Prediction of the solar radiant flux and heating rates in a polluted atmosphere

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ABSTRACT

A direct method for the solution of the spherical harmonics approximation to the equation of radiative transfer in an inhomogeneous, absorbing, emitting, non-isotropically scattering plane-parallel atmosphere is presented. The P_1 and P_3-approximations of the spherical harmonics method are compared with other more detailed methods for determining the solar radiative flux and the flux divergence in an absorbing and scattering atmosphere with an arbitrary phase function. The solar heating rates due to absorption by the pollution aerosols were predicted for a model atmosphere as a function of aerosol composition, concentration distribution, and solar elevation angle. The solar heating rates for nitrogen dioxide, ozone, and sulfur dioxide are also determined. It is found that a small amount of absorbing particles can significantly affect the radiative transfer in the atmosphere.

I. Introduction

Accurate prediction of the solar radiant fluxes and heating rates in the atmosphere has been hindered by difficulties in accounting for the atmospheric aerosol. For this reason the optical properties of the aerosols have received considerable attention and still are a subject of current research efforts. A very important topic has been the absorption of solar radiation by the aerosols. The first evidences of absorption came from measurements of the solar radiant flux by British (see Robinson, 1970) and Soviet (see Kondratyev, 1972) investigators in the 1940's and 1950's. These inferences of aerosol absorption have been further substantiated by direct measurement of the bulk absorption properties of collected samples (Fischer, 1970; Hänel, 1972) as well as indirect determination of the absorption characteristics by optical methods (Eiden, 1966 and others). Recent measurement of the spectral solar fluxes in the atmosphere during the CAENEX program (Kondratyev, 1973) has revealed the spectral dependency of aerosol absorption. The absorption has been found to be strongest in urban atmospheres and heating rates as high as 10°C/day have been estimated (Roach, 1961). This amount of absorption could possibly modify the temperature distribution over an urban area. It is therefore of interest to determine the solar heating rates which can occur above an urban area. Accurate prediction of radiant energy transfer is however quite difficult, since the gases and aerosols in the earth's atmosphere in general and in a polluted atmosphere in particular are capable of absorbing, emitting, and nonisotropically scattering radiation. Exact solutions are extremely time consuming and impractical, and thus for most applications approximate methods are employed.

The purpose of this paper is to present a direct method for the solution of the spherical harmonics approximation to the equation of radiative transfer in an absorbing, emitting, and nonisotropically scattering plane-parallel atmosphere. The lower orders of this method are then compared with other schemes for predicting the solar radiant flux and flux divergence in a polluted atmosphere. The solar heating rates are determined for various pollution aerosol compositions, vertical distributions, concentrations, and solar elevation angles. The solar

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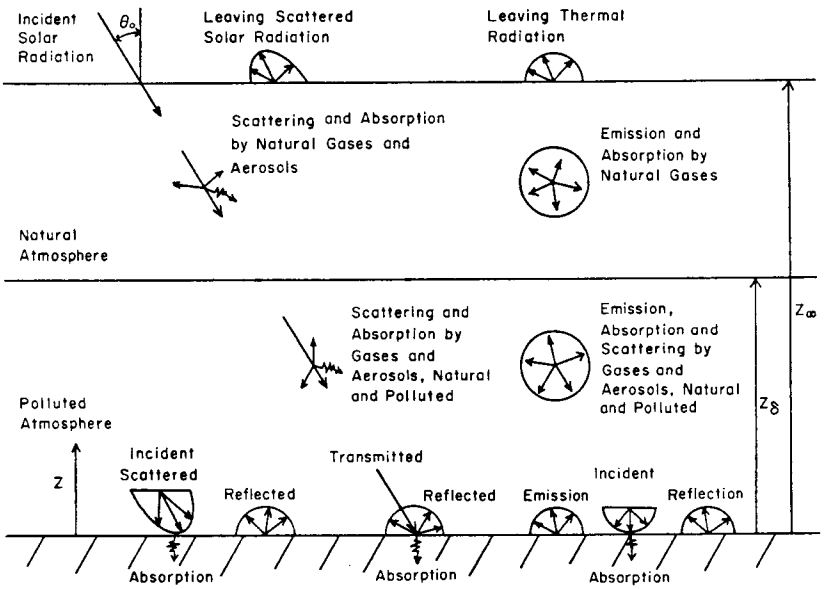

Fig. 1. Physical model for solar and atmospheric thermal radiation.

heating rates are also presented for three common air pollutant gases.

2. Analysis

2.1. *Atmospheric radiation model*

The idealized model of the atmosphere is shown schematically in Fig. 1. The atmosphere is assumed to be plane-parallel and consist of only two layers: (1) the natural atmosphere, and (2) the polluted atmosphere (the planetary boundary layer). The incident solar radiation is absorbed and scattered in the natural atmosphere by gases such as ozone and water vapor and scattered by the natural aerosol. In the polluted region the solar radiation is absorbed and scattered by natural and pollutant gases and aerosols. In this model the natural aerosol is considered nonabsorbing and the absorption accounted for by the pollution aerosol. Thermal radiation is emitted and absorbed by natural atmospheric gases in both layers and by pollutant gases and aerosols in the lower layer. Pollutant aerosols also scatter thermal radiation. At the surface solar radiation is absorbed and reflected diffusely, while the surface emits and reflects thermal radiation diffusely.

