

**ACTINOMETRIC METHOD FOR THE DETERMINATION OF THE TOTAL
NUMBER OF AEROSOL PARTICLES IN THE
VERTICAL ATMOSPHERIC COLUMN**

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RESUMEN

Se describe un método para la evaluación de la cantidad total de partículas suspendidas en una columna vertical de aire de sección unitaria. El método está basado en un modelo de extinción de radiación solar por aerosoles en la atmósfera. Los resultados se comparan con el coeficiente de turbiedad de Ångström. Usando la concentración de partículas en superficie y suponiendo un decaimiento exponencial de la concentración de partículas, se establece la escala de altura para la distribución vertical del aerosol. Por último, aprovechando el modelo de atenuación, se realizaron algunos experimentos numéricos que permiten evaluar el efecto de los diferentes componentes atmosféricos considerados en el modelo, tales como la cantidad de partículas, el agua precipitable, la abundancia de bióxido de carbono y la cantidad total de ozono atmosférico.

ABSTRACT

A method is described for the evaluation of the total amount of suspended particles in a unitary vertical air column. The method is based in an aerosol and atmospheric extinction model, its results are compared with the Ångström turbidity coefficient. Taking into account the surface concentration of particles and assuming an exponentially decay of the concentration, the height scale of the aerosol vertical distribution was established. Finally, taking advantage from the attenuation model, some numerical experiments that permit the evaluation of the effect of the different atmospheric components considered in the model were made, such as total amount of particles, precipitable water, the carbon dioxide abundance and the total atmospheric ozone.

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INTRODUCTION

The atmospheric turbidity has been described traditionally by the Ångström turbidity coefficient (Ångström, 1929, 1981) which is directly related to the total number of aerosol particles. Sometimes an additional parameter is applied related to the exponent in Junge's particle distribution, this coefficient being based on the particles radii distribution function that takes into account their relative abundance with respect to the radii that are lineally dependent upon themselves in a log-log coordinate system ($dN/d \log(r)$ vs. $\log(r)$) (Junge, 1952; Iqbal, 1983). However, the distribution features could be improved by fitting a second order curve in the region of small radii ($0.03 < r < 0.10 \mu\text{m}$), in order to obtain a better estimation of the atmospheric turbidity in terms of total amount of particles in the air column, which is a parameter with a simple physical meaning.

In this paper a method is proposed for the determination of the total amount of particles in the vertical air column. This method consists in the solution of an integral equation which is stated in terms of solar direct measurements under an atmospheric attenuation model.

To test the applicability of the method, actinometric determinations at three sites with different climatological characteristics were made: the first one was Ciudad Universitaria, at Mexico City, where the atmosphere shows a serious pollution problem due mainly to suspended particles. The second site was San Felipe Tlalmimilopan, State of Mexico; this is a semirural zone. Finally, the test was made at El Realejo, State of San Luis Potosí, a mountainous zone with a very clear atmosphere.

Some numerical experiments were made to evaluate the attenuation efficiency of the atmospheric components considered in the model (aerosol particles, air, water vapor, uniformly mixed gases and ozone).

THE MODEL

The model used considers an atmosphere which contains pure air, water vapor, aerosol particles, ozone and uniformly mixed gases (CO_2 , N_2O , CO , etc.).

The solar irradiance spectral distribution at the top of the atmosphere, $I_{0,\lambda_i} = I_0(\lambda_i)$ has been calculated according to Labs and Neckel (1968), with solar constant value equal to 1365 w/m^2 in the spectral interval $0.285 - 4.000 \mu\text{m}$; this spectral

window has been divided into 102 intervals ($i = 1, \dots, 102$) with a length $0.01 \mu\text{m}$ for $0.285 \leq \lambda_i \leq 1.0 \mu\text{m}$ and $0.1 \mu\text{m}$ for $1.0 \leq \lambda_i \leq 4.0 \mu\text{m}$. The solar radiation variability incident at the top of the atmosphere due to earth distance is also taken into account.

The basic expression for the solar irradiance (I) is the extinction integral equation, which in discrete form can be written as:

$$I = \frac{1}{R} \sum_{i=1}^n I_{0,i} \exp\left(-\sum_{j=1}^5 m_j \tau_{i,j}\right) \quad (1)$$

here R is the correction factor for Sun-Earth distance; n ($n = 102$) the number of intervals in which the solar spectrum ($0.285 - 4.0 \mu\text{m}$) is divided; j is the subscript for each atmospheric component: air with water vapor as scatter factor, $j = 1$; ozone, $j = 2$; carbon dioxide, $j = 3$; water vapor as solar radiation absorption factor, $j = 4$, and the atmospheric aerosol, $j = 5$. The relative optical mass (m_j) for each component represents the ratio of the radiation path length to the corresponding layer depth. The optical thicknesses ($\tau_{ij}(z)$) are defined as follows:

$$\tau_{ij}(z) = \int_0^{\infty} (\sigma_{ij}(z) + \kappa_{ij}(z)) dz \quad (2)$$

where $\sigma_{ij}(z)$ and $\kappa_{ij}(z)$ are respectively the scattering and absorption coefficients for j-th component at z height for λ_i wavelength (at midpoint of i-th spectral interval). Thus, the transmission function for each component is expressed as:

$$T_{ij} = \exp(-m_j \tau_{ij}) \quad (3)$$

The formula for the transmission of uniformly mixed gases (umg) and water vapor is taken from Leckner's (1978) work. These expressions are described in detail by Iqbal (1983).

Since certain gas absorption coefficients depend on pressure and temperature, we resorted to effective depths; that is, depths that would have a corresponding gas layer at a given pressure and temperature (Goody, 1964, p. 234).

The ozone transmission functions were calculated with the aid of the absorption coefficients reported by Bauer (1970), taking into account the absorption in the

Hartley - Huggins bands ($\lambda_i < 0.340 \mu\text{m}$) and the Chapuis band ($0.44 \mu\text{m} \leq \lambda_i < 0.70 \mu\text{m}$). Since the absorption coefficients vary in these bands, mean values were introduced for them in sufficiently narrow bands in order to obey the exponential law:

$$T_{i2} = \exp(-m_2 \kappa_{i2} \Omega) \quad (4)$$

here κ_{i2} is the mean ozone absorption coefficient corresponding to the i -th spectral interval; Ω is the ozone layer depth (in atm-cm).

The air transmission function (in terms of Rayleigh scattering) was computed according to Fröhlich and Shaw (1980) formula in which water vapor scattering effect is included.

To obtain the aerosol transmission function, we use the formal expression for the extinction (scattering + absorption) aerosol coefficient which can be written as:

$$\gamma_{s, \lambda_i}(z) = \sigma_{is}(z) + \kappa_{is}(z) = \int_{s_1}^{s_2} \pi r^2 Q_{\lambda_i}(s, \tilde{m}) f(z, s) ds \quad (5)$$

where S_1, S_2 define active optical particles interval (Leyva, 1980), $Q_{\lambda_i}(s, \tilde{m})$ is the Mie extinction efficiency factor (Mie, 1908; Van de Hulst, 1957) for wavelength λ_i , particle radius $r = \text{antilog}(s)$ and complex refractive index $\tilde{m} = \nu - i \cdot \chi$; furthermore, $dN = f(z, s) \cdot dS$ gives particle number per unit volume per unit radius within differential interval $(s, s + ds)$.

