Effect of Aerosols on the Estimation of Total Ozone in an Atmospheric Column from the Measurements of Its Ultraviolet Radiance

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ABSTRACT

Values of the total ozone content of an atmospheric column are estimated from the simulated five-wavelength measurements of the intensity of radiation backscattered by a horizontally homogeneous earth-atmosphere system along the nadir direction, and after making use of an estimation procedure similar to the one developed by Mateer et al. (1971) for obtaining total ozone values from the backscatter ultraviolet (BUV) spectrometer data aboard the Nimbus 4 satellite. Model atmospheres with different ozone contents and with different kinds of aerosols but with a fixed surface pressure (1000 mb) are assumed to rest on a Lambert surface of 0.2 reflectivity. In general, estimated ozone amounts are found to differ significantly from the corresponding input to the models with the difference between these two values depending on several factors such as solar zenith angle, aerosol parameters and the wavelength pair used in the estimation. For an aerosol attenuation optical thickness of about 0.2 at 0.3800 μm (average haziness under clear sky conditions), this difference is about 0.006 atm·cm provided the analysis is restricted to cases with solar zenith angles < 85°. For strong hazy conditions represented by the aerosol attenuation optical thickness of about 1.0 at 0.3800 μm, this difference is found to be as large as 0.025 atm·cm.

It is also shown that the effect of tropospheric particulate pollutants on total O$_3$ estimation can be reduced by a very significant factor in many cases if an additional spectral measurement is made available in the region of very weak O$_3$ absorption. Such an additional measurement assists in estimating the spectral dependence of the effective albedo of the earth-lower troposphere system.

1. Introduction

Ozone is perhaps the single most critical stratospheric constituent because its vertical distribution directly affects the deposition of energy in the stratosphere, while its total amount determines the ultraviolet radiation reaching the biosphere. Within the past decade, under the probability of increasing pollutant concentrations due to commercial stratospheric aviation and vertical transport of chlorofluorocarbons as well as of oxides of nitrogen, the stability of the stratospheric ozone layer has been questioned repeatedly in scientific journals as well as in the press (Wofsy et al., 1975). Because of this, considerable attention is currently being given to the problem of reliable and continuous monitoring of atmospheric O$_3$ on a global scale.

One of the techniques well suited for this purpose is a thorough analysis of the spectral signature of the earth-atmosphere system in the UV part (0.25–0.38 μm; 1 μm = 10$^{-6}$ cm) of the electromagnetic spectrum. This particular technique was first proposed and discussed in the open literature by Singer and Wentworth (1957). Dave and Mateer (1967) looked into the feasibility of estimating the total O$_3$ content of an atmospheric column from satellite measurements of its UV radiance. This study of Dave and Mateer was restricted to models obeying Rayleigh’s law of scattering only. They recommended the use of a pair of wavelengths, one within 0.31–0.32 μm (moderate absorption by ozone), and the second within 0.33–0.34 μm (weak absorption by ozone). According to these investigators, the total O$_3$ content of an atmospheric column can be estimated within 5% provided the spectral measurements are made within 1% accuracy, surface reflectivity within 0.05, and pressure at the lower boundary within 50 mb. Furthermore, when the zenith angle of the sun exceeds 60°, a significant fraction of the emergent radiation originates from the stratosphere and hence it is imperative to have correct information about the high-level O$_3$ profile at that time.

Mateer et al. (1971) developed a procedure for estimating the total O$_3$ amount of an atmospheric column from nadir measurements of the backscattered radiation at 0.3125, 0.3175, 0.3312, 0.3398 and 0.3800 μm taken with the backscatter ultraviolet (BUV) double monochromator aboard the Nimbus 4 meteorological satellite. They then used their procedure for estimating total O$_3$ for a sample of 320 cases of approximate coincidences (in space as well as in time) of the BUV data, and ground-truth measurements of total O$_3$ with
the Dobson spectrophotometer. They found that, on the average, the satellite values were lower than the corresponding Dobson values by about 0.025 atm-cm. Some cases were also reported where the difference between these two values was as large as 0.060 atm-cm. They believed that a part of this difference was due to lack of a perfect coincidence between the Dobson and satellite data. Results of the analysis of BUV data for a two-year period are also reported by these investigators (Heath et al., 1973).

The main purpose of this paper is to report the results of an investigation aimed at estimating the effect of stratospheric as well as tropospheric particulate pollutants on total ozone estimates derived from the analysis of simulated BUV data. Computational details for the generation of these simulated observations are given in Section 2. Section 3 contains information about the estimation procedure used by us for obtaining total ozone values from our simulated observations. In principle, our estimation procedure is similar to the one used by Mateer et al. (1971). Results of our investigation are reported in Sections 5 and 6 after providing information about various aerosol parameters in Section 4.

An additional spectral measurement at 0.3600 μm is planned with the total ozone mapping spectrometer (TOMS) instrument aboard the Nimbus G satellite which is scheduled to be launched during 1978. Therefore, the five-wavelength total-ozone estimation procedure of Section 3 is modified to include the additional simulated measurement so as to permit accounting of the spectral dependence of the effective albedo of the earth-lower troposphere system. Estimates of the total O₃ amounts obtained with the five-wavelength and six-wavelength procedures are compared in Section 7.

2. Computational details

The total ozone estimation procedure described in Section 3, and the generation of the simulated observations used for our investigations, require computations of the following quantities:

1) $I(\lambda, \Omega_{\text{in}}, R=0.0, \theta_0)$: Intensity (also referred to as the radiance) of the scattered radiation emerging at the top of the atmospheric model along the local nadir direction ($\theta=0^\circ$) where $\lambda$ is the wavelength of the incident radiation, $\Omega_{\text{in}}$ the total ozone content (atm-cm) of an atmospheric column of 1 cm$^{-2}$ cross section, $\theta_0$ the solar zenith angle (deg), and $R$ the surface reflectivity (see below).

