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AERONOMIC PROBLEMS OF MOLECULAR OXYGEN PHOTODISSOCIATION—III. SOLAR SPECTRAL IRRADIANCES IN THE REGION OF THE O_2 HERZBERG CONTINUUM, SCHUMANN–RUNGE BANDS AND CONTINUUM

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Abstract—Retrospective evaluation of spectral irradiances obtained during the last 10 years at wavelengths relevant to the photodissociation of molecular oxygen provides an indication of the accuracy and precision of the information currently available. In the spectral region of wavelengths less than 175 nm corresponding to the O_2 Schumann–Runge continuum which is absorbed in the thermosphere, all the observational results are not reliable since the global accuracy is of the order of $\pm 50\%$. In the spectral region 175–200 nm of the Schumann–Runge bands, mainly absorbed in the mesosphere, the uncertainties of all available data are not less than 20–30% and make it impossible to determine the exact solar activity effect on the O_2 photodissociation rate. The available measurements for the spectral region associated with the O_2 Herzberg continuum, 200–240 nm, relevant to the stratosphere have typical uncertainty limits reaching $\pm 10\%$ with additional random errors of $\pm 10\%$ for 1 nm intervals. The general accuracy is not yet sufficient to infer the exact part in the irradiance changes associated with solar variability. A consistent reference spectrum for a better assessment must be, therefore, adopted to describe the complex behavior displayed by the spectral solar irradiances in the spectral ranges of the photodissociation of O_2.

1. INTRODUCTION

The direct photodissociation of molecular oxygen in the Earth's atmosphere depends on the solar irradiance corresponding to the O_2 Schumann–Runge continuum, to the predissociated O_2 Schumann–Runge bands, and to the O_2 Herzberg continuum.

Since the absorption cross-section in the Schumann–Runge continuum beginning at 57,000 cm^{-1} (175.4 nm) has an increasing value from at least 2×10^{-19} cm^2 up to not less than 10^{-17} cm^2 at the peak, the solar radiation is absorbed in the thermosphere. The absorption in the 57,000–49,500 cm^{-1} region (175–202 nm) corresponding to the Schumann–Runge bands decreases from about $1\text{–}2 \times 10^{-19}$ cm^2 near 56,500–57,000 cm^{-1} to about 10^{-23} cm^2 near 49,500–50,000 cm^{-1}. The solar radiation is, therefore, mainly absorbed in the mesosphere and partly in the upper stratosphere. Finally, the absorption in the Herzberg continuum (49,500–41,250 cm^{-1}) beginning in the lower mesosphere is typically a stratospheric process since the O_2 cross-section is less than 10^{-23} cm^2 and decreases to 10^{-24} cm^2 at the threshold near 242 nm.

In the present paper, we attempted to determine a consistent set of ultraviolet irradiances related only to this spectral range of direct photodissociation of O_2. Since the first publications 35 years ago by Tousey *et al.* (1951) and Friedman *et al.* (1951), the successive compilations at 10 year intervals (Detwiler *et al.*, 1961; Ackerman, 1971; Brasseur and Simon, 1981) show that it is difficult to determine the absolute values of the solar spectral irradiances with sufficient accuracy. Nevertheless, comparisons with the most recent observations must be made to establish again a reference spectrum. In our analysis we have considered the results of observations of the solar irradiances by Heath (1980), Mentall *et al.* (1981), Mount and Rottman (1983, 1985 and unpublished data), Anderson *et al.* (1987), Mentall and Williams (1988), Labs *et al.* (1987) and the recent tables published in *Atmospheric Ozone*, Chapter 7, *Radiative Processes: Solar and Terrestrial* (WMO, 1985). We have adopted (Figs 1, 2 and 3) as a reference spectrum the solar irradiances of *Spacelab* 2 using SUSIM, the Solar Ultraviolet Spectral Irradiance Monitor (VanHoosier *et al.*, 1987).

2. THE SOLAR IRRADIANCE IN THE SPECTRAL REGION OF THE O_2 SCHUMANN–RUNGE CONTINUUM

Since almost all oxygen atoms produced by photodissociation of O_2 in the thermosphere move down