The function $f(z, s)$ must satisfy the following condition:

$$N(z) = \int_{s_1}^{s_2} f(z, s) ds \quad (6)$$

where $N(z)$ is the number of optically active particles per unit volume at z height.

If we define $f'(s)$ as the normalized distribution function (Fig. 1) such that:

$$\int_{s_1}^{s_2} f'(s) ds = 1 \quad (7)$$

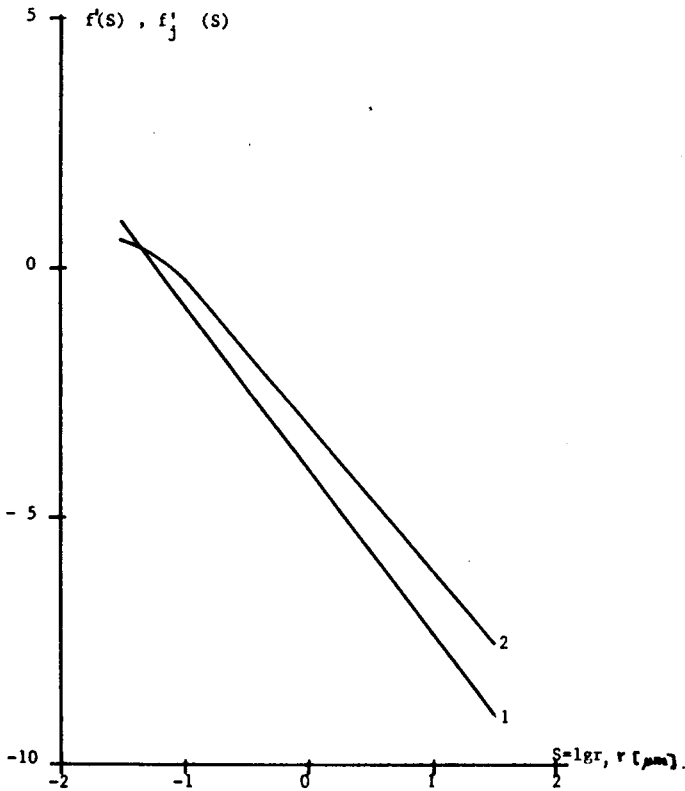


Fig. 1. Aerosol particles normalized distribution.

1) Junge's potential: $f'_j(s) = 8.52 \cdot 10^{-5.00 - 3.30s}$; $-1.5 \leq s \leq 1.5$

2) Dry standard : $f'(s) = \begin{cases} 3.72 \cdot 10^{-1.60(s+1.70)^2} ; & -1.5 \leq s \leq -1 \\ 6.92 \cdot 10^{-4.00 - 2.95s} ; & -1 \leq s \leq 1.5 \end{cases}$

then, assuming that

$$f(z, s) = N(z) f'(z) \tag{8}$$

we have:

$$\gamma_{s, \lambda_1}(z) = N(z) \int_{s_1}^{s_2} \pi r^2 Q_{\lambda_1}(s, \tilde{m}) f'(s) ds \tag{9}$$

and the optical aerosol thickness:

$$\tau_{s,\lambda_i} = N_t \gamma'_{s,\lambda_i} \quad (10)$$

where:

$$\gamma'_{s,\lambda_i} = \int_{s_1}^{s_2} \pi r^2 Q_{\lambda_i}(s, \tilde{m}) f'(s) ds \quad (11)$$

is defined as the "Normalized Aerosol Extinction Coefficient" (Muhlia, 1984) calculated according to Leyva *et al.* (1985), and:

$$N_t = \int_0^{\infty} N(z) dz \quad (12)$$

is the number of aerosol particles in a vertical column of air with unit base (1 cm^2), which is used here to determine quantitatively the atmospheric turbidity.

Again, assuming that the concentration of particles varies exponentially with height (Penndorf, 1954) we have:

$$N_t = \int_0^{\infty} N_0 \exp(-z/H) dz = N_0 H \quad (13)$$

i.e. the hypothesis of an exponential vertical distribution of $N(z)$ is equivalent to that of an homogeneous aerosol layer with H height. If N_0 is the concentration of particles at $z = 0$ then we can write:

$$\tau_{s,\lambda_i} = N_0 H \gamma'_{s,\lambda_i} = N_0 \tau'_{s,\lambda_i} \quad (14)$$

From the aforementioned, equation (1) most include five parameters to calculate solar direct radiation at a given time: uniformly mixed gases concentration G (PPM); ozone layer thickness, Ω (atm-cm); amount of precipitable water, w (cm); atmospheric pressure, P (mb) and total amount of aerosol particles in a unit vertical column, N_t (part/cm²).

The value of G is obtained from McClatchey *et al.* (1972), included implicitly in the Leckner's (1978) formula for umg transmittance (mainly for carbon dioxide, and molecular oxygen). The values of the ozone layer thickness (Ω) correspond to

those observed with the No 98 Dobson ozone spectrophotometer. The water vapor effective depth layer was calculated on the basis of the precipitable water content (w) by means of surface level dew point temperature using an empirical model proposed by Baruch (personal communication, 1986):

$$w = 1.16 T_{\text{dew}}(\text{°C}) + 9.42 \quad [\text{mm H}_2\text{O}] \quad (15)$$

In this manner it was possible to obtain more realistic results than the relation of Hann (1901) which, apparently, overestimates the precipitable water. Atmospheric pressure was used for calculating absolute optical air mass $m = m_r P/P_0$ where m_r is the relative optical mass of the air, and $P_0 = 1013.2$ mb. The aerosol transmission function calculation was made using the continental aerosol standard normalized distribution based on that proposed by Valley (1980), Fig. 1, and the optical constants (ν and χ) of a dry atmospheric aerosol, from Zuev *et al.* (1973); the dependence of the optical constant from wavelength and the normalized extinction coefficient of the standard continental aerosol calculated here are shown in Figs. 2 and 3, respectively.

The total amount of particles is implicit in equation (1) in such a way that knowing I it is possible to find the corresponding N_t that satisfies the mentioned equation. To perform this equation inversion a numerical method was used (Appendix 1).

NUMERICAL EXPERIMENTS

On the basis of equation (1), it is possible to calculate the relative importance that the different atmospheric components have on the solar radiation attenuation. This is made by calculating the relative attenuation of direct beam of total solar radiation through the logarithmic derivative:

$$\delta_x = \frac{\bar{x}}{\bar{I}} \frac{\Delta I}{\Delta x} \quad (16)$$

where \bar{x} is the mean value of any atmospheric attenuation factors and \bar{I} is the total irradiance, corresponding to \bar{x} .

The optical depths due to scattering and absorption of umg are directly proportional to atmospheric pressure. For this reason, variations in P are equivalent to variations in the Rayleigh scattering and absorption by CO_2 and O_2 due to reduction (or increasing) of the column of umg above the point of observation.