2) $T(\lambda, \Omega_{\text{in}}, \theta_0)$: A quantity representing the total (direct plus diffuse) transmission along the local nadir direction when the atmospheric model is illuminated from below, isotropically, by a Lambertian reflection of all energy incident upon the ground.

3) $S(\lambda, \Omega_{\text{in}})$: Diffuse flux reflectivity of the model for the case of its isotropic illumination from below.

In addition to these parameters, the aforementioned quantities are also dependent on pressure at the lower boundary (assumed 1000 mb), and various aerosol parameters to be represented by a two-part symbol to be described in Section 5.

Intensity $[I(\lambda, \Omega_{\text{in}}, R, \theta_0)]$ of the diffuse radiation emerging along the local nadir direction for models resting on a Lambert surface of reflectivity $R$ is then given by

$$I(\lambda, \Omega_{\text{in}}, R, \theta_0) = I(\lambda, \Omega_{\text{in}}, R=0.0, \theta_0) + \frac{R}{1-RS(\lambda, \Omega_{\text{in}})} T(\lambda, \Omega_{\text{in}}, \theta_0).$$

The quantity appearing on the left-hand side of Eq. (1) will be denoted by the symbol $I_s(\lambda, \Omega_{\text{in}}, R, \theta_0)$ when its computed values are to be used for generation of the $N_e(\lambda, \Omega_{\text{in}}, R, \theta_0)$ vs $N_e$ curves (see Section 3), and by the symbol $I_n(\lambda, \theta_0)$ when its computed values are to be used as simulated measurements (without any random error) for obtaining information about the total O₃ amount in the atmospheric column under scrutiny.

Our basic atmospheric model is that of a plane-parallel atmosphere of homogeneous character and infinite extent along the horizontal directions. It is of finite optical extent along the vertical direction to which any nonhomogeneity due to scattering and/or absorption is confined. In order to permit meaningful calculations for $\theta_0$ up to 90$^\circ$, the sphericity of the atmosphere is partly accounted for by computing the attenuation suffered by the incoming solar radiation arriving at various atmospheric levels for the actual terrestrial geometry. Thus, our model can be thought of as a pseudo-spherical model of the terrestrial atmosphere. It should be emphasized that refraction of the incoming beam is not taken into account. Sphericity of atmosphere is also taken account in such an approximate manner in the work of Mateer et al. (1971).

The computation algorithm of the aforementioned quantities makes use of the method of the direct numerical solution of the spherical harmonics approximation to the scalar form of the appropriate equation of radiative transfer (Dave and Canosa, 1974; Dave, 1974; Dave and Armstrong, 1974). This algorithm is for an atmospheric model consisting of 32 nonhomogeneous basic layers. The height $h$ of the base of these basic layers above the 100 mb level is given as $h=0$ (1) 25 km, 25 (5) 50 km and 50 (10) 60 km. The top of the atmosphere is assumed to be located at a height of 70 km. Provision is made for assigning $O_3$ amount and aerosol number contents of a 1 cm$^2$ cross-section column to each of these basic layers. Two different kinds of aerosol (assumed to exist in the form of a spherical polydispersion) height distributions can be accommodated by this algorithm. After computing total optical thicknesses of all basic layers, basic layers with a total
optical thickness greater than 0.02 are further divided into sublayers so that the total optical thickness of no sublayer exceeds 0.02. This is necessary for maintaining a reasonably high degree of accuracy during computations of the intensity moments and integrals. For some $\lambda$, $\Omega_\text{in}$ and aerosol parameter combinations, it was found necessary to divide the model into as many as 300 sublayers.

For all wavelengths, computations are carried out after assuming that $\pi \cos \theta_0$ units of solar energy are incident normal to a unit horizontal area located at the top of the model in each spectral band.

Computations of $I(\lambda, \Omega_\text{in}, R=0.0, \theta_0)$ and $T(\lambda, \Omega_\text{in}, \theta_0)$ are performed for 10 different values of the parameter $\theta_0$, viz., $0^\circ$, $45^\circ$, $60^\circ$, $70^\circ$, $75.6^\circ$, $79.6^\circ$, $82.5^\circ$, $84.7^\circ$, $86.7^\circ$ and $90^\circ$. This set of values was used by Mateer et al. (1971) in preparation of the basic tables for their total ozone estimation investigations.

3. Total ozone estimation procedure

The backbone of the total ozone estimation procedure is a set of basic tables of the computed quantities $I(\lambda, \Omega_\text{in}, R=0.0, \theta_0)$, $T(\lambda, \Omega_\text{in}, \theta_0)$ and $S(\lambda, \Omega_\text{in})$ for terrestrial atmospheric models free of any particles comparable to, or larger than, the wavelength of incident radiation. These tables are prepared for the 10 different values of $\theta_0$ listed in Section 2 and for 10 different values of the parameter $\Omega_\text{in}$ given by $\Omega_\text{in} = 0.200$, 0.050, 0.650 atm-cm. Ozone profiles for three of these $\Omega_\text{in}$ values (viz., 0.200, 0.400 and 0.600) as used for our study are shown in Fig. 1. These O$_3$ profiles correspond to those observed in midlatitudes and are based on data provided by Mateer (private communication). In brief, various O$_3$ profiles are obtained by varying O$_3$ contents of the tropospheric and lower stratospheric layers. The origin of these data is the balloon, rocket and Umkehr ozone soundings of the atmosphere. In addition, the aforementioned quantities are computed for six wavelengths whose normal Rayleigh scattering optical thickness for the standard atmosphere and absorption coefficient of 1 atm-cm of ozone to the base $e$, as used in our work, are given in Table 1.

