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Aeronomic problems of the molecular oxygen photodissociation

I. the O_2 herzberg continuum

by

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FOREWORD

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AERONOMIC PROBLEMS OF THE MOLECULAR OXYGEN PHOTODISSOCIATION

I. THE O₂ HERZBERG CONTINUUM

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Abstract

An analysis of the aeronomic conditions related to oxygen photodissociation has been made by including updated absorption cross sections in the region of the Herzberg continuum at wavelengths greater than 200 nm. This analysis demonstrates that there is a decrease in the production of odd oxygen atoms compared with that obtained by using the O₂ cross sections adopted before 1980. Quantitative results show that the photodissociation frequency $J(O_2)$ cannot be greater, at mesospheric and stratopause levels, than about $5 \times 10^{-10} \text{ s}^{-1}$ for the 200 - 240 nm. The possible errors ($\pm 10\%$) in these O₂ photolysis rates decrease with decreasing altitudes to reach isophotolytic levels ($\pm 0\%$) in the middle stratosphere, from about 25 km to 30 km for increasing solar zenith angles, respectively. In the lower stratosphere, at 20 km and below, there is a rapid increase in the computed errors where the transmittance is less than 10^{-2} . Fuller information will be obtained after an estimation of the possible errors associated with uncertainties in the absorption cross-sections of ozone molecules.

Résumé

Des données récentes, expérimentales et théoriques, sur la section efficace d'absorption de l'oxygène au-delà de 200 nm dans le continuum de Herzberg de l'oxygène moléculaire ont conduit à une nouvelle analyse des conditions aéronomiques liées à la photodissociation. Il est certain que ces nouveaux résultats démontrent que la production des atomes d'oxygène est inférieure à celle qui était admise avant 1980. Le calcul montre que la fréquence de photodissociation, $J(O_2)$, correspondant à l'intervalle spectral 200 - 240 nm ne peut être supérieur à $5 \times 10^{-10} \text{ s}^{-1}$ dans la mésosphère et au niveau de la stratopause. L'incertitude au départ de $\pm 10\%$ dans la production des atomes d'oxygène diminue dans la stratosphère jusqu'à atteindre ($\pm 0\%$) à des niveaux isophotolytiques allant d'environ 25 à 30 km pour des distances zénithales solaires croissantes. Dans la stratosphère inférieure, c'est-à-dire à 20 km et en-dessous, l'erreur croît rapidement lorsque la transmittance atmosphérique devient inférieure à 10^{-2} . Une information plus complète requiert encore une estimation des erreurs pouvant résulter des incertitudes existant dans les valeurs de la section efficace d'absorption de l'ozone.

Samenvatting

Recente experimentele en theoretische gegevens, betreffende de werkzame absorptiedoorsnede van zuurstof boven de 200 nm in het Herzberg continuüm van moleculaire zuurstof, lieten toe een nieuwe analyse van de aëronomische toestanden in verband met de fotodissociatie te maken. Het is zeker dat deze nieuwe resultaten aantonen dat de produktie van de zuurstofatomen kleiner is dan deze aangenomen voor 1980. De berekening toont dat de fotodissociatiefrequentie, $J(O_2)$, overeenstemmend met het spectraal interval 200-240 nm niet groter mag zijn dan $5 \times 10^{-10} \text{ s}^{-1}$ in de mesosfeer en ter hoogte van de stratopauze. De mogelijke fouten ($\pm 10\%$) in de produktie van de zuurstofatomen wordt kleiner met dalende hoogte en bereikt isofotolytische niveaus ($\pm 0\%$) in het midden van de stratosfeer, gaande van ongeveer 25 km tot 30 km voor toenemende zenitafstanden van de zon. In de lage stratosfeer, op 20 km en lager, is er een vlugge toename in de berekende fouten wanneer de atmosferische doorlaatbaarheid kleiner wordt dan 10^{-2} . Teneinde nauwkeuriger informatie te bekomen moet nog een schatting van de vergissingen gemaakt worden die kunnen ontstaan uit de onzekerheden die bestaan in de waarden van de werkzame absorptiedoorsnede van ozon.

Zusammenfassung

Rezente Daten, experimentelle und theoretische, über dem O_2 Absorptionsquerschnitt über die 200 nm in Herzberg Kontinuum von Molekularsauerstoff, haben geführt zu einer neuen Analyse des aëronomischen Zustandes der mit der Fotodissoziation verbunden ist. Es ist gewiss dass diese neuen Resultaten zeigen dass die Produktion von Sauerstoffatomen geringer ist als diese angenommen vor 1980. Die Berechnung zeigt dass die Fotodissoziationfrequenz, $J(O_2)$, übereinstimmend mit dem Spektralintervall 200-240 nm nicht grösser mag sein als $5 \times 10^{-10} s^{-1}$ in der Mesosphäre und auf der Höhe der Stratopause. Die Ungewissheit im Anfang von $\pm 10\%$ in der Produktion der Sauerstoffatomen wird geringer in der Stratosphäre zu ($\pm 0\%$) auf isofotolytischem Niveau von ungefähr 25 zu 30 km für zunehmende Zenitabstanden der Sonne. In der Nieder-stratosphäre, dass heisst auf 20 km und geringer, nimmt der Rechenfehler schnell zu wenn die atmosphärische Transparenz geringer wird als 10^{-2} . Um genauere Information zu bekommen muss noch eine Schätzung der Rechenfehler gemacht werden die entstehen können aus den Ungewissheiten die existieren in den Werten des Absorptionsquerschnittes der Ozon.

INTRODUCTION

The atmospheric ozone production depends on the dissociation of molecular oxygen. The origin of mesospheric and stratospheric ozone is directly related to the O_2 photodissociation in the spectral ranges of the Herzberg continuum and of the predissociated bands of the Schumann-Runge system at wavelengths less than 240 nm. At wavelengths greater than 200 nm the photodissociation depends almost exclusively on the Herzberg continuum and from 175 nm to 200 nm it is associated with the Schumann-Runge bands and their underlying continua.

Since the publication by Herman and Mentall (1982) of observational results on the stratospheric transmittance in the region 200 - 220 nm, there is now a general agreement that the laboratory cross sections used in the modelling calculations before 1982 for the Herzberg continuum absorption were too large (see, for example, Nicolet, 1983; Bucchia, Megie and Nicolet, 1985). New laboratory measurements and recent theoretical determinations (Johnston, Paige and Yao, 1984; Jenouvrier, Coquart and Merienne-Lafore, 1986; Cheung, Yoshino, Parkinson, Freeman, 1986; Saxon and Slanger, 1986) show that the absorption cross section of the Herzberg continuum may be determined with a relatively good accuracy between 240 nm and 215 nm but that there may still be inconsistencies in the spectral region of the Schumann-Runge bands. However, since the 240-200 nm spectral interval leads to not less than 60% of the O_2 photodissociation at the stratopause level (50 km for an overhead sun) to about 90% in the lower stratosphere (20 km) a detailed and separate study of O_2 photodissociation processes at wavelengths greater than 200 nm is first required. The purpose of this paper is, therefore, to determine the various parameters of the aeronomic photodissociation of molecular oxygen in this spectral region. The analysis of the Schumann-Runge bands will be the subject of other publications and the effect of the (0-0) and (1-0) will not be discussed in this paper.

DATA BASE DESCRIPTION

At present, it is clear that the O_3 number density calculated with the available parameters in the photochemical region, i.e. in the neighbourhood of the stratopause does not correspond to the measured O_3 concentrations. Two recent analyses by Froidevaux, Allen and Yung (1985) and by Rush and Eckman (1985) establish that several aspects of the ozone problem must be clarified and that a complete understanding of the stratopause chemistry requires a detailed study of the principal aeronomic parameters. Since the principal object of this paper is the O_2 absorption cross sections in the Herzberg continuum, we adopt here without any detailed discussion, for intervals of 500 cm^{-1} , the spectral solar irradiances, the atmospheric molecular scattering cross sections and the ozone cross sections. Spectral irradiances accepted here are deduced (Nicolet, unpublished) from the satellite data obtained by Heath (1981) and from rocket measurements of Mentall, Frederick and Herman (1981). The general agreement between these data considered in intervals of 500 cm^{-1} (within $\pm 3\%$, except near 212.5 nm) indicates that a fairly accurate reference spectrum may be considered for the O_2 photodissociation. However, if the results of Mount and Rottman (1982) are also taken into account, our adopted reference spectrum should be perhaps increased by about 5% . The spectral solar irradiances for intervals of 500 cm^{-1} are given in Table 1. It is clear that these irradiances based on observations made in 1980 are $40 \pm 30\%$ less than those obtained in the years before 1970 (see Ackerman, 1971).

The atmospheric Rayleigh scattering has been determined recently with great accuracy by Bates (1984) and the corresponding values of the atmospheric scattering cross section are calculated using for formula (Nicolet, 1984), with an accuracy better than $\pm 1\%$,

$$\sigma_{MS} = 4.02 \times 10^{-28} \lambda^{-(4+x)} \quad (1)$$

TABLE 1.- Solar spectral irradiances for $\Delta\nu = 500 \text{ cm}^{-1}$ and corresponding attenuation cross sections in the spectral region of the Herzberg continuum of O_2 .

