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OZONE PROFILE INTERCOMPARISON BASED ON SIMULTANEOUS OBSERVATIONS BETWEEN 20 AND 40 KM

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Abstract—The vertical distribution of stratospheric ozone has been simultaneously measured by means of five different instruments carried on the same balloon payload. The launches were performed from Gap during the intercomparison campaign conducted in June 1981 in southern France. Data obtained between altitudes of 20 and 40 km are compared and discussed. Vertical profiles deduced from Electrochemical Concentration Cell sondes launched from the same location by small balloons and from short Umkehr measurements made at Mt Chiran (France) are also included in this comparison. Systematic differences of the order of 20% between ozone profiles deduced from solar u.v. absorption and *in situ* techniques are found.

INTRODUCTION

Many measurement techniques have been used over the past several years to determine ozone profiles in the stratosphere. They include standard techniques for routine observations, such as the ground-based Umkehr observations for solar elevation angles of less than 30° and the small balloon techniques based on two types of electrochemical instruments, the Brewer–Mast (BM) and the Electrochemical Concentration Cell (ECC) type.

Other techniques have been used with large research balloon payloads. They are based on chemiluminescent, u.v. absorption and mass spectrometer *in situ* measurements or on u.v. absorption measurement by means of filter radiometers and solar spectrometers using the Sun as a source.

These observing programs have been developed for the study of the long-term variation in stratospheric ozone and for the validation of satellite ozone profile measurements. Consequently, the various experimental techniques with their own uncertainties should be compared within a specific campaign in order to understand differences between measurements, to improve the reliability of the existing data and to insure the continuity in future observations.

The purpose of this paper is to present the intercomparison results of ozone profiles deduced from simultaneous measurements obtained during the intercomparison campaign conducted in June 1981 in southern France. In this paper, only data obtained between 20 and 40 km will be discussed. Intercomparisons at lower and higher altitudes are discussed elsewhere (Megie and Pelon, 1983; Monnanteuil *et al.*, 1983).

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

CURRENT CONCLUSIONS OF PREVIOUS INTERCOMPARISON CAMPAIGNS

Until now, only a few intercomparison campaigns of ozone profile measurements above 20 km of altitude have been performed. The first campaign took place during the summer of 1953 in Arosa (Switzerland). Data obtained from four different techniques are published in Brewer *et al.* (1960). Balloon measurements, namely the Brewer's electrochemical sonde and the Paetzold's opticale sonde, provided vertical distributions below an altitude of 25–30 km. More recently, intercomparison results between 20 and 40 km based on the Dasibi instrument from NASA JSC and the mass spectrometer from the University of Minnesota have been published by Mauersberger *et al.* (1981). Other results obtained from balloon campaigns, namely the last flight of the Stratcom series, the SABE 2 (Solar Absorption Balloon Experiment) flight and the LIP (LIMS Instrument Package) flight series, and from the rocket campaign conducted jointly by WMO, FAA and NASA in 1979, have been reported in the NASA–WMO report on the stratosphere (WMO, 1982). From these intercomparisons, systematic differences have been found between several experimental techniques.

From both the Stratcom and the SABE 2 flights, u.v. radiometers gave larger ozone density values than those derived from the various *in situ* observations, 30%

for instance at 35 km during the SABE 2 flight. It should be pointed out that, since atmospheric pressure is measured independently by each ozone sensor, differences observed between some instruments could reflect error in both ozone and pressure measurements.

Data obtained in September 1980 from a mass spectrometer and Dasibi instrument (Mauersberger *et al.*, 1981) agreed over the whole altitude range of measurement. Lower density values above 35 km were obtained previously in 1978 with the u.v. absorption instrument. This resulted from the need to correct for a pressure difference between the absorption cell and the outside ambient pressure measurement as mentioned by Mauersberger *et al.* (1981).

Umkehr-derived profiles were generally lower during the SABE 2 flight than the values from the u.v. radiometers for altitudes above 31 km but this method also gives lower values than generally observed around the main maximum by other routine ozone sounding techniques (WMO, 1982).