2.2. *Definitions*

In order to give a brief presentation of the governing equations it is necessary to first define the following quantities:

$I_\nu(z, \mu, \phi)$ = Spectral radiative intensity at frequency ν, height z, and direction μ, ϕ
θ, ϕ = Zenith and azimuthal angles with $\mu = \cos\theta$,
$I_{b\nu}$ = Planck's function,
$F_{\infty,\nu}$ = Solar flux at top of the atmosphere, z_∞, in direction μ_0, ϕ_0 and at frequency ν,
$\varkappa_\nu$ = Spectral absorption coefficient,
σ_ν = Spectral scattering coefficient,
β_ν = Spectral extinction coefficient, $\varkappa_\nu + \sigma_\nu$,
$\omega_\nu = \sigma_\nu/\beta_\nu$ = Albedo for single scattering,
$p_\nu(z, \mu', \phi' \to \mu, \phi)$ = Scattering probability function from direction μ', ϕ' to direction μ, ϕ,
$\varepsilon_\nu(\mu, \phi)$ = Spectral emittance function,
$r_\nu(\mu', \phi' \to \mu, \phi)$ = Reflection distribution function
τ_ν = optical depth, $\int_0^z \beta_\nu(z)dz$,
$\tau_{\infty,\nu}$ = optical thickness, $\int_0^{z\infty} \beta_\nu(z)dz$.

2.3. *Basic equations*

The fundamental concepts of radiant energy transfer are well documented (Kondratyev,

1969; Goody, 1964; Chandrasekhar, 1960) and need not be repeated here. The divergence of the radiative flux physically represents the net rate of loss (or gain) of radiant energy per unit of volume. The conservation of radiant energy equation is

$$\frac{d\mathcal{J}}{dz} = \int_0^\infty \{\kappa_\nu[4\pi I_{b\nu}(z) - G_\nu(z)]\}\, d\nu \qquad (1)$$

where the spectral incident radiant energy is defined as

$$G_\nu(z) \equiv \int_0^{2\pi} \int_{-1}^{+1} I_\nu(z, \mu, \phi)\, d\mu\, d\phi \qquad (2)$$

and the radiant flux as

$$\mathcal{J}_\nu(z) \equiv \int_0^{2\pi} \int_{-1}^{+1} I_\nu(z, u, \phi)\, \mu d\mu d\phi \qquad (3)$$

The first term on the right-hand-side of Eq. (1) represents emission and the second term accounts for absorption of radiant energy. The heating rate, H, is defined as

$$H \equiv -\frac{1}{\varrho c_p}\frac{d\mathcal{J}}{dz} \qquad (4)$$

The spectral intensity $I_\nu(z, \mu, \phi)$ is defined by the quasisteady equation of transfer (Chandrasekhar, 1960)

$$\mu\frac{\partial I_\nu}{\partial z} = -\beta_\nu(z) I_\nu + \kappa_\nu(z) I_{b\nu}(z) + \frac{\sigma_\nu(z)}{4\pi}\int_{\mu'}\int_{\phi'} p_\nu(z, \mu', \phi' \to \mu, \phi) \times I_\nu(z, \mu', \phi')\, d\mu' d\phi' \qquad (5)$$

In writing this equation it was assumed that the atmosphere was in local thermodynamic equilibrium, the index of refraction is equal to unity and the radiative transfer is quasi-steady, i.e., $(1/c)\partial/\partial t \ll \mu(\partial/\partial z)$. The first term on the right-hand-side of Eq. (5) represents the attenuation due to absorption and scattering, the second emisson, and the third multiple scattering from direction μ', ϕ' into direction μ, ϕ.

The formulation of the problem is completed by specifying the boundary conditions on the intensity at the top and bottom of the atmosphere. It is assumed that the sun is the only source of radiation at the top of the atmosphere, and at the surface of the earth the reflection and emission characteristics of the surface are taken as known. This is written as

$$I_\nu^-(z_\infty, \mu, \phi) \equiv F_{\infty,\nu}\,\delta(\mu - \mu_0)\delta(\phi - \phi_0), \quad \mu < 0 \qquad (6a)$$

and

$$I_\nu^+(0, \mu, \phi) = \int_{\mu'}\int_{\phi} r_\nu(\mu', \phi' \to \mu, \phi)\, I_\nu^-(0, \mu', \phi')\, \mu' d\mu' d\phi' + \varepsilon_\nu(\mu, \phi) I_{b\nu}(0), \quad \mu > 0 \qquad (6b)$$

For the solar spectrum the intensity can be divided into the direct transmitted portion, I_ν^t, and a scattered portion, I_ν^s (Chandrasekhar, 1960). The directly transmitted intensity can be immediately calculated as

$$I_\nu^t(\tau_\nu, \mu_0, \phi_0) = F_{\infty,\nu}(\tau_\nu, \mu_0, \phi_0) \times \exp[-(\tau_{\infty,\nu} - \tau_\nu)/\mu_0]\,\delta(\mu - \mu_0)\delta(\phi - \phi_0) \qquad (7)$$

An additional source term is added to the equation of transfer for the scattered intensity I_ν^s to account for radiation which is scattered from the directly transmitted solar beam. The equation of transfer for the scattered intensity when emission is neglected becomes

$$\mu\frac{dI_\nu^s}{d\tau_\nu} = -I_\nu^s + \frac{\omega_\nu}{4\pi}\int_{\mu'}\int_{\phi'} I_\nu^s(\tau_\nu, \mu', \phi')\, p_\nu(\mu', \phi' \to \mu, \phi)\, d\mu' d\phi' + \frac{\omega_\nu}{4\pi}\int_{\phi_0}\int_{\mu_0} I_\nu^t(\tau_\nu, \mu_0, \phi_0)\, p_\nu(\mu_0, \phi_0, \to \mu, \phi)\, d\mu_0 d\phi_0 \qquad (8)$$

The advantages of solving the equation of transfer in this manner is that the scattered intensity may be orders of magnitude smaller than the directly transmitted intensity and that the latter is assumed to be a collimated beam (plane wave) which does not have a continuous angular variation.

