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Aeronomical aspects of mesospheric photodissociation : Processes resulting from the solar H Lyman alpha line

by

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## FOREWORD

This article entitled "Aeronomical aspects of mesospheric photodissociation : Processes resulting from the solar H Lyman alpha line" gives the various numerical parameters to be used in the mesosphere. It will be published in Planetary and Space Science.

## AVANT-PROPOS

L'article intitulé "Aeronomical aspects of mesospheric photodissociation : Processes resulting from the solar H Lyman alpha line" explicite les paramètres qui doivent intervenir dans les applications mésosphériques. Il sera publié dans Planetary and Space Science.

## VOORWOORD

Het artikel "Aeronomical aspects of mesospheric photodissociation : Processes resulting from the solar H Lyman alpha line" formuleert de verschillende parameters die moeten gebruikt worden in de mesosfeer. Het zal verschijnen in Planetary and Space Science.

### VORWORT

Der Artikel "Aeronomical aspects of mesospheric photodissociation : Processes resulting from the solar H Lyman alpha line" formuliert die verschiedenen Parameter die müssen benützt werden in der Mesosphäre. Er wird herausgegeben werden in Planetary and Space Science.

## AERONOMICAL ASPECTS OF MESOSPHERIC PHOTODISSOCIATION : PROCESSES RESULTING FROM THE SOLAR H LYMAN ALPHA LINE

by

## Marcel NICOLET

#### Abstract

An analysis is made of the photodissociation and photoionization processes in the mesosphere due to the solar H Lyman-alpha line. The irradiance of the line and its variation with solar activity are considered in the determination of the photodissociation of  $CH_4$ ,  $CO_2$ ,  $H_2O$  and  $O_2$ , and of the photoionization of NO. Lyman-alpha contributes directly to these processes in the mesosphere after its absorption, which depends on wavelength and temperature, by molecular oxygen. The H Lyman-alpha radiation considered for mesospheric processes is characterized by a profile of an emission line with a central reversal, and wings extending to about  $\pm 1.75$  A where the intensity reaches about 1% of that of the peak. Simple formulas are deduced for the photodissociation optical depths and frequencies and these take into account, the various solar activity conditions and the different spectral characteristics of each molecule.

#### Résumé

Une analyse détaillée des processus de photodissociation et de photoionisation mésosphériques a été effectuée pour le domaine spectral de la radiation solaire H-Lyman alpha. L'irradiance de cette raie et ses variations liées à l'activité solaire ont été considérées en vue de la détermination de la photodissociation mésosphérique de CH<sub>4</sub>, CO<sub>2</sub>, H<sub>2</sub>O et O2 et également de la photoionisation de NO. L'oxygène moléculaire dont l'absorption est fonction à la fois de la longueur d'onde et de la température détermine la pénétration atmosphérique de la radiation Lyman-alpha. Mais, la contribution de cette radiation aux processus de photodissociation mésosphérique est déterminée par le profil de l'émission solaire caractérisée par un renversement central et une extension des ailes atteignant 1.75 A de chaque côté du centre lorsque l'intensité est réduite à 1%. Des formules ont été déduites à la suite d'une analyse détaillée pour chaque molécule et permettent de déterminer simplement les épaisseurs optiques et les fréquences de photodissociation tout en fixant le facteur de transmission atmosphérique.

## Samenvatting

Er werd een gedetailleerde analyse gemaakt van de processen van mesosferische fotodissociatie en foto-ionisatie voor het spectraal gebied van de H-Lyman alpha lijn. De uitstraling van deze lijn en haar variaties verbonden aan de zonneaktiviteit werden beschouwd om de mesosferische fotodissociatie van  $CH_{4}$ ,  $CO_{2}$ ,  $H_{2}O$  en  $O_{2}$  te bepalen, alsook de foto-ionisatie van NO. De moleculaire zuurstof waarvan de absorptie tegelijkertijd functie is van de golflengte en van de temperatuur, bepaalt de atmosferische doordringing van de Lyman-alpha straal. De mate waarin deze straling echter bijdraagt tot de processen van mesosferische fotodissociatie wordt bepaald door het profiel van de zonne-emissie gekenmerkt door een centrale omkering en een uitstrekking van de vleugels tot ± 1.75 A aan weerszijden van het centrum wanneer de intensiteit verminderd is tot op 1%. Formules werden afgeleid ten gevolge van een gedetailleerde analyse voor iedere molecule en deze laten toe gewoonweg de optische dikten en de frequenties van fotodissociatie te bepalen terwijl de atmosferische transmissiefactor eveneens vastgelegd wordt.

### Zusammenfassung

Man hat eine detaillierte Analyse gemacht von den mesosphärischen Fotodissoziations- und Foto-Ionisierungsprozessen für das Spektralgebiet der H-Lyman alpha Linie. Die Ausstrahlung dieser Linie und ihre Variationen verbunden der Sonnenaktivität wurden an betrachtet zur Bestimmung der mesosphärische Fotodissoziation von  $CH_4$ ,  $CO_2$ ,  $H_2O$  und  $O_2$ , und auch der Foto-Ionisierung von NO. Der Molekularsauerstoff wovon die Absorption zugleicherzeit Funktion ist der Wellenlänge und der Temperatur, bestimmt die atmosphärische Durchdringung der Lyman-alpha Linie. Aber, der Beitrag dieser Strahlung in den mesosphärischen Fotodissoziationsprozessen wird bestimmt durch das Profil der Sonnenemission gekennzeicht durch eine zentrale Umkehrung und eine Ausstreckung der Flügeln bis zu 1.75 A auf beiden Seiten des Zentrums wenn die Intensität vermindert ist bis zu 1%. Formeln wurden abgeleitet infolge eine detaillierte Analyse für jede Molekül und diese Formeln erlauben einfeich die optische Dicken und die Fotodissoziationshäufigkeiten zu bestimmen während des atmosphärischen Transmissionsfaktores auch fixiert wird.

## INTRODUCTION

About 40 years ago (Nicolet, 1945), the suggestion was made that the solar H Lyman-alpha line at 121.56 nm might be responsible for the photoionization of nitric oxide in the D region. Since then, the objective of many observations of the Sun and of the atmosphere has been to study this radiation in all its aspects. It is worth recalling that the above suggestion was based on a triple hypothesis which has since been confirmed by observations : the Lyman-alpha line can pass through a window, in the terrestrial atmosphere, which occurs between two molecular oxygen bands; its irradiance is much greater than that of the nearby solar continuum; and the NO molecule exists in the D region of the ionosphere. However, besides causing photoionization of NO, Lyman-alpha radiation can also cause photodissociation of  $H_2O$ ,  $CH_{\dot{A}}$ ,  $CO_2$  and  $O_2$  (Fig. 1) and other molecules. Since  $O_2$  is one of the main constituents in the terrestrial atmosphere and since it absorbs strongly to beyond 200 nm, the photodissociation processes here are quite different from those in the atmospheres of other planets where oxygen is only a minor constituent.

The experimental data published recently by an Australian (Carver et al., 1977; Lewis al., 1983) permit the et group determination, subject to the accuracy of the data, of the absorption in molecular oxygen as a function of wavelength (at intervals of 0.1 A) and of mesospheric temperature. Besides this, the observations of the profile and the spatial distribution of the solar H Lyman-alpha line that have been made in various circumstances have made it possible to choose a profile appropriate to aeronomic studies (Purcell and Tousey, 1960; Bruner and Parker, 1969; Bruner and Rense, 1969; Prinz, 1973, 1974; Vidal-Madjar, 1977; Lemaire et al., 1978; Barsi et al., 1979; Roussel-Dupré 1979, 1983). This is a composite profile which represents the whole of the solar disk as seen from the top of the Earth's atmo-

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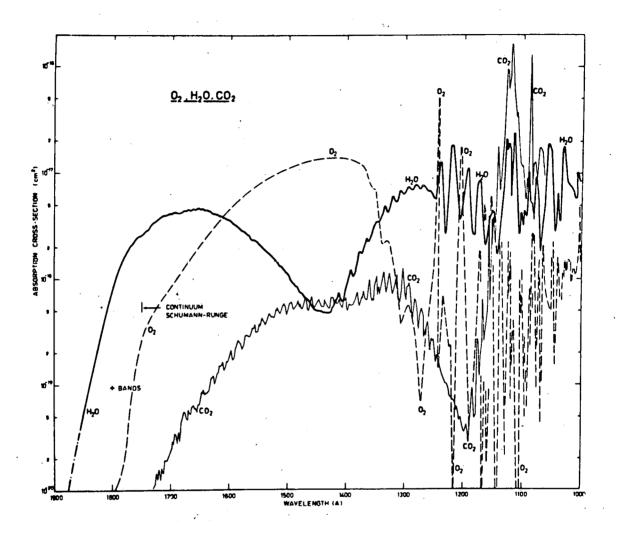


Fig. 1.- Absorption cross sections of  $O_2$ ,  $H_2O$  and  $CO_2$  from 190 to 100 nm taken from Nicolet (1964). The H Lyman-alpha line at 121.6 nm is almost at the centre of a "window" between two  $O_2$  bands. The  $H_2O$ cross section is at least 1000 times greater than that of  $O_2$ .