$\nu(\text{cm}^{-1})$ (1)	$\lambda(\mu\text{m})$ (2)	$q_{\infty}(\text{cm}^{-2}\text{s}^{-1})$ (3)	$\sigma(\text{O}_3)$ (4) (cm^2)	$\sigma_{\text{MS}}(\text{O}_2)$ (5) (cm^2)	$\sigma_{\text{MS}}(\text{air})$ (6) (cm^2)	$\sigma_{\text{MS}}(\text{O}_2)_{\text{EQU}}$ (7) (cm^2)
41250	0.24242	2.04×10^{13}	9.00×10^{-18}	0.14×10^{-24}	0.14×10^{-24}	0.70×10^{-24}
41750	0.23952	1.33	7.97	0.15	0.15	0.74
42250	0.23669	1.51	6.86	0.16	0.16	0.78
42750	0.23392	1.26	5.79	0.17	0.17	0.83
43250	0.23121	1.49	4.83	0.18	0.18	0.87
43750	0.22857	1.31	4.00	0.19	0.19	0.92
44250	0.22599	1.26	4.24	0.21	0.20	0.97
44750	0.22346	1.52	2.55	0.22	0.21	1.02
45250	0.22099	1.11	1.97	0.23	0.22	1.08
45750	0.21855	1.06	1.52	0.25	0.23	1.13
46250	0.21622	7.91×10^{12}	1.17	0.26	0.25	1.19
46750	0.21390	8.19	8.57×10^{-19}	0.28	0.26	1.26
47250	0.21164	7.12	6.50	0.30	0.27	1.32
47750	0.20942	4.37	5.10	0.31	0.29	1.39
48250	0.20725	2.54	4.15	0.33	0.30	1.46
48750	0.20513	2.07	3.51	0.36	0.32	1.54
49750	0.20304	1.69	3.18	0.38	0.34	1.61
49750	0.20100	1.49	3.18	0.40	0.35	1.69

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- (1) Mean wavenumber (cm^{-1}) for a spectral range of 500 cm^{-1} between $41250 \pm 250 \text{ cm}^{-1}$ and $49750 \pm 250 \text{ cm}^{-1}$.
(2) Corresponding mean wavelength to the wavenumber in μm (vacuum) for calculation of the various mean cross sections.
(3) Solar irradiances adopted at the top of the earth's atmosphere ($\Delta\nu = 500 \text{ cm}^{-1}$).
(4) Ozone absorption cross section. See text.
(5) Molecular scattering cross section of O_2 . From formula (5). See text.
(6) Molecular scattering cross section of air. From formula (1). See text.
(7) Equivalent scattering cross section (0) for atmospheric application, formula (3). To be compared with the Herzberg continuum cross section in aeronomic applications.

where

$$x = 0.389 \lambda + 0.09426 \lambda^{-1} - 0.3228, (\lambda \text{ in } \mu\text{m}), \quad (2)$$

The atmospheric scattering cross section was converted for use with the total number of O_2 molecules so as to allow easier comparisons with O_2 direct atmospheric absorption, i.e. an equivalent cross section.

$$\sigma_{MS}(O_2)_{\text{EQU.}} = [n(M)/n(O_2)] \sigma_{MS}(M) = 4.8 \sigma_{MS}(M) \quad (3)$$

where $n(M)$ and $n(O_2)$ are the atmospheric total and molecular oxygen concentrations (cm^{-3}), respectively. The numerical values of $\sigma_{MS}(M)$ and $\sigma_{MS}(O_2)_{\text{EQU}}$ for intervals of 500 cm^{-1} are given in Table 1 (columns 6 and 7, respectively).

In a laboratory measurement of O_2 absorption cross sections, the correction for molecular scattering (deduced from Bates' formulas, 1984) is given by the following formula

$$\sigma_{MS}(O_2) = 1.52 \times 10^{-25} \left(\frac{0.24}{\lambda} \right)^{4+x} \text{ cm}^2 \quad (4)$$

where

$$x = 64.329 \lambda + 3.464 / \lambda - 28.681 \quad (5)$$

with a precision better than $\pm 0.25\%$ from $\lambda = 0.24 \mu\text{m}$ to $0.20 \mu\text{m}$; this formula is extended to $0.19 \mu\text{m}$ excluding the regions of the rotational lines of the Schumann-Runge bands. Table 1, column 5, gives also the numerical values of $\sigma_{MS}(O_2)$ from $0.242 \mu\text{m}$ to $0.190 \mu\text{m}$ to be recommended for correction of the Rayleigh scattering in all laboratory measurements of the O_2 absorption cross sections outside of the action of the rotational lines of the bands of the O_2 Schumann-Runge system.

The O_3 cross sections used in this calculation were taken from Ackerman (1971) between 242.4 nm and 213.9 nm and directly deduced from laboratory data of Inn and Tanaka (1953) at wavelengths less than 214 nm. Measurements of Molina and Molina (not yet published 1986) indicate that between 216 and 242 nm the differences are not greater than $\pm 2\%$, and may reach -5% near 210 ± 5 nm. The adopted O_3 absorption cross sections are also given in Table 1, column 4, for intervals of 500 cm^{-1} .

Thus the total atmospheric transmittance T_{Atm} is given by

$$T_{\text{Atm}} = T_{O_2} \times T_{\text{MS}} \times T_{O_3} \quad (6)$$

with the optical depths,

$$\tau_{O_2} = \sigma_{\text{HER}} N(O_2) \quad (7)$$

$$\tau_{\text{MS}} = 4.8 \sigma_{\text{MS}} N(O_2) \quad (8)$$

and

$$\tau_{O_3} = \sigma_{O_3} N(O_3)$$

where $N(O_2)$ and $N(O_3)$ are the total number of oxygen and ozone molecules (cm^{-2}) corresponding to a vertical column above a certain level and deduced from the standard model (Table 2). For various solar zenith angles (χ), equation (6) can be written :

$$(T_{\text{Atm}})^{f(\chi)} = (T_{O_2} \times T_{\text{RS}} \times T_{O_3})^{f(\chi)} \quad (9)$$

where $f(\chi)$ is the air mass corresponding for relatively low solar zenith angles to $\sec \chi$.

TABLE 2.- Adopted Standard Model in the Homosphere.

Altitude (km)	Temperature (K)	Pressure (mb)	$n(O_2)$ (cm^{-3})	$n(O_3)$ (cm^{-3})	$N(O_2)$ (cm^{-2})	$N(O_3)$ (cm^{-2})
85	181	4.12×10^{-3}	3.46×10^{13}	5.25×10^7	1.79×10^{19}	3.28×10^{13}
80	181	1.04×10^{-2}	8.71	7.00	4.64	6.34
75	200	2.49	1.89×10^{14}	2.21×10^8	1.13×10^{20}	1.29×10^{14}
70	220	5.52	3.81	7.00	2.51	3.38
65	239	1.14×10^{-1}	7.26	2.21×10^9	5.20	9.98
60	256	2.25	1.33×10^{15}	7.00	1.02×10^{21}	3.09×10^{15}
55	266	4.28	2.44	2.15×10^{10}	1.94	9.58
50	271	7.98	4.47	6.61	3.61	2.95×10^{16}
45	264	1.49×10^0	8.56	2.03×10^{11}	6.74	9.08
40	250	2.87	1.74×10^{16}	6.07	1.30×10^{22}	2.78×10^{17}
35	237	5.75	3.69	1.40×10^{12}	2.59	7.65
30	227	1.20×10^1	8.02	2.52	5.39	1.73×10^{18}
25	222	2.55	1.75×10^{17}	4.32	1.15×10^{23}	3.45
20	217	5.33	3.87	4.77	2.48	5.80
15	217	1.21×10^2	8.48	2.57	5.43	7.66
10	223	2.65	1.80×10^{18}	1.13	1.19×10^{24}	8.67
5	256	5.40	3.21	5.67×10^{11}	2.41	9.01

In order to perform calculations of the solar flux penetration and of the various photodissociation rates, O_2 absorption cross sections based on recent experimental and theoretical data must be adopted for the Herzberg continuum.

THE HERZBERG CONTINUUM

The absorption cross sections of the Herzberg continuum at wavelengths greater than 200 nm based on laboratory measurements made at relatively high pressures before 1980 and extrapolated to low pressure values applicable to the stratosphere must be replaced now by experimental and theoretical results obtained recently (1984-1986).

The determination of the O_2 absorption cross sections by Johnston et al. (1984) for various temperatures between 206 and 327 K leads to the following values : $3.5 \pm 0.2 \times 10^{-24} \text{ cm}^2$, $4.2 \pm 0.2 \times 10^{-24} \text{ cm}^2$, $5.2 \pm 0.2 \times 10^{-24} \text{ cm}^2$, $6.6 \pm 0.3 \times 10^{-24} \text{ cm}^2$ at 225, 220, 215, 210 and 205 nm, respectively, when the O_2 Rayleigh scattering (see Table 1) is deduced from the total absorption cross section. Such values can be obtained to within $\pm 5\%$ by an expression independent of the temperature of the same form as given by Johnston et al. (1984), λ in nm,

$$\sigma_{\text{HER}}(O_2) = 7.5 \times 10^{-24} (199/\lambda) \exp \{- 50 [\ln(199/\lambda)]^2\} \text{ cm}^2 \quad (10)$$

This formula is chosen here to determine the mean absorption cross sections of the Herzberg continuum for intervals of 500 cm^{-1} from the threshold at 242 nm to 210 nm. The deduced numerical values are given in Table 3 (column 4) and are in agreement (better than $\pm 3\%$, except at 234 nm) with the experimental data based on averaging the lowest cross sections obtained by Cheung et al. (1986) with the largest values deduced by Jenouvrier et al. (1986). It should be emphasized that the differences, as shown in Table 3, column 3, between the experimental values may reach $\pm 10\%$ near 230 nm. Such significant differences should

Table 3. - Mean absorption cross sections in the O₂ Herzberg continuum in intervals of 500 cm⁻¹ for wavelengths greater than 200 nm.