Having obtained simulated observations $I_m(\lambda, \theta_0)$ at $\lambda=0.3125$, 0.3175, 0.3312, 0.3398 and 0.3800 $\mu$m for a given $\theta_0$, the first step is the computation of the effective albedo ($R'$) using the $I_m(0.3800, \theta_0)$ measurement [the appropriate section of the basic tables are described in Table 1.

<table>
<thead>
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<th>Wavelength ($\mu$m)</th>
<th>$\tau_0^{(R,R)}$</th>
<th>$\alpha$</th>
</tr>
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<td>1.0200</td>
<td>1.6700</td>
</tr>
<tr>
<td>0.3175</td>
<td>0.9570</td>
<td>0.9100</td>
</tr>
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<td>0.8000</td>
<td>0.1750</td>
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<tr>
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<td>0.5634</td>
<td>0.0012</td>
</tr>
<tr>
<td>0.3800</td>
<td>0.4494</td>
<td>0.0000</td>
</tr>
</tbody>
</table>
parameters. It is given by the following equation provided the same value for the extraterrestrial flux is assumed at both wavelengths:

$$N_e(\lambda_i, \lambda_j; \Omega_{in}, R, \theta_0) = 100 \log_{10} \left[ \frac{I_e(\lambda_i, \Omega_{in}, R, \theta_0)}{I_e(\lambda_j, \Omega_{in}, R, \theta_0)} \right].$$ \hspace{1cm} (2)

It will be defined for two wavelength pairs, viz., $\lambda_i = 0.3312 \mu m$ and $\lambda_j = 0.3125 \mu m$ which will be referred to as the A pair, and $\lambda_i = 0.3398 \mu m$ and $\lambda_j = 0.3175 \mu m$ which will be referred to as the B pair. Variations of $N_e(\lambda_i, \lambda_j; \Omega_{in}, 0.0, \theta_0)$ vs $\Omega_{in}$ for all 10 values of the parameter $\theta_0$ listed earlier are shown in Figs. 2 and 3 for the A and B pairs, respectively. For high sun, the A pair is more sensitive to changes in ozone amounts than the B pair. As for example, at $\Omega_{in} = 0.250$ atm-cm and $\theta_0 = 0^\circ$, $[\Delta N_e/\Delta \Omega_{in}]_A = 118$ log unit (atm-cm)$^{-1}$ and $[\Delta N_e/\Delta \Omega_{in}]_B = 71$ log unit (atm-cm)$^{-1}$. The sensitivity of the A pair generally decreases rapidly with increase in $\Omega_{in}$ and $\theta_0$. At $\theta_0 = 82.5^\circ$, the quantity $N_e$ for the A pair changes very little with an increase in $\Omega_{in}$ if $\Omega_{in} > 0.400$ atm-cm. For $\theta_0 = 84.7^\circ$ and $86.7^\circ$, $N_e$ vs $\Omega_{in}$ curves for the A pair show a distinct maximum. This loss of sensitivity of the A pair to changes in $\Omega_{in}$ at low sun is due to the rise of the contribution function of $I_e(0.3125, \Omega_{in}, 0.0, \theta_0)$ through the lower stratosphere (Dave and Mateer, 1967). The B pair also loses its sensitivity to changes in $\Omega_{in}$ at $\theta_0 = 86.7^\circ$, if $\Omega_{in}$ is large, and at $\theta_0 = 90^\circ$ for over nearly the entire range of $\Omega_{in}$ considered here. Variations of $N_e(0.3398, 0.3175; \Omega_{in}, R, 84.7^\circ)$ vs $\Omega_{in}$ are shown in Fig. 4 for $R = 0.0, 0.5$ and 1.0. Such diagrams assist in understanding the errors introduced in the estimated ozone amount due to a specified error in $R$.

The second step of the total $O_3$ estimation procedure...
is then construction of $N_e$ vs $\Omega_{in}$ curves for both pairs, but for the required value of $\theta_0$ and $R'$. For this purpose, we have to assume that $R''$ is independent of $\lambda$.

The third step is computation of the measured logarithmic ratio values given by

$$N_m(x=A \text{ or } B, \theta_0) = 100 \log_{10} \frac{I_m(\lambda, \theta_0)}{I_m(\lambda_0, \theta_0)},$$

and obtaining of the first estimates of total ozone amounts $\Omega_A^*$ and $\Omega_B^*$ using curves of the pairs A and B, respectively.

The fourth step consists of estimating of a new value of the effective albedo ($R''$) by making use of the $I_m(0.3398, \theta_0)$ measurement, value of $\Omega_A^*$ or $\Omega_B^*$ depending on which pair has greater sensitivity in the region of immediate interest and Eq. (1) with the appropriate section of the basic tables. The reason for this (Mateer et al., 1971) is that if there is some dependence of the effective albedo on $\lambda$, $R''$ would be closer than $R'$ to the real effective albedo.

Finally, we construct the required $N_e$ vs $\Omega_{in}$ curves using the $R''$ value (repeat step 2), and obtain new estimates of the total ozone amounts in the atmospheric column under investigation (repeat step 3). Again, these new estimates using A(B) pair data can be denoted by $\Omega_A^*$ ($\Omega_B^*$). However, for convenience, we will use $\Omega_A^*$ in the place of $\Omega_A^*$ and $\Omega_B^*$ in the place of $\Omega_B^*$; $\Omega_A^*$ is the total ozone estimate obtained from the observations at the xth pair.

Mateer et al. (1971) select the so-called best total ozone estimate value from those of $\Omega_A^*$ and $\Omega_B^*$ obtained after using two different sets of basic tables and after examining slopes of $N_e$ vs $\Omega_{in}$ curves in the region of immediate interest. Since the pressure at the lower boundary at the time and place of a BUV measurement is unknown, they developed an elaborate procedure based on several assumptions and intuition. However, we have no need of introducing these aspects in our investigation where the information about the surface pressure is available.