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sphere. The effective width of the line, to be used in aeronomic calculations, includes the principal central region, but not the distant wings. The profile has a width of 3.5 A and extends from 1213.9 A to 1217.3 A; the intensity at these limits is only 1% of that near the centre, and thus the profile covers more than 99% of the total irradiance.

In aeronomic work it is important also to know the absolute value of the irradiance of Lyman-alpha and how this varies during the solar cycle. This question has been discussed elsewhere by, for example, Bossy and Nicolet, 1981; Bossy, 1983 and Nicolet, 1983a, b. However, for the purposes of the present study of mesospheric conditions, the following expression has been adopted for the irradiance  $q_{n}$  (Ly  $\alpha$ ) of Lyman-alpha at the top of the atmosphere and for the mean distance of the Earth from the Sun :

$$q_{\chi}(Ly \alpha) = 3 \times 10^{10} [1 + 0.2 \frac{F_{10.7}^{-65}}{100}] \text{ photons } \text{cm}^{-2} \text{ s}^{-1}$$
 (1)

where  $3 \times 10^{10}$  photons cm<sup>-2</sup> sec<sup>-1</sup> is a conventional value for the irradiance of the quiet Sun (at the minimum of the solar cycle); this value is further discussed in Section IX. In 98% of cases, the expression in brackets represents the effect of solar activity with an accuracy of  $\pm 10\%$ ; F<sub>10.7</sub> is the radio solar flux at 2800 MHz, published by the National Research Council in Ottawa, and is used as an index of solar activity.

The aim of the present work is to investigate the process of photodissociation by solar Lyman-alpha for a number of molecules (CH<sub>4</sub>, H<sub>2</sub>O, CO<sub>2</sub> and O<sub>2</sub>) bearing in mind their individual characteristics, and to obtain simple results that can be applied directly to the terrestrial mesosphere.

#### II. DETERMINATION OF RATES OF PHOTODISSOCIATION

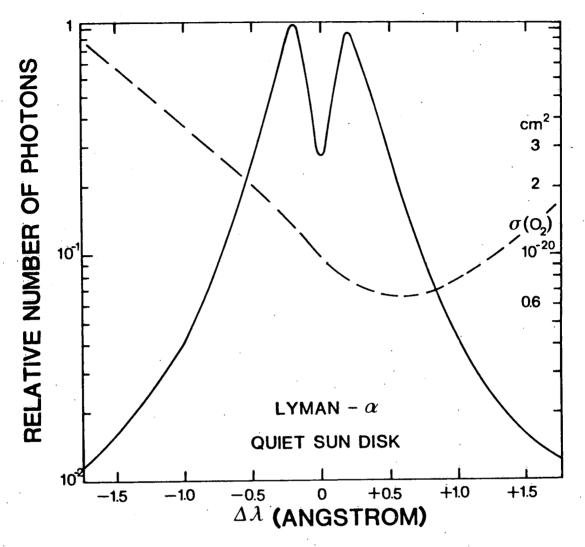
The simplest expression for the rate of dissociation J(XY) of a molecule XY is :

$$J(XY) = \sigma_{D}(XY) q_{\infty} \exp(-\tau_{A}) s^{-1}$$
(2)

where  $\sigma_D$  is the absorption cross-section (cm<sup>2</sup>), assumed constant, of the molecule XY;  $q_{\infty}$  is the total number of photons cm<sup>-2</sup> s<sup>-1</sup> available at the top of the Earth's atmosphere for the same spectral range, and exp(- $\tau_A$ ) is the atmospheric transmittance with  $\tau_A = \sigma_A N$ , where  $\sigma_A$  is the constant absorption cross section (cm<sup>2</sup>) for N molecules (cm<sup>-2</sup>). In this particular case of the solar H Lyman-alpha line, the formula becomes

$$J_{Ly\alpha}(XY) = q_{\infty}(Ly\alpha) \Sigma \sigma_{i}(XY) (q_{i}/q_{\infty}) e^{-\tau_{i}(O_{2})}$$
(3)

where  $q_{\omega}(Ly\alpha)$  is the total number of Lyman-alpha photons as shown in Fig. 2. The <u>photodissociation-absorption</u> cross-section  $\sigma_i(XY)$  will depend on the molecule considered : it is constant for CH<sub>4</sub> and CO<sub>2</sub>, but varies with wavelength for H<sub>2</sub>O, and with temperature and wavelength for O<sub>2</sub>. The source function  $q_i/q_{\omega}$  is adapted to the profile of the solar H Lyman-alpha line which represents the spectral irradiance for a wavelength interval of ± 1.75 A; its minimum value is 1.56 x 10<sup>-3</sup> at 1213.9 A and 1217.3 A, and its maximum is 0.13 at 1215.4 A and 1215.8 A. The optical depth  $\tau_i(O_2)$  for  $\Delta\lambda = 0.1$  A is deduced from O<sub>2</sub> absorption cross sections adapted to temperatures from 170 K to 250 K corresponding to mesospheric conditions; these cross sections are obtained from experimental data published by Lewis et al. (1983).





Mean profile of the solar H Lyman-alpha line ( $\Delta\lambda = 3.5 \text{ A}$ ), for the full disk as seen from the Earth, adopted for the calculation. The corresponding variation of the  $0_2$  absorption cross section at T = 203 K, according to Lewis <u>et al</u>. (1983), is also given. The number of photons q(Ly $\alpha$ ) varies by a factor of 100, and  $\sigma(0_2)$  by a factor of 10.

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When the photodissociation-absorption cross-section  $\sigma^{}_D(XY)$  remains constant over the whole spectral range, Equation (3) can be written

$$J_{Ly\alpha}/J_{Ly\alpha,\infty} = q(Ly\alpha)/q(Ly\alpha,\infty) = \Sigma(q_i/q_{\infty}) e^{-\tau_i(O_2)}$$
(4)

Eq. 4 corresponds to the mesospheric transmittance of the solar H Lyman-alpha irradiance, i.e. to the reduction factor of the number of Lyman-alpha photons (cm<sup>-2</sup> s<sup>-1</sup>) which depends on the total number of  $O_2$  molecules, N cm<sup>-2</sup>, and also on the temperature.

When the mesospheric transmittance of the solar H Lymanalpha line, as defined by Eq. (4), is adopted for the determination of a photodissociation process, the photodissociation-absorption cross section  $\sigma_D(XY)$  is generally an equivalent cross section which may depend on the wavelength (H<sub>2</sub>O) or on the temperature (O<sub>2</sub>).

## III. PENETRATION OF LYMAN-ALPHA INTO THE TERRESTRIAL

## MESOSPHERE

At temperatures representative of the whole of the mesosphere (170K to 250K), the experimental values for the absorption by O<sub>2</sub> of Lyman-alpha radiation in the band 121.550 ± 0.175 nm lead to the following expression for the mean absorption cross section  $\sigma(O_2)_{\tau} = 0$  for zero optical depth  $\tau = 0$  and  $\Delta \lambda = \pm 1.75$  A :

$$\sigma(0_2)_{\tau=0} = (2.35 \pm 0.01) \times 10^{-20} \text{ cm}^2$$

(5)

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where 2.35 is adapted to the temperature 230 K : the value, assumed to be the mean temperature for the mesosphere, which will be used here as the reference temperature.