Mean λ (nm)	Experimental ⁽¹⁾ Mean value (10 ⁻²⁴ cm ²)	Maximum ⁽²⁾ Difference (%)	Adopted ⁽³⁾ Formula (10 ⁻²⁴ cm ²)	Formula/ Experimental ⁽⁴⁾ (%)
242.42			0.80	
239.52	1.01	+ 1	1.00	- 1
236.69	1.27	+ 3	1.26	+ 0
233.92	1.63	+ 6	1.73	+ 6
231.21	2.04	+ 10	2.10	+ 3
228.57	2.47	+ 11	2.51	+ 2
225.99	3.05	+ 8	2.96	- 3
223.46	3.54	+ 7	3.42	- 3
220.99	4.01	+ 7	3.92	- 2
218.58	4.47	+ 6	4.39	- 2
216.22	5.03	+ 6	4.90	- 3
213.90	5.51	+ 4	5.38	+ 1
211.64	5.90	+ 5	5.84	- 1
209.42	6.30	+ 4	6.26	- 1
207.25	6.60	+ 4	6.64	+ 0
205.13	6.87	+ 5	6.95	+ 1
203.04	.		7.21	
201.00	.		7.39	

(1) The experimental mean value corresponds to the averaged cross section deduced from laboratory data obtained at Harvard Observatory, Cambridge, Mass. and at Reims University, France.

(2) The positive and negative percentages are for Harvard and Reims, respectively.

(3) Formula (10). See Text.

(4) Differences between columns 4 and 2.

be resolved and, for reasons of convenience, cause us to accept the results deduced from formula (10) as given in Table 3, column 4. This formula used as a reference indicates that important differences may still exist at wavelengths less than 210 nm. It can be seen (in Figure 1) that formula (10) leads to the largest values of the O_2 cross section below 210 nm while the theoretical determination of Saxon and Slanger (1986) corresponds to significantly lower cross sections for the Herzberg continuum underlying the Schumann-Runge bands. The adoption (see Figure 1), at wavelengths less than 210 nm, of cross sections corresponding to the median values of the maximum cross section deduced from formula (10) and of the minimum cross section deduced from the theoretical determination of Saxon and Slanger (1986) leads to an increasing difference reaching $\pm 10\%$ at 201 nm (Table 3). Such differences increase up to about $\pm 20\%$, as illustrated in Figure 1, if an extrapolation is made to 190 nm corresponding to the spectral range of the 2-0 to 6-0 Schumann-Runge bands. Thus, the available sets of Herzberg continuum cross sections derived from laboratory measurements seem to reach an acceptable agreement within their experimental accuracy. But the errors of the experimental or theoretical determinations prevent a sufficiently precise extrapolation for the Herzberg continuum underlying the Schumann-Runge bands between 190 and 200 nm. A special laboratory determination at low temperature (≤ 200 K) is needed without introducing line-wing corrected cross sections.

The stratospheric determinations of Herman and Mentall (1982) in the region 205-220 nm are also shown in figure 1; they suggest that the largest Herzberg continuum cross sections considered in our analysis are not in disagreement. A more precise judgment is difficult since the quoted errors have several possible interpretations. Nevertheless, an estimation of the O_2 photodissociation frequency and also of various rates can be made at wavelengths greater than 200 nm.

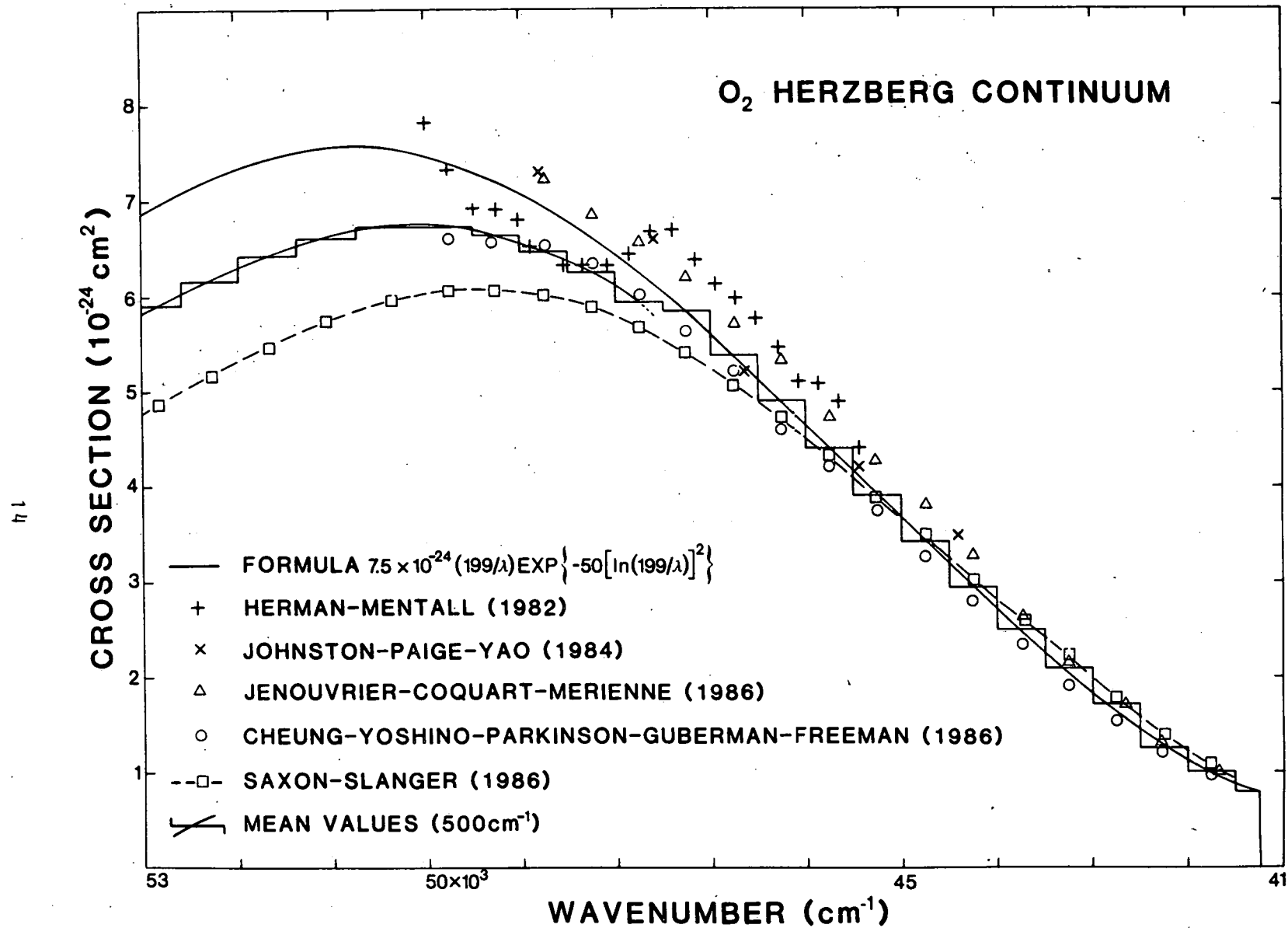


Fig. 1.- Recent experimental and theoretical absorption cross sections of molecular oxygen in the spectral region of the Herzberg continuum.

O₂ PHOTODISSOCIATION RATES

On the basis of experimental and theoretical data available at present, we adopt (cf. Table 3 and Figure 1) the absorption cross sections for the O₂ Herzberg continuum at wavelengths greater than 200 nm as given in Table 4 with their minimum and maximum values for spectral intervals of 500 cm⁻¹. It is possible, therefore, to determine the changes in the rate of photodissociation (between 240 nm and 200 nm) of molecular oxygen in the stratosphere with the parameters of the standard atmosphere (Table 2) and the various other aeronomic parameters (Table 1).

Figure 2 illustrates the variation with height of the stratospheric transmittance leading to a rapid decrease of the spectral irradiances at long wavelengths. The maximum transmittance, as it is well known, occurs near 210 nm (see curve at 30 km).

The O₂ photodissociation frequencies for wavelengths greater than 200 nm are collected in Table 5 based on the absorption cross sections given in Table 4. They yield a value of $5.06 \times 10^{-10} \text{ s}^{-1}$ at the top of the earth's atmosphere with an accuracy of $\pm 5\%$ corresponding to the general variation of the O₂ cross sections indicated in Table 4. It can be seen (Table 5) that there is an isophotolytic level where the photodissociation frequency does not vary with the variation of the O₂ absorption cross section. This isophotolytic level (crossing point from \pm to $\bar{\pm}$) is between 20 and 25 km for an overhead sun, at 25 km for a solar zenith angle of 45°, between 25 and 30 km for 60° and at 30 km for 70°. Such an isophotolytic level, which corresponds to a mean transmittance related to about $N(\text{O}_2) = 1.5 \times 10^{23} \text{ cm}^{-2}$ and $N(\text{O}_3) = 5 \times 10^{18} \text{ cm}^{-2}$, does not depend of the extreme values which are adopted ($\pm 10\%$ instead of $\pm 5\%$, for example) for the O₂ cross sections. In order to illustrate the differences that arise, depending on the solar zenith angle, various O₂ photodissociation frequencies are shown between 20 km and 55 km in

