

SOLAR IRRADIANCE AND ITS SPECTRAL DISTRIBUTION THROUGH THE TERRESTRIAL  
ATMOSPHERE

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Summary

A review of the general subject of solar UV irradiance of importance to the photochemistry of stratospheric and mesospheric ozone is given. An analysis is made of the spectral solar irradiance and of its atmospheric transmissivity in the various regions associated with the absorption of molecular oxygen and of ozone.

1. Introduction

A review of the general subject of solar UV irradiance of importance to the photochemistry of stratospheric and mesospheric ozone requires the simultaneous analysis of the solar spectral irradiance of wavelengths greater than 100 nm and of its atmospheric transmittance which depends essentially on the absorption of molecular oxygen and ozone. It can be assumed that molecular nitrogen plays a role in the atmospheric transmittance only by its scattering cross section which is of the order of  $5.6 \times 10^{-24} \text{ cm}^2$  at Lyman-alpha (Dalgarno et al., 1967). The total scattering cross section ( $\sigma_{RS}$ ) in the homosphere can be determined by a simple formula (Nicolet, 1984), based on a recent theoretical determination by Bates (1984), ( $\lambda$  in  $\mu\text{m}$ ),

$$\sigma_{RS} = 4.02 \times 10^{-28} / \lambda^4 + x \text{ cm}^2 \quad (1)$$

where

$$x = 0.389 + 0.09426/\lambda - 0.3228 \quad (2)$$

The formula accounts for the degree of depolarisation which varies with the wavelength and, as an example, leads to the following values for the total cross section

( $\mu\text{m}$ )	0.190	0.195	0.200	0.205	0.210	0.220
$\sigma \times 10^{-25}$	4.63	4.07	3.60	3.19	2.84	2.54
$\sigma \times 10^{-24} (*)$	2.22	1.95	1.73	1.53	1.37	1.22

The last line (\*) corresponds to the scattering cross section related to the total number of  $\text{O}_2$  molecules  $\text{cm}^{-2}$  in order to lead to an immediate comparison of the role of the molecular scattering with that of the  $\text{O}_2$

absorption in the determination of the atmospheric optical depth.

Thus, the degree of penetration of solar radiation is determined by the absorption of  $O_2$  and  $O_3$  (mainly) and by the molecular scattering of the air molecules. In consequence, the way in which ozone chemistry in the mesosphere and stratosphere can be influenced by variations (with wavelength and time) in the solar radiation flux is controlled primarily by  $O_2$  and its direct product  $O_3$ . The solar spectrum must, therefore, be divided in various spectral regions related to the variations of the  $O_2$  and  $O_3$  absorptions.

## 2. Spectral regions from 100 nm to the visible

First,  $O_2$  is photodissociated in various spectral ranges of wavelengths less than 242 nm. The solar radiation is successively absorbed :

- (1) in the mesosphere, at the Lyman alpha line (121.57 nm) since the absorption cross section is only of the order of  $10^{-20} \text{ cm}^2$ .
- (2) in the thermosphere, at wavelengths less than 175 nm in the region of the Schumann-Runge continuum with absorption cross sections between  $10^{-18}$  and  $10^{-17} \text{ cm}^2$ .
- (3) in the whole mesosphere down to the upper stratosphere, in the region (200 to 175 nm) of the Schumann-Runge bands with a cross section varying from about  $10^{-19}$  to  $10^{-22} \text{ cm}^2$ .
- (4) in the stratosphere, at wavelengths less than 242 nm in the region of the Herzberg continuum with low absorption cross sections of the order or less than  $10^{-23} \text{ cm}^2$ .

Second,  $O_3$  is important because of its absorption of solar radiation in three spectral regions :

- (5) in the stratosphere, in the spectral region of the Hartley band at wavelengths less than 315 nm with a cross section varying between  $10^{-17} \text{ cm}^2$  and about  $10^{-19} \text{ cm}^2$ . At wavelengths less than 242 nm there is a simultaneous absorption by  $O_2$  and  $O_3$ . The Hartley band leads to the ozone photodissociation with  $O_2^*$  and  $O^*$  in their first excited level.
- (6) in the lower stratosphere and in the troposphere, in the spectral regions of the Huggins bands where the cross sections vary between  $10^{-19}$  and  $10^{-22} \text{ cm}^2$  and depend strongly on the temperature. These bands corresponds to the limit of the  $O_3$  ultraviolet absorption.
- (7) in the stratosphere from the ozone peak to ground level, in the visible region (450 - 850 nm) with the Chappuis bands with a low absorption cross section from about  $5 \times 10^{-21}$  to  $10^{-22} \text{ cm}^2$ . These bands play an important role in the photodissociation of  $O_3$  not only in the troposphere, but also in the major part of the stratosphere.