4. Aerosol parameters

Aerosols of a unit volume of air in our models are assumed to exist in the form of a spherical polydispersion of known size distribution characteristics and made of a material of known refractive index ($m = 1.5 - n_2$) with respect to air. There is some evidence in the published literature (Holland and Gagne, 1970) suggesting that the scattering by a group of randomly oriented irregular particles may not be approximated to that due to equivalent spheres of the same refractive index at all times. However, this sphericity assumption cannot be expected to modify our conclusions in any significant manner.

The size distribution characteristics of spherical polydispersions used in our investigations are represented by three modified gamma distributions [Haze H, L and M (Deirmendjian, 1969)], and by a discontinuous function called the Junge size distribution function (Bullrich, 1964). This fourth function, to be referred to herein as Haze J, has a constant particle number density in the radius range 0.02–0.1 $\mu$m, and it decreases as $(\text{radius})^{-4}$ in the radius range 0.1–7.0 $\mu$m. Further information about the rationale behind the use of these size distribution functions for representing size distribution characteristics of the terrestrial aerosols can be found in the respective references.

In Fig. 5, we have shown variations of the scattering cross section per average particle as a function of the wavelength $\lambda$ ($\mu$m) of the incident radiation for four different spherical polydispersions. $m = 1.5-0.0i$, presented by three modified gamma distributions [Haze H, L and M (Deirmendjian, 1969)], and by a discontinuous function called the Junge size distribution function (Bullrich, 1964). This fourth function, to be referred to herein as Haze J, has a constant particle number density in the radius range 0.02–0.1 $\mu$m, and it decreases as $(\text{radius})^{-4}$ in the radius range 0.1–7.0 $\mu$m. Further information about the rationale behind the use of these size distribution functions for representing size distribution characteristics of the terrestrial aerosols can be found in the respective references.

In Fig. 5, we have shown variations of the scattering cross section per average particle (cm$^{-2}$ per average particle) as a function of the wavelength for four spherical polydispersions listed in the preceding paragraph. The results presented in this diagram are those of aerosol particles made from a substance with a refractive index of 1.5–0.0i. The scattering cross section per average particle of the spherical polydispersion called Haze M or L increases with an increase of wavelength in the spectral region of concern to our present study. On the other hand, the curve for the Haze H spherical polydispersion exhibits a broad maximum around 0.35 $\mu$m wavelength, and that for the Haze J case shows a decrease in scattering cross section with an increase in $\lambda$.

Variations of the scattering cross section (solid curves) and the absorption cross section (broken curves) per average particle as a function of the imaginary part ($n_2$, range 0.0–0.1) of the refractive index of the aerosol material are shown in Fig. 6 for $\lambda = 0.3398 \mu$m and for the four polydispersions listed earlier. From the analysis of all data, we find that the changes in cross sections for a given $n_2$ size distribution combination, are not more than 15% over the 0.31–0.38 $\mu$m spectral range.
are presented only for the angular range of immediate interest, *viz.,* 90–180°. For observations along the local nadir, the direction of interest varies from Θ = 180° to Θ = 90°, as the solar zenith angle (θ₀) is changed from 0° to 90°. For m = 1.5–0.0i, the L and M polydispersions are, relatively speaking, more efficient scatterers in the angular range 150–180° than the other two. An increase in the imaginary part of the refractive index results in the disappearance of this difference among these four cases. P(\cos Θ) vs Θ curves for the other five wavelengths also depict the same general features.

For studying the effect of aerosols on total O₃ estimations, we have chosen two aerosol-content versus height curves which together are expected to represent vertical profiles of aerosols encountered in the terrestrial atmosphere under average conditions. One of these aerosol-content height profiles is void of any particles in the lower atmosphere, and is therefore referred to as the stratospheric distribution. The other, with its aerosol content confined to the lower parts of the atmosphere only, is referred to as the tropospheric distribution.

In Fig. 8, we have shown variations of the pressure thickness (mb km⁻¹) and the aerosol number-density (number cm⁻² km⁻³) for the stratospheric as well as tropospheric distributions as a function of height (km).

**Fig. 6.** Variations of the scattering cross section (solid curves) and of the absorption cross section (dashed curves) per average particle as a function of the imaginary part (n₂) of the refractive index (m = n₁ − in₂) of the aerosol material. Different curves are for different spherical polydispersions (*viz.,* Haze H, J, L and M).

**Fig. 7.** Variations of the normalized phase function of a unit volume of four different spherical polydispersions (Haze H, J, L, and M) as a function of the scattering angle. The upper (lower) set of curves is for the polydispersions made from a material with a refractive index of 1.5–0.0i (1.5–0.1i). An integration of a given phase function over a solid angle of 4π yields a value of 4π.

**Fig. 8.** Variations of the pressure thickness (scale at the top) and of the stratospheric as well as tropospheric aerosol number-density (number cm⁻² km⁻³) as a function of height as used in our investigations. This figure is for the models with 1×10⁶ stratospheric and/or 40×10⁶ tropospheric particles in a column of 1 cm² cross section. Curves for the models with aerosol contents greater than those presented here are obtained after shifting the appropriate curve to the right as necessary.
Results presented in this diagram are for the models with \(1 \times 10^6\) and \(40 \times 10^6\) average particles in the stratospheric and tropospheric cases, respectively. Similar data for the models with total aerosol contents different from those presented here are obtained after multiplying the aerosol-content vector of a given distribution by the appropriate scalar factor. In other words, for our present study, a change in the total aerosol content of the atmospheric column for a given type of aerosol implies no change in the relative vertical profile of that aerosol distribution.

The aerosol attenuation optical thickness of a given model can be obtained by multiplying the sum of the scattering and absorption cross sections of the appropriate size distribution function and refractive index, by the total aerosol content of the aerosol height distribution used in the model.