AVAILABLE DATA

Table 1 lists the different ozone sensors that provided data for the intercomparison of ozone profiles between 20 and 40 km. During the first flight on 19 June 1981, data were obtained from four vertical excursions of the gondola as described by Chanin (1983). The first ascent started just before dawn in order to allow the comparison with the Lidar observations. Sunrise occurred when the balloon reached an altitude of about 15 km. The two u.v. radiometers were not designed to provide reliable observations for large zenith angles and consequently no profiles were reported from these two instruments for the first ascent. On the other hand, because of possible degradation of the reactive solution used in the Brewer–Mast sonde on board the large balloon, only data obtained during the first ascent have been taken into account. In addition, the Dasibi instrument unfortunately failed after reaching the first ceiling and only provided measurements during that first ascent. Consequently, intercomparison between *in situ* and u.v. radiometer measurements is not straightforward. Nevertheless, assuming that over the longitudinal range covered during this flight there is no significant change in the ozone profile above 22 km—this seems to be confirmed by the 3 profiles obtained by Krueger and Simeth (1983) and by Aimedieu (1983)—the intercomparison of all experimental techniques will be discussed below. Umkehr and ECC data were not obtained from the same location but these soundings were made at sufficiently close distances to be taken into account here. During the second flight on 26 June 1981, only two vertical distributions up to 40 km of altitude are available. The Brewer–Mast sonde failed above 25 km of altitude and the optical sonde data were obtained only during the sunlit part of the flight, especially during the payload descent under parachute. These latter observations are however more difficult to analyze and the preliminary results will not be discussed in this work.

RESULTS AND DISCUSSION

The conditions of observation during this intercomparison campaign are described extensively in Chanin (1983).

The experimental techniques and results for each instrument are published separately in the same volume (Aimedieu, 1983; Krueger and Simeth, 1983; Robbins, 1983; Roeland *et al.*, 1983; Marché *et al.*, 1983). From the profile intercomparison below 25 km, published by

TABLE 1. AVAILABLE DATA FOR OZONE PROFILES BETWEEN 20 AND 40 km

Sensor	Location	Flight 9 June 1981				Flight 26 June 1981
		1st a*	1st d†	2nd a*	2nd d†	
Short Umkehr (Univ. Reims)	Ground-based Mt Chiran	×				
Chemilumiscent sonde (Service d'Aéronomie)	Large balloon	×	×	×	×	×
Brewer–Mast sonde (EERM)	Large balloon	×				
Dasibi (NASA–JSC)	Large balloon	×				×
U.v. solar radiometer (IASB)	Large balloon		×	×		
U.v. solar radiometer (NASA–GSFC)	Large balloon		×	×	×	
ECC sondes (NASA–WFC)	Small balloons Gap	×	×		×	

a* = ascent
d† = descent

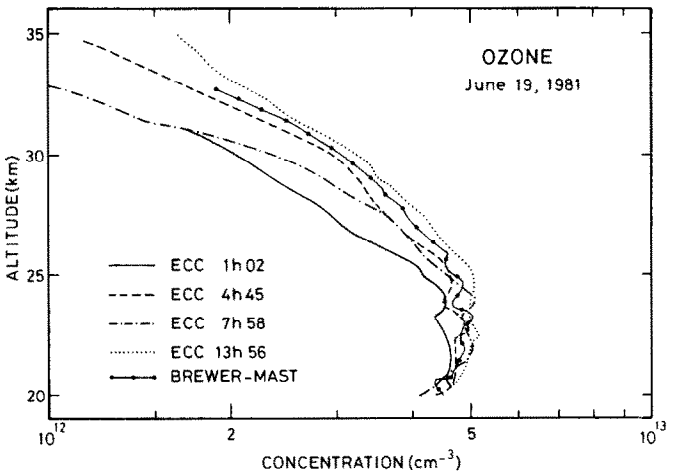

FIG. 1. OZONE VERTICAL DISTRIBUTION FOR ALTITUDES BETWEEN 20 AND 35 km AS MEASURED ON 19 JUNE 1981, BY FOUR ECC SONDES.
The Brewer–Mast data are also given for comparison purposes.

Mégie and Pelon (1983), it appears clear that the electrochemical instruments, namely the ECC sondes, provide reliable data up to 24 km. Figure 1 shows that the discrepancies between all these soundings increase with the altitude, reaching a factor 2 at 33 km. Consequently ECC soundings are only valid for altitudes below 25 km.