2.3. *Solution of the equation of transfer*

The solution of Eq. (8) is obtained using the method of spherical harmonics. For brevity,

the equation of transfer is solved for an absorbing, emitting, and scattering medium with the solar source term included. The superscript s denoting the scattered intensity has been dropped for simplicity. The derivation of the spherical harmonics equations staring with Eq. (5) is a very well-known procedure (Cheng, 1965; Kourganoff, 1963) and only the outline of the steps is included here for completeness. The details are given elsewhere (Bergstrom & Viskanta, 1972).

The basic assumption of the method of spherical harmonics is that the intensity and the scattering distribution can be expanded into a series of spherical harmonics which for one-dimensional problems is a series of Legendre polynomials and can be written as

$$p_\nu(\cos\theta) = \sum_{l=0}^{N} p_l P_l(\cos\theta) \tag{9}$$

where

$$\cos\theta = \mu'\mu + (1-\mu'^2)^{\frac{1}{2}}(1-\mu^2)^{\frac{1}{2}}\cos(\phi'-\phi).$$

If Eq. (9) for the scattering distribution function is substituted into the Eq. (5), the addition theorem of spherical harmonics (Cheng, 1965) is employed, and the resulting equation integrated over ϕ from 0 to 2π, there results

$$\mu\frac{d}{d\tau_\nu}\int_0^{2\pi} I_\nu(\tau_\nu,\mu,\phi)\,d\phi = -\int_0^{2\pi} I_\nu(\tau_\nu,\mu,\phi)\,d\phi + 2\pi(1-\omega_\nu)I_{b\nu}(\tau_\nu) + \frac{\omega_\nu}{2}\int_{\mu'}\int_{\phi'} I_\nu(\tau_\nu,\mu',\phi')\,d\phi'\sum_{l=0}^{N} p_l P_l(\mu')\,d\mu' P_l(\mu) + \frac{\omega_\nu}{2}\hat{I}^t_\nu(\tau_\nu,\mu_0,\phi_0)\sum_{l=0}^{N} p_l P_l(\mu) P_l(\mu_0) \tag{10}$$

Defining an azimuthally integrated intensity as

$$\hat{I}_\nu(\tau_\nu,\mu) \equiv \int_0^{2\pi} I_\nu(\tau_\nu,\mu,\phi)\,d\phi \tag{11}$$

and substituting Eq. (11) into Eq. (10) results in

$$\mu\frac{d}{d\tau_\nu}\hat{I}(\tau_\nu,\mu) = -\hat{I}_\nu(\tau_\nu,\mu) + 2\pi(1-\omega_\nu)I_{b\nu}(\tau_\nu) + \frac{\omega_\nu}{2}\int_{\mu}\hat{I}_\nu(\tau_\nu,\mu')\sum_{l=0}^{N} p_l P_l(\mu')\,d\mu' P_l(\mu) + \frac{\omega_\nu}{2}\hat{I}^t_\nu(\tau_\nu,\mu_0)\sum_{l=0}^{N} p_l P_l(\mu) P_l(\mu_0) \tag{12}$$

Assuming a solution for $\hat{I}_\nu(\tau_\nu,\mu)$ in the form

$$\hat{I}_\nu(\tau_\nu,\mu) = \sum_{k=0}^{N} a_k(\tau_\nu) P_k(\mu) \tag{13}$$

substituting in Eq. (13) and manipulating the resulting equation yields

$$\left(\frac{n}{2n-1}\right)\frac{da_{n-1}}{d\tau_\nu} + \left(1+\omega_\nu\frac{p_n}{2n+1}\right)a_n + \left(\frac{n+1}{2n+3}\right)\frac{da_{n+1}}{d\tau_\nu} = \delta_{0,n}2\pi(1-\omega_\nu)I_{b\nu}(\tau_\nu) + (\omega_\nu/2)\hat{I}^t_\nu(\tau_\nu,\mu_0)p_n \tag{14}$$

for $n = 0, 1, 2, \ldots N$. Equation (14) is a system of ordinary, first order differential equations which are entirely analogous to the original integro-differential equation for $N\to\infty$.

Since there are N first order differential equations, N boundary conditions are required to define the problem. The boundary condition at the top of the atmosphere ($z = z_\infty$) is

$$I^-_\nu(\tau_{\infty,\nu},\mu,\phi) = 0 \quad \text{for } \mu < 0 \tag{15a}$$

and at the earth's surface ($z = 0$) is written as

$$I^+_\nu(0,\mu,\phi) = \int_{\mu'}\int_{\phi'} r_\nu(\mu',\phi'\to\mu,\phi)I^-(0,\mu',\phi')\times\mu'd\mu'd\phi + \varepsilon_\nu(\mu,\phi)I_{b\nu}(0) + \int_{\mu'}\int_{\phi'} r_\nu(\mu',\phi'\to\mu,\phi)I^t(0,\mu_0,\phi_0)\times\mu'd\mu'd\phi' \quad \text{for } \mu>0 \tag{15}$$

where the transmitted solar intensity is given by

$$I^t_\nu(0,\mu_0,\phi_0) = F_{\infty,\nu}\exp(-\tau_{\infty,\nu}/\mu_0)\,\delta(\mu-\mu_0)\,\delta(\phi-\phi_0) \tag{16}$$