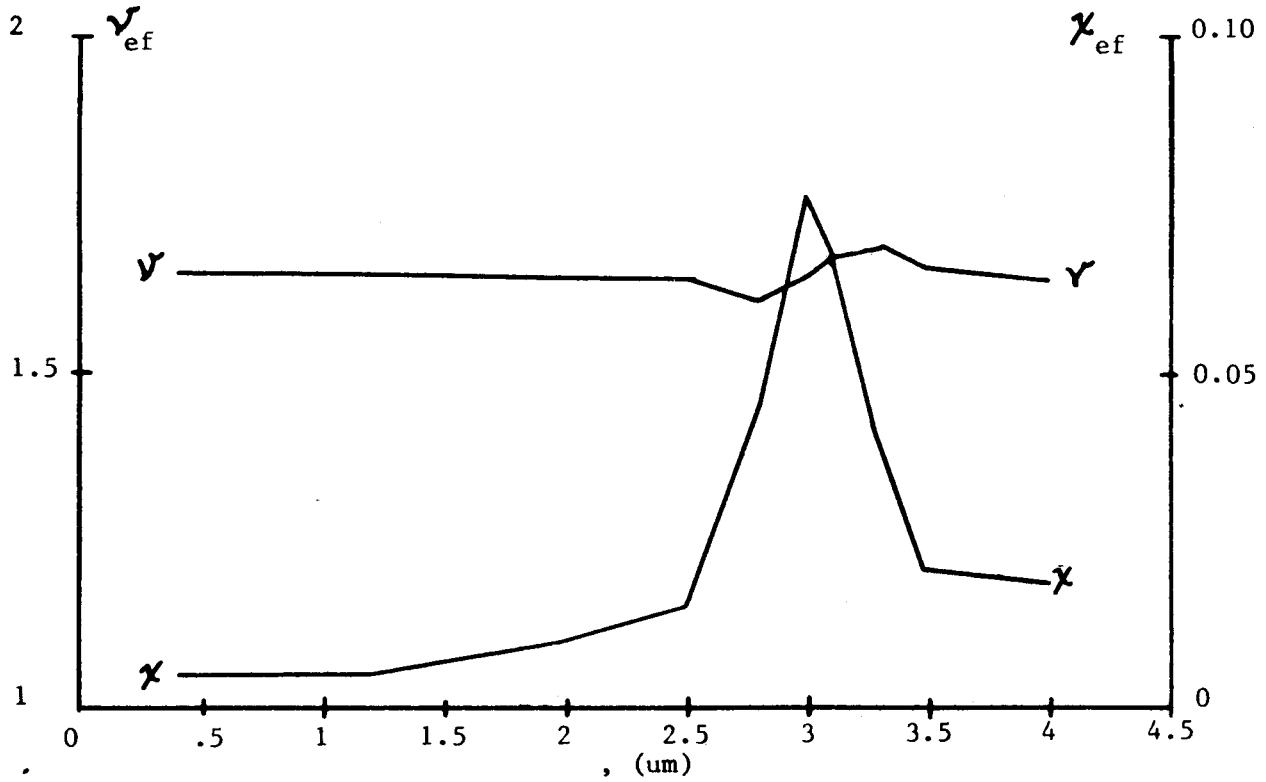


Fig. 2. Wavelength dependency of real (ν) and imaginary (χ) parts of the complex refractive index of aerosol particles (Zuev *et al.*, 1973).

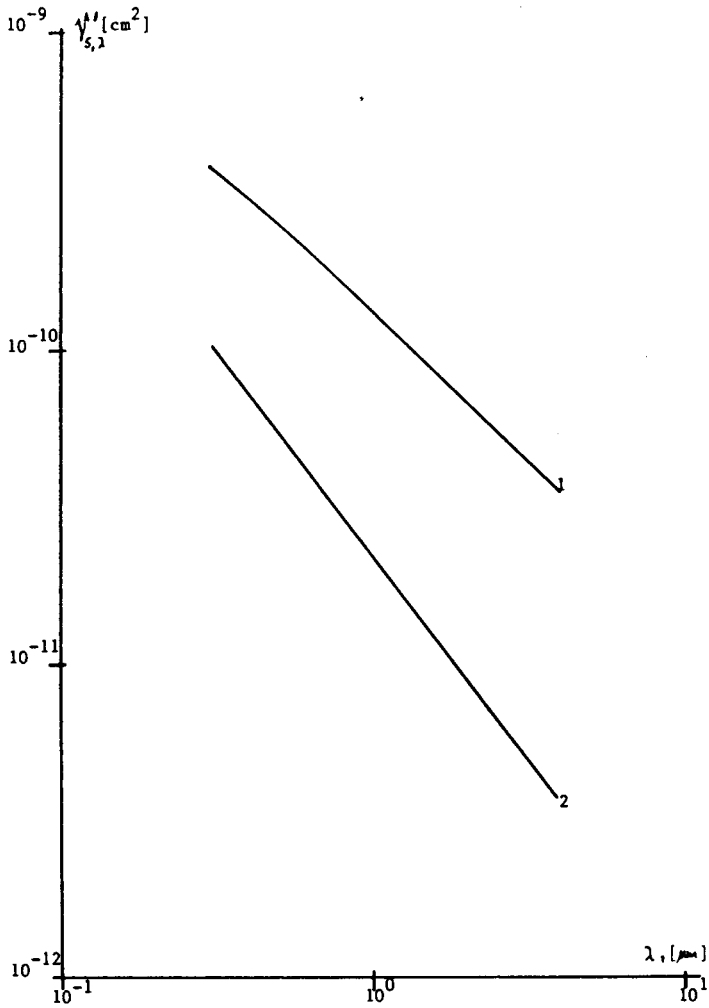


Fig. 3. Normalized extinction coefficient (cm^2) of aerosol particles.
1 - with continental dry standard distribution (Valley, 1980).
2 - with Junge's potential distribution (Muhlia, 1984).

It must be noted that δ_x refers to the relative variation in irradiance per 100% variation of the corresponding parameter. That is, the importance of any factor in the attenuation of direct beam depends on the magnitude of its variability under actual conditions. In this form, the strong variability of aerosol particle content justifies the important role that aerosol particles play in the modulation of radiative fluxes in the atmosphere.

According to the results of the numerical experiments, $\delta_N = 0.099$ for $0 \leq N_t \leq 10^9$ part/cm², and corresponds to main attenuation: *i.e.* the total number of particles in the vertical column of air. In the case of the atmospheric pressure, δ_p is equal to 0.082 for $700 \leq P \leq 1100$ mb. For water vapor values of δ_w equal to 0.062 for $0 \leq w \leq 0.5$ cm, and approximately equal to 0.050 for $0.5 \leq w \leq 3.0$ cm were obtained. For ozone a value of δ_Ω equal to 0.008 was obtained.

Additionally, we can use δ_N to estimate the relative error of N_t in terms of the relative error of I (assuming no error in the other parameters) as follows:

$$\frac{\Delta N_t}{N_t} \approx \left| \frac{\Delta I}{I} \right| \cdot 10$$

This relation is not greatly modified when the possible errors in the other parameters are included (mainly w and Ω). Then the relative error in the determination of N_t is approximately ten times the intensity relative error.