## 3. The Schumann-Runge continuum at $\lambda < 175 \text{ nm}$

All the oxygen atoms that are produced in the thermosphere by absorption of solar radiation at wavelengths less than 175 nm are transported downward by diffusion before recombining by a three-body association a few km above the mesopause. Recent analysis (Nicolet, 1981 and references contained therein) shows that the minimum solar irradiance corresponding to the quiet sun is of the order of  $6 \times 10^{11} \text{ photons cm}^{-2} \text{ s}^{-1}$  (Radiation temperature about 4500 K) and that the maximum observed for active sun conditions is about  $1.5 \times 10^{12} \text{ photons cm}^{-2} \text{ s}^{-1}$  ( $T_R = 4750 \text{ K}$ ).

All recent observations by Mount and Rottman (1983a, b; 1984) confirm that the total number of oxygen atoms produced from a minimum to a maximum of a solar activity cycle varies from about  $(1.25 \pm 0.25) \times 10^{12}$  to  $(3.0 \pm 0.5) \times 10^{12}$  oxygen atoms  $\text{cm}^{-2} \text{sec}^{-1}$ . These thermospheric atoms produced by photodissociation lead to variations with solar activity on the upper boundary conditions near the mesopause.

#### 4. The Lyman-alpha radiation at 121.57 nm

The solar hydrogen Lyman alpha radiation which is of great importance in mesospheric chemistry has been subject of many astrophysical and aeronomical studies (see Nicolet 1984a, d and references contained therein). The irradiance of the line and its variation with solar activity must be considered in the determination of the photoionization of NO and of the photodissociation of  $\text{CH}_4$ , of  $\text{CO}_2$  and particularly of  $\text{H}_2\text{O}$  and of  $\text{O}_2$ . Its mesospheric absorption by molecular oxygen, and therefore its atmospheric transmissivity, depend on wavelength and temperature. The solar H Lyman-alpha line is characterized by a profile with a central reversal, and wings extending to about  $\pm 0.175 \text{ nm}$  where the intensity reaches about 1% of that the peak (Figure 1). The unit effective optical depth for solar Lyman alpha is reached near  $N = 10^{20}$   $\text{O}_2$  molecules  $\text{cm}^{-2}$  (near 75 km for an overhead sun, or near 70 km for a solar zenith angle of  $60^\circ$ ). This depth rises to about 8 near 60 km for an overhead sun, namely for  $N = 10^{21}$ . With a mean temperature of  $T = 230 \text{ K}$ , the solar H Lyman alpha transmittance

$$T_{\text{O}_2}(\text{Ly } \alpha) = \exp[-2.115 \times 10^{-18} N^{0.8855}] \quad (3)$$

This formula can be used as a basis for all calculations when appropriate photodissociation-absorption cross sections are simultaneously adopted. For  $\text{CH}_4$ ,  $\text{CO}_2$  and NO constant photodissociation cross sections must be adopted since there is no detailed analysis (high resolution) of the spectrum at Lyman alpha. As far as  $\text{H}_2\text{O}$  is concerned, a mean cross section can be determined (Nicolet, 1984d), with an excellent accuracy,

$$\sigma_{\text{D}}(\text{H}_2\text{O})_{\text{Ly}\alpha} = (1.57 \pm 0.05) \times 10^{-17} \text{ cm}^2 \quad (4)$$

if the experimental data of Lewis et al. (1983) are adopted. Since the absorption of Lyman alpha by oxygen depends strongly on both temperature and wavelength, its effective absorption-photodissociation cross section  $\sigma_{\text{D}}(\text{O}_2)$  is given for  $T = 230 \text{ K}$  by the following expression :

$$\sigma_{\text{D}}(\text{O}_2)_{\text{Ly}\alpha} = 1.15 \times 10^{-18} / N^{0.1175} \text{ cm}^2 \quad (5)$$

A global analysis, covering more than a solar cycle, of observations made by various satellites for the irradiance of H Lyman-alpha at the top of the earth's atmosphere, leads to the expression (photons  $\text{cm}^{-2} \text{s}^{-1}$ )

$$q_{\infty}(\text{Ly}\alpha) = 2.5 \times 10^{11} \left[ 1 + 0.2 \frac{F_{10.7}^{-65}}{100} \right] \quad (6)$$

if  $2.5 \times 10^{11}$  photons  $\text{cm}^{-2} \text{sec}^{-1}$  is accepted for the irradiance of a quiet sun.