These two boundary conditions can be rewritten respectively as (Cheng, 1965)

$$\int_0^1 \hat{I}_\nu^-(z_\infty, \mu) P_{2i-1}(\mu) d\mu = 0, \quad i = 1, 2, \ldots \tfrac{1}{2}(N+1) \tag{17a}$$

and

$$\int_0^{-1} \hat{I}_\nu^+(0, \mu) P_{2i-1}(\mu) d\mu = 2\pi \int_0^{-1} P_{2i-1}(\mu) \times \left[\int_0^1 r_\nu(\mu', \mu) \hat{I}^-(0, \mu) \mu' d\mu' + \int_0^1 r(\mu_0, \mu) \hat{I}^t(0, \mu_0) \mu' d\mu' + \varepsilon_\nu(\mu) I_{b\nu}(0) \right] d\mu,$$

$$i = \tfrac{1}{2}(N+3), \ldots N. \tag{17b}$$

If p_n and ω are independent of τ Eq. (14) has constant coefficients and the solution is immediate. For inhomogeneous atmospheres the height can be divided into M layers in each of which the coefficients can be considered constant. The layers are coupled by simply requiring that the intensity be continuous at the boundaries (Shettle & Weinman, 1970). Thus at $\tau = \tau_m$

$$\hat{I}^{m-1}(\tau_m, \mu) = \hat{I}^m(\tau_m, \mu) \tag{18a}$$

and therefore

$$\int_0^1 \hat{I}^m(\tau_m, \mu) P_{2i-1}(\mu) d\mu = \int_0^1 \hat{I}^{m-1}(\tau_m, \mu) P_{2i-1}(\mu) d\mu, \tag{18b}$$

$$i = 1, 2, \ldots \tfrac{1}{2}(N+1)$$

$$\int_0^{-1} \hat{I}^m(\tau_m, \mu) P_{2i-1}(\mu) d\mu = \int_0^{-1} \hat{I}^{m-1}(\tau_m, \mu) P_{2i-1}(\mu) d\mu, \tag{18c}$$

$$i = \tfrac{1}{2}(N+3), \ldots N$$

In the solar part of the spectrum the emission is negligible in comparison to the incident radiation field and the solution of Eq. (14) takes the form

$$a_{n,\nu}(\tau_\nu) = \sum_{i=1}^{N} R_{n,i} C_i \exp(\lambda_i \tau_\nu) + \alpha_n \exp(-\tau_\nu/\mu_0) \tag{19}$$

The unknowns are the N values of the C_i's. The coefficients $R_{n,i}$, α_n, and λ_i are found in a straight forward manner from Eq. (14). The procedure is outlined by Bergstrom & Viskanta (1972).

For the thermal part of the spectrum the first term on the right-hand side of Eq. (14) predominates over the second, and the solution becomes

$$a_{n,\nu}(\tau_\nu) = \sum_{i=1}^{N} R_{n,i} \int_0^{\tau_\nu} f_i I_{b\nu} \exp[+\lambda_i(\tau_\nu - t)] dt + \sum_{i=1}^{N} R_{n,i} C_i \exp(+\lambda_i \tau_\nu). \tag{20}$$

Again the unknowns are the N values of the constants C_i, and the coefficients f_i, $R_{n,i}$, and λ_i are found by substituting the solution into Eq. (14). The details also are described elsewhere (Bergstrom & Viskanta, 1972).

For the P_1 and P_3-approximations exact relationships can be found for the coefficients, $R_{n,i}$, α_n, and λ_i. For higher order approximations the equations are usually solved numerically as sets of differential equations (Weinberg & Wigner, 1958).

2.4. *Comparison of different methods of solution*

Since the properties of aerosols are not well known and vary considerably, it is usually very difficult to compare the predicted solar flux and its divergence to experimental data. Therefore, it was decided to examine the results of various methods of prediction against each other for typical conditions of interest in order to determine their relative agreement. The techniques chosen were the P_1 and P_3-approximations of spherical harmonics method, the 20th order of the discrete ordinates method (Mudgett & Richard, 1971; Chandrasekhar, 1960) and an iterative method (Herman & Browning, 1965).

For simplicity, the conditions selected for the comparison were a homogeneous layer containing only pollution aerosols at a typical concentration. The effects of the natural atmosphere were thus for the moment ignored. The predicted normalized flux and flux divergence for a value of the cosine of the solar zenith angle, μ_0, of 0.636 are presented in Table 1. More extensive comparison of results obtained using the three methods of solution are given elsewhere (Berg-

Table 1. *Comparison of different methods for predicting the flux and flux divergence for a uniform layer of composition haze (Bergstrom, 1972)*

$\mu_0 = 0.636$, $C_p = 200$ μg/m^3, $\lambda = 0.5$ μm, $r_s = 0.1$, and $z_\delta = 1$ km

	Spherical harmonics		20th order Gaussian discrete ordinate	Iterative (Herman & Browning, 1965)
τ/τ_δ	P_1	P_3		
(*a*) Normalized fluxes, $\mathcal{F}_\nu/F_{\infty,\nu}$				
1.0	0.553	0.561	0.557	0.572
0.9	0.548	0.556	0.550	0.564
0.8	0.543	0.551	0.547	0.555
0.7	0.538	0.545	0.541	0.548
0.6	0.533	0.540	0.536	0.542
0.5	0.528	0.535	0.531	0.536
0.4	0.523	0.530	0.525	0.532
0.3	0.518	0.525	0.520	0.526
0.2	0.513	0.520	0.515	0.523
0.1	0.509	0.514	0.510	0.517
0.0	0.504	0.509	0.505	0.515
(*b*) *Normalized flux divergences* $[d(\mathcal{F}_\nu/F_{\infty,\nu})/d(z/z_\delta)]$				
1.0	0.0517	0.0515	0.0529	0.0529
0.9	0.0513	0.0517	0.0532	0.0534
0.8	0.0508	0.0518	0.5333	0.0537
0.7	0.0504	0.0518	0.0532	0.0538
0.6	0.0499	0.0518	0.0529	0.0539
0.5	0.0494	0.0517	0.0525	0.0538
0.4	0.0589	0.0515	0.0520	0.0536
0.3	0.0484	0.0513	0.0513	0.0534
0.2	0.0479	0.0510	0.0505	0.0531
0.1	0.0474	0.0507	0.0495	0.0526
0.0	0.0469	0.0503	0.0484	0.0520