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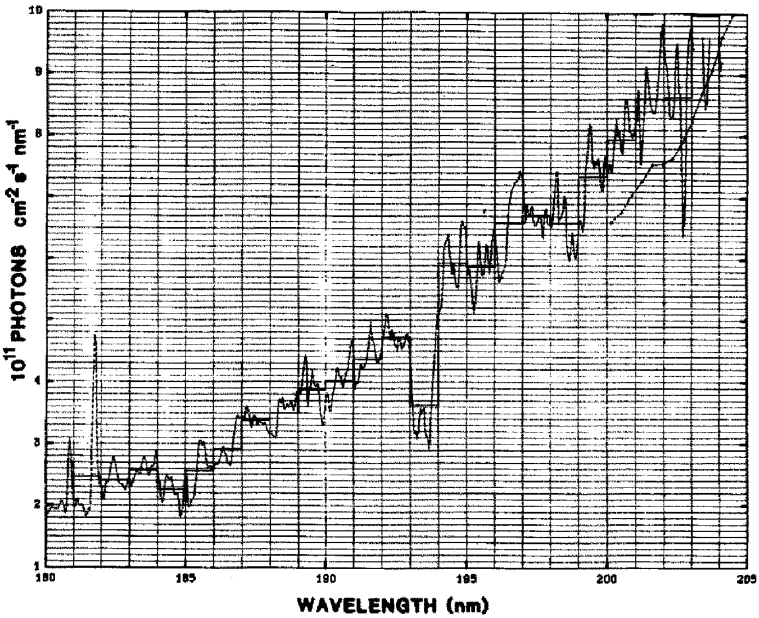

FIG. 1. SPECTRAL SOLAR IRRADIANCES (PHOTONS cm^{-1} s^{-1} nm^{-1}) FROM 180 TO 205 nm. The dashed lines reproduce at each 0.05 nm the irradiances from the SUSIM experiment (VanHoosier *et al.*, 1987) in photons cm^{-2} s^{-1}. The solid lines represent averaged values in 1 nm intervals. The dotted curve beginning at 200 nm is used to connect (generally three values indicated by $\times$ for each nanometer) the irradiances given by Labs *et al.* (1987) at specific wavelengths.

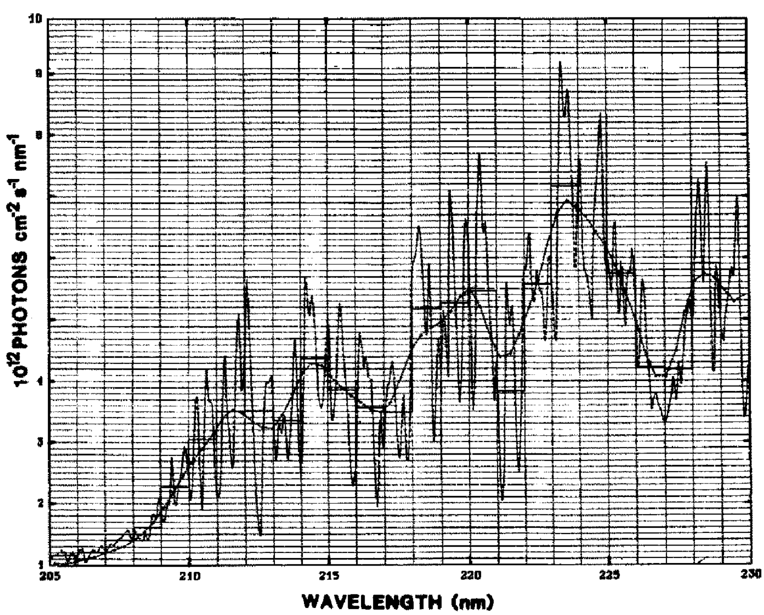

FIG. 2. SPECTRAL SOLAR IRRADIANCES (PHOTONS cm^{-2} s^{-1} nm^{-1}) FROM 205 TO 230 nm. As in Fig. 1.

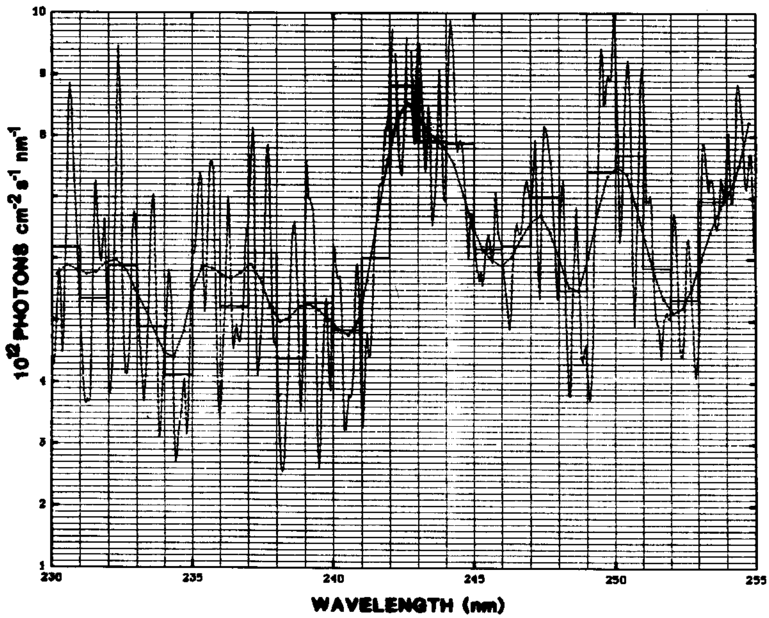

FIG. 3. SPECTRAL SOLAR IRRADIANCES (PHOTONS cm^{-2} s^{-1} nm^{-1}) FROM 230 TO 255 nm. As in Figs 1 and 2.