To conclude, solar H Lyman-alpha plays a leading role in the photo-dissociation of CO<sub>2</sub> and of H<sub>2</sub>O. At its absorption peak, the life time of water vapor is only of a few days and its concentration must decrease rapidly with height. The observations from Spacelab (Lippens et al. 1984) show that the H<sub>2</sub>O number density (Figure 2) decreases by about a factor of ten between 70 and 75 km and between 75 km and 80 km.

### 5. The Schumann-Runge band region at $\lambda < 200$ nm

The study of the absorption of the Schumann-Runge bands poses a difficult problem arising from the rotational structure

TABLE I.- Cross sections ( $\sigma$  cm<sup>2</sup>) and transmittance T(O<sub>2</sub>) in molecular oxygen (N = cm<sup>-2</sup>) for the (5-0) band of the Schumann-Runge system (mean wavelength 193.3 nm).

Temperature	190	210	230	250	270	300 K
$\sigma_{MAX}$ (10 <sup>-21</sup> )	1.00	1.01	1.02	1.04	1.06	1.12
N = 10 <sup>21</sup> ; $\sigma_A$ (10 <sup>-22</sup> )	2.35	2.48	2.64	2.83	3.08	3.58
N = 10 <sup>22</sup> ; $\sigma_A$ (10 <sup>-23</sup> )	6.99	7.55	8.27	9.18	10.30	12.22
N = 10 <sup>21</sup> ; T(O <sub>2</sub> )	0.79	0.78	0.77	0.75	0.73	0.70
N = 10 <sup>22</sup> ; T(O <sub>2</sub> )	0.50	0.47	0.44	0.40	0.36	0.30

related not only to the oscillator strengths and rotational line widths of all bands, but also to the temperature. Figure 3 illustrates differences in the optical thickness that are due to the line width and temperature. The (5-0) band is characterized by a strong sensitivity to the temperature which is greater than that caused by changes of line widths. Another problem in the spectral region of the Schumann-Runge bands is that of the solar irradiance. At the present time it is not yet possible, from the limited number of observations, to deduce, with any certainty, the absolute irradiance and the exact effect of changing solar activity. An increase by a factor of 1.11 + 0.04 in the mesosphere may be accepted for the photodissociation of O<sub>2</sub> if the solar activity action is characterized by increases of 20%, 15%, 10% and 5% for the four groups of bands (19-0) to (15-0), (14-0) to (10-0), (9-0) to (6-0) and (5-0) to (2-0), respectively (Nicolet 1984b). A limitation to an accurate determination derives also from the fact that in the region of the (v',0) bands with v' < 6 which are stratospherically important, the Herzberg continuum plays a role. In their studies of the atmospheric absorption, all authors (1970-1980, see Nicolet 1981) used the theoretical values of Jarman and Nicholls which are too high. In practice, parameterisations of detailed calculations (Frederick and Hudson 1980a, b; Nicolet and Peetermans, 1980; Nicolet, 1981; Allen and Frederick, 1982) can provide simple expressions for application to atmospheric models. A new attempt to resolve these problems requires new observational data such as those given by Solar Mesosphere Explorer and laboratory studies as obtained by Yoshino et al. (1982) with high resolution to insure that exact rotational line widths can be determined.