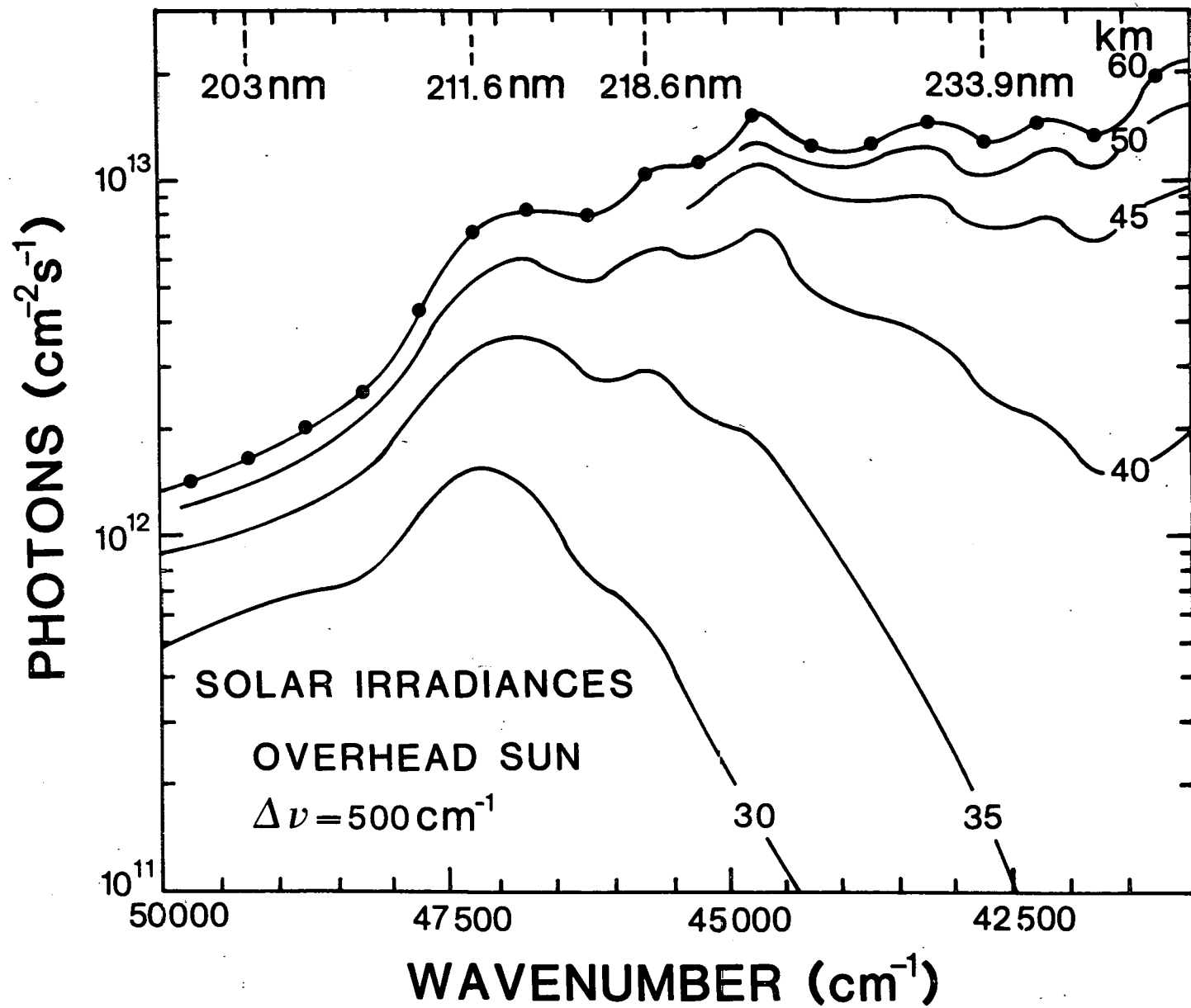


Fig. 2.- Spectral solar irradiances. Number of photons $\text{cm}^{-2} \text{s}^{-1}$ in intervals of 500 cm^{-1} between 60 km and 30 km for overhead sun between 240 and 200 nm.

Table 4.- Adopted absorption cross sections in the Herzberg continuum of O₂ for intervals of 500 cm⁻¹ for the determination of the O₂ photodissociation frequency and of photodissociation rate at various altitudes and solar zenith angles in the region of wavelengths greater than 200 nm.

Mean wavelength (A)	Mean wavenumber (cm ⁻¹)	O ₂ cross section 10 ⁻²⁴ cm ²	Minimum value x	Maximum value x
2424	41250	0.80	0.95	1.05
2395	41750	1.00	0.95	1.05
2367	42250	1.26	0.95	1.05
2339	42750	1.73	0.95	1.05
2312	43250	2.10	0.95	1.05
2285	43750	2.51	0.95	1.05
2259	44250	2.96	0.95	1.05
2234	44750	3.42	0.95	1.05
2209	45250	3.92	0.95	1.05
2186	45750	4.39	0.95	1.05
2162	46250	4.90	0.95	1.05
2139	46750	5.38	0.95	1.05
2116	47250	5.84	0.95	1.05
2094	47750	5.94	0.95	1.05
2072	48250	6.25	0.94	1.06
2051	48750	6.47	0.93	1.07
2030	49250	6.63	0.91	1.09
2010	49750	6.72	0.90	1.10

TABLE 5.- Photodissociation frequency (s^{-1}) of molecular oxygen for wavelengths greater than 200 nm in the Herzberg continuum. Values at the top of the earth's atmosphere and between 65 km and 10 km for various solar angles χ . Standard atmospheric model with attenuation due to atmospheric molecular scattering and ozone absorption.

Altitude (km)	sec χ = 1 $J(O_2)_{HER}$	sec χ = 1.414 $J(O_2)_{HER}$	sec χ = 2 $J(O_2)_{HER}$	sec χ = 3 $J(O_2)_{HER}$
∞	$5.06 \times 10^{-10} (\pm 5\%^*)$	$5.06 \times 10^{-10} (\pm 5\%)$	$5.06 \times 10^{-10} (\pm 5\%)$	$5.06 \times 10^{-10} (\pm 5\%)$
65	5.03	5.02	5.01	4.98
60	5.00	4.97	4.93	4.87
55	4.89	4.82	4.73	4.58
50	4.62	4.45	4.23	3.89
45	3.95	3.60	3.17	2.59
40	2.69	2.18	1.68	1.13
35	1.32	9.11×10^{-10}	5.73×10^{-11}	2.85×10^{-11}
30	$4.92 \times 10^{-11} (\pm 4\%^{**})$	$2.65 \times 10^{-11} (\pm 3\%)$	$1.20 \times 10^{-11} (\pm 2\%)$	$3.45 \times 10^{-12} (\pm 0\%)$
25	$1.14 \times 10^{-11} (\pm 2\%)$	$3.95 \times 10^{-12} (\pm 0\%)$	$9.71 \times 10^{-13} (\pm 3\%)$	$1.10 \times 10^{-13} (\pm 9\%)$
20	$1.32 \times 10^{-12} (\pm 4\%)$	$2.15 \times 10^{-13} (\pm 9\%)$	$1.81 \times 10^{-14} (\pm 18\%)$	$4.16 \times 10^{-16} (\pm 30\%)$
15	$5.97 \times 10^{-14} (\pm 18\%)$	$2.82 \times 10^{-15} (\pm 30\%)$	$4.11 \times 10^{-17} (\pm 50\%)$	$3.39 \times 10^{-20} (\pm 100\%)$
10	$2.70 \times 10^{-16} (\pm 60\%)$	1.30×10^{-18}	7.88×10^{-22}	2.35×10^{-27}

(*) ($\pm x\%$) or ($\bar{\pm} x\%$) corresponds to the variation of $\pm y\%$ in the O_2 absorption cross section as indicated in Table 4 of O_2 cross sections.

(**) Decreases of $J(O_2)_{HER}$ to approximately 10^{-1} , 10^{-2} , 10^{-3} , and 10^{-4} correspond to about $\pm 4\%$, $\pm 0\%$, less than $\pm 10\%$ and less than $\bar{\pm} 20\%$, respectively, with the variation of the O_2 absorption cross sections as indicated in Table 4 ($\pm 5\%$ to $\pm 10\%$).

Figure 3. It is evident that near the stratopause level the variation is small and corresponds to the atmospheric region where the photo-equilibrium conditions for ozone can be applied. In the neighbourhood of the stratopause the total ozone content can be produced in less than a day. Table 6, which gives the O_2 photodissociation rates ($cm^{-3} s^{-1}$) at various heights between 5 and 85 km for certain solar zenith angles ($\lambda > 200$ nm), shows that, near the stratopause, the overhead O_3 concentrations ($10^{11} - 10^{10} cm^{-3}$) are produced in less than 6 hours. Table 6 shows that there is also an isophotolytic production of oxygen atoms corresponding to the isophotolytic level of the O_3 photodissociation frequency. These numerical results are illustrated in Figure 4 which shows that the production of oxygen atoms reaches its maximum in the middle stratosphere as it is already shown.

Since the essential features of the calculations are, in the stratosphere, very sensitive to the aeronomic parameters related to the wavelength, the spectral distributions of the solar irradiance and of the various transmittances and the associated O_2 photodissociation are given in various tables (7 to 16) for various altitudes and overhead sun conditions.

At 60 km (Table 7) where $N(O_2)$ is of the order of $10^{21} cm^{-2}$ and $N(O_3)$ of the order of $3 \times 10^{15} cm^{-2}$ for overhead sun, the optical atmospheric depth is negligible since it is generally around 0.01 except near 240 nm where it reaches about 0.03. The solar irradiance for spectral intervals of $500 cm^{-1}$ varies from about $2 \times 10^{13} photons cm^{-2} s^{-1}$ at 242.4 nm to about $1.5 \times 10^{12} photons cm^{-2} s^{-1}$ at 201 nm, but the O_2 photodissociation frequency for each spectral interval is always at least 2% and not more than 10% of the O_2 total frequency for wavelengths greater than 200 nm. Furthermore, even if the optical depth is small, it can be seen that the various percentages corresponding to the O_2 absorption, the molecular scattering attenuation and the O_3 absorption (in the three last columns of Table 7, respectively) give a clear indication of their relative roles; there is a dominant effect of O_3 at