TABLE 1. SOLAR FLUX (Q) AND BRIGHTNESS TEMPERATURE (T_s). TOTAL NUMBER OF PHOTONS cm^{-2} s^{-1} FOR WAVELENGTHS LESS THAN 175 nm AT THE TOP OF THE EARTH'S ATMOSPHERE

T_s (K)	Q (cm^{-2} s^{-1})	T_s (K)	Q (cm^{-2} s^{-1})
4400	3.6×10^{11}	4750	1.6×10^{12}
4450	4.5×10^{11}	4800	1.9×10^{12}
4500	5.6×10^{11}	4850	2.3×10^{12}
4550	6.9×10^{11}	4900	2.7×10^{12}
4600	8.6×10^{11}	4950	3.3×10^{12}
4700	1.3×10^{12}	5000	3.9×10^{12}

to heights near the mesopause to become reattached and to form oxygen molecules again, the basic aeronomic parameter is the total production of oxygen atoms. Knowledge of the total number of photons (cm^{-2} s^{-1}) at wavelengths less than 175 nm is, therefore, required as the basic solar irradiance parameter. It should lead to a determination of the variations of the upper boundary conditions (number density of oxygen atoms at the mesopause level) which must be related to solar activity conditions.

The first value of the total irradiance at $\lambda < 175$ nm was used by Nicolet and Mange (1954) and based on observations of Friedman *et al.* (1951) corresponding to a brightness temperature of about 4500 K, i.e. a total number of photons (Table 1) of the order of 5×10^{11} cm^{-2} s^{-1} equivalent to a total production of oxygen atoms of the order of 10^{12} cm^{-2} s^{-1}. Ten years later, higher observational values led to a total irradiance of the order of 2.7×10^{12} photons cm^{-2} s^{-1} corresponding to a brightness temperature of the order of 4900 K, equivalent to a difference of about 400 K in the solar continuum. A strange result!

Table 2 provides, for various solar radio fluxes at 10.7 cm, the total number of photons observed at wavelengths less than 175 nm; it can be concluded that there is no direct possibility of understanding the differences since it seems that there is a lack of agreement. The low values of Heroux and Higgins (1977) of the order of 6×10^{12} photons cm^{-2} s^{-1} for low solar activity correspond to only 50% of the values deduced from VanHoosier *et al.* (1987) for almost the same conditions of solar activity and also 50% of the values adopted by Ackerman (1971). How can a real solar activity effect be deduced when there are unquestionable differences of $\pm 50\%$? A conventional value should be $(1.0 \pm 0.25) \times 10^{12}$ photons cm^{-2} s^{-1}, indicating that the observational accuracy must be improved. On the other hand, the variation with solar activity is not known, but should be less than a factor of 2.

In conclusion, a determination of the total thermospheric production of oxygen atoms requires more observations at wavelengths less than 175 nm with better accuracy to fix the atomic oxygen concentrations near the mesopause and its variations with solar activity.

TABLE 2. TOTAL NUMBER OF PHOTONS (Q_∞ cm^{-2} s^{-1}) IN THE SPECTRAL REGION OF THE SCHUMANN–RUNGE CONTINUUM (175–135 nm)

Date	Solar flux (10.7 cm)	Q_∞	Reference
1951	—	$\simeq 5 \times 10^{11}$	Friedman *et al.* (1951)
1961	—	$\simeq 2.5 \times 10^{12}$	Detwiler *et al.* (1961)
1971	—	$\simeq 1.2 \times 10^{12}$	Ackerman (1971)
1981	—	$\simeq 6.5 \times 10^{11}$	Brasseur and Simon (1981)
2 Nov. 1973	84	5.7×10^{11}	Heroux and Higgins (1977)
23 Apr. 1974	74	5.2	
13 Dec. 1972	111	6.2	Rottman (1974)
		8.7	Rottman (1981)
30 Aug. 1973	91	6.0	Rottman (1974)
		7.7	Rottman (1981)
28 Jul. 1975	75	8.2	Rottman (1981)
18 Feb. 1976	70	1.0×10^{12}	
9 Mar. 1977	80	1.0	
16 Nov. 1978	132	8.0×10^{11}	Mentall *et al.* (1985)
5 Jun. 1979	224	1.5×10^{12}	Mount *et al.* (1980)
22 May 1980	270	8.4×10^{11}	Mentall *et al.* (1985)
15 Jul. 1980	211	1.4×10^{12}	Mount and Rottman (1981a,b)
16 Oct. 1981	304	6.7×10^{11}	Mentall *et al.* (1985)
17 May 1982	139	8.0	Mount and Rottman (1983)
23 Jul. 1983	136	7.2	Mount and Rottman (1985)
29 Jul–6 Aug. 1985	70–85	1.25×10^{12}	VanHoosier *et al.* (1987)

