active channel	303.1	2.8
reference channel	342.0	5.2

June 1981. They were placed on the top of the gondola as described by Chanin (1983). The normal to the plane of the front diffusers made an angle of 53° with respect to the horizontal plane. This angular position was set in order to adequately cover the solar zenith angles encountered during the flight, after the first ascent of the balloon. The gondola is roughly stabilized by a sail. Nevertheless, it is periodically animated by some oscillations and rotations in the horizontal plane. Its vertical axis does not vary by more than 1°. These movements specially occurred during the balloon vertical excursions. Therefore, the measured signals look like those reported in Fig. 4. They become negligible when the Sun is outside the instrument field of view.

A complete description of the flight characteristics and of ancillary data is given by Chanin (1983).

DATA REDUCTION

Penetration of u.v. solar irradiance into the stratosphere is driven by the Beer–Lambert's law. Assuming that the attenuation in the 250–350 nm wavelength interval is only due to ozone absorption and Rayleigh scattering above 20 km altitude, the solar irradiance I which reaches the altitude z is given by:

$$I(\lambda, z, \chi) = I_0(\lambda)\exp-\tau(\lambda, z)\sec\chi \tag{1}$$

with the optical depth τ defined as follows in this particular case:

$$\tau(\lambda, z) = N_{O_3}(z)\sigma_{O_3}(\lambda, T)+N_{Air}(z)\sigma_R(\lambda, T) \tag{2}$$

where $I_0(\lambda)$, is the solar extraterrestrial irradiance, χ, the zenith angle, $\sigma_{O_3}(\lambda, T)$, the absorption cross-section of ozone, $\sigma_R(\lambda, T)$, the extinction cross-section for Rayleigh scattering, N_{O_3}, the ozone column density, N_{Air}, the air column density, λ, the wavelength, and T, the absolute temperature.

Expression 1 is only valid for narrow wavelength intervals (monochromomatic light).

The ozone column content can be deduced from stratospheric irradiance measurements by using equations 1 and 2. In order to reduce the errors linked to the inaccuracies in our knowledge of the absolute value of the solar extraterrestrial irradiances, to the instrument spectral change during observations and to other possible absorbing layers like aerosols, a differential absorption method is used. In addition, the correction factors due to the angular response of the filter radiometer are minimized by using the ratio between the signal measured through the active channel and the reference channel. As both channels are defined by interference filters having a non-negligible spectral-width (see Table 1), the measured output signal i from one detector corresponds to the integration over the spectral bandpass of one channel of all spectral-dependent parameters:

$$i = C\Sigma I(\lambda, z, \chi)R(\lambda)\,\Delta\lambda \tag{3}$$

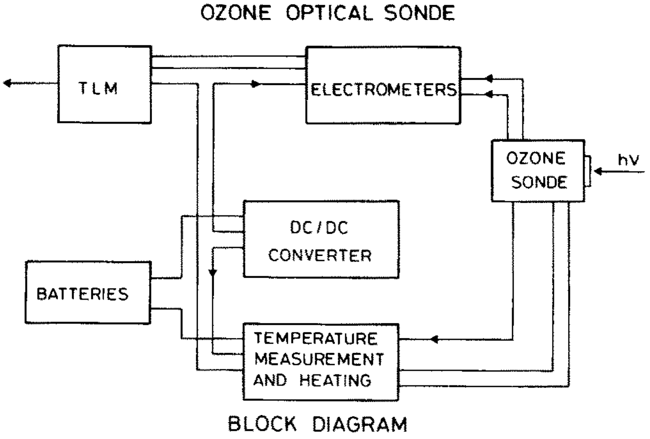

FIG. 3. BLOCK DIAGRAM OF THE U.V. RADIOMETER.