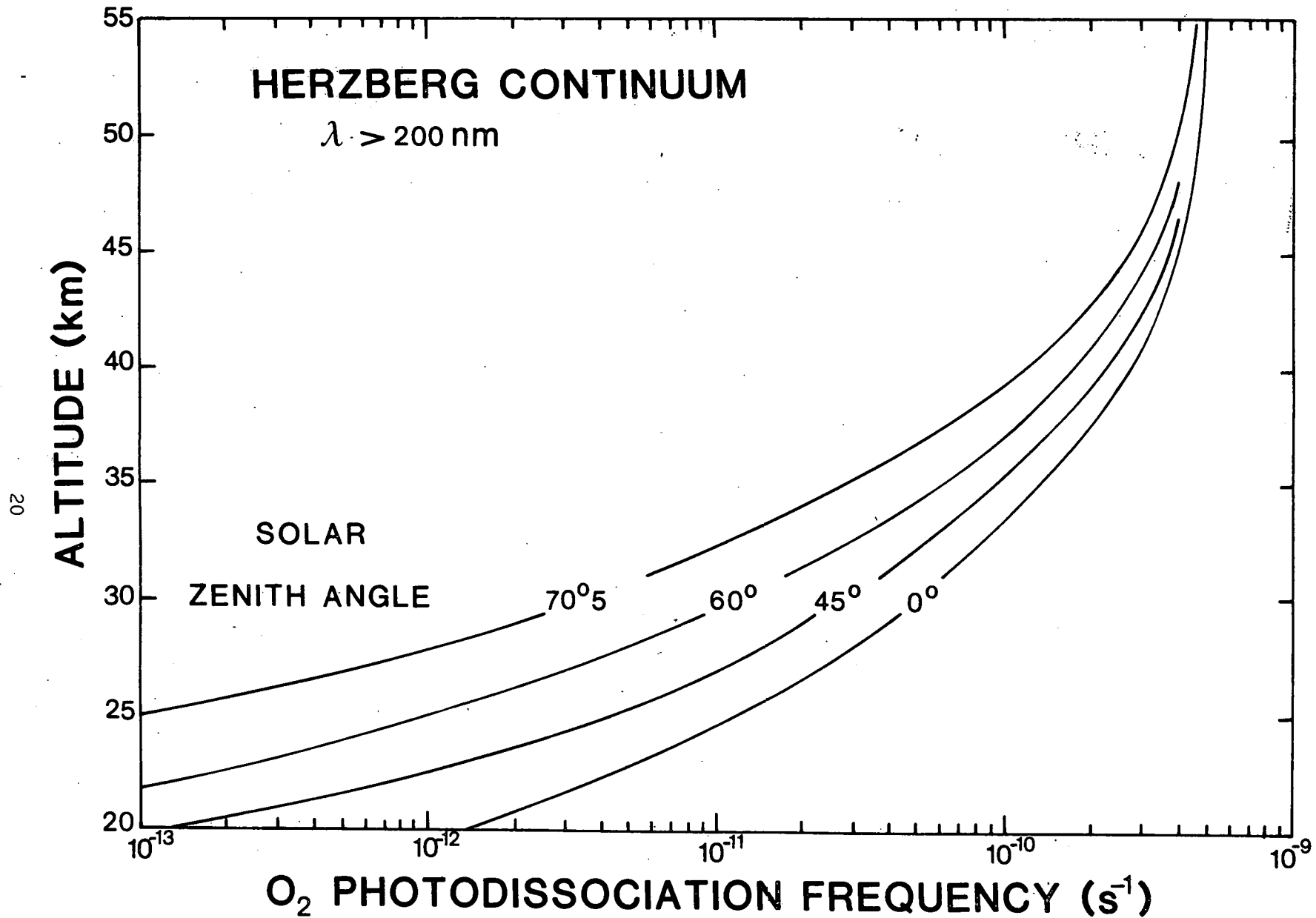


Fig. 3.- Photodissociation frequency of molecular oxygen $J(\text{O}_2)$, (s^{-1}), at stratospheric levels, for the spectral region of wavelengths greater than 200 nm and various solar zenith angles.

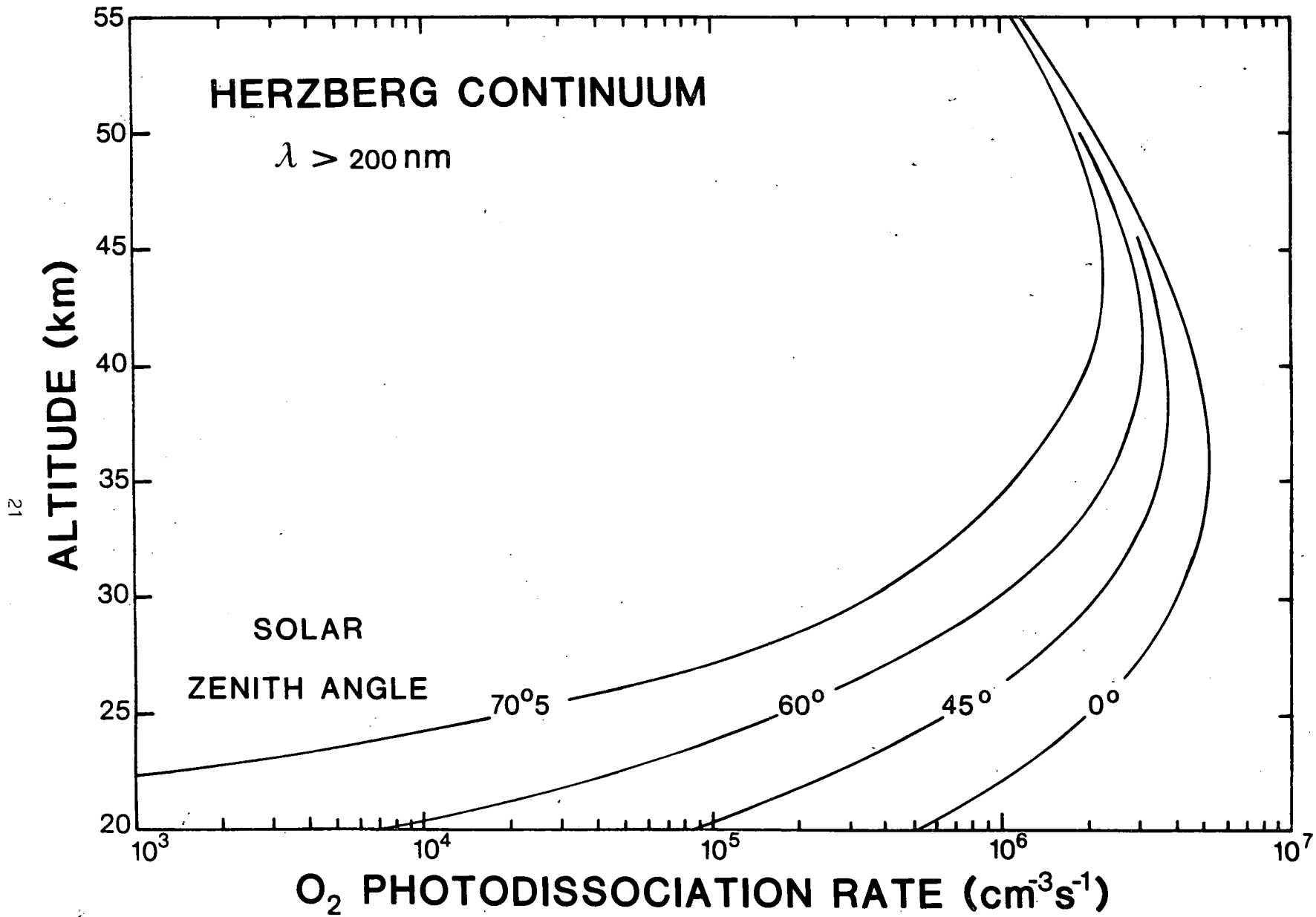


Fig. 4.- Photodissociation rates of molecular oxygen (cm⁻³ s⁻¹) in the stratosphere at various solar zenith angles for the spectral region of wavelengths between 200 and 240 nm.

Table 6.- Photodissociation rates ($\text{cm}^{-3} \text{s}^{-1}$) of molecular oxygen for wavelengths greater than 200 nm in the Herzberg continuum. Values between 85 km and 10 km for various solar zenith angles. Standard atmospheric model with attenuation due to atmospheric molecular scattering and ozone absorption.

Altitude (km)	$\sec \chi = 1$ ($\text{cm}^{-3} \text{s}^{-1}$)	$\sec \chi = 1.414$ ($\text{cm}^{-3} \text{s}^{-1}$)	$\sec \chi = 2$ ($\text{cm}^{-3} \text{s}^{-1}$)	$\sec \chi = 3$ ($\text{cm}^{-3} \text{s}^{-1}$)
85	^(*) 1.75×10^4 ($\pm 5\%$)	1.75×10^4 ($\pm 5\%$)	1.75×10^4 ($\pm 5\%$)	1.75×10^4 ($\pm 5\%$)
80	4.40	4.40	4.40	4.40
75	9.50	9.50	9.50	9.50
70	1.92×10^5 ($\pm 5\%$)	1.92×10^5 ($\pm 5\%$)	1.92×10^5 ($\pm 5\%$)	1.92×10^5 ($\pm 5\%$)
65	3.65	3.64	3.63	3.61
60	6.64	6.61	6.56	6.48
55	1.19×10^6 ($\pm 5\%$)	1.18×10^6 ($\pm 5\%$)	1.15×10^6 ($\pm 5\%$)	1.12×10^6 ($\pm 5\%$)
50	2.07	1.99	1.89	1.74
45	3.38	3.06	2.71	2.22
40	4.68	3.80	2.92	1.97
35	4.87	3.35	2.11 ($\pm 4\%$)	1.05 ($\pm 3\%$)
30	3.98 ($\pm 4\%$)	2.12 ($\pm 3\%$)	9.35×10^5 ($\pm 2\%$)	2.76×10^5 ($\pm 0\%$)
25	1.98 ($\pm 2\%$)	6.88×10^5 ($\pm 0\%$)	1.74 ($\pm 3\%$)	1.75×10^4 ($\pm 9\%$)
20	5.11×10^5 ($\pm 4\%$)	8.31×10^4 ($\pm 9\%$)	7.02×10^3 ($\pm 18\%$)	1.17×10^2 ($\pm 30\%$)
15	5.06×10^4 ($\pm 18\%$)	2.39×10^3 ($\pm 30\%$)	3.48×10^1 ($\pm 50\%$)	< 1
10	4.86×10^2 ($\pm 60\%$)	2.34	< 1	< 1

(*) ($\pm x\%$) or ($\bar{x}\%$) in the values of the O_2 photodissociation rates corresponds to the variation ($\pm 5\%$ to $\pm 10\%$) of the O_2 absorption cross section as indicated in table 4.