In the case of Lyman-alpha, it is necessary to take account simultaneously of the line profile and the variation in the  $O_2$  absorption cross section, both of which are illustrated in Fig. 2. The expression for the effective cross section of molecular oxygen at solar H Lyman-alpha then becomes :

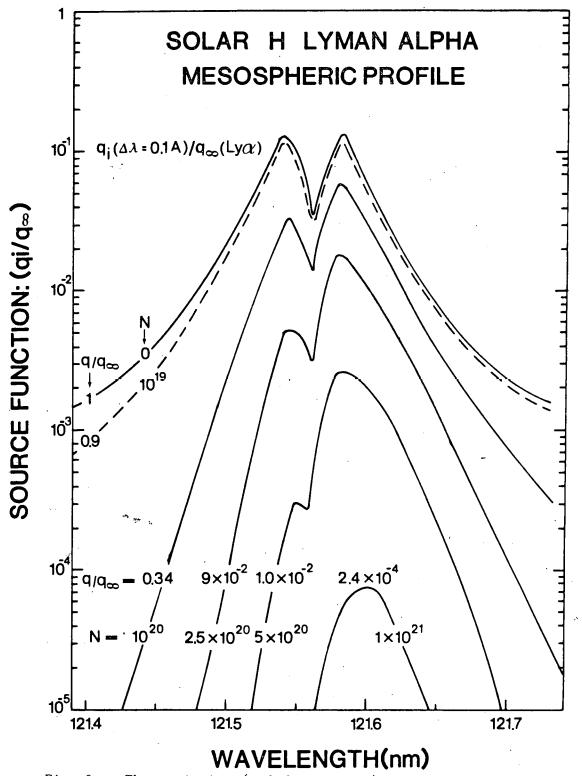
$$\sigma(0_2)_{Ly\alpha,\tau=0} = (2.17 \pm 0.01) \times 10^{-20} \text{ cm}^2$$
 (6)

at  $\tau(O_2) = 0$  and at T = 230 K. Again the variation ± 0.01 covers the temperature range 250 > T > 170 K. This value is less than that given by Eq. (5), since the atmospheric transmittance is adapted to the profile of Lyman-alpha. The calculation of Eq. (4) is, therefore, based on

$$q(Ly\alpha)/q_{\infty}(Ly\alpha) = \sum_{1}^{35} q_{i}e^{-\tau} (O_{2})^{35} -\tau (O_{2})^{-\tau} (O_{2})^{$$

where  $\tau_q(O_2) = \sigma_q(O_2)N$  is the effective optical depth at solar H Lymanalpha : that is, the product of the effective absorption cross section  $\sigma_q(O_2)$  and of the total number N of  $O_2$  molecules per cm<sup>2</sup>. The results of a detailed calculation are illustrated by various curves in Figure 3. It can be seen that there is a continuous deformation of the solar H Lyman-alpha profile due to the differences in the absorption of  $O_2$ molecules at various wavelengths. The maximum of the emission is displaced towards 121.6 nm where the  $O_2$  absorption cross section is least. It is important to note also that the transmittance decreases by a

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The variation (and deformation) of the profile of the solar H Lyman-alpha line, i.e. of the source function defined by  $q_{i,Ly\alpha}(\Delta\lambda = 0.1 \text{ A})/q_{\infty,Ly\alpha}(\Delta\lambda =$ 3.5 A), with increasing number N of O<sub>2</sub> absorbing molecules : (q = number of photons cm<sup>-2</sup> s<sup>-1</sup>). Six curves are shown for N = 0, 10<sup>19</sup>, 10<sup>20</sup>, 2.2 x 10<sup>20</sup>, 5 x 10<sup>20</sup> and 10<sup>21</sup> cm<sup>-2</sup>. Approximate values of the effective transmittance of the H Lyman-alpha irradiances, i.e. the ratio q(Ly\alpha)/q<sub>∞</sub>(Lyα), are also given for the various profiles. factor of about  $10^4$  when the number of O<sub>2</sub> molecules (N) increases from  $10^{19}$  to  $10^{21}$  cm<sup>-2</sup>. For this reason the calculations have not been extended beyond an optical thickness  $\tau_q(O_2) = 10$ ; this implies that N =  $10^{21}$  molecules O<sub>2</sub> cm<sup>-2</sup> is considered to be the practical limit in calculations relating to the effect of solar H Lyman-alpha.

The numerical results show that the effect of temperature on the absorption cross section of  $O_2$  is unimportant for small optical thickness, that is for N <  $10^{20}$  cm<sup>-2</sup>. Starting from the conventional value adopted here for the irradiance at the top of the atmosphere,  $q_{\infty}(Ly\alpha) = 3 \times 10^{10}$  photons cm<sup>-2</sup> s<sup>-1</sup>, the reduced values as the radiation penetrates into the mesosphere are as follows :

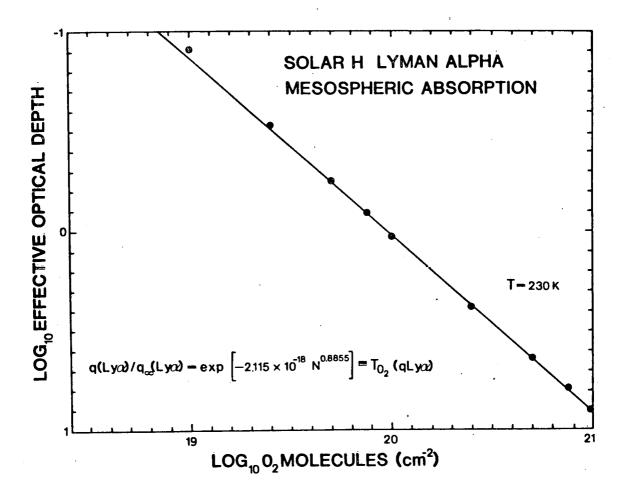
 $\begin{array}{ccccc} N(cm^{-2}) & 0 & 10^{18} & 10^{19} & 2.5 \times 10^{19} & 5 \times 10^{19} & 10^{20} \\ q(cm^{-2} s^{-1}) & 3 \times 10^{11} & 2.96 & 2.65 & 2.22 \pm 0.01 & 1.68 \pm 0.01 & 1.01 \pm 0.02 \end{array}$ 

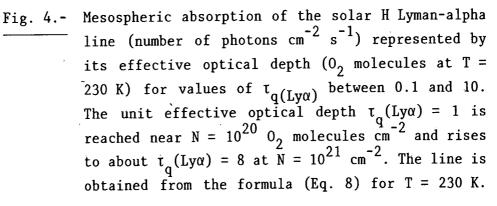
For N =  $10^{20}$  cm<sup>-2</sup>, the effect of temperature in the range 170 K to 250 K is only 2%, but it increases by a factor of more than 2 at N =  $10^{21}$  cm<sup>-2</sup>, and hence for N >  $10^{20}$  cm<sup>-2</sup> the effect of temperature must be considered. Nevertheless, the adoption of a reference temperature of 230 K, adjusted by ± 20 K for  $10^{20}$  < N <  $10^{21}$  cm<sup>-2</sup>, leads to the following simple expression for the mesospheric transmittance T<sub>O<sub>2</sub></sub> (Lya) of solar H Lyman-alpha, the accuracy of which is better than ± 2%

$$\Gamma_{\alpha}$$
 (Ly  $\alpha$ ) = q(Ly  $\alpha$ )/q<sub>\alpha</sub>(Ly  $\alpha$ ) = exp [- 2.115 x 10<sup>-18</sup> N<sup>0.885</sup>] (8)

Figure 4 shows how the effective optical depth  $\tau_q(Ly \alpha)$  increases from 0.1 to 10 as the number of O<sub>2</sub> absorbing molecules increases from 10<sup>19</sup> to 10<sup>21</sup> cm<sup>-2</sup>. The corresponding effective O<sub>2</sub>

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absorption cross section decreases from  $(2.17 \pm 0.01 \times 10^{-20} \text{ cm}^2 \text{ at N} = 0$  (Eq. 6) to  $(1.24 \pm 0.01) \times 10^{-20} \text{ cm}^2$  at N =  $10^{19} \text{ cm}^{-2}$ , and to  $(1.09 \pm 0.02) \times 10^{-20} \text{ cm}^2$  at N =  $10^{20} \text{ cm}^{-2}$  corresponding to about the unit optical depth (see Figure 4). But for N >  $10^{20} \text{ cm}^{-2}$ , the temperature effect increases rapidly, as shown in Figure 5 which illustrates the effect of temperatures between 250 K and 170 K on the effective O<sub>2</sub> absorption cross section of the solar H Lyman-alpha line.