strom & Viskanta, 1972). The computation time ranged from 1 to 2 seconds on a CDC 6400 computer for the P_1 and P_3-approximations of the spherical harmonics method, 20 seconds for the 20th order Gaussian quadrature of the discrete ordinates method, and 200 sec using the iterative procedure. The agreement between the predicted fluxes is surprisingly good while the discrepancy is slightly greater for the flux divergence. In this example the aerosol concentration are typical of "ordinary" levels of pollution and the total thickness for the transmitted beam is $0.281/\mu_0$. The solar transmitted flux is only reduced by 35% for $\mu_0 = 0.636$. Thus, most of the energy is still in the directly transmitted solar beam.

While it is difficult to assess the absolute accuracy of each method, the small relative difference gives some degree of confidence in the reliability of the techniques. More extensive comparisons (Bergstrom & Viskanta, 1972) show that the P_1 and P_3-approximations of the spherical harmonics method agree well with the 20th order discrete ordinates Gaussian quadrature scheme which several investigators (Merriam, 1968; Love, 1963; Sarofim et al., 1971) have claimed to give accurate results. In the subsequent computations the P_3-approximation is used. The order of the approximation could be readily increased if this was warranted in future studies.

2.5. *Parameters for solar radiant flux and heating rate computations*

The input parameters include the pollutant aerosol composition, concentration and vertical distribution, solar angle, natural aerosol conditions, humidity, and surface reflectance (albedo) The composition of the aerosol was assumed to be of variable parts of an absorber (carbon) and a nonabsorber (quartz). The properties of the urban air pollution aerosol (size distribution and indices of refraction) were taken from the model proposed by Bergstrom (1972). The recently reported experimental work (Hänel, 1972) shows that the absorption by the aerosol is a function of location and the relative humidity. However, the model (Bergstrom, 1972) used in the present computations corresponds to a dry aerosol with a mean imaginary part of the index of refraction somewhere between 0.01 to 0.1, depending on the percent of absorbing particles. This absorption increases with wavelength and can approximate, for example, the aerosol found over the industrial sector of Mainz, West Germany (Hänel, 1972).

For urban areas the surface reflectance is believed to be from 0 to 0.2 (Ludwig et al., 1969). For this range the reflectance has a minor influence on the heating rates and an obvious influence on the net solar flux at the surface. The amount of the natural aerosol present was also found to have a minor effect on the heating rates while, as expected, increases in the natural aerosol decreased the surface solar fluxes. The humidity was found, of course, to affect the heating rates, but since the dependence is fairly straightforward and has been discussed elsewhere (McDonald, 1960) it need not be repeated here. Typical solar heating rates due to water vapor are from 0.5 to 2 C/day. The influences of pollutant concentration, composition and vertical distribution, and solar angle on the

heating rates and solar fluxes are discussed below.

The vertical pollutant distributions investigated were the Gaussian, the uniform, and the exponential variations. The three distributions are written as the following:

Gaussian

$$C_p(z) = C_1 \exp\left[-(z_e - z)^2/2\sigma_z^2\right] \tag{21a}$$

Exponential

$$C_p(z) = C_1 \exp\left(-z/h\right) \tag{21b}$$

Uniform

$$C_p = C_1,\ 0 \leqslant z \leqslant z_\delta$$
$$= 0,\ z > z_\delta \tag{21c}$$

where C_p is the pollutant concentration, z_e the "effective stack height", σ_z is the standard deviation, h is the "scale height", and z_δ is the thickness of the polluted layer (planetary boundary layer). The Gaussian variation has been often used to describe plume diffusion (Hoffert, 1972). The uniform distribution is representative of conditions with limited vertical mixing (elevated inversions). The exponential decrease is typical of good mixing conditions where the scale height, h, was taken to be 1.25 km (Bullrich, 1964). The distributions are normalized with respect to the total amount of pollution. This quantity has the units of mass per unit area [μg/m^3 (km)] and is defined as

$$M_A \equiv \int_0^{z_\infty} C_p(z)\,dz \tag{22}$$

Physically, it simply represents the total amount of aerosol in a vertical column above a region divided by the cross-sectional area of the column, and has also been termed the "vertical mass loading" (Griggs, 1972). Assuming that the radiative properties of the pollutant are the same throughout the vertical extent of the layer, then the vertical mass loading is proportional to the optical thickness of the pollution since

$$\tau_{\infty,\nu} = \int_0^{z_\infty} \beta_\nu\,dz = \beta'_\nu \int_0^{z_\infty} C_p(z)\,dz = \beta'_\nu M_A \tag{23}$$

where β'_ν is the extinction coefficient per unit mass.

For the computations the data for the ozone absorption, Rayleigh scattering, and natural aerosol extinction in the free atmosphere were obtained from tabulations (Elterman, 1970). The spectral distribution of the solar energy was taken from the *Handbook of Geophysics*, 1960. The natural aerosol was assumed to have a real index of refraction of 1.5 (Yamamoto & Tanaka, 1969) for computations of scattering distribution.