APPLICATION OF THE METHOD AND RESULTS

To apply the algorithm, series of actinometric observations were made at the following sites: Ciudad Universitaria (19°20'N and 99°11'W); San Felipe (19°18'N and 99°40'W) and El Realejo (22°37'N and 100°24'W), that correspond to different types of climatological conditions (polluted, semirural and rural atmospheres), as it was mentioned.

The measurements of direct solar radiation were made with a Linke and Feussner type pyrliometer, referenced against the standard pyrliometer Ångström No.166, that was referenced against the World Standard Group of instruments (WRC, 1981).

In Table 1 mean meteorological parameters measured for each site of observation

TABLE 1

Daily average (\pm standard deviation) of the main parameters obtained for each site in which we realize the observational experiment.

* Measured surface particles concentration. (number in parenthesis are exponents of ten).

CIUDAD UNI- VERSITARIA	\bar{P} (mb)	$\bar{\Omega}$ (atm-cm)	\bar{w} (cm)	\bar{N}_0 (cm $^{-3}$) *	\bar{N}_t (cm $^{-2}$)
310185	775 $^{\pm}$ 1.8	0.266 $^{\pm}$ 0.011~	1.8 $^{\pm}$ 0.1	1.19(4) $^{\pm}$ 0.85(4)	1.14(9) $^{\pm}$ 0.49(9)
010285	775 $^{\pm}$ 1.6		1.5 $^{\pm}$ 0.2	6.97(3) $^{\pm}$ 4.45(3)	5.21(8) $^{\pm}$ 1.78(8)
040285	777 $^{\pm}$ 1.0		1.9 $^{\pm}$ 0.1	8.71(3) $^{\pm}$ 6.59(3)	7.39(8) $^{\pm}$ 4.88(8)
060285	776 $^{\pm}$ 2.0		1.6 $^{\pm}$ 0.1	7.97(3) $^{\pm}$ 1.28(3)	9.28(8) $^{\pm}$ 3.51(8)
070285	777 $^{\pm}$ 2.0		1.6 $^{\pm}$ 0.2	10.44(3) $^{\pm}$ 6.28(3)	9.00(8) $^{\pm}$ 2.63(8)
SAN FELIPE					
060385	740.4 $^{\pm}$ 1.6	0.245 $^{\pm}$ 0.005~	1.3 $^{\pm}$ 0.1	6.35(3) $^{\pm}$ 1.93(3)	8.80(8) $^{\pm}$ 3.61(8)
070385	740.7 $^{\pm}$ 1.6		0.9 $^{\pm}$ 0.3	6.40(3) $^{\pm}$ 2.40(3)	9.84(8) $^{\pm}$ 4.43(8)
080385	740.3 $^{\pm}$ 1.4		0.6 $^{\pm}$ 0.4	8.47(3) $^{\pm}$ 2.40(3)	11.18(8) $^{\pm}$ 5.66(8)
EL REALEJO					
310584	801.9 $^{\pm}$ 1.1	0.306 $^{\pm}$ 0.005	1.9 $^{\pm}$ 0.2	8.76(2) $^{\pm}$ 1.50(2)	2.29(8) $^{\pm}$ 0.90(8)
010684	800.4 $^{\pm}$ 0.8	0.303 $^{\pm}$ 0.004	1.1 $^{\pm}$ 0.2	4.46(2) $^{\pm}$ 1.98(2)	1.08(8) $^{\pm}$ 0.80(8)
020684	801.7 $^{\pm}$ 1.1	0.300 $^{\pm}$ 0.003	1.5 $^{\pm}$ 0.6	2.83(3) $^{\pm}$ 1.57(3)	3.43(8) $^{\pm}$ 1.59(8)

~ This value is taken for all days.

are reported. As shown in this table, the atmospheric pressure and the ozone, as expected, do not show appreciable diurnal variations. The amount of precipitable water was similar for Ciudad Universitaria and El Realejo stations. For San Felipe station, the w parameter varies distinctively during the day, and from day to day.

During the field experiments the numerical concentration of aerosol particles (N_0 part/cm³) was measured using a TSI model 3030 aerosol size analyzer. With the values of N_t , determined actinometrically, and N_0 , it was possible to find the scale height, H , supposing an exponential decreasing of aerosol concentration. The values obtained for H averaged for the three stations are shown in Table 2.

The scale height at the early hours, at the three sites, is different from that of the afternoon hours (Table 2). The afternoon scale heights are significantly greater (at 5% confidence, using the Kruskal-Wallis test) than those before noon in all cases, as could be expected. This is probably a result of convective turbulent vertical motions, because of the soil radiative heating.

TABLE 2

H average values for three time intervals and for the three sites.

(* Considering noon at 12 true solar time).

STATION	Daily average [km]	Morning average* [km]	Afternoon average* [km]
Ciudad Univer- sitaria	1.101 [±] 0.550	0.589 [±] 0.241	1.511 [±] 0.300
San Felipe	1.578 [±] 0.739	0.967 [±] 0.424	2.087 [±] 0.516
El Realejo	2.161 [±] 1.376	1.579 [±] 1.155	2.614 [±] 1.423

For comparison purposes, the Ångström turbidity coefficient was calculated using basically the same numerical algorithm described in Appendix I, and substituting $\beta\lambda^{-\alpha}$ for $N_t\gamma'_{s,\lambda_i}$, with $\alpha = 1.3$ as a mean value (Ångström, 1961; Iqbal, 1983). The coefficient can be expressed in terms of the total number of aerosol particles in an air vertical column $N_t^{(J)}$ having a normalized potential Junge's distribution ($f'(s) = 8.52 \times 10^{\uparrow(-3.5 s - 5.0)}$; $-1.5 \leq s = \log_{10}(r) \leq 1.5$, Fig. 1) independent from height (z). In fact, the Ångström's expression for aerosol optical depth $\beta\lambda^{-\alpha}$ can be obtained through the general expression for the optical thickness:

$$\tau_{\lambda}^{\text{Å}} = \int_0^{\infty} \int_{s_1}^{s_2} \pi r^2 Q_{\lambda_i}(s, \tilde{m}) f'_j(s) N_t^{(J)}(z) dz ds \quad (17)$$

where $N_t^{(J)}(z)$ is Junge's total number of aerosol particles at z level.

After some calculations we get:

$$\tau_{\lambda}^{\text{Å}} = N_t^{(J)}(2.18 \cdot 10^{-11} \lambda^{-1.3}) \quad (18)$$

that is:

$$\beta = 2.18 \cdot 10^{-11} N_t^{(J)} \quad (19)$$

In the analogue equation (2) the term $2.18 \cdot 10^{-11} \cdot \lambda^{-1.3} = \gamma_{s\lambda}^{\text{Å}}$ must be interpreted as an Ångström normalized extinction coefficient (Fig. 3).

The results indicate that $N_t^{(J)}$ is almost five times N_t and that the associated scale height to $N_t^{(J)}$ will be greater by the same amount than those associated to N_t .