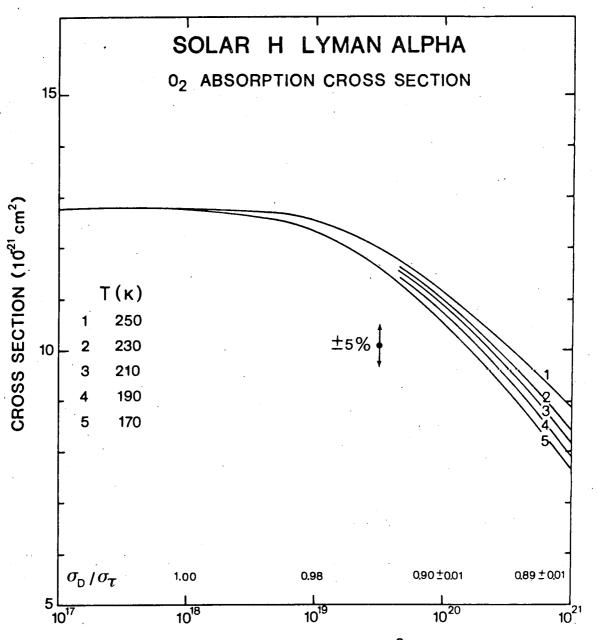
The effect of temperature can be introduced into the determination of the transmittance of Lyman-alpha if a correction is applied for N >  $10^{20}$  cm<sup>-2</sup>. Figure 6 shows the percentage variation in the number of Lyman-alpha photons for a temperature change of 10 K; it falls gradually from 15% for N =  $10^{21}$  cm<sup>-2</sup> to only 0.5% for N =  $10^{20}$  cm<sup>-2</sup>. Thus, in cases where it is considered to be necessary to take the temperature into account when N >  $10^{20}$  cm<sup>-2</sup>, corrections can be applied to Eq. 5 which refers to the mean mesospheric temperature, 230 K.

## IV. MESOSPHERIC PHOTODISSOCIATION OF CH4

The photodissociation of  $CH_4$  in the mesosphere results almost exclusively from the action of solar Lyman-alpha radiation. The experimental work of Watanabe et al. (1953), which has been discussed by Nicolet (1964, 1980), indicates that, to a first approximation, the absorption cross section is constant in the spectral region centred on the Lyman-alpha line. Hence the photodissociation frequency of  $CH_4$  at solar H Lyman-alpha is given by :

$$J_{Ly \ \alpha}(CH_4) = \sigma_D(CH_4) \ q_{\infty}(Ly \ \alpha) \ e^{-\tau_q (O_2)} \ s^{-1}$$
 (9)

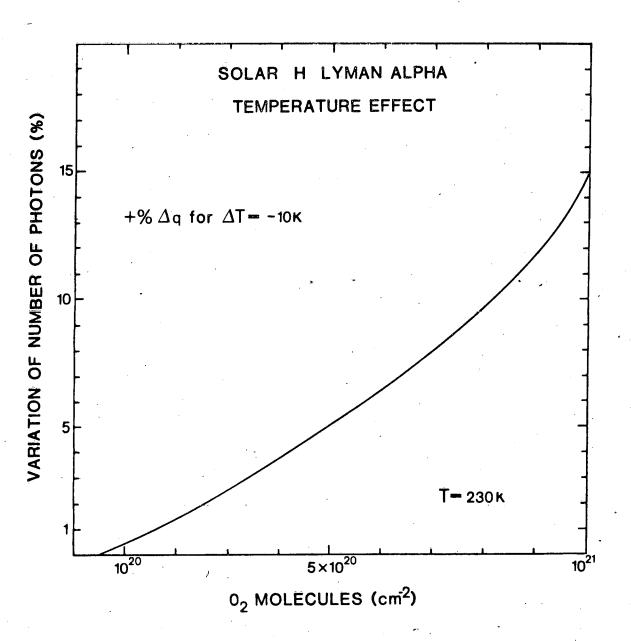
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## 0<sub>2</sub> MOLECULES (cm<sup>2</sup>)

Fig. 5.-

cross effective absorption The section of molecular oxygen for the solar H Lyman-alpha line at various temperatures between 250 K and 170 K. As cross section decreases there is an 0, the increase in the differences between the values for the various temperatures which may be taken into account at N >  $10^{20}$  cm<sup>-2</sup>. For  $10^{20} \le N \le 10^{21}$ cm<sup>-2</sup>, the mesospheric temperature T = 230  $\pm$  20 K is adopted. The ratio  $\boldsymbol{\sigma}_{D}$  is based on the effective cross sections of molecular oxygen as photodissociation-absorption cross section  $(\boldsymbol{\sigma}_{D})$  and as transmission-absorption ( $\sigma_{_{T}}$ ) at solar H Lymanalpha; it decreases from 1 to 0.89 as N increases from  $10^{18}$  to  $10^{21}$  cm<sup>-2</sup>.





The effect of temperature on the effective transmittance  $T_0$  (Ly $\alpha$ ) of the solar H Lyman-alpha line,  $q(Ly\alpha)/q_{\infty}(Ly\alpha)$ . The variation (%) of the number of Lyman-alpha photons  $\Delta q$  is given for differences of temperatures  $\Delta T$  related to T = 230 K.

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where  $\sigma_D = 1.85 \times 10^{-17} \text{ cm}^2$  is the mean photodissociation-absorption cross-section of CH<sub>4</sub> and  $\tau_q(O_2)$  is the effective optical depth at Lymanalpha, as given by Eqs (7) and (8). This direct application follows from the assumption that the mean cross section of CH<sub>4</sub> in the range Lymanalpha ± 1.75 A is constant. Finally, by using the parameters introduced in the present work, we can write :

$$J_{LY \alpha}(CH_4) = 5.55 \times 10^{-6} [1 + 0.2 \frac{F_{10.7} - 65}{100}]$$
  
x exp [- 2.115 x 10<sup>-8</sup> N<sup>0.885</sup>] s<sup>-1</sup> (10)

The effect of temperature can be introduced by using the information given in Fig. 6. Starting from T = 230 K, for each negative  $10^{\circ}$ K step in temperature, the graph gives the corresponding positive percentage change in J(CH<sub>4</sub>), instead of q, for N >  $10^{20}$  cm<sup>-2</sup>; that is for optical thicknesses greater than  $\tau_{\alpha}(O_2) = 1$ .

## V. PHOTODISSOCIATION OF CO2 BY SOLAR H LYMAN-ALPHA

The absorption spectrum of  $CO_2$  has been studied by a number of investigators (see Nicolet, 1980). At Lyman-alpha, the measurements of Nakata et al. (1965), with a resolution of 0.2 A, lead to an absorption cross-section about 10% higher than previous values : namely  $\sigma_{Ly \ \alpha}(CO_2) = 8.14 \times 10^{-20}$  cm<sup>2</sup>. However, since the measurements indicate that at 1215.7 A there is small peak, this value must be considered as a maximum value. Therefore, if a photodissociation frequency of the order of

$$J_{\infty}(CO_2) = 2.2 \times 10^{-8} \text{ s}^{-1}$$

is adopted at the top of the Earth's atmosphere for quiet sun conditions,  $q_{\infty}(Ly \alpha) = 3 \times 10^{10}$  photons cm<sup>-2</sup> sec<sup>-1</sup>, the accuracy cannot be better than  $\pm 10\%$ .

Thus, a formula similar to equation (10), which refers to  $CH_4$ , can be adopted if the 5.55 x  $10^{-6}$  is replaced by 2.2 x  $10^{-8}$ . Even through the diffuse character of the bands in the spectral region of Lyman-alpha may be due to predissociation, new measurements with higher resolution and varying temperature between 170 K and 250 K are required so as to improve the accuracy of the  $CO_2$  photodissociation frequency due to solar H Lyman-alpha in the mesosphere.

## VI. PHOTODISSOCIATION OF H2O BY SOLAR H LYMAN-ALPHA

Photodissociation of  $H_2O$  in the mesosphere is caused by Lyman-alpha and also by radiation in the region associated with the absorption of the Schumann-Runge bands of  $O_2$ , and has recently been discussed by Nicolet (1984). The photodissociation frequency must therefore be considered as a function of wavelength. Figure 7 indicates how the absorption cross-section of  $H_2O$  varies over the 3.5 A band centred on Lyman-alpha. It is not possible to use the mean value of  $(1.5 \pm 0.1) \times 10^{-17}$  cm<sup>2</sup> without a detailed analysis of how it varies with N and T. Thus the calculation was made using 35 steps 0.1 A wide centred on Lyman-alpha, and the following expression :

$$I_{Ly\alpha}(H_20) = J_{Ly\alpha,\infty}(H_20) e^{-\tau_{H_2}0(0)}$$

(11)

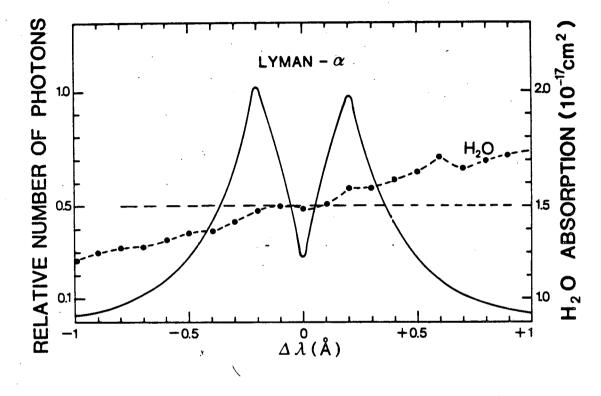


Fig. 7.- The absorption cross section of  $H_2^0$ , according to Lewis et al. (1983) (dashed line) and the profile of solar H Lyman-alpha (full line).