Table 7 .- Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 60 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	1.98x10 ¹³	2	0.97	0.03	3	2	95
2395	1.30	3	.97	0.03	4	3	93
2367	1.48	4	.98	0.02	6	3	91
2339	1.23	4	.98	0.02	8	4	88
2312	1.46	6	.98	0.02	11	5	84
2285	1.29	6	.98	0.02	16	6	78
2259	1.24	7	.99	0.01	21	7	72
2234	1.50	10	.99	0.01	28	8	64
2209	1.10	9	.99	0.01	35	10	55
2186	1.05	9	.99	0.01	43	11	46
2162	7.83x10 ¹²	8	.99	0.01	51	12	37
2139	8.11	9	.99	0.01	58	14	28
2116	7.05	8	.99	0.01	64	15	21
2094	4.33	5	.99	0.01	68	15	17
2072	2.43	3	.99	0.01	71	16	13
2051	2.05	3	.99	0.01	73	16	11
2030	1.67	2	.99	0.01	74	16	10
2010	1.47	2	.99	0.01	74	17	10

23

(1) N(O₂) = 1.01 x 10²¹ cm⁻² ; (2) N(O₂) = 3.08 x 10¹⁵ cm⁻²

the longest wavelengths and an increasing effect of O_2 and molecular scattering towards the shortest wavelengths; There is an increasing effect on the transmittance of ozone with decreasing altitudes (Table 8 to 13 and Figure 2) down to 30 km. Below this level (Tables 14, 15 and 16) the O_2 absorption takes an increasing part in the total atmospheric transmittance.

At the stratopause, the optical depth between 240 nm and 200 nm is less than unity for all solar zenith angles less than 70° (Tables 9 and 9a), but at 45 km (Tables 10 and 10a) unit optical depths are reached at the longest wavelengths particularly at large solar zenith angles. It should be stressed that the transmittance for a certain zenith angle, corresponds to the transmittance for a vertical column T_{atm} to a certain power, i.e. T_{atm}^2 for $\sec \chi = 2$ and T_{atm}^3 for $\sec \chi = 3, \dots$, and that the atmospheric optical depth varies accordingly.

Unit optical depth is reached at 30 km for overhead sun (Table 13) at wavelengths less than 210 nm, between 1 and 2 in the spectral interval 200 - 215 nm. The spectral region 210 ± 5 nm corresponds to a peak in the solar flux reaching the lower atmosphere. Furthermore, the spectral region of wavelengths greater than 220 nm is unimportant in the photo-dissociation of molecular oxygen.

Thus, if the accuracy in the absorption cross section of the Herzberg continuum at wavelengths greater than 200 nm is better than $\pm 5\%$ between 200 nm and 240 nm in the neighbourhood of the stratopause (50 ± 10 km), it is only required between 200 and 230 nm in the middle stratosphere (30 ± 5 km) and only needed in the lower stratosphere (20 km) at wavelengths less than 215 nm (Table 15).

It can be also said that the solar irradiance corresponding to the number of photons $cm^{-2} s^{-1}$ in spectral intervals of $500 cm^{-1}$ depending on atmospheric transmittance less than 10^{-2} , i.e. optical depths greater

Table 8 .- Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 55 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	1.87x10 ¹³	2	0.92	0.09	2	1	97
2395	1.23	3	0.92	0.08	2	2	96
2367	1.41	4	0.93	0.07	4	2	94
2339	1.19	4	0.94	0.06	5	3	92
2312	1.41	6	0.95	0.05	8	3	89
2285	1.25	6	0.96	0.04	11	4	85
2259	1.21	7	0.96	0.04	15	5	80
2234	1.47	10	0.97	0.03	20	6	74
2209	1.08	9	0.97	0.03	27	7	66
2186	1.03	9	0.98	0.02	34	9	58
2162	7.73x10 ¹²	8	0.98	0.02	41	10	49
2139	8.02	9	0.98	0.02	49	12	39
2116	6.98	8	0.98	0.02	57	13	30
2094	4.28	5	0.98	0.02	61	14	25
2072	2.40	3	0.98	0.02	66	14	20
2051	2.03	3	0.98	0.02	68	15	17
2030	1.66	2	0.98	0.02	69	16	15
2010	1.46	2	0.98	0.02	69	16	15

(1) N(O₂) = 1.93 × 10²¹ cm⁻²; (2) N(O₃) = 9.57 × 10¹⁵ cm⁻²

Table 9.- Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 50 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	1.56x10 ¹³	1	0.76	0.27	1	1	98
2395	1.04	2	0.79	0.24	2	1	97
2367	1.22	3	0.81	0.21	2	1	92
2339	1.05	4	0.84	0.18	3	2	95
2312	1.28	6	0.86	0.15	5	2	93
2285	1.15	6	0.88	0.13	7	3	90
2259	1.13	7	0.90	0.11	10	3	87
2234	1.39	10	0.91	0.09	14	4	82
2209	1.03	9	0.93	0.08	19	5	76
2186	9.94x10 ¹²	9	0.94	0.06	25	6	69
2162	7.48	8	0.94	0.06	32	8	60
2139	7.80	9	0.95	0.05	41	9	50
2116	6.81	9	0.96	0.04	49	10	41
2094	4.19	6	0.96	0.04	54	12	34
2072	2.35	3	0.96	0.04	59	12	29
2051	1.99	3	0.96	0.04	62	13	25
2030	1.62	3	0.96	0.04	64	14	22
2010	1.43	2	0.96	0.04	64	14	22

(1) N(O₂) = 3.61 x 10²¹ cm⁻²; (2) N(O₃) = 2.95 x 10¹⁶ cm⁻²

Table 9a. - Spectral distribution in the range of the O₂ Herzberg continuum (242-200 nm) of the solar irradiance, of the atmospheric transmittance and of the atmospheric optical depth at various solar zenith angles χ .

Altitude 50 km.

Mean wave- length (Å)	$\chi = 45^\circ$			$\chi = 60^\circ$			$\chi = 70^\circ 5$		
	Solar irradiance	Atm.	Opt.	Solar irradiance	Atm.	Opt.	Solar Irradiance	Atm.	Opt.
	Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500$ cm ⁻¹)	Trans.	Depth	Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500$ cm ⁻¹)	Trans.	Depth	Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500$ cm ⁻¹)	Trans.	Dept
2424	1.39x10 ¹³	0.68	0.38	1.19x10 ¹³	0.58	0.54	9.05x10 ¹²	0.44	0.81
2395	9.45x10 ¹²	0.71	0.34	8.21x10 ¹²	0.62	0.48	6.45	0.48	0.72
2367	1.12x10 ¹³	0.74	0.30	9.93	0.66	0.42	8.05	0.53	0.63
2339	9.76x10 ¹²	0.77	0.25	8.80	0.70	0.36	7.35	0.58	0.54
2312	1.20x10 ¹³	0.81	0.22	1.10x10 ¹³	0.73	0.31	9.42	0.63	0.46
2285	1.09	0.83	0.18	1.01	0.77	0.26	8.86	0.68	0.39
2259	1.08	0.86	0.16	1.01	0.80	0.22	9.06	0.72	0.33
2234	1.33	0.88	0.13	1.27	0.83	0.18	1.16x10 ¹³	0.76	0.27
2209	9.95x10 ¹²	0.90	0.11	9.53x10 ¹²	0.86	0.15	8.83x10 ¹²	0.80	0.23
2186	9.65	0.91	0.09	9.31	0.88	0.13	8.73	0.82	0.19
2162	7.28	0.92	0.08	7.06	0.89	0.11	6.68	0.84	0.17
2139	7.62	0.93	0.07	7.42	0.91	0.10	7.07	0.87	0.15
2116	6.66	0.94	0.06	6.51	0.91	0.09	6.22	0.88	0.13
2094	4.10	0.94	0.06	4.01	0.92	0.08	3.85	0.88	0.13
20.72	2.30	0.94	0.06	2.25	0.92	0.08	2.16	0.88	0.12
2051	1.95	0.94	0.06	1.91	0.92	0.08	1.83	0.88	0.12
2030	1.59	0.94	0.06	1.56	0.92	0.08	1.49	0.88	0.12
2010	1.40	0.94	0.06	1.37	0.92	0.08	1.31	0.88	0.13

Table 10. - Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 45 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	8.92x10 ¹²	< 1	0.44	0.83	1	< 1	99
2395	6.37	2	0.48	0.74	1	1	98
2367	7.98	2	0.53	0.64	1	1	98
2339	7.33	3	0.58	0.54	2	1	97
2312	9.42	5	0.63	0.46	3	1	96
2285	8.90	6	0.39	0.39	4	2	94
2259	9.14	7	0.73	0.73	6	2	92
2234	1.17x10 ¹³	10	0.77	0.26	9	3	88
2209	8.97x10 ¹²	9	0.81	0.21	12	4	84
2186	8.90	10	0.84	0.18	17	4	79
2162	6.83	8	0.86	0.15	22	5	77
2139	7.25	10	0.88	0.12	30	7	64
2116	6.40	9	0.90	0.10	38	8	54
2094	3.96	6	0.91	0.10	43	10	47
2072	2.23	4	0.91	0.09	49	11	40
2051	1.89	3	0.91	0.09	53	12	35
2030	1.55	3	0.91	0.09	55	12	33
2010	1.36	3	0.91	0.09	55	13	32

(1) N(O₂) = 6.74 x 10²¹ cm⁻²; (2) N(O₃) = 9.08 x 10¹⁶ cm⁻²

Table 10a.- Spectral distribution in the range of the O₂ Herzberg continuum (242-200 nm) of the solar irradiance, of atmospheric transmittance and of the atmospheric optical depth at various solar zenith angles χ .