For the first flight on 19 June 1982, the ozone profiles obtained during the first ascent and reported by Aimedieu (1983) and Robbins (1983) disagree by about 15% near the main ozone maximum around 23 km of altitude (Fig. 2). The agreement above 27 km is better. In addition, the detailed structures of the ozone profile are not always well correlated. Table 2 lists the vertical distribution of ozone obtained with the Brewer–Mast sonde on board the same gondola. Figure 2 shows that these data are in good agreement with those of Robbins (1983) even above 25 km. Unfortunately it is not possible, with only one comparison, to conclude on the reliability of the Brewer–Mast sondes above 25 km. Further simultaneous measurements, with several sondes, are needed to confirm the result obtained during this campaign. The Umkehr profile obtained from Mt Chiran (Marché *et al.*, 1983) and reported on Fig. 2 gives lower values around 30 km and higher values below 22 km. The cross-over is situated in layer number 6, between 23.6 and 28.4 km. In addition, the altitude of the main maximum, situated in layer number 5, seems too low when compared to the other three profiles. Between 10 and 38 km, the estimated error in the short Umkehr measurements is between ± 5 and $\pm 9\%$. Robbins (1983) claims a total rms uncertainty of $\pm 1.6\%$ in the same altitude range and Aimedieu (1983) an accuracy varying from $\pm 8\%$ at 16 km to $\pm 15\%$ at 38 km.

The error budget analysis made by Robbins (1983) shows that, in the case of *in situ* u.v. absorption measurements, the uncertainty on the ozone absorption cross-section is the main contribution to the total rms uncertainty. In the case of the chemiluminescent measurements, the error is mainly due to the value of the air-reactant mixture flow and to the laboratory calibration performed by means of a Dasibi instrument. Considering the quoted accuracies for both techniques, it appears that a large amount of the data obtained simultaneously by *in situ* instruments are consistent. Nevertheless, discrepancies as large as 15% around the main ozone maximum are significant. They have to be explained by further analysis of the data reduction techniques and the experimental problems associated with measurements in the stratosphere and in the laboratory. On the other hand, comparison with the Umkehr results is in contradiction with the previous conclusions reported in the NASA–WMO report on the stratosphere (WMO, 1982), claiming that *in situ* techniques yield in general higher concentration near the ozone maximum than the Umkehr observations. As discussed by Mégie and Pelon (1983), the presence of aerosol particles due to high volcanic activity since May 1980 could explain the higher ozone concentrations deduced by the Umkehr observations in layers 4 and 5, situated around 20 km.

Figures 3 and 4 show the ozone profiles obtained respectively from Krueger and Simeth (1983) and by Roeland *et al.* (1983). In both figures, the ozone vertical distribution measured with the *in situ* u.v. absorption

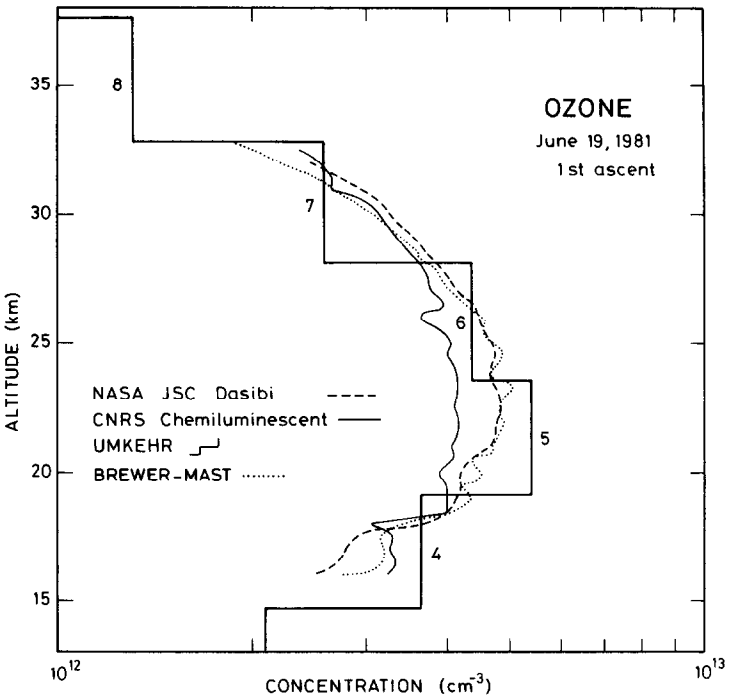

FIG. 2. OZONE VERTICAL DISTRIBUTION FOR ALTITUDES BETWEEN 15 AND 40 km AS MEASURED ON 19 JUNE 1981, BY THE THREE *in situ* INSTRUMENTS.
The short Umkehr profile obtained at Mt Chiran is also given for comparison purposes.