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corresponding to the general Eq. (3), where

$$J_{Ly\alpha,\infty}(H_2 0) = 3 \times 10^{11} \Sigma \sigma_i(H_2 0) [q_{j\alpha}/q_{\infty}] \text{ sec}^{-1}$$
 (12)

In Eq. 12  $\sigma_i(H_2O)$  is the <u>photodissociation-absorption</u> crosssection for  $\Delta\lambda = 0.1$  A as given by Lewis et al. (1983). Equation (12) leads to the H<sub>2</sub>O photodissociation frequency at the top of the earth's atmosphere for a quiet sun

$$J_{Ly\alpha,\infty}(H_2 0) = 4.53 \times 10^{-6} \text{ sec}^{-1}$$
 (13)

With  $q_{\infty}(Ly\alpha) = 3 \times 10^{11}$  photons cm<sup>-2</sup> s<sup>-1</sup>, the effective absorption cross section at zero optical depth is, therefore :

$$\sigma_{D,\tau=0}(H_2 0) = 1.51 \times 10^{-17} \text{ cm}^2$$
 (14)

which is close to the mean value for the spectral range of the solar H Lyman-alpha line.

Adopting again the 230 K temperature for the  $O_2$  absorption cross section, the  $H_2O$  photodissociation frequency at Lyman-alpha is given by the simple formula :

$$J_{Ly\alpha}(H_2 0) = J_{Ly\alpha,\infty}(H_2 0) \exp \left[-1.727 \times 10^{-18} N^{0.8895}\right]$$
 (15)

with an accuracy better than  $\pm 2\%$ . If 170 < T < 250 K, there is practically no temperature effect (less than  $\pm 2\%$ ) for N <  $10^{20}$  cm<sup>-2</sup>. The following values are obtained for quiet sun conditions

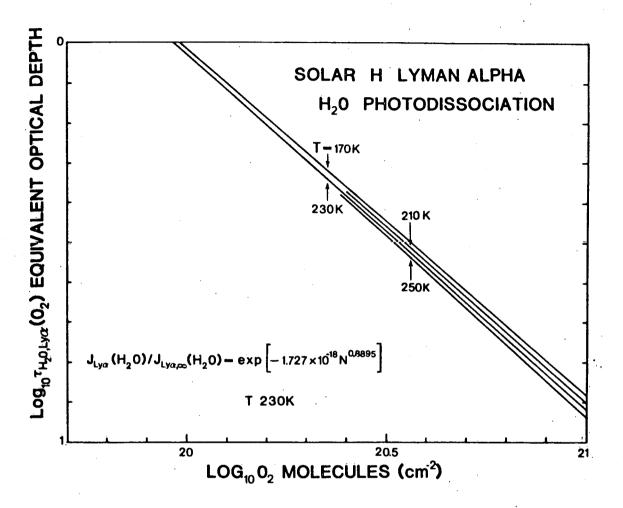
 $\begin{array}{ccccc} N(cm^{-2}) & 10^{18} & 10^{19} & 5x10^{19} & 7.5x10^{19} & 10^{20} \\ J_{\rm H \ 0}(s^{-1}) & 4.47x10^{-6} & 4.02\pm0.01 & 2.58\pm0.02 & 2.00\pm0.03 & 1.56\pm0.03 \end{array}$ 

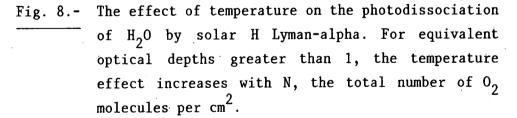
The temperature effect is detected for  $N \ge 10^{20}$ , as can be seen from Figure 8 when the equivalent optical depth  $\tau_{H 0,Ly\alpha}(O_2)$ , introduced in Eq. (11), becomes greater than unity. For a temperature change of 20 K, the magnitude of the effect is of the order of 1% and 10% at N =  $10^{20}$  and  $10^{21}$  cm<sup>-2</sup>, respectively. Whether effects of this order are to be considered or neglected will depend on the nature of the problem being investigated. In the study of the photodissociation of H<sub>2</sub>O in the mesosphere, where both Lyman-alpha and the region of the Schumann-Runge bands are important, Eq. (14) or any other simple expression may be used; but for a different discussion, see Nicolet (1984).

When it is necessary to take the temperature effect into account, Fig. 9 provides the required information; for example, between N =  $10^{21}$  cm<sup>-2</sup> and  $10^{20}$  cm<sup>-2</sup>, a decrease in the temperature of 10 K at 230 K results in a reduction in the rate of dissociation of H<sub>2</sub>O from about 15% to 0.5%. The effect is about the same as that shown in Fig. 6 for the variation in the number of Lyman-alpha photons. This similarity is due to the variation in the effective cross section itself  $\sigma_{\rm D}({\rm H_2O})$  which is found from the following expression :

$$J_{Ly\alpha}(H_20) = \sigma_D(H_20) q_{\infty}(Ly\alpha) e^{-\tau_q(O_2)}$$
 (16)

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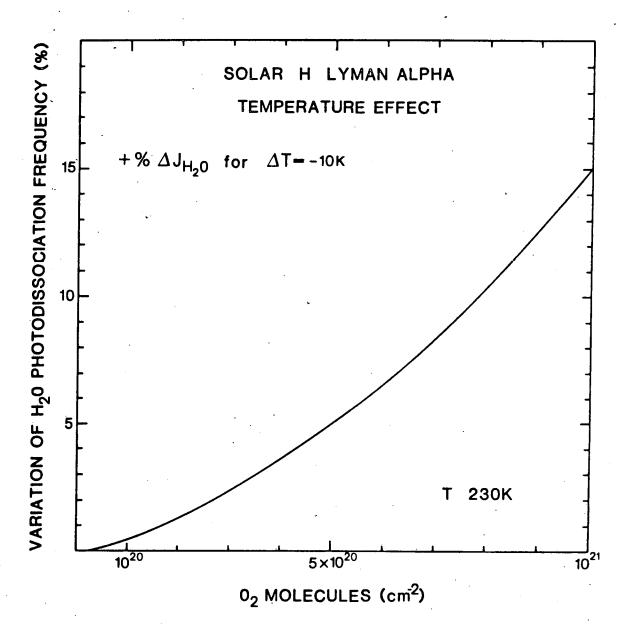


Fig. 9.- The effect of temperature on the equivalent transmittance of the photodissociation frequency of  $H_2O$  resulting from the solar H Lyman alpha line. The variation  $\Delta J(H_2O)$  is given for differences of  $\Delta T$  related to T = 230 K.

where  $\tau_q(O_2)$  is the effective optical depth at solar H Lyman-alpha as given by Eq. (7). The effective absorption cross section  $\sigma_D(H_2O)$  increases from  $1.51 \times 10^{-17}$  cm<sup>2</sup>, corresponding to an optical depth  $\tau_q(O_2) = 0$  (see Eq. (12)) to  $1.62 \times 10^{-17}$  cm<sup>2</sup> for N =  $10^{21}$  cm<sup>-2</sup>? corresponding to an optical depth of about 8. In order to cover all possible values of N from  $10^{19}$  to  $10^{21}$  cm<sup>-2</sup>, a mean value :

$$\sigma_{\rm D,Ly\alpha}({\rm H_20}) = (1.57 \pm 0.05) \times 10^{-17} {\rm cm}^2$$
 (17)

can be adopted with a precision of ± 3.3%. Thus, when a correct mean value has been determined for the effective absorption cross-section at Lyman-alpha, the following simple expression fo  $J_{Ly\alpha}(H_2O)$  in the mesosphere can be used, assuming a mean temperature of 230 K :

$$J_{Ly\alpha}(H_2 0) = 5.7 \times 10^{-6} [1 + 0.2 \frac{F_{10.7}^{-65}}{100}]$$
  
x exp [- 2.115 x 10<sup>-8</sup> N<sup>0.8855</sup>] (18)

The final conclusion is that the photodissociation of  $H_2O$  can be expressed in the same form as that for  $CH_4$  if the mean effective absorption-photodissociation cross section is adapted to the conditions found in the mesosphere. The effect of the temperature appears only in the exponential term in Eq. (18), that is in the transmittance of the mesosphere for solar H Lyman-alpha radiation (see Fig. 9).