Altitude 45 km

Mean wave-length (A)	$\chi = 45^\circ$			$\chi = 60^\circ$			$\chi = 70^\circ 5$		
	Solar irradiance Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500 \text{ cm}^{-1}$)	Atm. Trans. (tot.)	Opt. Depth (tot.)	Solar irradiance Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500 \text{ cm}^{-1}$)	Atm. Trans. (tot.)	Opt. Depth (tot.)	Solar irradiance Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500 \text{ cm}^{-1}$)	Atm. Trans. (tot.)	Opt. Depth (tot.)
2424	6.33x10 ¹²	0.31	1.17	3.90x10 ¹²	0.19	1.65	1.71x10 ¹²	0.08	2.48
2395	4.70	0.35	1.04	3.06	0.23	1.47	1.46	0.11	2.21
2367	6.14	0.41	0.90	4.23	0.28	1.27	2.24	0.15	1.91
2339	5.85	0.46	0.77	4.26	0.34	1.08	2.48	0.20	1.63
2312	7.80	0.52	0.65	5.96	0.40	0.92	3.77	0.25	1.37
2285	7.59	0.58	0.55	6.05	0.46	0.77	4.11	0.31	1.16
2259	8.01	0.63	0.45	6.63	0.53	0.64	4.81	0.38	0.96
2234	1.05x10 ¹³	0.69	0.37	9.01	0.59	0.52	6.94	0.46	0.78
2209	8.29x10 ¹²	0.74	0.30	7.26	0.55	0.42	5.87	0.53	0.64
2186	8.27	0.78	0.25	7.47	0.70	0.35	6.27	0.59	0.53
2162	6.42	0.81	0.21	5.89	0.75	0.29	5.08	0.64	0.44
2139	6.89	0.84	0.17	6.41	0.78	0.24	5.67	0.69	0.37
2116	6.12	0.86	0.15	5.75	0.81	0.21	5.16	0.72	0.31
2094	3.81	0.87	0.14	3.59	0.82	0.20	3.26	0.75	0.29
2072	2.15	0.88	0.13	2.04	0.83	0.18	1.86	0.76	0.28
2051	1.82	0.88	0.13	1.73	0.84	0.18	1.58	0.76	0.27
2030	1.49	0.88	0.12	1.42	0.84	0.17	1.30	0.77	0.26
2010	1.31	0.88	0.13	1.24	0.84	0.17	1.14	0.76	0.27

Table 11. - Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 40 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	1.65x10 ¹²	< 1	0.08	2.51	< 1	< 1	99
2395	1.43	< 1	0.11	2.23	1	< 1	99
2367	2.20	1	0.15	1.93	1	1	98
2339	2.46	1	0.19	1.64	1	1	98
2312	3.77	3	0.25	1.38	2	1	97
2285	4.14	4	0.32	1.15	3	1	96
2259	4.88	5	0.39	0.95	4	1	95
2234	7.08	9	0.47	0.76	6	2	92
2209	6.03	9	0.54	0.61	8	2	90
2186	6.48	10	0.61	0.49	11	3	86
2162	5.29	10	0.67	0.40	16	4	80
2139	5.93	12	0.72	0.32	21	5	74
2116	5.42	12	0.76	0.27	28	7	65
2094	3.44	8	0.79	0.24	34	7	59
2072	1.97	5	0.80	0.22	39	9	52
2051	1.68	4	0.81	0.21	43	10	47
2030	1.38	4	0.82	0.20	46	10	44
2010	1.21	3	0.81	0.20	46	11	43

(1) N(O₂) = 1.29 × 10²² cm⁻²; (2) N(O₃) = 2.77 × 10¹⁷ cm⁻²

Table 12.- Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 35 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	2.03x10 ¹⁰	< 1	9.9x10 ⁻⁴	6.91	< 1	< 1	99
2395	2.88	< 1	2.2x10 ⁻³	6.13	< 1	< 1	99
2367	7.58	< 1	5.0	5.29	1	< 1	99
2339	1.42x10 ¹¹	< 1	1.1x10 ⁻²	4.49	1	< 1	99
2312	3.45	< 1	2.3	3.77	1	1	98
2285	3.64	1	4.3	3.14	2	1	97
2259	9.57	2	7.6	2.58	3	1	96
2234	1.93x10 ¹²	5	0.13	2.06	4	1	95
2209	2.16	6	0.19	1.63	6	2	92
2186	2.88	10	0.27	1.30	9	2	89
2162	2.76	10	0.35	1.05	12	3	85
2139	3.58	14	0.44	0.83	17	4	79
2116	3.60	16	0.51	0.66	23	5	72
2094	2.43	11	0.56	0.59	28	6	66
2072	1.45	7	0.59	0.52	33	7	60
2051	1.27	7	0.61	0.49	37	8	55
2030	1.05	6	0.62	0.47	40	9	51
2010	9.24x10 ¹¹	5	0.62	0.48	40	9	51

(1) N(O₂) = 2.59 x 10²² cm⁻²; N(O₃) = 7.64 x 10¹⁷ cm⁻²

than 4.5 can be neglected in the determination of the total effect on the O_2 photodissociation when the optical depth reaches unity near 205 nm (see Table 13).

At 25 km, where the optical depth is of the order of 2 near 205 nm for overhead sun conditions, the solar irradiances with transmittance less than 2.5×10^{-3} , i.e. optical depths greater than 6, can be neglected (Table 14) in the calculation of $J(O_2)_{HER}$.

At 20 km, where the optical depth is not less than 4 near 205 nm for overhead sun (Tables 15), the accuracy of the total solar irradiances at wavelengths greater than 200 nm depends only on the accuracy of the observational and experimental data between 200 nm and 215 nm, i.e. a very short spectral interval. At 15 km (Table 16) in the same conditions, the transmittance is less than 10^{-3} corresponding to atmospheric optical depths greater than 7.

A more detailed analysis of the various tables may lead to additional conclusions on the various roles of the photolysis parameters related to the spectral region of wavelengths greater than 200 nm related to the O_2 Herzberg continuum.

Finally, it may be also useful to consider the sensitivity of the atmospheric transmittance to uncertainties in the O_2 cross sections corresponding to the Herzberg continuum at various stratospheric altitudes. After fixing the values of the molecular scattering attenuations and of the O_3 absorption cross sections, a variation of $\pm 10\%$ is adopted for the O_2 absorption cross sections and introduced in the calculation of the atmospheric transmittance corresponding to 500 cm^{-1} intervals. The results given in Table 17 show that, for overhead sun, there is virtually no difference in the transmittances at the stratopause level and in the upper stratosphere. In the middle stratosphere at 30 km, the differences reach only $\pm 3\%$ to 4% at wavelengths less than 215 nm. The larger percent changes in the transmittance observed at short wave-

Table 13. - Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 30 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	3.56x10 ⁶	-	1.8x10 ⁻⁷	> 10	< 1	< 1	100
2395	1.35x10 ⁷	-	1.0x10 ⁻⁶	> 10	< 1	< 1	99
2367	1.62x10 ⁸	-	6.7x10 ⁻⁶	> 10	1	< 1	99
2339	5.22	-	4.1x10 ⁻⁵	10.10	1	< 1	99
2312	3.14x10 ⁹	-	2.1x10 ⁻⁴	8.46	1	1	98
2285	1.12x10 ¹⁰	<< 1	8.6x10 ⁻⁴	7.06	2	1	97
2259	3.88	< 1	3.1x10 ⁻³	5.78	3	1	96
2234	1.49x10 ¹¹	1	9.8x10 ⁻³	4.62	4	1	95
2209	2.86	2	2.6x10 ⁻²	3.66	5	2	93
2186	5.77	5	5.4x10 ⁻²	2.91	8	2	90
2162	7.62	7	9.6x10 ⁻²	2.34	11	3	86
2139	1.31x10 ¹²	14	0.16	1.83	16	4	80
2116	1.58	19	0.22	1.46	21	5	74
2094	1.20	15	0.28	1.29	26	6	68
2072	7.76x10 ¹¹	10	0.32	1.14	31	7	62
2051	7.17	10	0.35	1.06	35	8	57
2030	6.09	9	0.36	1.02	38	9	53
2010	5.29	8	0.36	1.04	38	9	53

(1) N(O₂) = 5.38 x 10²² cm⁻²; N(O₃) = 1.72 x 10¹⁸ cm⁻²

Table 14. - Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 25 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2424	< 1	-	-	> 10	< 1	< 1	100
2395	1.35	-	1.0x10 ⁻¹²	> 10	< 1	< 1	99
2367	6.75x10 ²	-	4.5x10 ⁻¹¹	> 10	1	< 1	99
2339	2.13x10 ⁴	-	1.7x10 ⁻⁹	> 10	1	< 1	99
2312	6.51x10 ⁵	-	4.4x10 ⁻⁸	> 10	1	1	98
2285	9.37x10 ⁶	-	7.2x10 ⁻⁷	> 10	2	1	97
2259	1.16x10 ⁸	-	9.2x10 ⁻⁶	> 10	3	1	96
2234	1.42x10 ⁹	<< 1	9.3x10 ⁻⁵	9.28	4	1	95
2209	7.16	< 1	6.5x10 ⁻⁴	7.35	6	2	92
2186	3.03x10 ¹⁰	1	2.9x10 ⁻³	5.86	9	2	89
2162	7.06	3	8.9x10 ⁻³	4.72	12	3	85
2139	2.01x10 ¹¹	9	2.5x10 ⁻²	3.71	17	4	79
2116	3.36	18	4.7x10 ⁻²	2.97	22	5	73
2094	3.16	17	7.2x10 ⁻²	2.63	27	6	67
2072	2.33	14	9.5x10 ⁻²	2.34	32	7	61
2051	2.35	14	0.11	2.18	36	8	56
2030	2.07	13	0.12	2.10	39	9	52
2010	1.77	11	0.12	2.13	40	9	51