The main input data and results are reported graphically in Figs. 4 (a, b, c), 5(a, b, c) and 6(a, b, c) for typical days of the series and for the three stations.

CONCLUSIONS

Using direct solar radiation determinations and a supposedly more realistic model of atmospheric aerosols it is possible to estimate the aerosol concentration in the vertical atmospheric column. The determination of particle concentration contributes to the study of the atmospheric pollution.

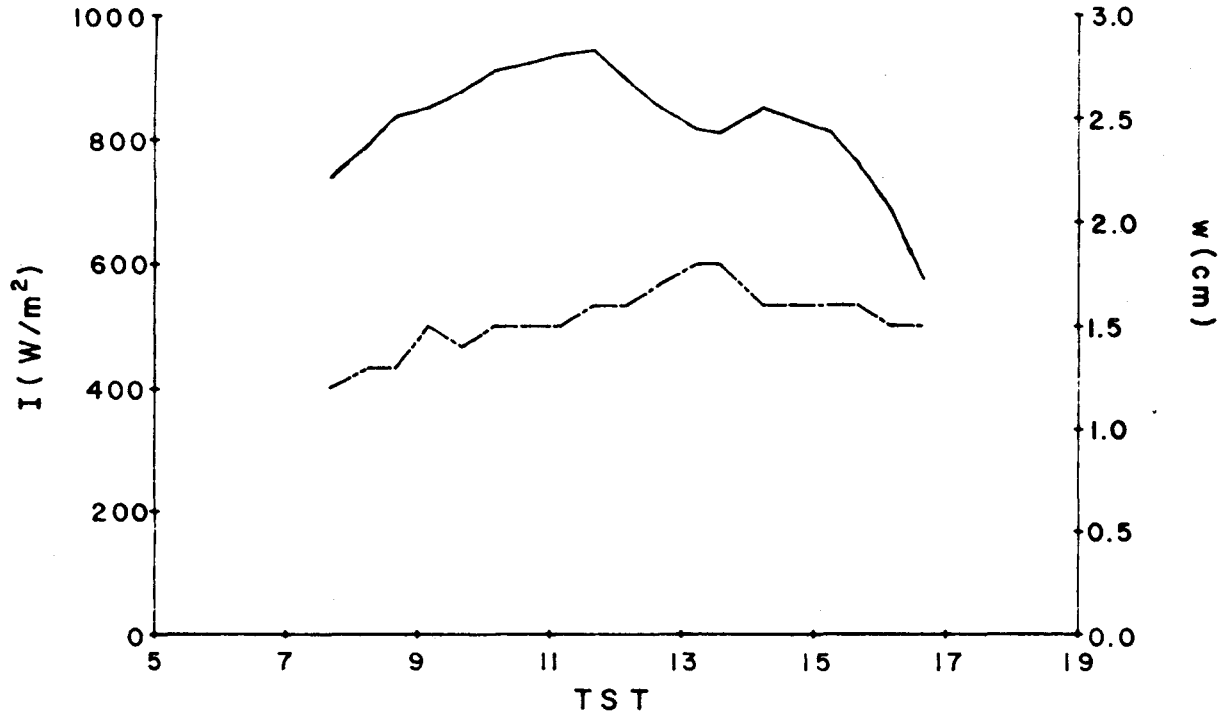


Fig. 4a. Ciudad Universitaria ($19^{\circ}20' N$; $99^{\circ}11' W$; date: Feb. 1, 1985.
 Input data: Direct solar intensity (—). Water vapor content (---).
 $P/P_0 = 0.770$ atm. $\Omega = 0.266$ atm-cm. $w = 1.5$ cm.

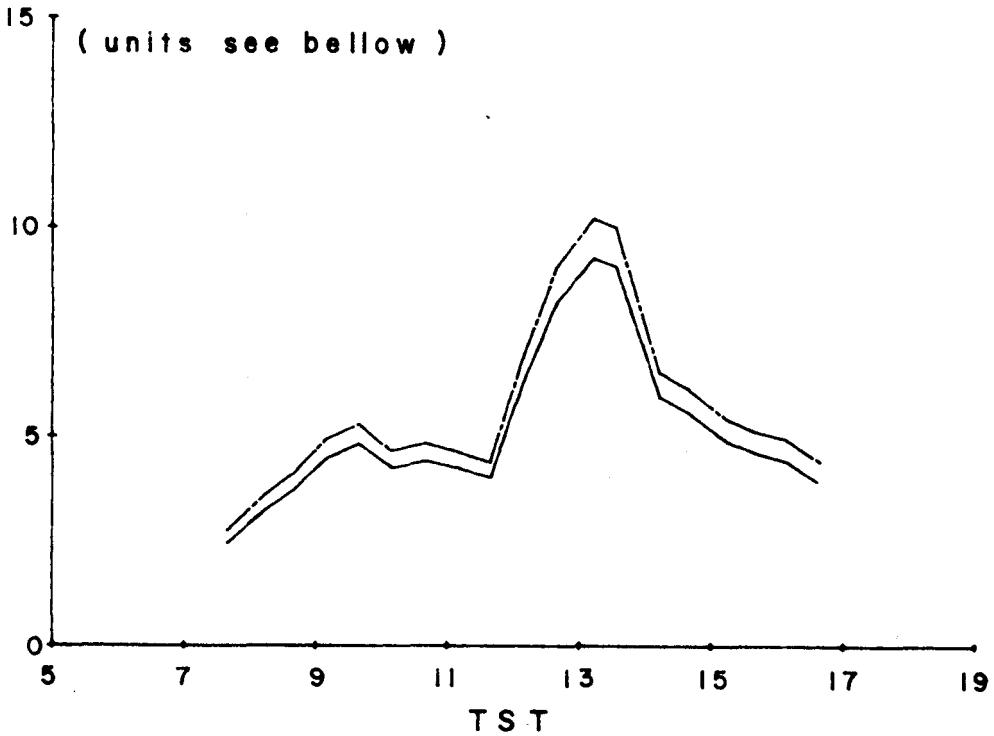


Fig. 4b. Results for Ciudad Universitaria (19°20 N; 99°11 W); date: Feb. 1, 1985.
 (—): N_t (10^8 cm^{-2}); (---): β (10^2).

The values of N_t obtained using this method are of the order of $2 \times 10^9 \text{ part/cm}^2$ in Ciudad Universitaria and in San Felipe Tlalmimilolpan and of the order of $2 \times 10^8 \text{ part/cm}^2$ at El Realejo, that, as it was mentioned earlier, correspond to a mountainous clean air area. The climatological value for the scale height H proposed by Pendorf and others (Pendorf, 1954; Shaw, 1982) are very close to that obtained here for Ciudad Universitaria station. An important finding in view of results presented here is that the use of Ångström's turbidity coefficient overestimates the total amount of particles N_t , because the linear (log-log) extrapolation from Junge's potential distribution overestimates particles with small radii between $10^{-1.5}$ and $10^{-1.0} \mu\text{m}$.