5. Effect of aerosols on intensities

For identification of all variable characteristics of aerosols in an atmospheric model we will use a two-part symbol. These two parts of the symbol are connected by a hyphen. The first (second) part of the symbol represents information about the stratospheric (tropospheric) aerosol. Each part of the symbol has the form \(n n Y\ mm\), if the kind represented by that part is present in the model. The numerals \(n n\) (sometimes \(n\) or \(nn\) or \(nnn\)) represent the total aerosol content (in millions of average particles per square centimeter) of the given type of aerosol in the model. The letter \(Y\) specifies the size distribution function, i.e., \(Y = H, J, L,\) or \(M\). The numerals \(n m\) are equal to 100\(nm\). Thus, the symbol \(0-200 L 05\) means that the model has no stratospheric aerosols, but it contains \(200 \times 10^6\) particles having its size distribution characteristics corresponding to that of the Haze \(I\), made from a material with \(m = 1.5-0.05\) and its vertical profile given by the curve marked "tropospheric" in Fig. 8.

In order to obtain some insight into the problem of the effect of aerosols on total \(O_s\) estimations, we will consider variations of the following two ratios, as a function of the wavelength of incident radiation, for several atmospheric models with only stratospheric and only tropospheric aerosols (results are presented for the case of a nonreflecting ground to bring out the atmospheric effects):

\[
\rho_{sl}(\lambda, \theta_0, nn \ H \ mm - 0) = \frac{I(\lambda, \Omega_{in}, 0,0, \theta_0) \text{ for the model } nn \ H \ mm - 0}{I(\lambda, \Omega_{in}, 0,0, \theta_0) \text{ for the model } 0 - 0},
\]

\[
\rho_{tp}(\lambda, \theta_0, 0 - nnn \ L \ mm) = \frac{I(\lambda, \Omega_{in}, 0,0, \theta_0) \text{ for the model } 0 - nnn \ L \ mm}{I(\lambda, \Omega_{in}, 0,0, \theta_0) \text{ for the model } 0 - 0}.
\]

Fig. 9. Variations, as a function of the wavelength (\(\mu m\)) of the changes in the intensity of radiation emerging along the local nadir direction due to the presence of stratospheric aerosols (Haze \(H\)) in the atmospheric model. Solid curves: \(m = 1.5-0.05\); dotted curves: \(m = 1.5-0.05\); dashed curves: \(m = 1.5-0.1\).

Parameters \(\Omega_{in}\) and \(R\) are omitted in the description of \(\rho_{sl}\) and \(\rho_{tp}\) as our discussion of these two ratios is restricted to \(\Omega_{in} = 0.250\) atm-cm, and \(R = 0.0\) cases only. These ratios represent increase, or decrease, in the intensity of the radiation backscattered along the local nadir direction due to the introduction of aerosols in the model.

Variations of \(\rho_{sl}(\lambda, \theta_0, nn \ H \ mm - 0)\) vs \(\lambda\) for \(\theta_0 = 0°, 79.6°\) and \(90°\) are shown in Fig. 9 for the atmospheric models \(20 H 00 - 0, 20 H 05 - 0\) and \(20 H 10 - 0\). The aerosol attenuation optical thickness for these models at \(0.380 \mu m\) is about 0.055. [Based on the work of Dave and Mateer (1968), we feel that a stratospheric aerosol content of \(20 \times 10^6\) particles in a \(1 \ cm^2\) cross-section column is very high for average atmospheric conditions as it would lead to the occurrence of bright purple twilights which are only observed after powerful volcanic eruptions. For average conditions, a value of \(5 \times 10^6\) for the stratospheric aerosol content seems to be more reasonable. However, it should be pointed out that stratospheric aerosol contents greater than \(20 \times 10^6\) particles in a \(1 \ cm^2\) cross-section column may be encountered under some special circumstances.] For \(\theta_0 = 0°\), the ratio \(\rho_{sl}(\lambda, \theta_0, nn \ H \ mm - 0)\) is independent of \(\lambda\) for all practical purposes. Its value decreases from 1.03, to 0.98 to 0.96 as the imaginary part of the refractive index is increased from 0.0 to 0.05 to 0.1. Thus, it can be concluded that for the case of the overhead sun,
non-absorbing stratospheric aerosols contribute a very small amount to the outgoing radiation while partly absorbing stratospheric aerosols essentially act as attenuators for the incoming solar and outgoing diffuse radiations.

For $\theta_0=79.6^\circ$, there is some dependence of $\rho_{\mu}(\lambda, \theta_0, 0 - \text{nnn} \text{L mm} - 0)$ on $\lambda$ at shorter wavelengths. For $\theta_0=90^\circ$, this ratio decreases very rapidly with an increase of $\lambda$ from 0.3175 to 0.3398 $\mu$m. This is because a significant fraction of the scattered radiation in the 0.31-0.32 $\mu$m region originates from the parts of the atmosphere located above the aerosol layer, while that in the 0.32-0.34 $\mu$m region originates entirely from the parts of the atmosphere located within and under the aerosol layer.

In Fig. 10 we have plotted values of $\rho_{\mu}(\lambda, \theta_0, 0 - 0)$ as a function of the wavelength $\lambda$ for $\theta_0=0^\circ$, 79.6$^\circ$ and 90$^\circ$, and for the atmospheric models 0 - 200 L 00, 0 - 200 L 05, and 0 - 200 L 10. The aerosol attenuation optical thickness for these models at 0.3800 $\mu$m is about 0.97. (Again, we feel that these models represent conditions of rather strong tropospheric particulate contamination. Average hazy conditions would be better characterized by about $40 \times 10^6$ particles in a tropospheric column of 1 cm$^2$ cross section.) The interesting feature of the data presented in this figure is the strong dependence of $\rho_{\mu}$ on $\lambda$ at $\theta_0=0^\circ$ and $n_2=0.0$ which decreases gradually with increase of $\theta_0$ and very rapidly with increase of $n_2$.