## 6. The Herzberg continuum at $\lambda < 242$ nm

Recent values of the solar irradiance at  $\lambda > 200$  nm obtained by satellites and rockets (see Nicolet, 1983, with references contained therein; Mount and Rottman, 1983a, b and 1984; Mentall et al. to be published) lead to mean values for  $\Delta\lambda = 1$  nm which differ as much as 10%. Such differences are due to difficulties of making accurate measurements and are unconnected with changes with solar activity. Nevertheless, the solar activity is detected around 240 nm by the 27-day variations observed in the irradiance. Such results show that the photodissociation of  $O_2$ , i.e. the formation of  $O_3$ , is related to solar activity when the photodissociation of  $O_3$  is practically independent of solar activity. Because the absorption cross section is very small in the Herzberg continuum, the various laboratory measurements are susceptible to large errors (Nicolet 1983, for references contained therein and Fig. 4). According to recent laboratory measurements (Cheung et al., 1984; Johnston et al. 1984; Ackerman and Biaumé, (unpublished) at wavelengths greater than 200 nm, measurements made in June 1969), there must be an reestimation of the first experimental cross sections. The cross section near 210 nm may be as low as  $5 \times 10^{-24}$  cm<sup>2</sup>. Stratospheric measurements (Frederick and Mentall, 1982; Herman and Mentall 1982; Anderson and Hall, 1983) lead to derived  $O_2$  cross sections which are smaller than the Herzberg continuum values generally used before 1982. (It may be added that nighttime stratospheric observations were made by Pirre, Rigaud and Huguenin in May 1983. These authors reject the new results and consider that old laboratory measurements are confirmed by their observations; according to a poster shown at the Symposium).

The existence of discrepancies in the absolute number densities of  $O_3$  or of  $N_2O$ ,  $CF_2Cl_2$ ,  $CFCl_3$ , ... in the upper stratosphere between photochemical models and observations made at mean latitudes is related to the discrepancies in the absolute absorption cross sections of  $O_2$  (and  $O_3$ ) in the region of the Herzberg continuum at  $210 \pm 20$  nm.

## 7. The Hartley band at $\lambda < 310$ nm

The spectral region from approximately 315 nm to about 200 nm corresponding to the  $O_3$  Hartley band is of prime importance for the whole of atmospheric chemistry, and considerable attention has been paid to its photolysis leading to products being formed in the first excited states ( $O_2^*$  and  $O^*$ , respectively). New measurements are being made (Bass and Paur, Brian et al., Freeman et al., Molina; and report by Hudson at this symposium) and therefore this problem cannot be discussed here. Nevertheless, it is important to show that previous measurements can be compared with the new measurements. Nicolet (1981) has tabulated, as representative of the best available data, the results of the analysis of Ackerman (1971) made in 1969 and based on the work of Vigroux (1953 and 1969), Inn and Tanaka (1959) and Hearn (1961). Since these values have been available for several years for atmospheric application ( $\Delta\nu = 500$  cm<sup>-1</sup>), a comparison has been made with the laboratory data obtained by Bass at 228 K (private communication). The comparison shown in Table II indicates that no significant difference is observed between 235 and 280 nm. At greater wavelengths the temperature effect leads to increasing differences reaching about 10% at 300 nm. Since there is practically no difference for a

TABLE II. Ozone absorption cross section ( $\Delta\nu = 500 \text{ cm}^{-1}$ ) in the Hartley band.

$\Delta\nu(\text{cm}^{-1} \times 10^{-2})$	$\Delta\lambda(\text{nm})$	$\sigma(\text{cm}^2)^{(*)}$	$\sigma(\text{cm}^2)^{(**)}$	Ratio
430 - 425	235 - 233	$5.79 \times 10^{-18}$	$5.92 \times 10^{-18}$	1.02
425 - 420	238 - 235	6.86	6.98	1.02
420 - 415	241 - 238	7.97	8.06	1.01
415 - 410	244 - 241	9.00	9.12	1.01
410 - 405	247 - 244	$1.00 \times 10^{-17}$	$1.00 \times 10^{-17}$	1.00
405 - 400	250 - 247	1.07	1.08	1.01
400 - 395	253 - 250	1.11	1.12	1.01
395 - 390	256 - 253	1.12	1.14	1.02
390 - 385	260 - 256	1.11	1.11	1.00
385 - 380	263 - 260	1.03	1.05	1.02
380 - 375	267 - 263	$9.43 \times 10^{-18}$	$9.57 \times 10^{-18}$	1.01
375 - 370	270 - 267	8.23	8.29	1.00
370 - 365	274 - 270	6.81	6.86	1.00
365 - 360	278 - 274	5.31	5.35	1.00
360 - 355	282 - 278	3.99	3.93	0.98
355 - 350	286 - 282	2.84	2.71	0.95
350 - 345	290 - 286	1.92	1.75	0.91
345 - 340	294 - 290	1.14	1.05	0.92
340 - 335	298 - 294	$6.60 \times 10^{-19}$	$6.04 \times 10^{-19}$	0.92
335 - 330	303 - 298	3.69	3.34	0.91
330 - 325	308 - 303	1.97	1.73	0.88