TABLE 2. OZONE VERTICAL CONCENTRATION (molecules cm^{-3}) VS ALTITUDE AS MEASURED BY THE FOUR ECC AND THE BREWER MAST SONDES ON 19 JUNE 1981

Altitude (km)	Brewer–Mast 1st ascent	ECC 01.02	ECC 04.45	ECC 07.58	ECC 13.56
20.0	4.55×10^{12}	4.31×10^{12}		4.11×10^{12}	4.35×10^{12}
20.5	4.36	4.58		4.42	4.69
21.0	4.71	4.58		4.50	4.87
21.5	4.80	4.61	4.68×10^{12}	4.98	4.93
22.0	4.88	4.57	4.73	5.04	5.02
22.5	4.83	4.54	4.79	4.95	5.16
23.0	4.93	4.50	5.00	4.88	5.04
23.5	4.97	4.48	4.71	4.86	4.98
24.0	4.72	4.54	4.46	5.08	5.02
24.5	4.91	4.39	4.66	4.82	5.07
25.0	4.68	4.17	4.60	4.54	5.05
25.5	4.58	4.00	4.51	4.30	4.92
26.0	4.53	3.62	4.27	4.10	4.64
26.5	4.28	3.41	3.99	3.96	4.44
27.0	4.08	3.07	3.77	3.79	4.30
27.5	3.97	2.90	3.62	3.57	4.16
28.0	3.78	2.78	3.46	3.32	3.94
28.5	3.61	2.57	3.37	3.07	3.76
29.0	3.53	2.40	3.25	2.88	3.51
29.5	3.23	2.22	3.11	2.60	3.43
30.0	3.08	2.05	2.96	2.33	3.26
30.5	2.84	1.89	2.72	2.07	3.01
31.0	2.67	1.73	2.44	1.76	2.78
31.5	2.46		2.21	1.44	2.53
32.0	2.20		2.01	1.27	2.40

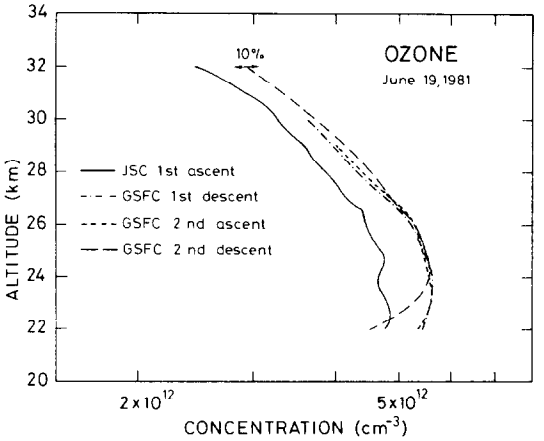

FIG. 3. OZONE VERTICAL DISTRIBUTION FOR ALTITUDES BETWEEN 22 AND 32 km AS MEASURED ON 19 JUNE 1981 BY THE NASA–GSFC U.V. FILTER RADIOMETER (KRUEGER AND SIMETH, 1983). The profile obtained by Robbins (1983) is given for comparison purposes.

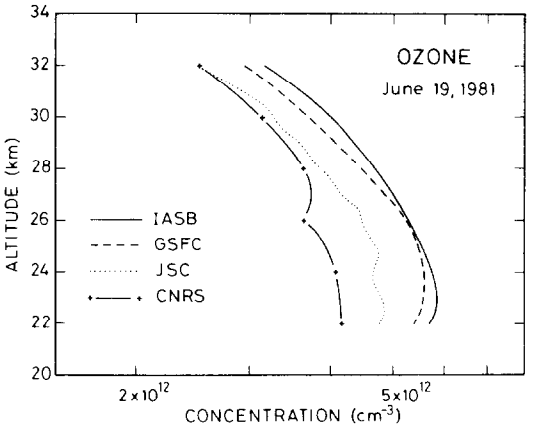

FIG. 5. AVERAGE OZONE VERTICAL DISTRIBUTION (22–32 km) AS MEASURED ON 19 JUNE 1981 BY THE TWO *in situ* INSTRUMENTS (JSC AND CNRS) AND THE TWO SOLAR U.V. ABSORPTION RADIOMETERS (IASB AND GSFC). Data below 24 km obtained by Krueger and Simeth (1983) during the 2nd descent are not taken into account.