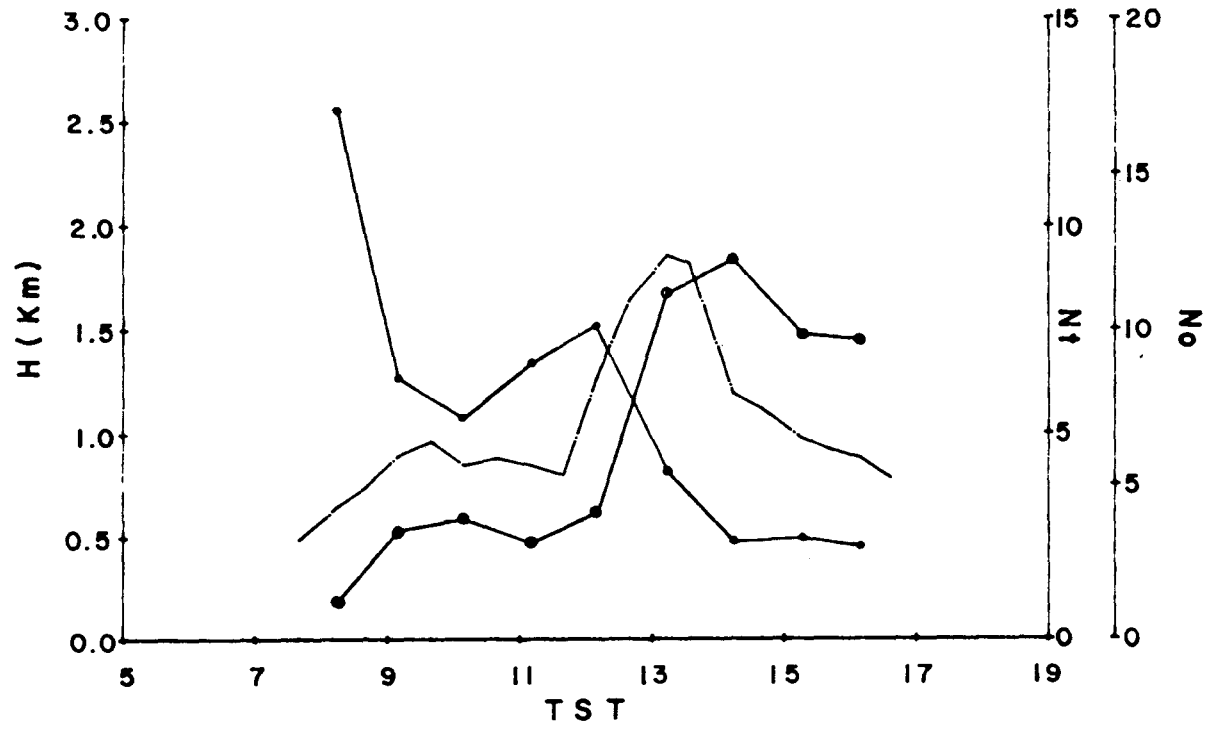


Fig. 4c. Ciudad Universitaria (19°20N: 99°11W); date: Feb. 1st., 1985.
 Scale height $H = Nt/No$: (-o-) with $Nt [10^8 \text{ cm}^{-2}]$: (---) and $No [10^3 \text{ cm}^{-3}]$: (-●-) measured.

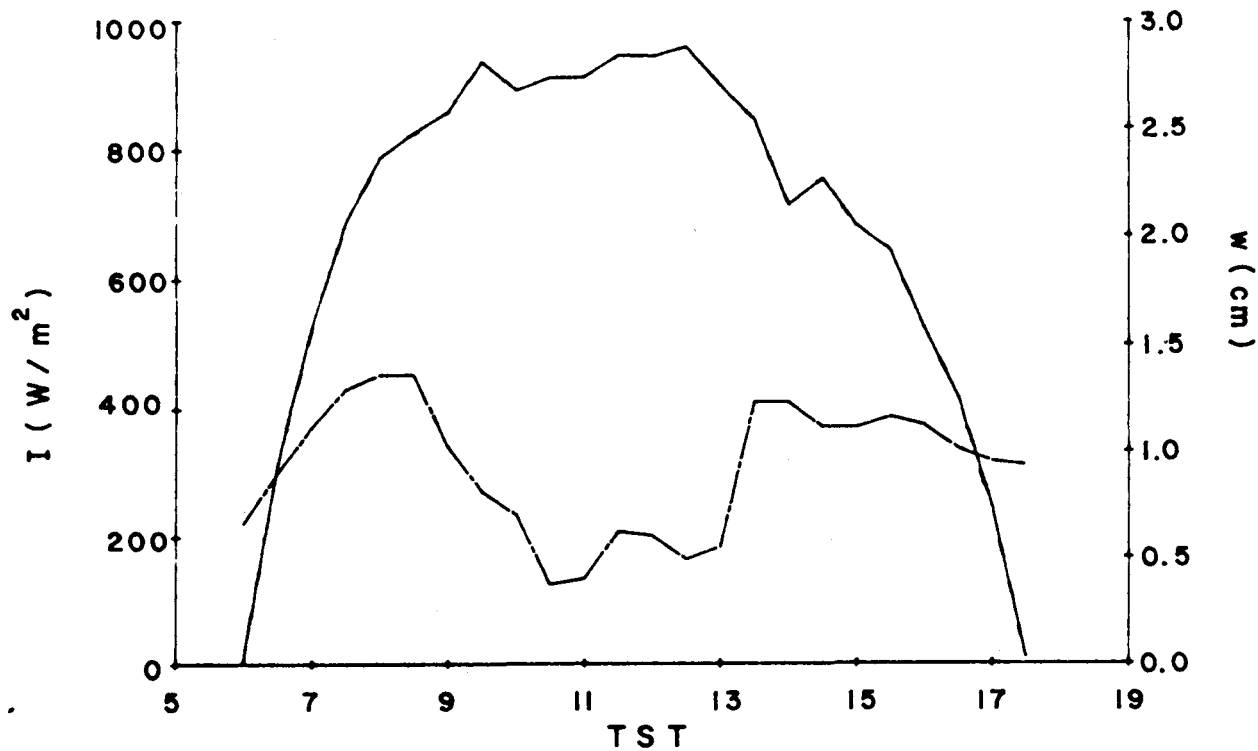


Fig. 5a. San Felipe ($19^{\circ}18'N$; $99^{\circ}40'W$); date: Mar. 7, 1985.
 Input data: Direct solar intensity (—). Water vapor content (---).
 $\bar{P}/P_0 = 0.730$ atm. $\bar{\Omega} = 0.245$ atm-cm. $\bar{w} = 0.9$ cm.