![Fig. 10. As in Fig. 9 except for Haze L.](image)

![Fig. 11. Pseudo-dependence of the estimated total $O_3$ amount on the solar zenith angle when the atmospheric models contain stratospheric aerosols of the characteristics mentioned in identification of the curves. Solid (dashed) curves: $\Omega_B$ ($\Omega_A$), simulated measurements at 0.3312 and 0.3125 (0.3398 and 0.3175) $\mu$m wavelengths, i.e., pair A (B).](image)

6. Five-wavelength configuration

In this section, we will examine the effects of stratospheric and/or tropospheric aerosols on the values of total ozone in an atmospheric column estimated from simulated measurements at five wavelengths (viz., 0.3125, 0.3175, 0.3312, 0.3398 and 0.3800 $\mu$m). Use of the total $O_3$ estimation procedure outlined in Section 3 yields two estimates, viz., $\Omega_A$ from the measurements for the A pair (0.3312 and 0.3125 $\mu$m), and $\Omega_B$ from the measurements for the B pair (0.3398 and 0.3175 $\mu$m). The 0.3800 $\mu$m measurement is used for the estimation of the effective albedo of the earth-lower troposphere system. We will exclude $\theta_0=90^\circ$ cases from our discussion as their proper handling requires lengthy and special treatment in many instances. Furthermore, for brevity, our discussion is restricted to models resting on a surface with 0.2 Lambert reflectivity. Some dependence of $\Omega_A$ and $\Omega_B$ on $R$ was also noticed.

a. Input ozone amount of 0.250 atm-cm

For the 0-0 model, values of $\Omega_A$ and $\Omega_B$ were found to be equal to that of $\Omega_{in}$, i.e., 0.250 atm-cm for all nine values of $\theta_0$.

Variances of $\Omega_A$ (solid curves) and $\Omega_B$ (broken curves), as a function of the solar zenith angle $\theta_0$, are shown in Fig. 11 for the atmospheric models 5 H 05 - 0 (topmost section), 20 H 05 - 0, 20 H 00 - 0 and 20 H 10 - 0 (bottom-most section). In general, presence of the stratospheric aerosols results in an overestimation
of total O3 unless the sun is very close to horizon (θo > 80°). The degree of this overestimation generally increases with an increase in the stratospheric aerosol content from 5 to 20 x 10^6 particles, and also with an increase in the imaginary part (n2) of the refractive index of the aerosol material from 0.0 to 0.1. For the cases of strong stratospheric particulate contamination (20 H 10 - 0 model) and average sun angles (60-75°), total O3 content of the atmospheric column is overestimated by about 0.004 atm-cm by the A pair, and by about 0.008 atm-cm by the B pair. For θo > 80°, values of Ω_a are closer to Ω_in compared to the corresponding values of Ω_b. As for example, for the θ_o = 86.7° case of the 20 H 05 - 0 model Ω_a = 0.226 atm-cm, while Ω_b = 0.242 atm-cm.

In Fig. 12, we have plotted values of Ω_a (solid curves) and Ω_b (broken curves), as a function of the solar zenith angle θ_o for four different atmospheric models with tropospheric aerosols only, viz., 0 - 40 L 05 (top-most section), 0 - 200 L 05, 0 - 200 L 00, and 0 - 200 L 10 (bottom-most section). Presence of even a fairly large amount of nonabsorbing tropospheric aerosols (0 - 200 L 00 model) has practically no effect on Ω_a, while Ω_b - Ω_in is, at the most, equal to -0.004 atm-cm. On the other hand, presence of partly absorbing tropospheric aerosols results in a very significant overestimation of total O3 which increases with increase in the tropospheric aerosol content as well as with increase in the imaginary part of the refractive index of the aerosol material, but decreases with an increase of the parameter θ_o. Again, total O3 content values estimated from the B pair data are greater than the corresponding values estimated from the A pair data. As for example, for the θ_o = 0° case of the 0 - 200 L 10 model Ω_a and Ω_b are equal to 0.259 and 0.272 atm-cm, respectively. An interesting feature of the results presented in Fig. 12 is that the presence of even fairly large amounts of tropospheric aerosols in the model has very little effect on Ω_a and Ω_b when the solar zenith angle exceeds 80°. This is because, under such circumstances, a large fraction of the emergent radiation originates from the parts of the atmosphere located above the tropospheric dust layer.

Unlike the stratospheric case, there is sufficient experimental evidence for representing, under some circumstances, the size distribution characteristics of the tropospheric aerosols by functions other than Haze L. The other types of analytic functions encountered in the aerosol literature are the Haze J and Haze M functions mentioned in Section 4. After examining the data presented in Figs. 5, 6 and 7, it was considered appropriate to look into the effects of a change in the size distribution function from Haze L to Haze J, on the results presented in Fig. 12. In order to make results of 0 - nmn J mm models comparable with those of 0 - nmn L mm models, it is necessary to adjust the aerosol content so that the aerosol attenuation optical thicknesses of the models whose results are to be compared, would be equal around the midpoint of the spectral region of interest. As for example, results of the 0 - 200 L 00, 0 - 200 L 05, and 0 - 200 L 10 models become comparable to those of the 0 - 1184 J 00, 0 - 1184 J 05 and 0 - 1184 J 10 models, respectively. The highest sensitivity of Ω_a and Ω_b to changes in the tropospheric size distribution function is noticed at θ_o = 0°. For this geometry, Ω_a and Ω_b are both equal to 0.249 atm-cm for the 0 - 200 L 00 model, but 0.247 and 0.244 atm-cm, respectively, for the 0 - 1184 J 00 model. On the other hand, these two quantities are 0.258 and 0.269 atm-cm, respectively, for the 0 - 200 L 05 model, but 0.256 and 0.265 atm-cm, respectively, for the 0 - 1184 J 05 model.