(1) N(O₂) = 2.48 x 10²³ cm⁻²; N(O₃) = 5.80 x 10¹⁸ cm⁻²

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Table 15. - Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 20 km and Overhead Sun (1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ (Δν = 500 cm ⁻¹)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2312	4.95	-	3.3x10 ⁻¹³	> 10	1	1	98
2285	4.70x10 ²	-	3.6x10 ⁻¹¹	> 10	3	1	96
2259	3.28x10 ⁴	-	2.6x10 ⁻⁹	> 10	4	1	95
2234	1.91x10 ⁶	-	1.3x10 ⁻⁷	> 10	5	2	93
2209	3.50x10 ⁷	-	3.2x10 ⁻⁶	> 10	8	2	90
2186	4.00x10 ⁸	<< 1	3.8x10 ⁻⁵	> 10	10	3	87
2162	1.97x10 ⁹	< 1	2.5x10 ⁻⁴	8.30	14	4	82
2139	1.10x10 ¹⁰	4	1.3x10 ⁻³	6.62	20	5	75
2116	2.78	15	3.9x10 ⁻³	5.41	27	6	67
2094	3.40	16	7.8x10 ⁻³	4.86	32	7	61
2072	2.96	16	1.2x10 ⁻²	4.39	38	8	54
2051	3.29	18	1.6x10 ⁻²	4.14	42	9	49
2030	3.00	17	1.8x10 ⁻²	4.03	44	10	46
2010	2.48	14	1.7x10 ⁻²	4.10	45	10	45

Table 16.- Spectral distribution in the range of the O₂ Herzberg continuum (242 nm - 200 nm) of the solar irradiance, of the O₂ photodissociation frequency, of the atmospheric transmittance and of the atmospheric optical depth with its divisions (%) into the O₂ absorption (1), the atmospheric scattering attenuation and the O₃ absorption (2).

Altitude 15 km and Overhead Sun^(1,2)

Mean wavelength (A)	Solar irradiance Photons cm ⁻² s ⁻¹ ($\Delta\nu = 500 \text{ cm}^{-1}$)	O ₂ Dissociation (%)	Total atmospheric transmittance	Atmospheric optical depth	O ₂ absorption (%)	Molecular scattering (%)	O ₃ absorption (%)
2259	2.49	<< 1	2.0×10^{-12}	> 10	6	2	92
2234	4.50×10^3	<< 1	3.0×10^{-10}	> 10	8	3	89
2209	2.06×10^5	<< 1	1.9×10^{-8}	> 10	12	3	85
2186	4.67×10^6	<< 1	4.4×10^{-7}	> 10	16	4	80
2162	3.74×10^7	< 1	4.7×10^{-6}	> 10	22	5	73
2139	3.16×10^8	3	3.9×10^{-5}	10.2	29	7	64
2116	1.01×10^9	14	1.4×10^{-4}	8.68	37	8	55
2094	1.39	17	3.2×10^{-4}	8.05	42	9	49
2072	1.26	17	5.2×10^{-4}	7.54	48	10	42
2051	1.41	19	6.8×10^{-4}	7.29	52	11	37
2030	1.24	17	7.4×10^{-4}	7.22	54	12	34
2010	9.51×10^8	13	6.4×10^{-4}	7.36	54	13	33

TABLE 17.- Spectral transmittances in the regions of the O_2 Herzberg continuum for intervals of 500 cm^{-1} and overhead sun conditions.

Altitudes : 50 to 20 km.

Wavelength (A)	50 km	45 km	40 km	35 km	30 km	25 km	20 km
2424	$0.76_{\pm 0\%}^{(*)}$	$0.44_{\pm 0\%}$	$8.11 \times 10^{-2}_{\pm 0\%}$	$9.93 \times 10^{-4}_{\pm 0\%}$			
2395	0.79	.48	0.11	2.17×10^{-13}			
2367	0.81	.53	0.15	5.02			
2339	0.84	.58	0.19	1.13×10^{-2}	$4.14 \times 10^{-5}_{\pm 0\%}^{(**)}$		
2312	0.86	.63	0.25	2.32	$2.11 \times 10^{-4}_{\pm 1\%}$		
2285	0.88	.68	0.32	4.31	8.55		
2259	0.90	.73	0.39	7.60	$3.08 \times 10^{-3}_{\pm 2\%}$		
2234	0.91	.11	0.47	9.80	9.80	$9.34 \times 10^{-5}_{\pm 4\%}$	
2209	0.93	.81	0.54	0.19	2.58×10^{-2}	6.45×10^{-4}	
2186	0.94	.84	0.61	0.27	5.44	$2.86 \times 10^{-3}_{\pm 5\%}$	$3.77 \times 10^{-5}_{\pm 12\%}$
2162	0.945	.86	0.67	0.35	9.63	$\pm 6\%$ 8.92	$\pm 6\%$ $2.49 \times 10^{-4}_{\pm 13\%}$
2139	0.95	.89	0.72	$0.44_{\pm 1\%}$	0.16	2.46×10^{-2}	$1.34 \times 10^{-3}_{\pm 14\%}$
2116	0.96	.90	0.76	0.51	0.22	4.73	3.90 $\pm 16\%$
2094	0.96	$.91_{\pm < 0.5\%}$	0.79 $\pm 1\%$	0.56	0.28	7.23 $\pm 7\%$	7.79 $\pm 17\%$
2072	0.96	.91	0.80	0.59	0.32	9.53 $\pm 8\%$	$1.21 \times 10^{-2}_{\pm 18\%}$
2051	0.96	.915	0.81	0.61	0.35 $\pm 4\%$	0.11	1.59 $\pm 19\%$
2030	0.96	.915	0.82	0.62	0.36	0.12 $\pm 9\%$	1.77 $\pm 20\%$
2010	0.96	.915	0.81	0.62	0.35	0.12	1.66 $\pm 20\%$ (***)

(*) The percentages (+x%) shown in the Table correspond to a variation of $\pm 10\%$ in the O_2 absorption cross sections and no variation in the O_3 absorption cross sections.

(**) Transmittances has less than 10^{-5} are not shown in the Table.

(***) Values of + x% at various solar zenith angles χ compared with the values of overhead sun conditions increase by a factor of at least $\sec \chi$.

lengths are associated with the relative increasing role of the O_2 absorption from 240 to 200 nm compared with that of O_3 . At 25 km the differences increase up to $\pm 9\%$. At 20 km, where the O_2 absorption plays a more important role than the O_3 absorption and where the total optical depth is greater than 5, the differences in the spectral transmittance (lower than 10^{-3}) may reach $\pm 20\%$ for a variation of $\pm 10\%$ in the O_2 absorption cross section.

CONCLUSIONS

The data presented here represent the parameters that have a significant role in the photolysis of oxygen at wavelengths greater than 200 nm. Further refinement in the calibration of cross sections in the Herzberg continuum cannot change the mean absolute values of the O_2 photodissociation rate for wavelengths longer than 200 nm by more than $\pm 10\%$ at altitudes above 20 km. The fact that there are isophotolytic levels in the middle atmosphere, between about 25 and 30 km for various solar zenith angles leads to the conclusion that much smaller differences are found for atmospheric optical depths greater than unity. Much larger differences than the difference in the O_2 cross sections occur only when the atmospheric optical depths are greater than 4 in the 205 ± 5 nm spectral region. In any case, the global behaviour will remain above the stratopause level, i.e. a low value of $J(O_2)$ of about $5 \times 10^{-10} \text{ s}^{-1}$ with a spread corresponding exactly to the differences in the O_2 absorption cross sections. Thus the uncertainties of this input parameter cannot modify the conclusion that the production of oxygen atoms in the neighbourhood of the stratopause is less than it was considered a few years ago. Other photodissociation processes such as the photodissociation of $^{16}O^{18}O$ molecules proposed by Cicerone and McCrumb (1980) or the photodissociation from the excited state $O_2(^1\Delta_g)$ suggested by Frederick and Cicerone (1985) cannot constitute significant sources of atomic oxygen (Blake et al., 1984, and Simonaitis and Leu, 1985, respectively). The production of odd oxygen atoms from the photodissociation of $^{16}O^{18}O$

molecules cannot be more than $3 \pm 1\%$ of the production by $^{16}\text{O}_2$ of oxygen atoms at mesopause levels near 70 ± 10 km. On the other hand, the photodissociation of metastable O_2 would require an absorption cross section which should be several thousand times greater than the actual cross sections of extremely weak transitions. A further inference resulting from the present analysis is that the spectral transmittances in the stratosphere down to at least 25 km, as shown in Table 17, are obtained with good accuracy and, therefore, that the uncertainties are small in the upper and middle stratosphere. This is important since it must be considered in the analysis of the uncertainties in the photodissociation rates of nitrous oxide and various chlorofluorocarbons.

Nevertheless, an uncertainty remains in the analysis of the treatment of ultraviolet radiation between 200 and 240 nm. For proper interpretation, in this spectral region, of the attenuated solar radiation field at stratospheric altitudes, a special series of calculation must be done to determine the importance of uncertainties which may result from possible differences occurring in the ozone absorption cross sections at wavelengths less than 240 nm. This will be the subject of another paper. In addition, since the molecular scattering cross section is known with high accuracy, it will be also possible to determine more precisely the amplification factor due to the additional solar flux available near 200 nm for photodissociation processes at stratospheric levels.

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