## VII. PHOTODISSOCIATION OF 02 SOLAR H LYMAN-ALPHA

Since the mean absorption cross section at Lyman-alpha by oxygen depends on both temperature and wavelength, the rate of dissociation  $J_{Ly\alpha}(O_2)$  defined by :

$$J_{Ly\alpha}(0_2) = \sigma_{D}(0_2) q_{\infty}(Ly\alpha) e^{-\tau_q(0_2)}$$
(19)

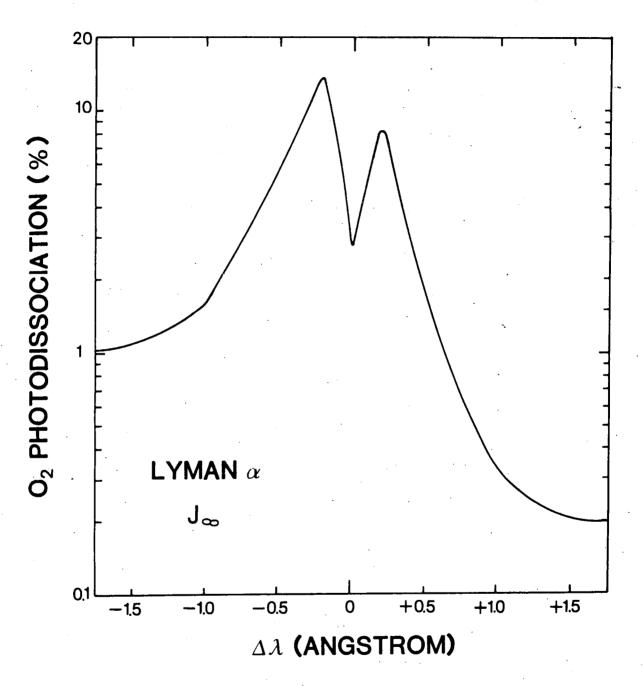
must be determined using a complete and detailed expression of the form of Eq. (3):

$$J_{Ly\alpha}(0_2) = q_{\infty}(Ly\alpha) \Sigma \sigma_i(0_2) (q_{i,\infty}/q_{\infty}) e^{-\sigma_i(0_2)N}$$
(20)

As in the preceding cases, the calculations are based on intervals of 0.1 A in the 3.5 A band centred on Lyman-alpha. For temperatures 170 K < T = 230 < 250 K and for quiet Sun  $q_{\infty}(Ly\alpha) = 3 \times 10^{11}$  photons cm<sup>-2</sup> sec<sup>-1</sup>, Eq. 20 leads to the following value for the photodissociation frequency of O<sub>2</sub> (Fig. 10) at the top of the terrestrial atmosphere and for the mean distance between the Sun and the Earth :

$$J_{Ly\alpha,\infty}(0_2) = (3.81 \pm 0.03) \times 10^{-9} \text{ sec}^{-1}$$
 (21)

It corresponds to an effective absorption-photodissociation cross-section



 $\begin{array}{c} \mbox{Fig. 10.-} \mbox{Structure of the photodissociation frequency of} \\ \mbox{molecular oxygen at solar H Lyman-alpha indicating} \\ \mbox{the effect of the variation with wavelength of the} \\ \mbox{absorption cross section of } 0_2. \end{array}$ 

$$\sigma_{\rm Lva}(0_2) = (1.27 \pm 0.01) \times 10^{-20} \text{ cm}^2$$
 (22)

with a precision better than  $\pm 1\%$  when T = 230 K is adopted to cover the temperature range 250 - 170 K. With the same temperature the ratio  $T_{J(O)} = J_{Ly\alpha}(O_2)/J_{Ly\alpha,\infty}(O_2)$  can be determined with the simple expression (Figure 11).

$$J_{Ly\alpha}(0_2)/J_{Ly\alpha,\infty(0_2)} = \exp \left[-4.923 \times 10^{-17} N^{0.8216}\right]$$
 (23)

with an accuracy better than  $\pm 3\%$  from N =  $10^{17}$  to N =  $10^{21}$  cm<sup>-2</sup>. The detailed calculations for various temperatures from T = 170 K to 250 K show that, for N <  $10^{20}$  cm<sup>-2</sup>,

$$J_{Ly\alpha}(0_2)_T > J_{Ly\alpha}(0_2)_T$$
(24a)

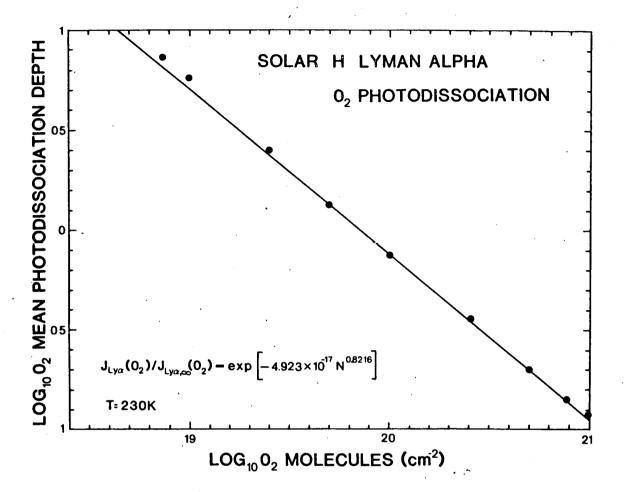
and for N >  $10^{20}$  cm<sup>2</sup>

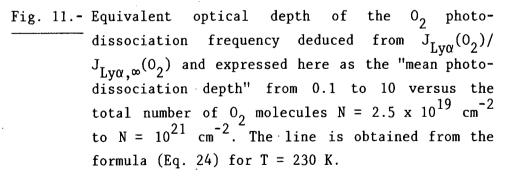
$$J_{Ly\alpha}(0_2)_T < J_{Ly\alpha}(0_2)_T$$

(24b)

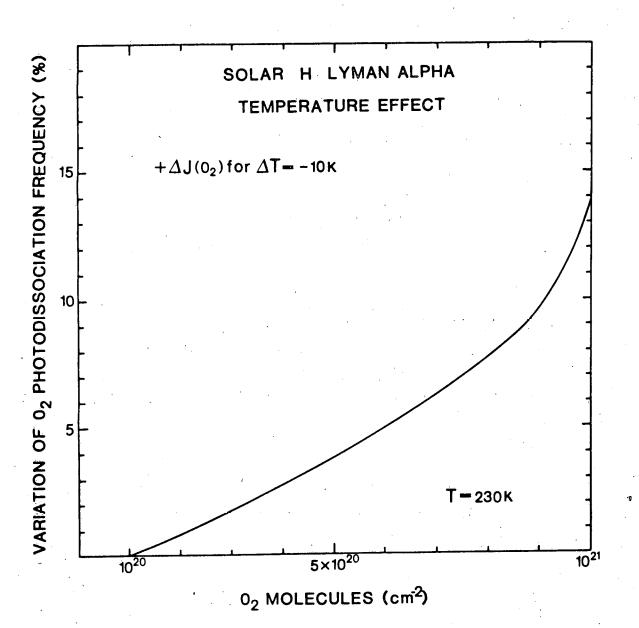
for  $T_2 < T_1$ .

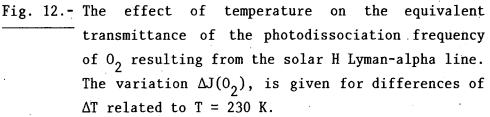
Finally, as in the cases studied earlier, for  $J_{Ly\alpha}(O_2)$  it is necessary to take the temperature effect into account for large optical thicknesses, i.e. for  $N(O_2) > 10^{20} \text{ cm}^{-2}$ . Figure 12 shows by how much  $J_{Ly\alpha}(O_2)$  increases for a temperature change of - 10 K; the increase varies from 15% for N =  $10^{21} \text{ cm}^{-2}$  to 1% at N =  $2.5 \times 10^{20} \text{ cm}^{-2}$ . At N =