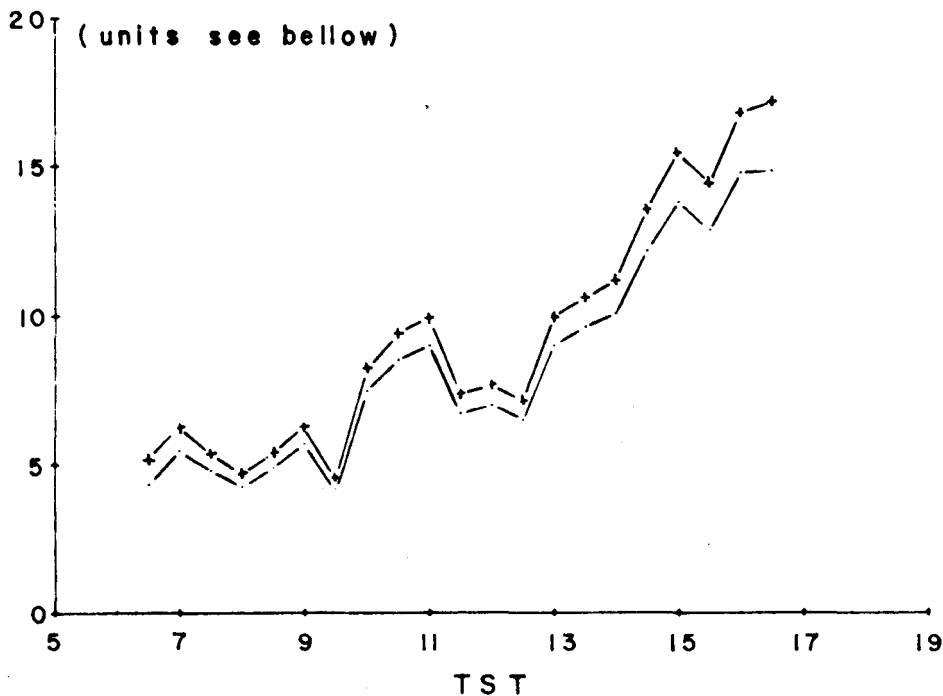


Fig. 5b. Results for San Felipe ($19^{\circ}18\text{N}$: $99^{\circ}40\text{W}$); date: Mar. 7, 1985.
 (---): $N_t [10^8 \text{ cm}^{-2}]$; (—): $\beta [10^{-2}]$.

On the other hand, parameters N_t and β may be considered both to describe the turbidity degree of the atmosphere. However, parameter N_t described here has an immediate physical meaning that can be considered as an inherent physical property of the atmospheric aerosol.

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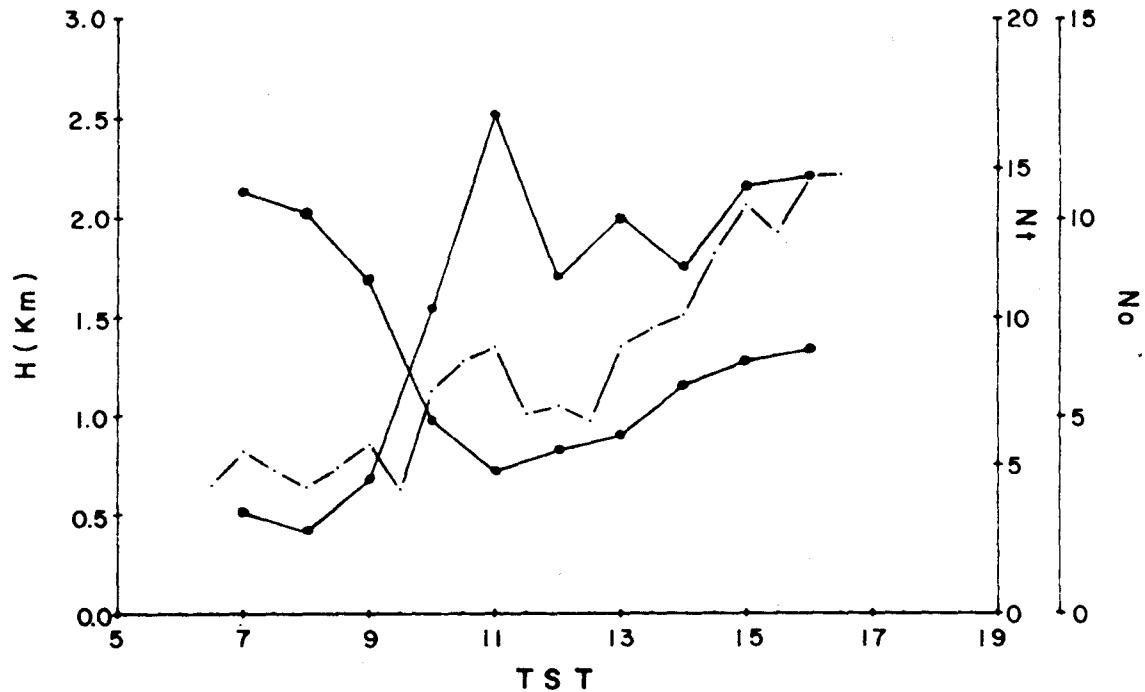


Fig. 5c. San Felipe ($19^{\circ}18\text{N}$; $99^{\circ}40\text{W}$); date Mar. 7, 1985.
 Scale height $H = N_t/N_o$: (---) with N_t [10^8 cm^{-2}]; (-.-) and N_o [10^3 cm^{-3}]; (-●-) measured.

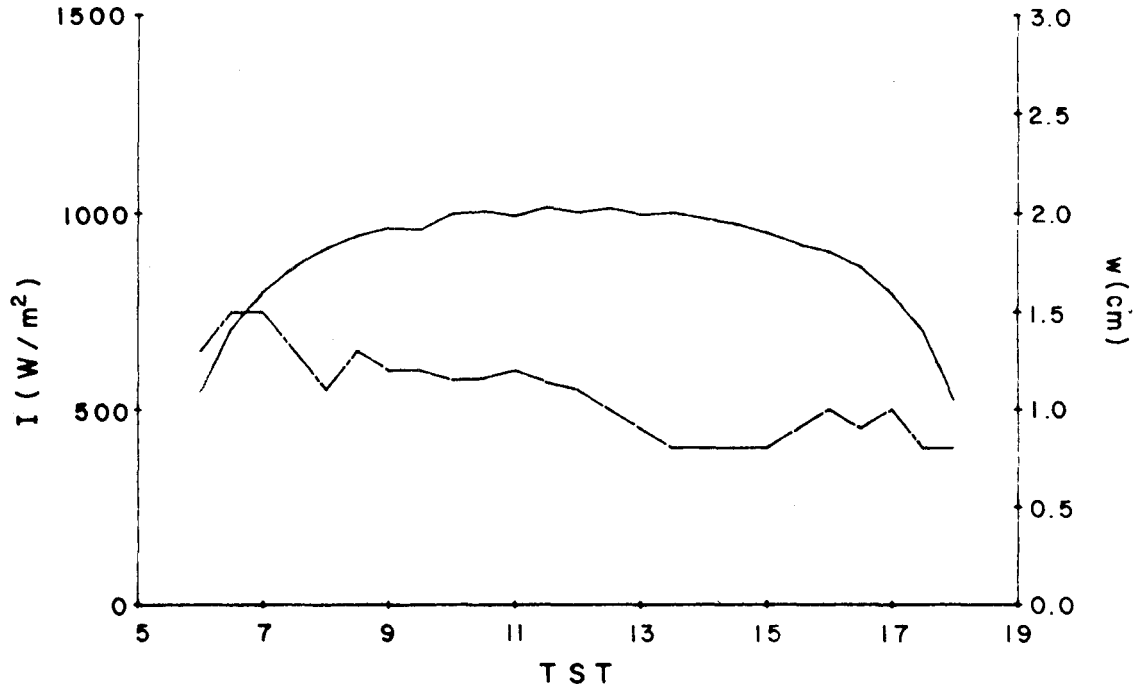


Fig. 6a. El Realejo ($22^{\circ}37' N$; $100^{\circ}24' W$); date Jun.1, 1984.
 Input data: Direct solar intensity (—); Water vapor content (---).
 $(\bar{P}/P_0 = 0.790 \text{ atm. } \bar{\Omega} = 0.303 \text{ atm-cm. } \bar{w} = 1.1 \text{ cm.})$