The absorption of solar radiation by the six water vapor bands of importance was accounted for by using the data reported by McDonald (1960). From the tabulated absorption, $A(u)$, polynomial functions of the form

$$A(u) = \sum_{i=0}^{3} a_i \ln u^i \tag{24}$$

were derived from a least square fit. In this equation the water vapor mass path length, u, is defined as

$$u \equiv -\int_{z_\infty}^{z} C_w(z)\,dz \tag{25}$$

where $C_w(z)$ is the water vapor concentration. Then for every sclar angle a mean absorption coefficient $\bar{\varkappa}$ was defined for each band and was computed for the layer from the relation

$$\bar{\varkappa} \equiv \ln\left[A(u_2) - A(u_1)\right]/\left[(u_2 - u_1)/\mu_0\right] \tag{26}$$

where u_1 and u_2 are the mass path lengths of water vapor for the top and bottom of the layer, respectively. This technique was found to yield heating rates which were in good agreement with the results obtained using the formula proposed by McDonald (1960) for conditions without pollutants or natural aerosol present in the atmosphere.

3. Results and discussion

The heating rates for the three vertical distributions are shown in Fig. 2*a* for a particular composition, vertical mass loading, solar elevation angle, and height of the polluted layer. It is interesting to note that the heating rates for all three distributions are significantly higher

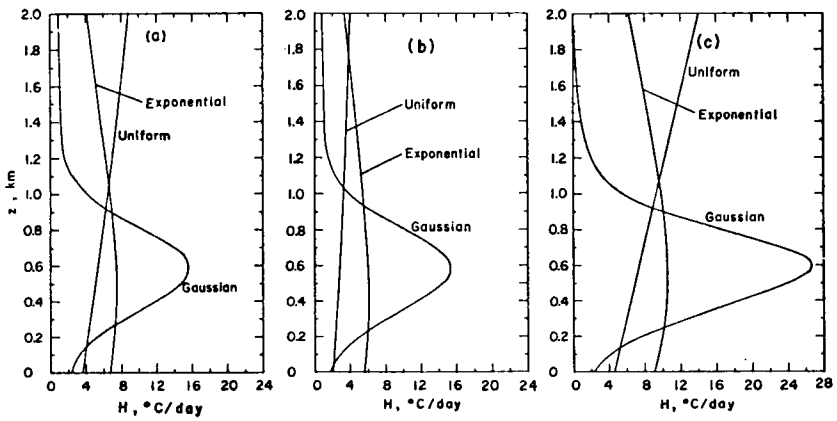

Fig. 2. Comparison of solar heating rates; (*a*) 20 % carbon, $M_A = 500$ (μg/m^3) (km), $z_\delta = 2$ km, $\mu_0 = 0.5$, (b) 20 % carbon, $M_A = 500$ (μg/m^3) (km), $z_\delta = 4$ km, $\mu_0 = 0.5$, (*c*) 30 % carbon, $M_A = 500$ (μg/m^3) (km), $z_\delta = 2$ km, $\mu_0 = 0.5$.

than those of water vapor (0.5–2.0°C/day). This illustrates the possible importance of the absorption of solar radiation by the aerosol. The heating rate is largest at the top for the homogeneous layer due to the decrease in radiant energy flux near the surface. This decrease in the radiant flux causes the point of maximum heating for the exponential distribution to be slightly above the surface (the point of maximum pollution concentration). For the same reason the point of maximum heating for the Gaussian distribution is somewhat above 0.5 km, the point of maximum concentration.

The increase in the height of the polluted layer (Fig. 2*b*) alters the heating rates for

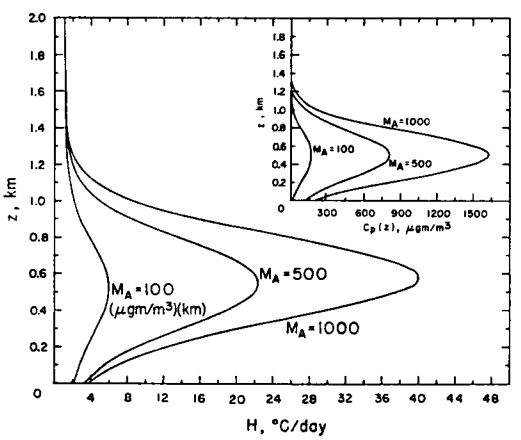

Fig. 3. Solar heating rates for the Gaussian distribution with M_A as a parameter, 20 % by weight carbon aerosol, $z_\delta = 2$ km and $\mu_0 = 0.5$.

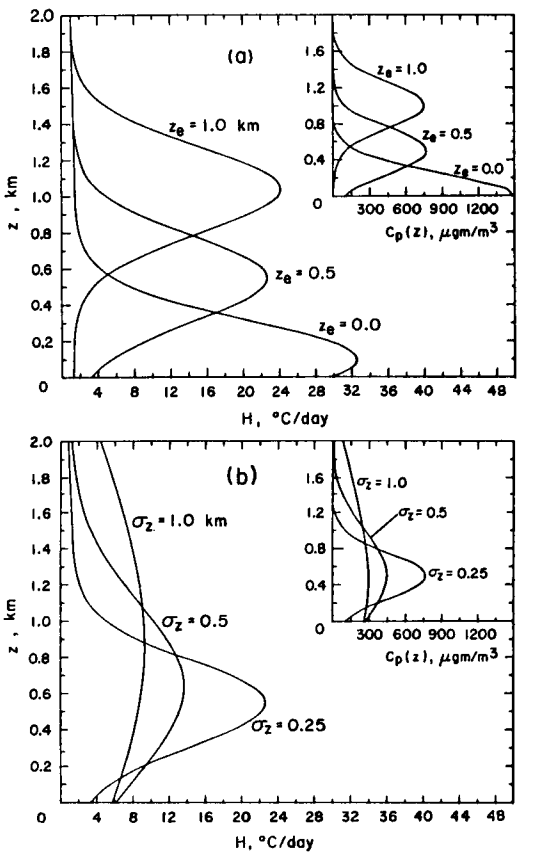

Fig. 4. Solar heating rates for the Gaussian distribution with σ_z and z_e as parameters; $M_A = 500$ (μg/m^3) (km), 20 % by weight carbon aerosol, $z_\delta = 2$ km and $\mu_0 = 0.5$.