3. THE SOLAR IRRADIANCE IN THE SPECTRAL REGION OF THE O_2 SCHUMANN-RUNGE BANDS

The spectral region of the Schumann–Runge bands of O_2 requires special attention since the published observational solar irradiances do not allow definitive conclusions to be reached not only about the spectral distribution but also about the absolute values in order to detect the effect of solar activity. Generally, the differences between the various sets of observations cannot be less than those arising from changes in solar activity.

The reference data deduced from the observations of the Naval Research Laboratory (VanHoosier *et al.*, 1987) were made between 120 and 400 nm from *Spacelab* 2 between 29 July and 6 August 1985 using SUSIM, the Solar Ultraviolet Spectral Irradiance Monitor. The irradiances measured in the SUSIM experiment with a 0.15 nm bandpass and given (mW m^{-2} nm^{-1}) at each 0.05 nm are reproduced (photons cm^{-2} s^{-1} nm^{-1}) in Figs 1, 2 and 3 from 180 to 205 nm, from 205 to 230 nm and from 230 to 255 nm, respectively. The averaged values in 1 nm intervals which may be compared with the individual irradiances (generally three in 1 nm intervals) shown at a certain number of wavelengths by Labs *et al.* (1987), are also depicted in the three figures from 200 to 255 nm.

Figure 4 compares the solar irradiance data for 500 cm^{-1} intervals between 57,000 and 50,000 cm^{-1}. The irradiances for such intervals as deduced from the SUSIM *Spacelab* 2 data lead to a different view of the solar emission in this region of the spectrum; this is illustrated in the figure where it can be seen that the values of the compilation of WMO (1985), those of Heath (1980) at high solar activity and of the complication of Brasseur and Simon (1981) are systematically below the values deduced from SUSIM. The various ratios are reproduced in Fig. 5: their values deduced from the Brasseur–Simon compilation and the NRL data are of the order of only 0.65 ± 0.15, from Heath, 0.75 ± 0.10, from WMO, 0.85 ± 0.10, and from Ackerman (1971), 1.00 ± 0.15.

If the comparisons (Fig. 6) are made for solar irradiances averaged in 1 nm intervals in the same region of the O_2 Schumann–Runge bands, the same conclusions are reached. The averaged ratio of the Brasseur–Simon compilation and the NRL data is still 0.65 ± 0.15 with a minimum at $\lambda < 180$ nm. The averaged values of Rottman between 1982 and 1984 correspond to a ratio of 0.80 ± 0.15 with a minimum near 180 nm. The recent values of Mentall and Williams (1988) in 1983 and 1984 lead to a different ratio 0.75 ± 0.05 at $\lambda < 188$ nm and 0.90 ± 0.05 at $\lambda < 190$ nm. Thus, the photodissociation rates of O_2 in the mesosphere depend, at the present time, on the

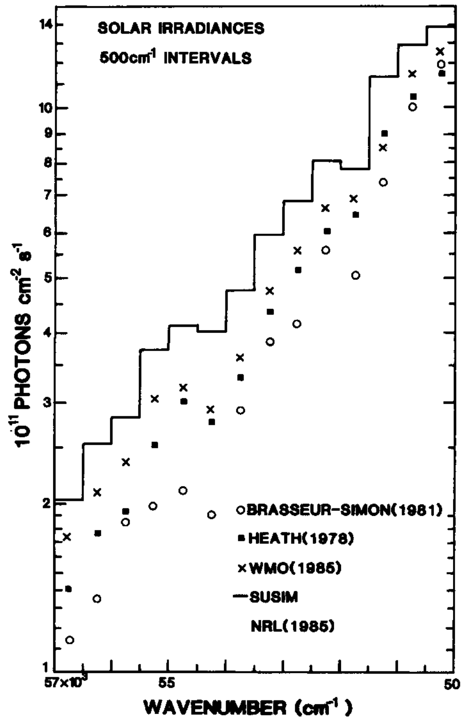

FIG. 4. SOLAR SPECTRAL IRRADIANCES (PHOTONS cm^{-2} s^{-1}) BETWEEN 57,000 AND 50,000 cm^{-1} IN 500 cm^{-1} INTERVALS.

adopted solar irradiances more than on possible solar activity effects which should be established according to the spectral region.