spectral region of 50 nm width (230-280 nm) where the cross section reaches values greater than  $10^{-18} \text{ cm}^2$ , the uncertainties involved in atmospheric modelling are not relevant to this spectral region. However, the use in atmospheric calculations of the  $\text{O}_3$  cross sections at wavelengths greater than 290 nm requires a careful analysis of the temperature effect associated with the quantum yield of the  $\text{O}(^1\text{D})$  production, particularly in the lower stratosphere. At 300 nm, the  $\text{O}_3$  cross section increases from  $T = 200 \text{ K}$  to  $240 \text{ K}$ ,  $270 \text{ K}$  and  $300 \text{ K}$  by 5%, 10% and 15%, respectively. This temperature dependence should be considered carefully for modelling applications. Additional work is needed since, in the ultraviolet region, at wavelengths greater than 300 nm, the Huggins bands (Figure 5) play a role in the absorption and the reported cross section variations with temperature are greater than the errors tolerable in the ozone observations.

#### 8. Note

Various other aspects have not been discussed here. As an example, the atmospheric conditions must be taken into account so that effects due to anisotropic scattering and the albedo in the solar radiation field can be treated.

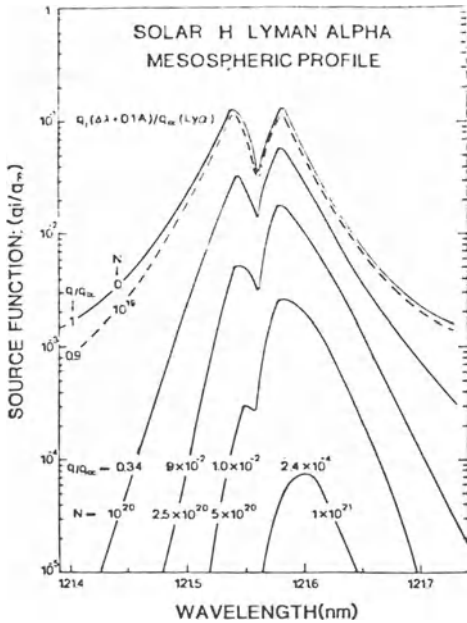


Figure 1

The variation (and deformation) of the profile of the solar H Lyman-alpha line, i.e. of the source function defined by  $q_i(\Delta\lambda = 0.1 \text{ \AA})/q_{\infty, \text{Ly}\alpha}(\Delta\lambda = 3.5 \text{ \AA})$ , with increasing number  $N$  of  $\text{O}_2$  absorbing molecules ( $q$  = number of photons  $\text{cm}^{-2} \text{ s}^{-1}$ ). Six curves are shown for  $N = 0, 10^{19}, 2.5 \times 10^{20}, 5 \times 10^{20}$  and  $10^{21} \text{ cm}^{-2}$ . Approximate values of the effective transmittance of the H Lyman-alpha irradiances, i.e. the ratio  $q(\text{Ly}\alpha)/q_{\infty}(\text{Ly}\alpha)$ , are also given for the various profiles.

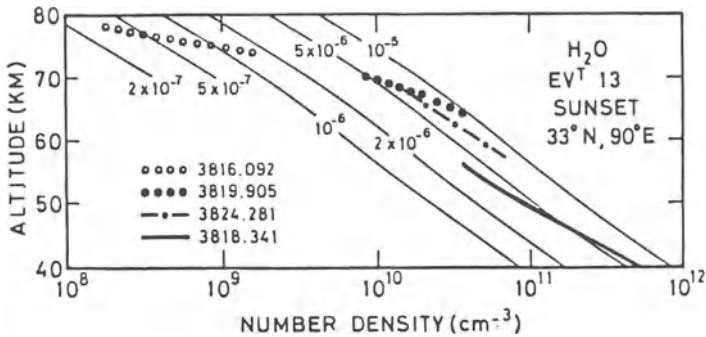


Figure 2

Water vapor number densities versus altitude retrieved using only lines in the regions where their curve of growth is quasi linear. The nearly parallel lines represent constant volume mixing ratios from  $10^{-7}$  to  $10^{-9}$ .