We will now look into the changes in values of Ω_a and Ω_b when stratospheric as well as tropospheric aerosols are present in the model. Values of Ω_a and Ω_b are plotted as a function of θ_o in the lower and upper sections of Fig. 13, respectively, for the atmospheric models 20 H 05 - 0 (dotted curves), 0 - 200 L 05 (broken curves) and 20 H 05 - 200 L 05 (solid curves). It can be seen that the errors in Ω_a introduced by the stratospheric and tropospheric aerosols individually are additive in the first approximation. As for example, for θ_o = 0°, Ω_a - Ω_in equals 0.005, 0.019 and 0.025 atm-cm for the models 20 H 05 - 0, 0 - 200 L 05 and 20 H 05 - 200 L 05, respectively. From this figure, it can be seen that the conditions of strong haziness and overhead sun can lead to an overestimation of total O3 in an atmospheric column by about 4%, and by about 10% when observations at the A and B pairs, respectively, are used in the analysis. On the other hand, at θ_o = 86.7°, such strong hazy conditions lead to an underestimation of

![Figure 12](image-url)
Fig. 13. Pseudo-dependence of the estimated total ozone amount (\(\Omega_e^A\) or \(\Omega_e^B\)) on the solar zenith angle when the atmospheric models with \(\Omega_{in} = 0.250\) atm-cm contain stratospheric aerosols (dotted curves), tropospheric aerosols (dashed curves) and stratospheric as well as tropospheric aerosols (solid curves).

the total O\(_3\) content by about 10\% at the A pair, but by about 2\% at the B pair. Since the degree of overestimation is directly proportional to the total aerosol content in the first approximation (see upper two sections of Figs. 11 and 12), we may conclude that estimates of total ozone based on the method outlined in Section 3 will contain about \(\pm 2\%\) error because of the particulate pollution encountered under average conditions.

b. Input ozone amount of 0.450 atm-cm

For the \(\theta_0 = 86.7^\circ\) case of the 0-0 model with \(\Omega_{in} = 0.450\) atm-cm, the total ozone estimation procedure outlined in Section 3 returns a value of 0.341 atm-cm for \(\Omega_e^A\). This is because of the possibility of retrieving two values of \(\Omega_e^A\) due to the presence of a distinct maximum in the \(N_e\) vs \(\Omega_{in}\) curve for this case (see Fig. 2). [The procedure of Mateer et al. (1971) contains a provision for discarding ozone estimates under such circumstances.] We will therefore restrict our discussion of results in this subsection to values of \(\Omega_e^A\) and \(\Omega_e^B\) for \(\theta_0\) up to 84.7\%.

Values of \(\Omega_e^A\) and \(\Omega_e^B\) are plotted as a function of \(\theta_0\) in the lower and upper sections of Fig. 14, respectively, for the atmospheric models 20 H 05 - 0 (dotted curves), 0 - 200 L 05 (broken curves) and 20 H 05 - 200 L 05 (solid curves). Intercomparison of the results presented in Fig. 13 for \(\Omega_{in} = 0.250\) atm-cm and those presented in Fig. 14 for \(\Omega_{in} = 0.450\) atm-cm, suggests a decrease in the degree of overestimation of total O\(_3\) with an increase in \(\Omega_{in}\) at high sun. As for example, \(\Omega_e^B - \Omega_{in}\)

= 0.025 atm-cm for the \(\theta_0 = 0^\circ\) case of the 20 H 05 - 200 L 05 model in both cases. On the other hand, at low sun, values of \(\Omega_e^A\) and \(\Omega_e^B\) are much more affected by aerosols at \(\Omega_{in} = 0.450\) atm-cm compared to those at \(\Omega_{in} = 0.250\) atm-cm. As for example, at \(\theta_0 = 82.5^\circ\), \(\Omega_e^B - \Omega_{in}\) equals -0.002 atm-cm, and -0.047 atm-cm for the 20 H 05 - 200 L 05 model with \(\Omega_{in} = 0.250\) atm-cm, and 0.450 atm-cm, respectively.

7. Six-wavelength configuration

We will now look into the quality of \(\Omega_e^A\) and \(\Omega_e^B\) values obtained with a total ozone estimation procedure based on measurements at six different wavelengths, viz., 0.3125, 0.3175, 0.3312, 0.3398, 0.3600 and 0.3800 \(\mu\)m. It was mentioned in Section 1 that there are plans to take such six wavelength measurements with the TOMS instrument aboard the Nimbus G satellite. For this purpose, we have made the following changes in the total ozone estimation procedure outlined in Section 3.

In the first step, we now compute the effective albedo \(K'_{0.3600}\) and \(K'_{0.3800}\) using values of \(I_m(\lambda, \beta_0)\) at \(\lambda = 0.3600\) and 0.3800 \(\mu\)m, respectively. We then obtain values of \(K'_{e}\) at the remaining four wavelengths by linear extrapolation. [The 0.3600 \(\mu\)m wavelength is very weakly affected by changes in \(\Omega_{in}\) (see Table 1) and hence, it is permissible to compute values of \(K'_{0.3600}\) using basic tables for \(\Omega_{in} = 0.400\) atm-cm.]

The \(N_e\) vs \(\Omega_{in}\) curves in the second step are then defined not for \(R'\) independent of \(\lambda\), but for \(R'\) dependent on \(\lambda\). (The third step is the same as the one in Section 3.)