4. THE SPECTRAL IRRADIANCES IN THE SPECTRAL REGION OF THE O_2 HERZBERG CONTINUUM

The uncertainties in the spectral region 200–240 nm are certainly less than the differences occurring at shorter wavelengths. Figure 7 illustrates for 500 cm^{-1} intervals the ratio of the compilation of Brasseur–Simon and the values deduced from the SUSIM data. The ratio is 0.98 ± 0.12 with a subdivision 0.90 ± 0.05 for $\nu > 48{,}000$ cm^{-1} and 1.05 ± 0.05 for $\nu < 48{,}000$ cm^{-1}. The WMO compilation corresponds to an almost identical ratio: 0.97 ± 0.17, with systematic differences <1 for $\nu > 45{,}000$ cm^{-1} and >1 for $\nu < 45{,}000$ cm^{-1}. The satellite measurements of Heath in 1978 (high solar activity) are systematically lower than the values deduced from SUSIM data (low solar activity); the mean ratio is 0.90 ± 0.07 and reaches 0.85 ± 0.05 at $\nu > 48{,}000$ cm^{-1} and 0.95 ± 0.05 at $\nu < 48{,}000$ cm^{-1}. Finally, the values which were adopted by Nicolet and Kennes (1987) are 10% lower than the SUSIM data. With an increase of a factor of 1.1 the ratio is 1.0 ± 0.05.

Various spectral irradiances are illustrated in Fig. 8 for intervals of 500 cm^{-1}. It is clear that the agreement is at least within $\pm 10\%$. Comparison between the irradiance deduced from *Spacelab* 1 (Labs *et al.*, 1987) and from SUSIM *Spacelab* 2 shows an astonishingly good agreement between 48,000 and 40,000 cm^{-1}. If there is a difference greater than 10% from 50,000 to 48,000 cm^{-1} (see Fig. 1), it is only 1.00 ± 0.04

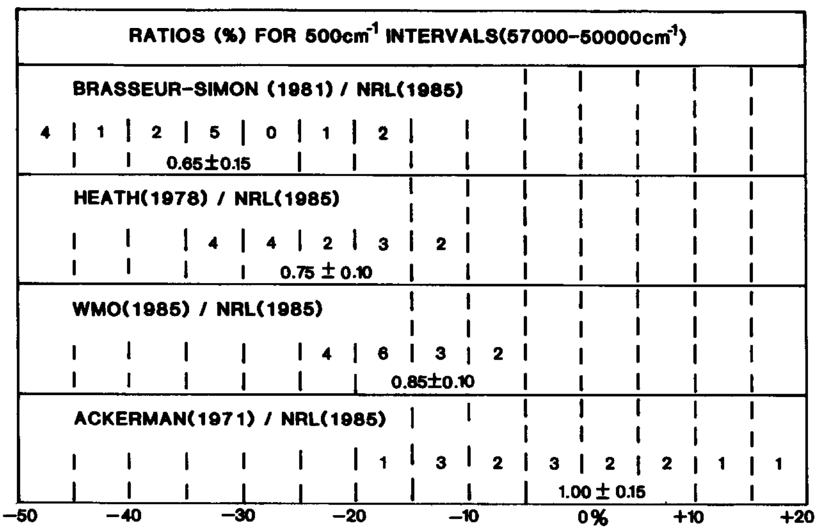

FIG. 5. DISTRIBUTION, BETWEEN 57,000 AND 50,000 cm^{-1}, OF RATIOS OF SOLAR IRRADIANCES DEPICTED IN FIG. 1.

Numbers of intervals by steps of 5% with the averaged values and their dispersions of the ratios. Reference spectrum: SUSIM.

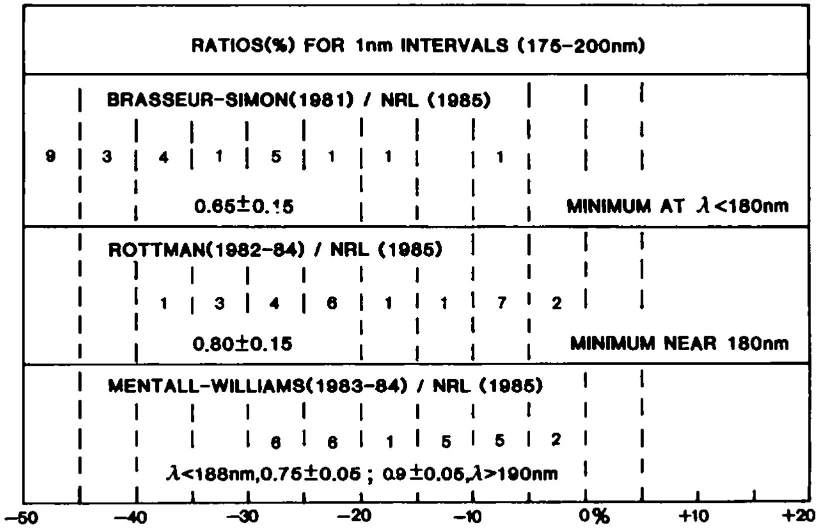

FIG. 6. DISTRIBUTION, BETWEEN 57,000 AND 50,000 cm^{-1} (175–200 nm), OF RATIOS OF SOLAR IRRADIANCES FOR 1 nm INTERVALS.