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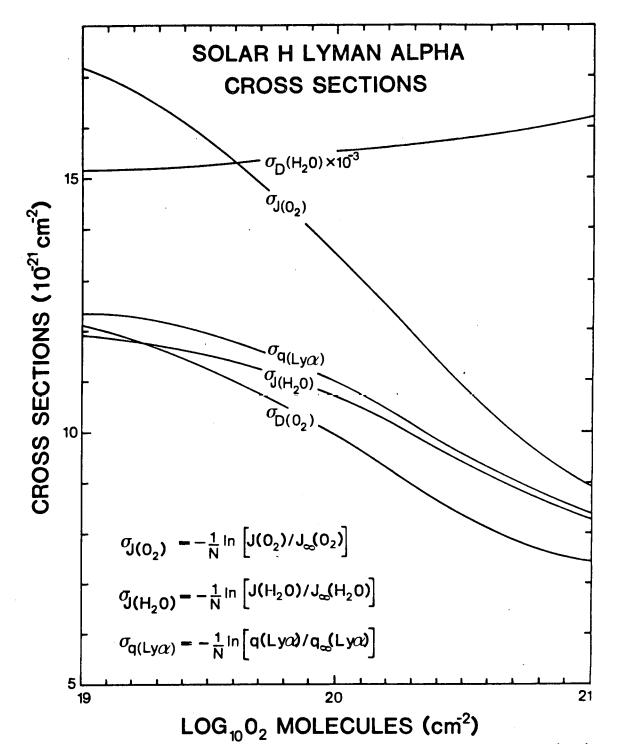
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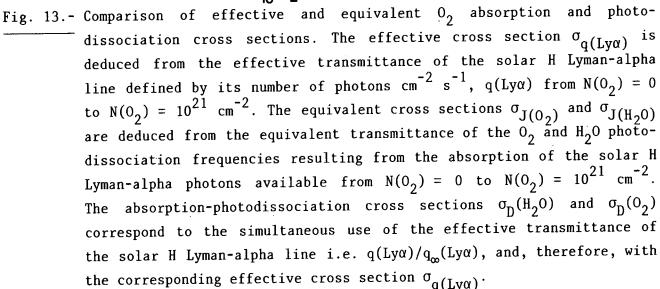
 $10^{20}$  cm<sup>-2</sup> there is no temperature effect, because of the inversion indicated in Eqs 24a and 24b, but at N =  $10^{19}$  cm<sup>-2</sup> the rate of photo-dissociation decreases to 0.84 ± 0.02, i.e. with an accuracy of ± 2%.

If Eq. (19) is used in conjunction with the result obtained in Eq. (23), the effective absorption-photodissociation cross-section  $\sigma_D(O_2)$  is obtained with the use of  $\tau_q(O_2)$ , the effective optical depth at Lyman-alpha as defined by Eq. (7). The cross-section decreases from 1.21 x  $10^{-20}$  cm<sup>2</sup> at N =  $10^{19}$  cm<sup>-2</sup> to  $9.9 \times 10^{-21}$  cm<sup>-2</sup> at N =  $10^{20}$  cm<sup>2</sup>, and is only 7.4 x  $10^{-21}$  cm<sup>-2</sup> at N =  $10^{21}$  cm<sup>-2</sup>; this is illustrated in Figure 13 which includes other information also.

The three curves  $\sigma_{q(Ly\alpha)}$ ,  $\sigma_{J(O_2)}$  and  $\sigma_{J(H_2O)}$  show the variation, with the total number of  $O_2$  molecules  $10^{19} < N < 10^{21}$  cm<sup>-2</sup>, of the equivalent absorption cross section corresponding to the mesospheric transmittance of the solar H Lyman-alpha photons  $q(Ly\alpha)$ , of the  $O_2$  photodissociation frequency  $J(O_2)$  and of the  $H_2O$  photodissociation frequency  $J(H_2O)$ , respectively. The two parameters  $\sigma_{q(Ly\alpha)}$  and  $\sigma_{J(H_2O)}$  are not very different since the effective absorption-photodissociation cross section  $\sigma_D(H_2O)$  at solar H Lyman-alpha shows only a small variation with N (Figure 13). As far as  $\sigma_{J(O_2)}$  is concerned, the variation is extremely important; it goes from about  $1.7 \times 10^{-20}$  cm<sup>2</sup> at N =  $10^{19}$  cm<sup>-2</sup> to only about  $0.9 \times 10^{-20}$  cm<sup>2</sup> at N =  $10^{21}$  cm<sup>-2</sup> (Figure 13). This is due to the double effect of  $\sigma_{q(L\alpha)}(O_2)$  and  $\sigma_{D(Ly\alpha)}(O_2)$ . Nevertheless, it is possible to use the atmospheric transmittance of the solar H Lyman-alpha line and to adopt Eq. (19) if the effective absorption-photodissociation cross section cross section  $\sigma_D(O_2)$  is calculated for T = 230 K using the following expression

$$\sigma_{\rm D}(0_2)_{\rm Ly\alpha} = 1.15 \times 10^{-18} / N^{0.1175} \, {\rm cm}^2$$
 (25)





with an accuracy better than  $\pm 3\%$  for N > 10<sup>19</sup> cm<sup>-2</sup>. Thus, J(O<sub>2</sub>) at Lyman-alpha in the mesosphere, is given by

$$J_{Lv\alpha}(0_2) = 1.15 \times 10^{-18} / N^{0.1175} q_{\omega} Ly\alpha) e^{-\sigma_q(Ly\alpha)}$$
(26)

which shows that the effective transmittance of the solar H Lyman-alpha line can always be used if the correct effective absorption-photodissociation cross sections of all the constituents are determined after a detailed calculation of their photodissociation frequencies.

## VIII. PHOTOIONIZATION OF NO BY SOLAR H LYMAN ALPHA

The calculation of the photoionization of nitric oxide in the D region is based on the absorption cross section and on the ionization yield determined by Watanabe (1958). If we accept the photoionization cross sections given by Watanabe at 1211.3, 1215.6 and 1217.4 A, i.e.  $2.01 \times 10^{-18}$ ,  $2.02 \times 10^{-18}$  and  $2.09 \times 10^{-18}$  cm<sup>2</sup>, respectively, a mean cross section corresponding to about  $2.02 \times 10^{-18}$  cm<sup>2</sup> can be adopted at the solar H Lyman-alpha line. Thus, the mesospheric photoionization frequency of nitric oxide,  $I_{NO}$ , is obtained from

 $I_{NO} = 2.02 \times 10^{-18} q_{\infty}(Ly\alpha)T_0$  (Lya) (27)

where  $T_O(Ly\alpha)$  is given by Eq. (8). A more detailed analysis of the NO photoionization frequency could require an experimental determination of the absorption cross section and of the ionization yield at high resolutions.

## IX. SUMMARY AND CONCLUSIONS

In the mesosphere, the photodissociation, or photoionization, by solar H Lyman-alpha radiation of all the constituents depends on several parameters whose precise values must still be the object of further experiments and observations. In particular, it is important to emphasise the importance of obtaining accurate absolute values for the irradiance of H Lyman-alpha. In the present work, the conventional value  $3 \times 10^{11}$  photons cm<sup>-2</sup> sec<sup>-1</sup> at 1 AU for a quiet Sun has been adopted provisionally pending the availability of a more accurate value. In fact, the many different values that have been proposed cover the range from  $2 \times 10^{11}$  to  $3.2 \times 10^{11}$ , and to cover all these, it would be necessary to write :

$$q_{\infty}(Ly\alpha) = (2.75 \pm 0.75) \times 10^{11} \text{ photons cm}^{-2} \text{ s}^{-1}$$
 (28)

In other words, the value adopted in the calculations has certainly introduced a systematic error, but this can easily be corrected in all the numerical expressions quoted in the preceding Sections. If the conventional value adopted here were reduced by 20%, the resulting -value would be :

$$J_{m}(Ly\alpha) = 2.5 \times 10^{11} \text{ photons cm}^{-2} \text{ s}^{-1}$$
 (

29)

with an accuracy of say  $\pm$  20%, and this may perhaps be closer to the true value.

The way in which the radiation from the solar disk varies with solar activity requires further attention, bearing in mind the limits indicated by those solar activity indices that are not subject to errors arising from drift in the calibration of the instruments. Direct observations of Lyman-alpha have proved be inadequate for to the determination of the long-term trend in solar activity, unless corrections are applied. Instrumental drifts and erratic changes are often so large that they completely mask the solar cycle variations.

The expression :

$$q_{\infty}(Ly\alpha)_{active Sun}/q_{\infty}(Ly\alpha)_{quiet Sun} = 1 + 0.2 \frac{F_{10.7}^{-65}}{100}$$
 (30)

which is based on Eq. (1), results from a global analysis, covering more than a solar cycle, of observations made by various satellites using sensors which were subject to various changes in sensitivity.

Finally, the variations in the profile of solar Lyman-alpha ought to be more accurately known. The composite profile for the irradiance as a function of solar activity represents the disk as a whole, including variations from the centre out to the limb, and also the fluctuations associated with the variable incidence of Lyman-alpha plages during the solar cycle. At present the only possible course is that adopted in the present work, namely to use a mean profile which represents the structure of the irradiance in the 3.5 A band centred on Lyman-alpha.