In the fourth step, we compute values of \(K'_{e}\) at \(\lambda = 0.3398\), 0.3600 and 0.3800 \(\mu\)m by making use of \(\Omega_e^A\) or \(\Omega_e^B\) depending on which wavelength pair has a
greater sensitivity in the $\Omega_{in}$ region of immediate interest. Values of $K^*_{ni}$ at the remaining three wavelengths are then obtained after quadratic extrapolation.

Thus, we have used the additional spectral measurement for taking into account the spectral dependence aspect of the effective albedo of the earth-lower troposphere system.

Values of $\Omega^A_{ni}$ and $\Omega^B_{ni}$ as obtained using the five-wavelength (broken curves) and six-wavelength total ozone estimation procedures are compared in Figs. 15 and 16, for the atmospheric models with $\Omega_{in}=0.250$ and 0.450 atm-cm, respectively. Each diagram consists of four sections with values of $\Omega^B_{ni}$($\Omega^A_{ni}$) being compared in the upper (lower) two sections. In all cases, results of different procedures are compared for the atmospheric models 20H 05 - 0 and 0 - 200 L 05. [Since the errors in $\Omega^A_{ni}$ and $\Omega^B_{ni}$ due to stratospheric and tropospheric aerosols are additive in the first approximation (see Figs. 13 and 14), we have not presented results for the atmospheric model 20H 05 - 200 L 05.] It can be seen that the use of the six-wavelength total $\Omega_5$ estimation procedure results in a very significant improvement in the quality of $\Omega^5_{ni}$ values for the models with tropospheric aerosols. This is especially true if the sun is high and the total $O_5$ content of the atmospheric column under study is low. For example, for the $\theta_0=0^\circ$ case of the 0 - 200 L 05 model with $\Omega_{in}=0.250$ atm-cm, we find that the quantity $\Omega^B_{ni}$ as obtained with the five-wavelength procedure is equal to 0.269 atm-cm, but is equal to 0.252 atm-cm when the additional measurement at 0.3600 $\mu$m is used in the analysis.

Reasons for this significant improvement can be found in the following physical foundation for the total ozone estimation procedure: Most of the total ozone content of the atmospheric column resides in the stratosphere, while most of the emergent radiation originates from the ground and lower troposphere. Thus, we are seeing a diffuse reflector (ground plus lower troposphere) through the ozone layer. In our models, we have assumed that the reflectivity ($R$) of the ground is independent of wavelength. However, the reflectivity of our diffuse reflector (effective albedo of the earth-lower troposphere system) is not necessarily spectrally independent. This spectral dependence of our diffuse reflector is accounted for, to a very significant extent, by the six-wavelength total $O_3$ estimation procedure.

8. Conclusion

In the preceding sections, we have examined the problem of estimating the total ozone amount of an atmospheric column in the presence of stratospheric and tropospheric aerosols. This examination is carried out by generating simulated spectral measurements for the five-wavelength Nimbus 4 BUV configuration, as well as for the six-wavelength Nimbus G TOMS configuration, and by analyzing such measurements with appropriate total $O_3$ estimation procedures. These simulated spectral measurements were generated for a
total of 40 different atmospheric models arrived at by varying the total O₃ content, and various aerosol parameters. Furthermore, a total of 10 different values of the solar zenith angle and eight different values of the Lambert reflectivity of the underlying surface were used for each model. However, results for a few selected atmospheric models resting on a ground with 0.2 Lambert reflectivity, are presented in this paper.

It has been shown that the values of total ozone in an atmospheric column (estimated with the five-wavelength procedure) can be in error by about ±2% due to particulate contamination encountered in the terrestrial atmosphere under average conditions. This error can be expected to rise to about ±8% under strong particulate contamination conditions, and still higher under very severe episodes for which no modeling is done at the present time. It has been shown that the errors due to tropospheric particulate contamination can be reduced significantly by taking appropriate advantage of the additional spectral measurement in the case of the forthcoming Nimbus G TOMS instrument. This improvement in the quality of the total O₃ estimates is very significant for the cases of low to moderate ozone conditions associated with solar zenith angles less than 75°.

It is necessary to monitor the O₃ content of the terrestrial atmosphere within ±1% accuracy over long periods of time, as such information plays an increasingly important role in our environmental, and hence in our political as well as economic decisions. It has been shown in the preceding sections that the current five-wavelength analysis of the UV observations taken under strong and severe particulate contamination conditions can yield pseudo-generations and destructions of atmospheric O₃. It is therefore necessary to make improvements on several fronts. Reliability and confidence level of the total O₃ estimates may be increased, to some extent, by establishing aerosol levels for various geographic and seasonal situations, and after including such information in the total O₃ estimation procedure.

Still better procedure would be that of obtaining aerosol information at the place and time of observation. In this respect, our study has shown that the six-wavelength TOMS configuration would be a very significant improvement over the corresponding five-wavelength BUV configuration in many instances. Before considering further instrumental changes, it is necessary to consider the lower boundary pressure parameter not varied in our present study. Dave and Mateer (1967) have already pointed out the need of simultaneous measurement of this lower-boundary pressure parameter within ±50 mb. Thus, the next instrumental improvement should be in the direction of making simultaneous measurements in the 8–13 µm window region or in the 0.758–0.768 µm oxygen band (Saided et al., 1967) for estimating the pressure at the lower boundary of the atmospheric column. After this, the next improvement can consist of simultaneous measurements of the intensity and the degree of polarization of the backscattered radiation in the 0.28–0.30 µm region for obtaining information about the stratospheric aerosols. Thus, it seems that an accurate monitoring of the terrestrial O₃ content on a global basis from ultraviolet satellite spectrophotometry would require a set of at least 10–12 simultaneous measurements. Design of such a next-generation instrument should follow an appropriate theoretical study for realistic models of the terrestrial atmosphere.

In the end, we would like to add that further information about various aspects of this investigation can be found in several contract reports by the author (Dave, 1976–1977).

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