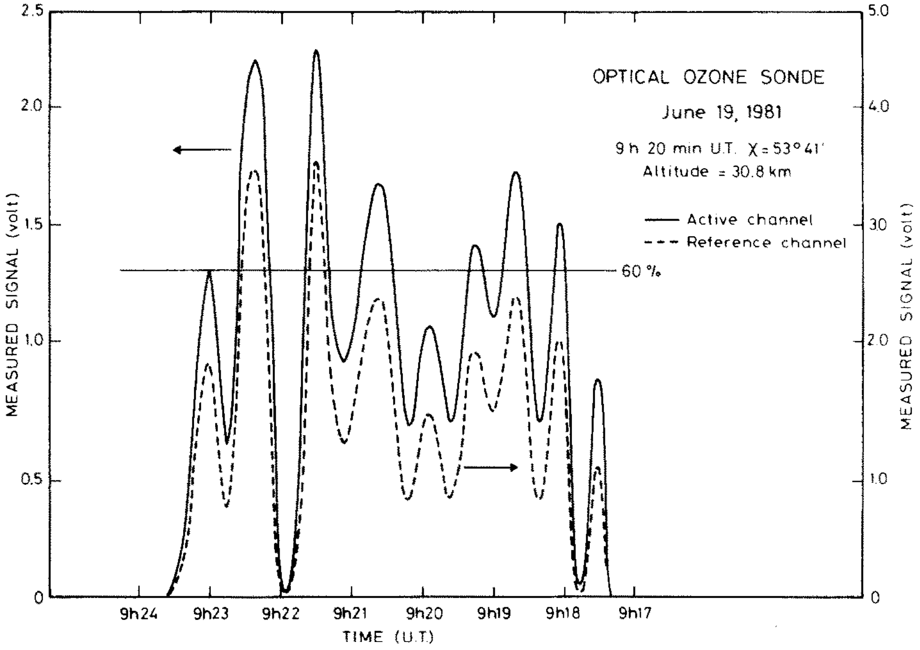

FIG. 4. EXAMPLE OF MEASURED SIGNALS THROUGH THE ACTIVE (SOLID CURVE) AND THE REFERENCE (DASHED CURVE) CHANNELS AROUND 09.20 U.T.

The line labelled 60% represents the lower limit, for the active channel, of the data selected, in this case, to deduce the ozone content. The variations in the measured signal correspond to gondola rotations.

where C denotes the calibration factor determined in the laboratory for each channel; $R(\lambda)$, the convolution between transmission of all optical components and detector quantum efficiency at a given wavelength; $I(\lambda, z, \chi)$, the incoming solar irradiance defined by equation 1.

The wavelength interval for integration ($\Delta\lambda$) has been fixed to 0.1 nm.

The signal from the two channels of each ozone sensor was sampled every half second. Pressure measurement and magnetometer signals provided by the gondola were selected for the same time intervals. The trajectory of the balloon was determined from radar tracing data and used to calculate the solar zenith angle with respect to the time. The vertical distribution of temperature was taken from the data provided by the ECC sonde launched on 19 June 1981 at 07.58 U.T. from Gap (France). Pressure measurements have been converted into altitude following the procedure described by Robbins (1983) and applied to all of the data obtained during the same balloon flight.

Before calculating the ratios between the active and the reference channels, corrections for the angular response determined in the laboratory were applied because of the non-symmetrical response of the radiometer with respect to the angle of incidence of the sunlight. Only data for angular deviations in azimuth of less than ±15° and for zenith angle less than 60° have been considered in order to eliminate data for which the correction factor was too large (greater than 1.66). In addition, it has been assumed that the filter spectral characteristics are not affected by the incidence angle of the sunlight by using a diffuser in front of the instrument.

For each channel, equation 3 is computed, introducing data coming from laboratory measurements, namely the calibration factor C and the instrument responsibility $R(\lambda)$. The solar extraterrestrial irradiance values were taken from Simon *et al.* (1982) and the absorption cross-section from Vigroux (1953). Temperature coefficients have been calculated by interpolation through the Vigroux's data for the temperature corresponding to each altitude of data. Air column density and Rayleigh scattering extinction cross-section have been calculated using standard techniques from the pressure measurement and the adopted temperature profile.

The ratios between the 304 nm and the 340 nm channels were first calculated for a given ozone column density model. By interation, this model is adjusted in order to fit the measured ratios to better than 0.1%.

Several thousand data points distributed along the balloon vertical excursions have been reduced, yielding ozone column content. These latter data are smoothed by fitting a 5th order polynomial function. The derivative of this function, for each profile, yields an ozone concentration in the same altitude range.

RESULTS AND DISCUSSION

The flight profile from 19 June 1981 has been described by Chanin (1983). Data were not available for the first ascent of the balloon during which the zenith angles were too large. Observations were reduced from 08.00 U.T., corresponding to a zenith angle of 49°27′. The first descent and the second ascent provided reliable data from which ozone profiles were deduced according to the method described in the preceding section. The optical depth during these two vertical excursions varied between 0.24 and 1.77 for the active channel. During the last descent, the temperature regulation of the filter radiometer failed. Consequently, the temperature of filters and detectors increased and data are not sufficiently reliable to give accurate results. The accuracy of the measurements is mainly limited by the uncertainties on the ozone absorption cross-section (±5%). Taking into account the errors introduced by the measurement technique in the stratosphere, the total estimated uncertainty increased up to ±7–±10%.