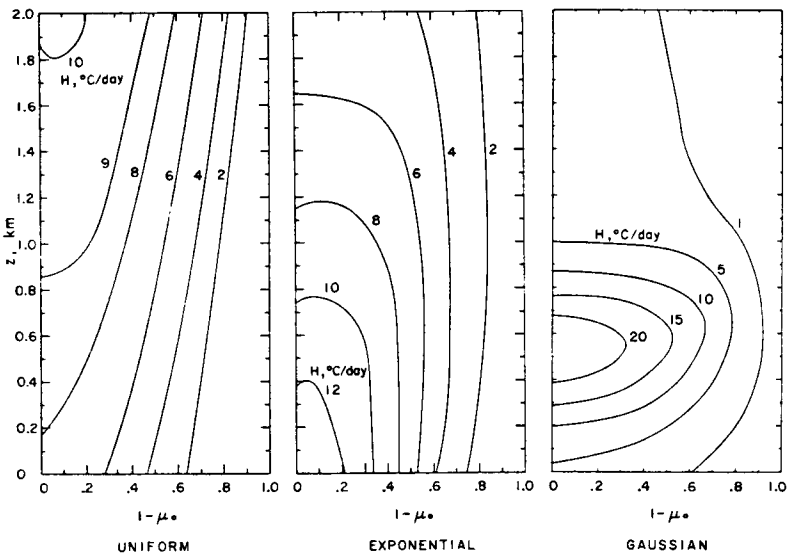

Fig. 5. Isopleths of heating rates; 20 % by weight carbon aerosol, $M_A = 500$ (μg/m^3) (km) and $z_\delta = 2$ km.

exponential and Gaussian distributions only slightly since for these variations there is little pollution above 2 km and hence increasing the height changes the concentrations only slightly. The heating rates for the 2 km thick uniform layer are almost twice those for the 4 km layer. Changing the aerosol composition from 20 % to 30 % "carbonlike" particles by mass increases the absorption coefficient and the heating rate by more than 50 %.

Fig. 3 illustrates the influence of increasing M_A (vertical mass loading) for a Gaussian distribution. The pollution concentration profiles are also given. As expected, the heating rate increases as the concentration is raised; however, the increase is not linear. This is due to the fact that for larger concentrations a greater fraction of the incident solar radiation is attenuated by the time it has reached a particular altitude. Due to this decrease in radiant energy the point of maximum heating increases slightly with altitude for the conditions $M_A = 500$ (μg/m^3) (km) and 1 000 (μg/m^3) (km). Since the concentration of carbon-like particles is 20 % by mass of the total aerosol, the ratio of the heating rate to the mass loading of absorbing carbon-like particles is about 15 C/day per 100 μg/m^3. The exact value of this ratio, of course, is also a function of the other parameters. This shows that a small amount of absorbing, carbon-like particles can cause very high heating rates.

The Gaussian plume distribution as defined by Eq. (21a) depends upon the effective stack height z_e and the standard deviation σ_z. The influence of these two parameters is shown in Figs. 4*a* and 4*b*. The pollution concentrations are also illustrated in the inset. For a surface source ($z_e = 0$) the heating rates in the lower few hundred meters are higher than for the two elevated sources since the maximum concentration for the surface source is much higher than that for the two elevated sources. The maximum heating rate for the highest effective stack height is larger than for the intermediate stack height although the maximum concentration is slightly less. The physical reason is that the radiant energy flux is larger at the point of maximum concentration for the highest effective stack height. Fig. 4*b* shows that for a small standard deviation the heating rates are more "peaked" than for larger deviations, similar to the concentration profiles.

The dependence of solar heating rate on the solar elevation angle for the three vertical distributions at constant M_A are illustrated in Fig. 5. Isopleths of the solar heating values are drawn for the exponential, uniform and Gaussian vertical distributions. The rates increase with the solar elevation angle, and the maximum heating occurs at different levels for the three distributions. The contours for the exponential and the Gaussian variations show