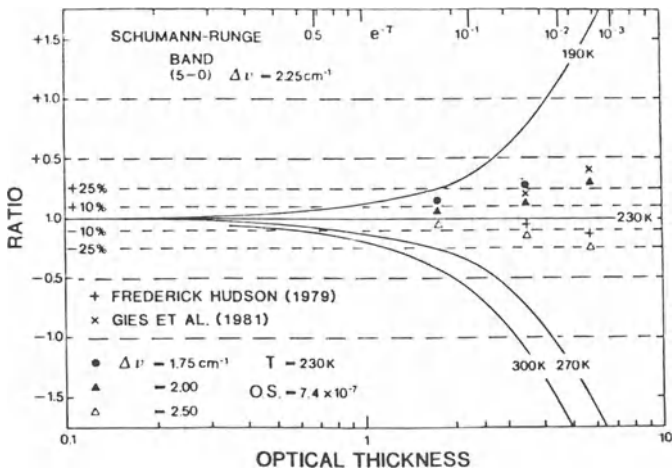


Figure 3

Illustration of differences (+ %) in the transmittance of the (5-0) Schumann-Runge band with the temperature and line width if  $T = 230$  K,  $\Delta\nu = 2.25 \text{ cm}^{-1}$ , and oscillator strength =  $7.4 \times 10^{-7}$ . Curves drawn for  $T = 190, 270,$  and  $300$  K show that the sensitivity to temperature is high and is greater than that caused by the variation of line widths.

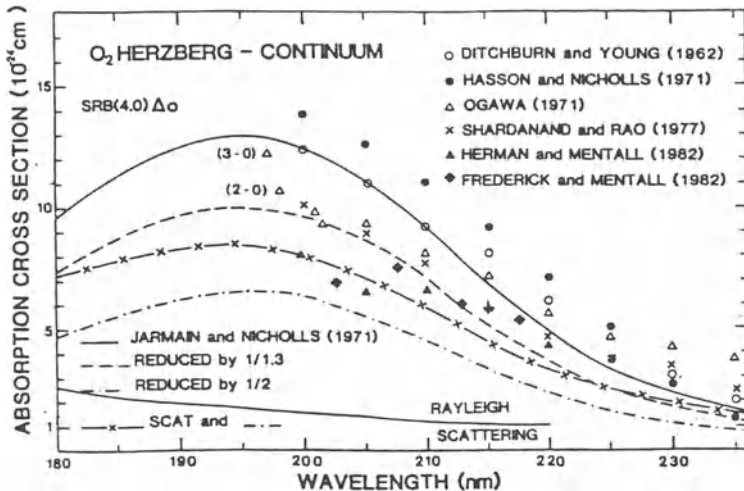


Figure 4

Experimental and theoretical values of the absorption cross-section in the Herzberg continuum of  $O_2$ . The extreme experimental values differ by a factor of 2. If absorption due to atmospheric Rayleigh scattering is taken into account, the theoretical value of Jarmain-Nicholls could be reduced by a factor of 2 to account for the observational cross-sections deduced by Herman-Mentall and Frederick-Mentall.



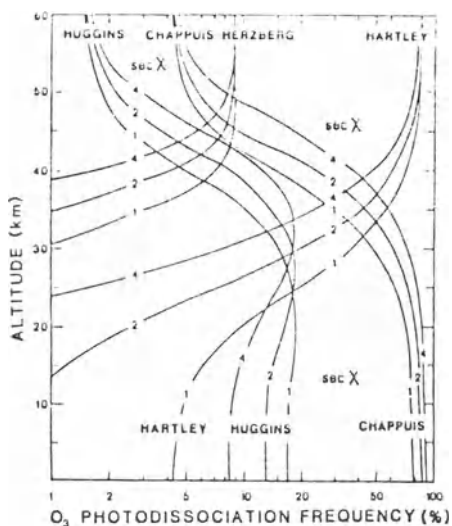


Figure 5

Photodissociation of ozone (%) as a function of altitude and solar zenith angle ( $\sec \chi = 1, 2$  and  $4$ ) for the main spectral regions: Herzberg continuum to 240 nm, Hartley band to 310 nm and Huggins bands beyond 310 nm. The Chappuis bands cover the visible region.

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