Number of intervals by steps of 5% with the averaged values and their dispersions of the ratios. Reference spectrum: SUSIM.

between 48,000 and 41,000 cm^{-1}. Of the 14 ratios, five correspond to 1.00, four to ± 1.02 and five to 1.04. An extension to 28,000 cm^{-1} for 500 cm^{-1} intervals shows an agreement of $\pm 5\%$ for at least 95% of the ratios, $\pm 4\%$ for 90%, $\pm 3\%$ for 80%, $\pm 2\%$ for 70% and $\pm 1\%$ for more than 50%. The number of lines which were considered from the measurements of Labs *et al.* (1987) for 500 cm^{-1} increases from 20 at 48,000–47,500 cm^{-1} to 60 at 28,500–28,000 cm^{-1}.

A more detailed comparison between various aver-

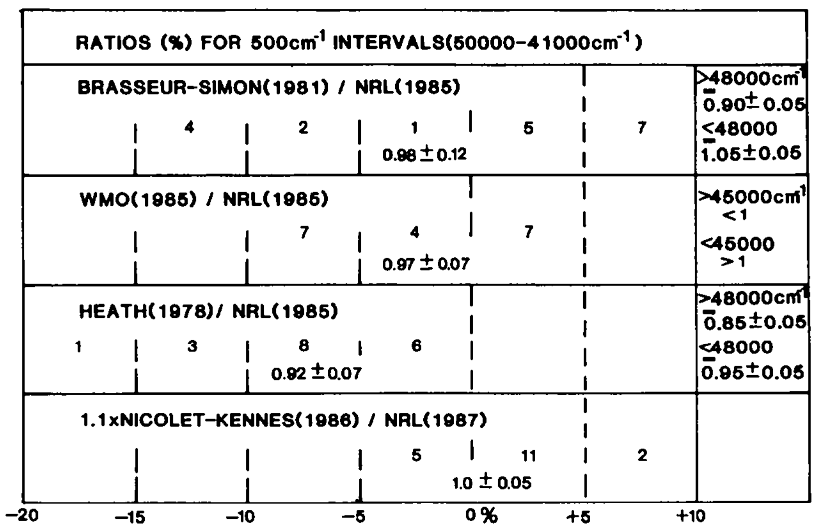

FIG. 7. DISTRIBUTION, BETWEEN 50,000 AND 41,000 cm^{-1}, OF RATIOS OF SOLAR IRRADIANCES FOR 500 cm^{-1} INTERVALS.

Number of intervals by steps of 5% with the averaged values and their dispersions of the ratios. Reference spectrum: SUSIM.

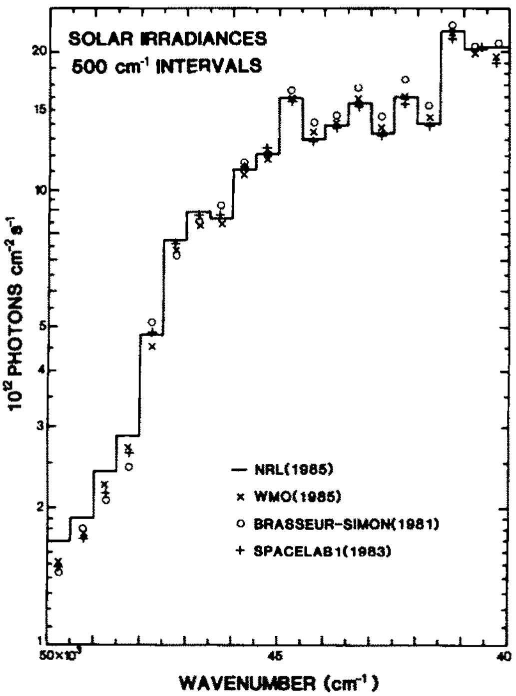

FIG. 8. SOLAR SPECTRAL IRRADIANCES (PHOTONS cm^{-2} s^{-1}) BETWEEN 50,000 AND 40,000 cm^{-1} IN 500 cm^{-1} INTERVALS.