instrument (Robbins, 1983) is reported for comparison purposes. The average profiles based on each instrument are shown in Fig. 5. Differences between the two *in situ* techniques have already been discussed. Taking as a reference the JSC Dasibi vertical distribution, it shows clearly that the two u.v. radiometers give ozone concentrations systematically higher by about 20%. The differences between the two u.v. radiometers are given in Table 3. They are generally less than 10%, that means within their uncertainties. The best agreement is found for altitudes corresponding to an optical thickness close to 1 as illustrated by Fig. 6 on which the optical depth encountered during the balloon vertical excursions are reported vs time. Further analysis and new measurements are needed to explain such disagreements between ozone profiles obtained by similar techniques during the same flight, but the estimated uncertainty for filter radiometers is mainly due to the error in the absorption cross-section ($\pm 5\%$). This method could also introduce larger errors in both ends of the altitude range because the data correspond to lower and higher optical depth (cf. Fig. 6). In addition, the u.v. differential absorption technique seems very sensitive at lower and higher altitudes to the measured values of the instrument spectral parameters as described by Roeland *et al.* (1983). It should be noted that the 4% of difference between results deduced from the two active channels used by Krueger and Simeth (1983) emphasis the need of intercomparison at several wavelengths.

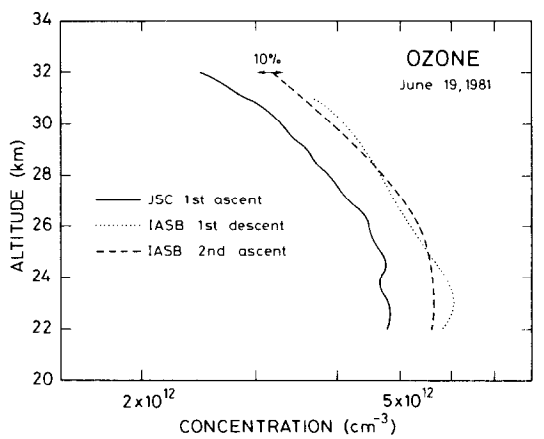

FIG. 4. OZONE VERTICAL DISTRIBUTION FOR ALTITUDES BETWEEN 22 AND 32 km AS MEASURED ON 19 JUNE 1981 BY THE IASB U.V. FILTER RADIOMETER (ROELAND *et al.*, 1983). The profile obtained by Robbins (1983) is given for comparison purposes.

For the second flight on 26 June 1981, the only available data are those obtained by Aimedieu (1983) and Robbins (1983) both using *in situ* techniques. They are represented in Fig. 7. The agreement is good, the maximum discrepancy being 10% around 26 km (data below 20 km are not considered here). Such a difference is not significant taking into account the inaccuracies in both measurements. Comparison with the data obtained during the first flight on 19 June 1981, indicates that the day-to-day variations above 25 km of altitude are not larger than the experimental uncertainties during this campaign.

In conclusion, the basic disagreement between *in situ*

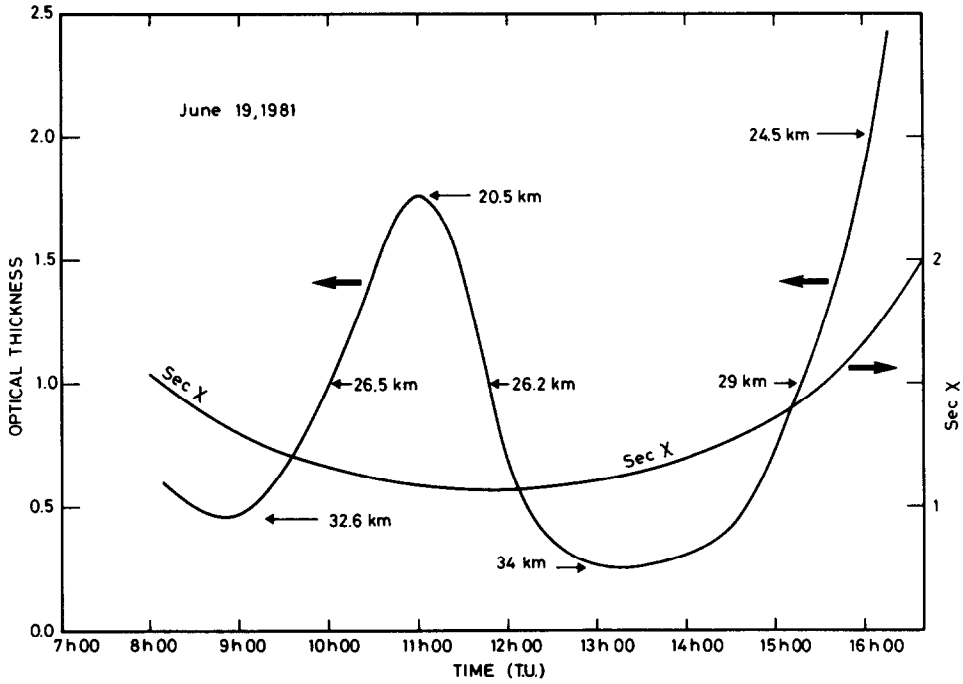

FIG. 6. OPTICAL THICKNESS OBSERVED DURING THE FLIGHT ON 19 JUNE 1981, VS TIME.
The secant of the solar zenith angle and typical altitudes are also indicated.