Table 2 and Fig. 5 gives the ozone concentrations from 22 to 32 km of altitude. Differences between the two profiles do not exceed 7.6%. The agreement is good between 24 and 29 km. It should be noted that the optical depth is close to 1 around 26 km for the two vertical profiles.

TABLE 2. OZONE CONCENTRATION FOR DESCENT 1(D1), ASCENT S(A2), MEAN AND RATIO

Altitude (km)	$[O_3]$ cm^{-3} D1	$[O_3]$ cm^{-3} A2	$[O_3]$ cm^{-3} mean	D1/A2
22.0	5.81 10^{12}	5.60 10^{12}	5.71 10^{12}	1.037
22.5	5.99	5.63	5.81	1.064
23.0	6.07	5.64	5.86	1.076
23.5	6.00	5.63	5.82	1.066
24.0	5.87	5.61	5.74	1.046
24.5	5.70	5.58	5.64	1.022
25.0	5.53	5.54	5.54	0.998
25.5	5.36	5.47	5.42	0.980
26.0	5.20	5.35	5.28	0.972
26.5	5.05	5.21	5.13	0.969
27.0	4.91	5.06	4.99	0.970
27.5	4.78	4.89	4.84	0.978
28.0	4.66	4.71	4.69	0.989
28.5	4.53	4.52	4.53	1.002
29.0	4.40	4.32	4.36	1.019
29.5	4.28	4.12	4.20	1.039
30.0	4.11	3.92	4.02	1.048
30.5	3.92	3.72	3.82	1.054
31.0	3.67	3.52	3.60	1.043
31.5	—	3.34	—	—
32.0	—	3.17	—	—

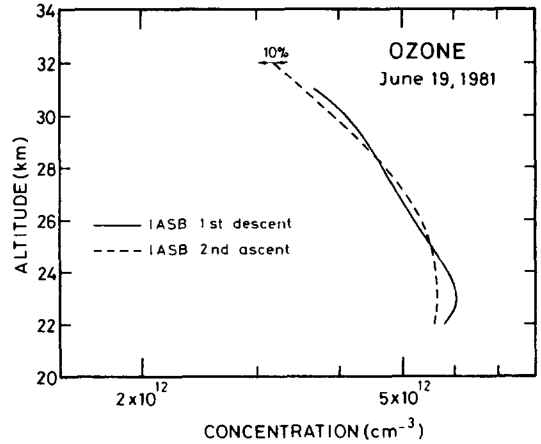

FIG. 5. OZONE CONCENTRATIONS DEDUCED FROM SOLAR U.V. ABSORPTION BETWEEN ALTITUDES OF 22 AND 32 km DURING THE FIRST DESCENT AND THE SECOND ASCENT OF THE GONDOLA ON 19 JUNE 1981.

Some parameters have been modified in the calculation in order to check the sensitivity of this reduction method. It appears that the choice of the detector quantum yield spectral distribution is not very critical. Using the same values as in Krueger *et al.* (1983), no difference in concentration larger than 2% has been found except above 31 km of altitude (3.5% lower). A more critical point is the spectral transmission in the wings of the active filter. Increasing by 10% all the transmission values lower than 0.01, the ozone concentration is increased by 9% at 22 km and decreased by 3% at 31 km. No significant changes (less than 2%) were found from 24 to 30 km of altitude where the agreement between the two profiles is good.

The weakness of these observations could be consequently the accuracy needed to measure the filter spectral characteristics for transmissions lower than 0.01 but also the non-symmetrical response of the instrument with respect of the incidence angle of sunlight. This latter problem can be solved by redesigning the optics.

On the other hand, comparison with spectral observations at lower spectral bandpasses (FWHM less than 1 nm) using a u.v. spectrometer pointed towards the Sun during the vertical excursions of the payload could certainly answer the question of the reliability of the filter radiometer measurements over a large altitude range. It could be possible that the measurement by differential absorption using one active wavelength is only reliable within 5 km of altitude, taking into account the current techniques used in the laboratory to characterize the instrument.

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