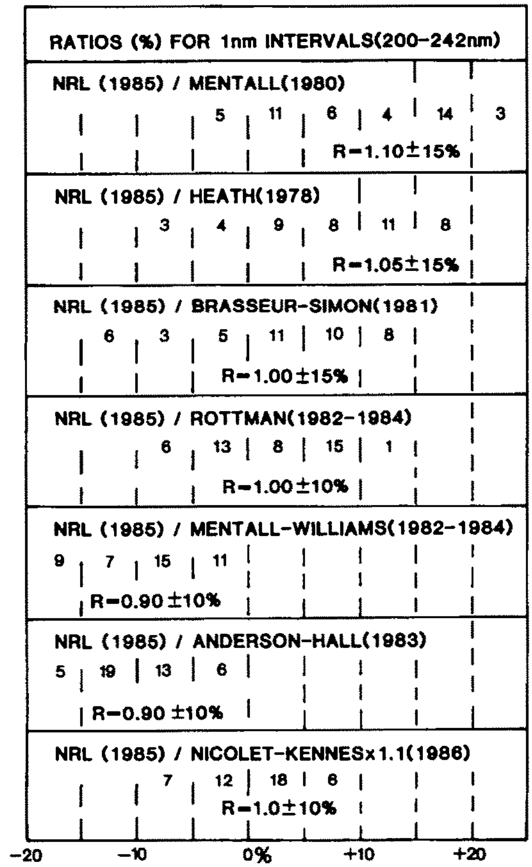

FIG. 9. DISTRIBUTION, BETWEEN 200 AND 242 nm, OF RATIOS OF SOLAR IRRADIANCES FOR 500 cm^{-1} INTERVALS.
Number of intervals by steps of 5% with the averaged values and their dispersions of the ratios R. Reference spectrum: SUSIM

aged values in 1 nm intervals indicates systematic differences as large as $\pm 10\%$ and random variations between $\pm 10\%$ and $\pm 15\%$. The results of the analysis are depicted in Fig. 9. The averaged values in 1 nm intervals deduced from the SUSIM data were again taken as the reference data.

Starting from the data of Mentall *et al.* (1981) corresponding to measurements of the solar irradiance on 15 September 1980 at low solar activity (solar flux at 10.7 cm = 70 units for the whole month), the comparison of the values NRL/Mentall gives a ratio $1.10 \pm 15\%$, i.e. a systematic difference of 10% with possible individual differences as large as 10–15%. However, individual differences greater than 10% may be explained by an error in the comparison of the order of 0.1 nm for intervals of 1 nm since the wavelength scale at some points in the spectrum may be different. It is not easy at low resolution to detect the errors, particularly when an averaged value of the irradiances is deduced from the complex blends exhibited in the solar spectrum at low resolution. The differences in this spectral region of 0.06–0.07 nm between the vacuum and air wavelengths must be considered also as possible errors in the comparisons between observational data. Figure 10 is an example of differences in 1 nm and 500 cm^{-1} intervals. The results of rocket observations of Mentall *et al.* (1981) are compared with the results of satellite observations made by Heath in 1978. Figures 11 and 12 explain the differences by a comparison of irradiances based on 1 nm running means of values of Mentall *et al.* and of those directly deduced from Heath's data with a resolution of about 1 nm. Near 215 ± 2.5 nm the irradiances of Mentall *et al.* are systematically low, but show a higher resolution depicted in the maxima and minima even with a running mean of 1 nm.

A comparison (Fig. 9) with the satellite data of Heath (1980) on 7 November 1978 at relatively high solar activity (solar radio flux at 10.7 cm, 175 units) indicates a ratio NRL/Heath = $1.05 \pm 15\%$, i.e. systematically lower values in the opposite variation of solar activity. The compilation of Brasseur and Simon (1981) shows an agreement (Fig. 9) with a ratio NRL/Brasseur–Simon = 1.00 but with differences

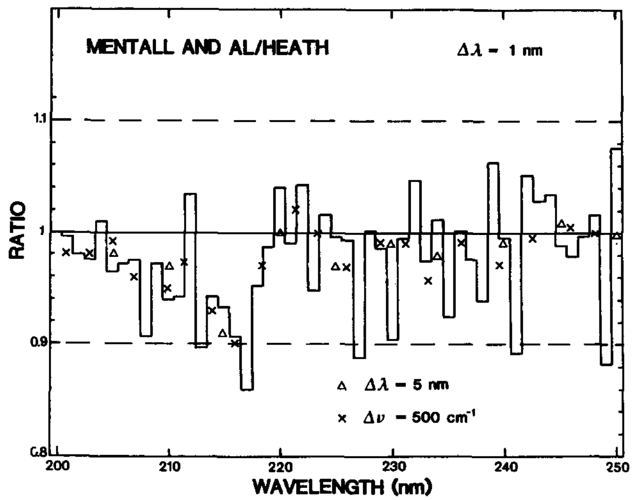

FIG. 10. RATIOS OF IRRADIANCES BETWEEN 200 AND 250 nm.
For 1 nm intervals the ratio of the averaged irradiances of Mentall *et al.* (1981) and Heath (1980) is about $0.95 \pm 10\%$ as can be seen in the figure. The agreement is better for 500 cm^{-1} intervals or 5 nm intervals.