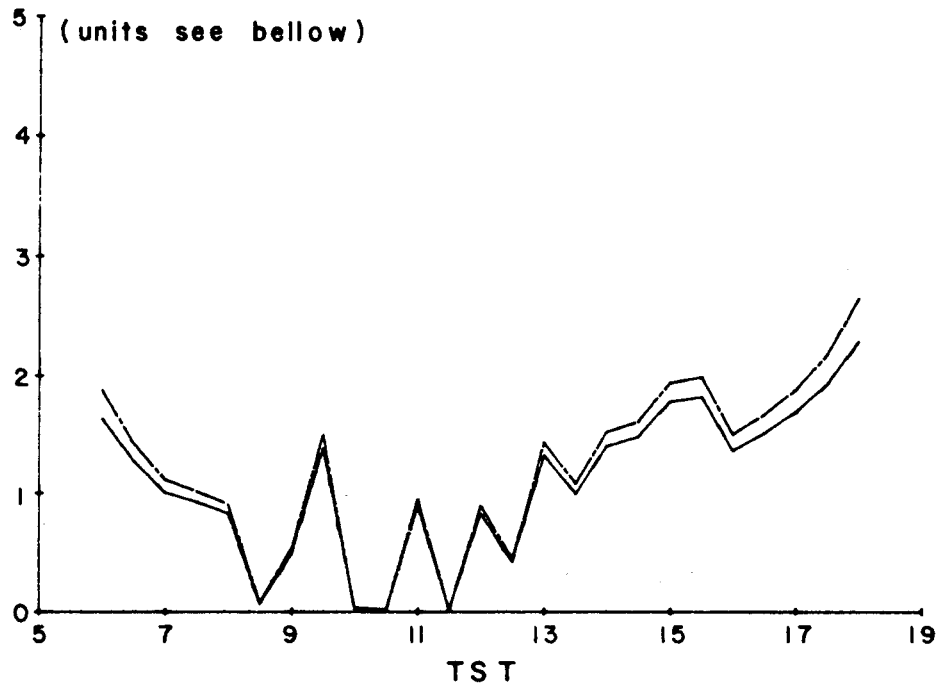


Fig. 6b. Results for El Realejo ($22^{\circ}37 \text{ N}$; $100^{\circ}24 \text{ W}$); date: Jun. 1, 1984.
 (—): N_t (10^8 cm^{-2}); (---): β (10^{-2}).

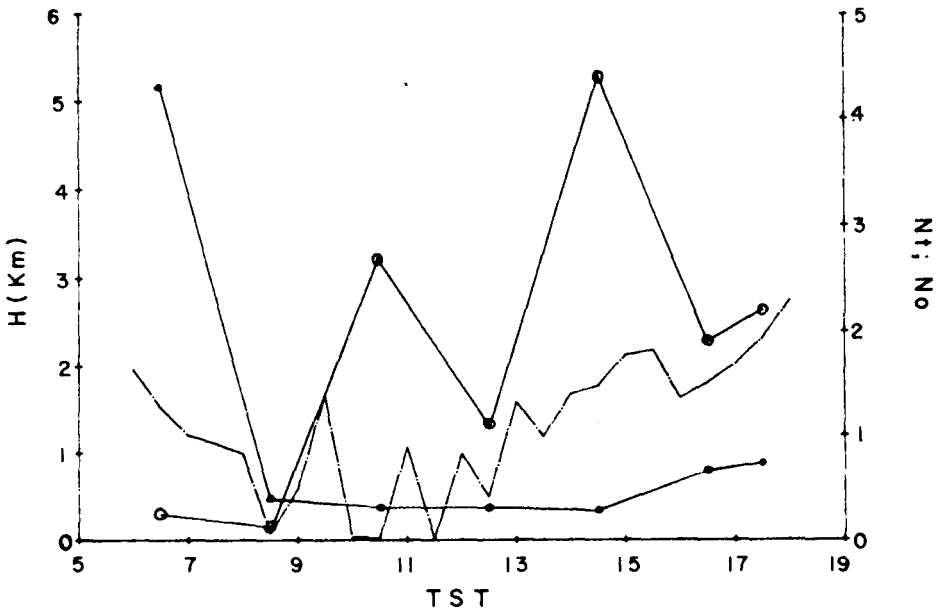


Fig. 6c. El Realejo ($22^{\circ}37' N$; $100^{\circ}24' W$); date: Jun. 1, 1984.
 Scale height $H = Nt/No$ (—●—) with Nt [10^8 cm^{-2}]: (---○---) and No [10^3 cm^{-3}]: (—○—) measured.

APPENDIX I

Brief description of the numerical algorithm to obtain N

The function $f(N)$ is defined as follows

$$f(N) = \frac{1}{R} \sum_{i=1}^n I_{0,i} T_i \exp(-mN\gamma'_{s,\lambda_i}) - I(m) \quad (1.1)$$

where: T_i represents, according to the model described in the text, the product of the air transmittances, ozone, water vapor, and uniformly mixed gases. $I(m)$ is the direct solar radiation intensity measured at a given time. It is easy to show that this

function is defined and continuously differentiable for all real N values, in addition, suppose that the derivative of $f(N)$ with respect to N is different from zero ($f'(N) \neq 0$) for finite values of N . Finally, assume that the equation:

$$f(N) \neq 0 \quad (1.2)$$

has necessarily a unique solution $N = N_t \in (a, b)$ where (a, b) is a numerical real interval. Under this hypothesis it is possible to construct and demonstrate that the following algorithm converge to some solution for an initial value N_0 sufficiently close to N_t (the experience shows that a good value for N_0 is 10^4). This method is known as the Newton-Raphson algorithm (Henrici, 1984).

Algorithm:

- 0) Choice N_0
- 1) Determine the sequence N_i using the recurrence relation

$$N_{i+1} = N_i - \frac{f(N_i)}{f'(N_i)} \quad (1.3)$$

- 2) Use the following criteria to obtain N_t with convenient approximation:

$$\text{if } |N_{i+1} - N_i| < \delta \quad \text{then } N_t = N_{i+1} \quad (1.4)$$

δ is a number sufficiently small such that it permits us to have N_t with certain exact figures *e.g.* if $\delta = 0.5$, then N_t has at least exact integer values.

- 3) Otherwise, go to 1.

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