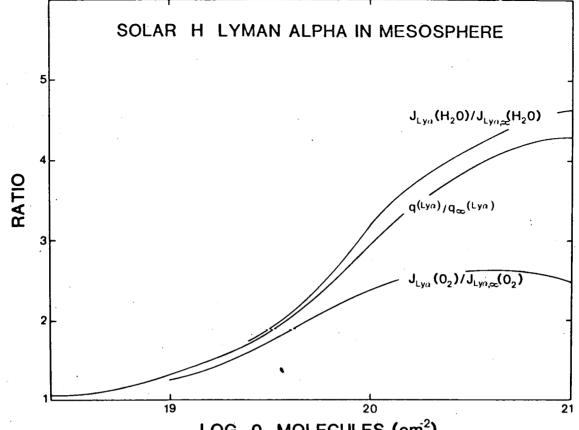
In another field, a fresh analysis of all the high-resolution absorption cross section near Lyman-alpha, and in the temperature range 190 - 250 K, would be very welcome. In order to demonstrate the variation with wavelength, near Lyman-alpha, of the various para-

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meters, Figures 14a and 14b show the results of calculations based on a number of different sets of conditions. In most studies, no account has been taken of the structure of Lyman-alpha and the irradiance has been assumed to remain constant over the spectral region considered. If this approximation is accepted, the transmittance relative to the constant value is as shown in Figure 14a. As can be seen, the relative transmittance of the irradiance determined for the real profile of Lymanalpha can reach values greater by a factor of four for large optical thicknesses. For the rate of dissociation of H<sub>2</sub>O by Lyman-alpha, the equivalent transmittance also increases by a factor of 4, while for  $O_2$  it rises by a factor of 2. It seems clear that, in the earlier publications, estimations of the penetration of solar H Lyman-alpha radiation are incorrect. Figure 14b takes account of the structure of the profile of Lyman-alpha, and allows other comparisons to be made if the experimental determinations of the absorption cross section of  $O_2$  are accepted. The equivalent transmittances for the rates of dissociation of  $H_2O$  and  $O_2$  are given as ratios to the transmittance of the Lyman-alpha irradiance. The variation of the effective photodissociation cross section of  $H_2O$ , which increases with the  $O_2$  optical depth, leads to a small increase of the equivalent transmittance for its photodissociation frequency, as compared with the mesospheric transmittance at solar H Lyman-alpha. On the other hand, the decrease in the effective photodissociation frequency of O2 with increasing optical depths leads to an important decrease in the equivalent transmittance of the O2 photodissociation frequency as compared with the solar H Lyman-alpha transmittance.

Before concluding the present critical analysis of the emission of solar H Lyman-alpha radiation and its absorption in the atmosphere, it seems important to refer to other limitations to the study of the effects of this radiation in the mesosphere. Figure 15 allows a comparison to be made between the error in the optical thickness (corresponding to the experimental cross sections) and that in the

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LOG<sub>10</sub>0<sub>2</sub> MOLECULES (cm<sup>2</sup>)

Fig. 14a.- Comparisons of the conditions of atmospheric transmittance in the spectral region of H Lymanalpha, if a constant flux with wavelength is adopted.

> The combination of the real profile of solar H Lyman-alpha, with its spectral structure, with that of the  $0_2$  absorption cross section leads to increase at high optical depths of its an transmittance which rises by more than a factor of 4. The effect is even greater for the relative  $H_20$ photodissociation frequency due to the variation with wavelength of its absorption cross section in  $H_2O$  and decrease in  $O_2$  cross (increase The relative 0, photodissociation sections). frequency is subject to the double effect of the decrease of the effective photodissociation and absorption cross section with increasing the optical depth; it increases only by a factor of 2 as compared with the assumption of a constant solar flux with wavelength.

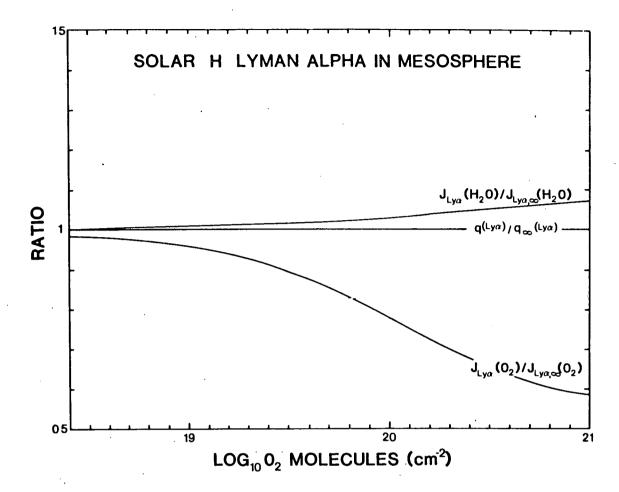


Fig. 14b.- Comparisons between the mesospheric transmittance of solar H Lyman-alpha, taken as unity, and the relative photodissociation frequencies of  $H_2O$  and  $0_2$ . The relative  $H_20$  photodissociation frequency is subject to a small increase with increasing depth which corresponds to а small optical increase in its absorption cross section towards longer wavelengths. The decrease of the relative  $0_2$  photodissociation frequency with increasing optical depths compared with the solar H Lymanalpha transmittance is due to the simultaneous strong decrease in its effective photodissociation cross section.

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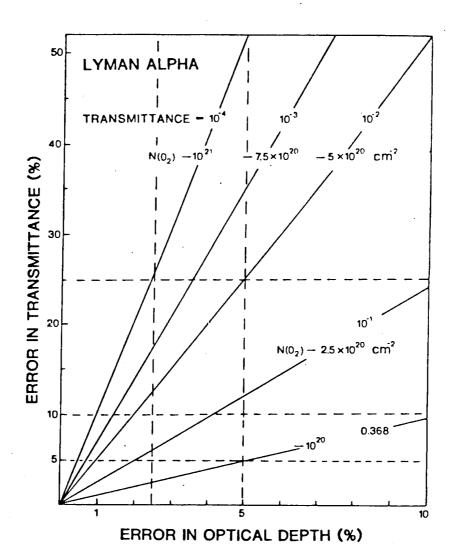


Fig. 15.- Errors in the mesospheric transmittance of solar H Lyman-alpha for various total numbers of absorbing  $0_2$  molecules, from N =  $10^{20}$  cm<sup>2</sup> (75 km for overhead sun, equivalent unit optical depth) to  $N = 10^{21} \text{ cm}^2$  (60 km for overhead sun, equivalent optical depth  $\cong$  9). An error of  $\pm$  5% in the absorption cross section would lead to an error of  $\pm$  50% for a transmittance of 10<sup>-4</sup>, i.e. 60 km for overhead an sun. The inaccuracy in the 0, absorption cross section leads to a limit for possible applications of the effect of temperature on the photodissociation frequencies at high optical depths.

transmittance; as can be seen, there are limits to the accuracy that can be achieved. For example, an error of 5% in the absolute value of the O<sub>2</sub> absorption cross section will result in an error of 25% in transmittance when N = 5 x 10<sup>20</sup> O<sub>2</sub> molecules cm<sup>-2</sup>, i.e. at 65 km for an overhead Sun, or 70 km for a solar zenith angle of 60°. In fact when N(O<sub>2</sub>) > 5 x 10<sup>20</sup> cm<sup>-2</sup>, the transmittance falls below 10<sup>-2</sup>. Figures 6, 9 and 12 show that the temperature effect is important (greater than 5% for T = 10 K) only when N(O<sub>2</sub>) > 5 x 10<sup>20</sup> cm<sup>-2</sup>; hence practical applications in the mesosphere correspond to transmittances greater than 10<sup>-2</sup> for Lyman-alpha, with a mean temperature of 230 K. Thus, Eq. (8),

$$T_0$$
 (Ly $\alpha$ ) = exp [- 2.115 x 10<sup>-18</sup> N<sup>0.8855</sup>] (31)

can be used as the basis for all calculations when the appropriate photodissociation-absorption cross-sections are simultaneously adopted, namely :

$$\sigma_{\rm D}({\rm CH}_4) = 1.85 \times 10^{-17} {\rm cm}^2$$
 (32)

$$\sigma_{\rm D}({\rm CO}_2) = 8.14 \times 10^{-20} \text{ cm}^2$$
 (33)

$$\sigma_{I}(NO) = 2.02 \times 10^{-18} \text{ cm}^{2}$$
 (34)

$$\sigma_{\rm D}({\rm H_20}) = (1.57 \pm 0.05) \times 10^{-17} {\rm cm}^2$$
 (35)

$$\sigma_{\rm D}(0_2) = 1.15 \times 10^{-18} / N^{0.1175} \, {\rm cm}^2$$
 (36)

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