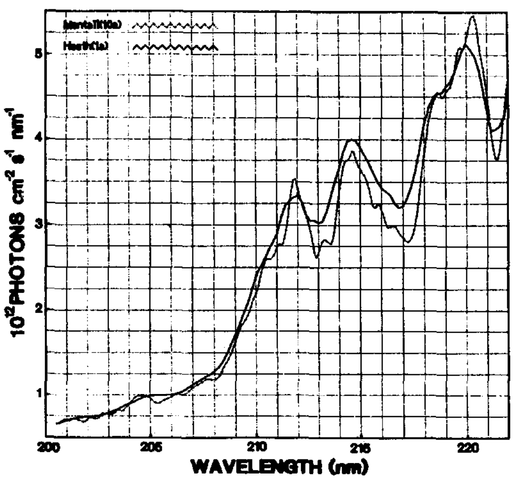

FIG. 11. COMPARISON BETWEEN SOLAR IRRADIANCES FROM 200 TO 222 nm.
The irradiances obtained by Mentall *et al.* (1981) represented by 1 nm running means and compared with the irradiances obtained by Heath in 1978. Agreement except near 215 ± 3 nm.

reaching 10–15%, i.e. a modest precision. The averaged values of four observations (Mount and Rottman, 1983, for a solar radio flux at 10.7 cm of 140 units on 17 May 1982; Mount and Rottman, 1985, for a solar radio flux at 10.7 cm of 140 units on 25 July 1983, and Rottman (unpublished) for solar radio fluxes of 102 on 7 December 1983 and 77 on 10 December 1984) give (Fig. 9) a ratio NRL/Rottman (1982–1984) of $1.00 \pm 10\%$, i.e. a ratio with a normal precision of $\pm 10\%$.

Two recent determinations by Mentall and Williams (1988) based on rocket flights on 7 December 1984 (solar radio flux at 10.7 cm = 102) and on 10 December 1984 (solar radio flux at 10.7 cm = 77) give a ratio NRL/Mentall–Williams = 0.90 ± 0.10, i.e. a systematic difference of 10% (Fig. 9) with a random precision of $\pm 10\%$ normal for comparison on 1 nm intervals. A recent analysis by Anderson and Hall (1988, unpublished) based on observations made at 40 km (April 1983) shows (Fig. 9) that the ratio NRL/Anderson–Hall corresponds to $0.90 \pm 10\%$.

In conclusion, in the region of the O_2 Herzberg continuum, 200–240 nm, the absolute accuracy of spectral solar irradiances is confined to $\pm 10\%$ for the recent observations (1978–1988); the precision of individual measurements is approximately $\pm 10\%$ for averaged values in intervals of 1 nm.

5. CONCLUSION

The SUSIM irradiances (NRL *Spacelab* 2), which were taken in the present investigation on the solar u.v. radiation as reference solar irradiances adapted to various spectral intervals, will be used in subsequent publications on the direct photodissociation of molecular oxygen in the mesosphere and stratosphere.

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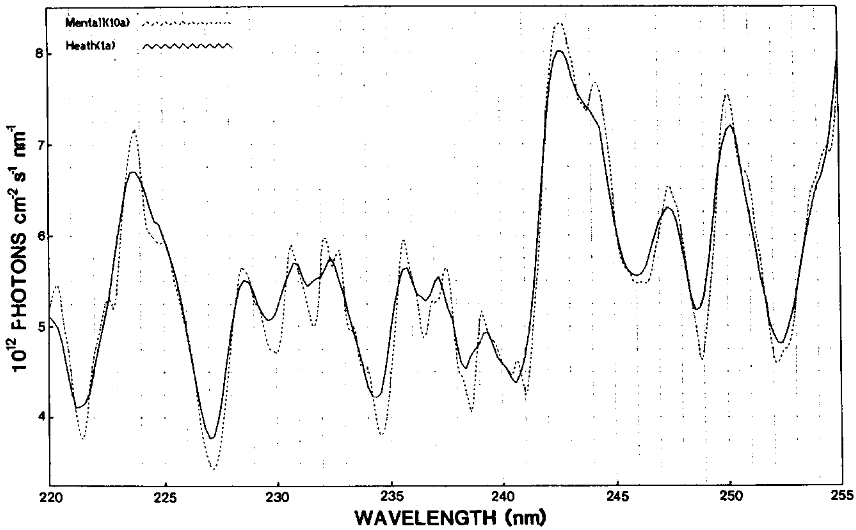

FIG. 12. COMPARISON BETWEEN SOLAR IRRADIANCES FROM 220 TO 255 nm.
Irradiances as in Fig. 11. Even with 1 nm running means the averaged values deduced from Mentall *et al.* (1981) have higher peaks and deeper minima than the irradiances obtained by Heath.

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