and solar u.v. filter radiometer measurements is confirmed. It appears that the solar absorption data give ozone concentrations that are 20% larger than the *in situ* techniques. Further investigations are needed to explain such divergences. Filter radiometers should fly with solar spectrometers which are able to provide data with a better accuracy covering the whole altitude range. Filter radiometers are obviously limited by the number of wavelengths used in the solar u.v. absorption measurements, which consequently increase the uncertainties. A sensitivity calculation for the parameters like the solar spectrum, filter transmissions, etc. should be performed. On the other hand, a careful analysis of wall-loss problems and laboratory calibrations for *in situ* techniques should also be made in order to identify the principal sources of errors in such measurement techniques. Nevertheless, the good agreement between *in situ* u.v. absorption and mass spectrometer measurements reported by Mauersberger *et al.* (1981) suggests that the wall-losses

TABLE 3. OZONE VERTICAL CONCENTRATION (molecules cm^{-3}) VS ALTITUDE AS MEASURED BY THE TWO SOLAR U.V. RADIOMETERS ON 19 JUNE 1981

Altitude (km)	Descent 1 IASB	Descent 1 GSFC	Descent 1 Ratio	Ascent 2 IASB	Ascent 2 GSFC	Ascent 2 Ratio
22	5.80×10^{12}	5.43×10^{12}	1.07	5.60×10^{12}	5.35×10^{12}	1.05
23	6.07	5.56	1.09	5.64	5.59	1.01
24	5.87	5.62	1.04	5.61	5.57	1.01
25	5.53	5.45	1.01	5.54	5.43	1.02
26	5.20	5.22	0.996	5.35	5.29	1.01
27	4.91	4.79	1.03	5.06	4.86	1.04
28	4.66	4.32	1.08	4.71	4.39	1.07
29	4.40	3.97	1.11	4.32	4.02	1.07
30	4.11	3.63	1.13	3.92		
31	3.67			3.52		
32				3.17		

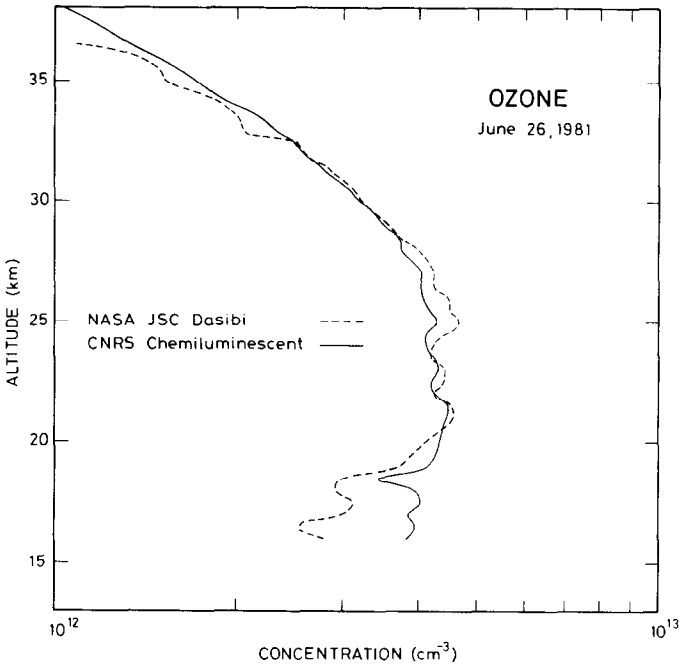

FIG. 7. OZONE VERTICAL DISTRIBUTION FOR ALTITUDES BETWEEN 15 AND 40 km AS MEASURED ON 26 JUNE 1981 BY THE TWO *in situ* INSTRUMENTS.

could not be the main source of uncertainties because this problem is not encountered in the mass spectrometer. On the other hand, the chemiluminescent instrument should also be calibrated using the balloon-borne Dasibi instrument to make future investigations more comparable. This campaign also shows that the ECC sondes provide reliable data only below 25 km. Differences as high as a factor of 2 have been found around 33 km, between the four soundings performed on 19 June 1981, from the same location.

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