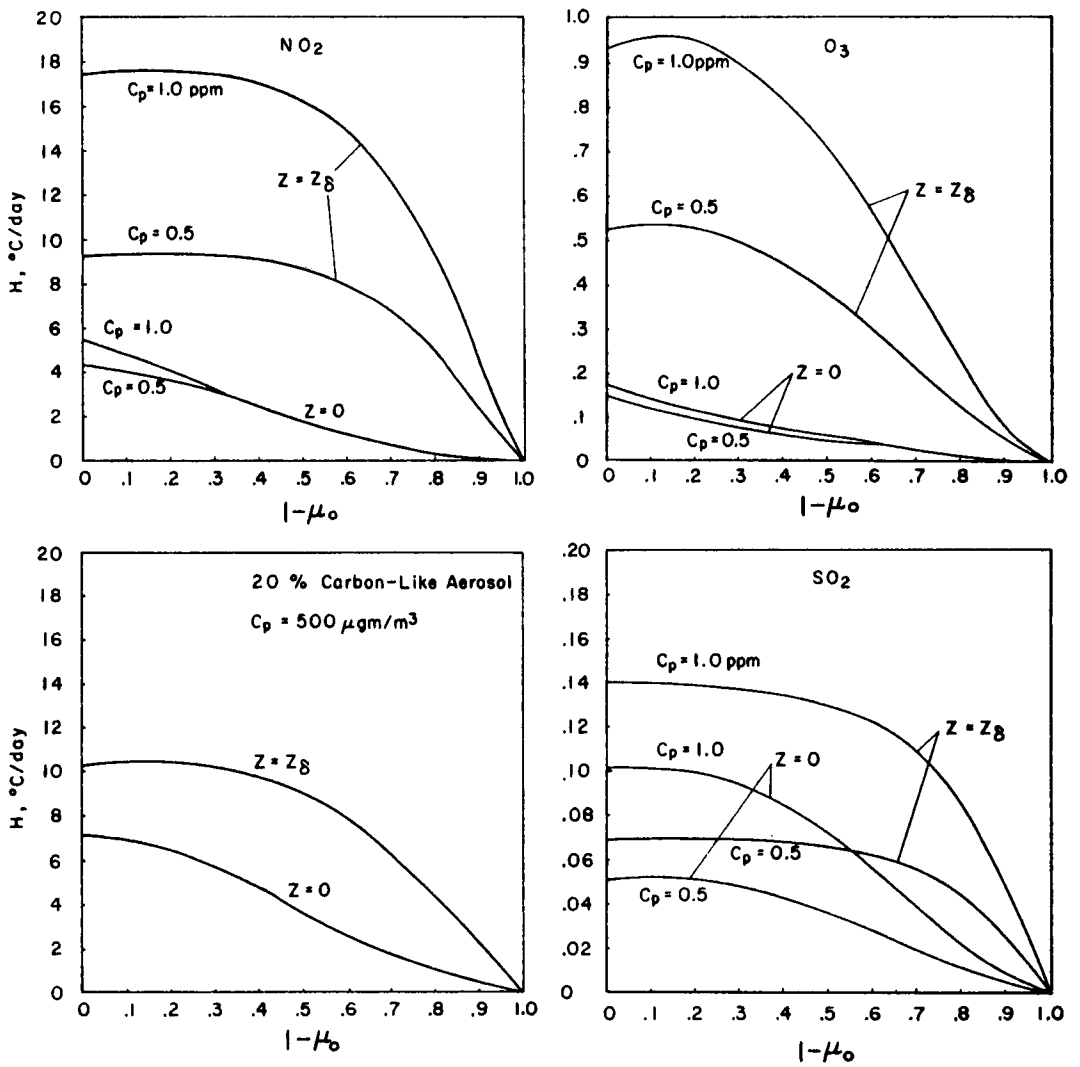

Fig. 6. Solar heating rates for uniform layer of pollutants with $z_\delta = 2$ km.

how the level of maximum heating increases away from the point of peak concentration with a decrease in the elevation angle as discussed earlier. The contours show that the maximum solar heating rate can occur at the top of a polluted layer for the uniform layer, near the effective stack height for the Gaussian, and near the surface for the exponential variations.

The heating rates due to three pollutant gases, nitrogen dioxide, sulfur dioxide, and ozone are shown in Fig. 6 for two concentrations as a function of the solar angle. The gases are assumed to be distributed uniformly through the two km thick layer. Water vapor, natural and polluted aerosol are also assumed to be present. The heating rate for the 20% carbon-like aerosol haze is shown for comparison. The values of the absorption coefficient for the three gases were taken from Leighton (1961). Absorption by nitrogen dioxide results in large heating rates and is comparable to the 20% by mass carbon-like aerosol. The solar heating rates for nitrogen dioxide and sulfur dioxide are lower than those computed by Atwater (1971). This

Table 2. *Variation of the solar flux at the surface,* $\theta_0 = 0$

(*a*) Effect of mass loading M_A for 20 % by weight carbon-like aerosol

$M_A(\mu g/m^3)$ (km)	0	100	500	1 000
$\mathcal{F}_{0,max}/F_\infty$	0.7360	0.7045	0.5865	0.4546

(*b*) Effect of aerosol composition for $M_A = 500$ ($\mu g/m^3$) (km)

Carbon-like particles, % by weight	0	20	30
$\mathcal{F}_{0,max}/F_\infty$	0.6833	0.5865	0.5279

may be due to the fact he did not account for the aerosol. His values for ozone heating are smaller than this study for an unknown reason.

The normalized solar fluxes at the earth's surface for $\mu_0 = 1$ are presented in Table 2*a* for four different mass loadings and in Table 2*b* for three different percentages by weight of absorbing particles. Table 2*b* is particularly interesting since it demonstrates that the increase in absorption overrides the decrease in scattering. This is due to the fact that some of the energy which is scattered eventually reaches the surface while the energy which is absorbed is removed from the solar radiation field.

The effects of mass loading and percentage of absorbing particles on the variation of the solar radiant flux at the surface with the cosine of the solar angle are illustrated in Figs. 7*a* and 7*b*. The fluxes have been normalized with respect to the flux for $\mu_0 = 1$ (see Table 2). The figure also illustrates the deviation of the fluxes from a linear dependence on μ_0 (the straight line drawn for comparison). The surface flux would vary linearly with μ_0 only for a transparent atmosphere. Thus, in order to accurately predict the solar flux at the surface the optical path and the amount of absorbing particles (or absorbing gases) must both be known. Fig. 7*b* also illustrates the influence of the albedo for single scattering on the surface flux. In the computations a mean albedo ω_ν was used for the polluted layer (Bergstrom & Viskanta, 1972). For the ranges of albedos studied this had only a slight effect on the heating rates and a few percent influence on the surface radiant flux. The assumption can be relaxed by including additional layers.

4. Conclusions

The lower orders of the spherical harmonics approximation to the equation of radiative transfer have been shown to compare well with other more detailed methods for the conditions

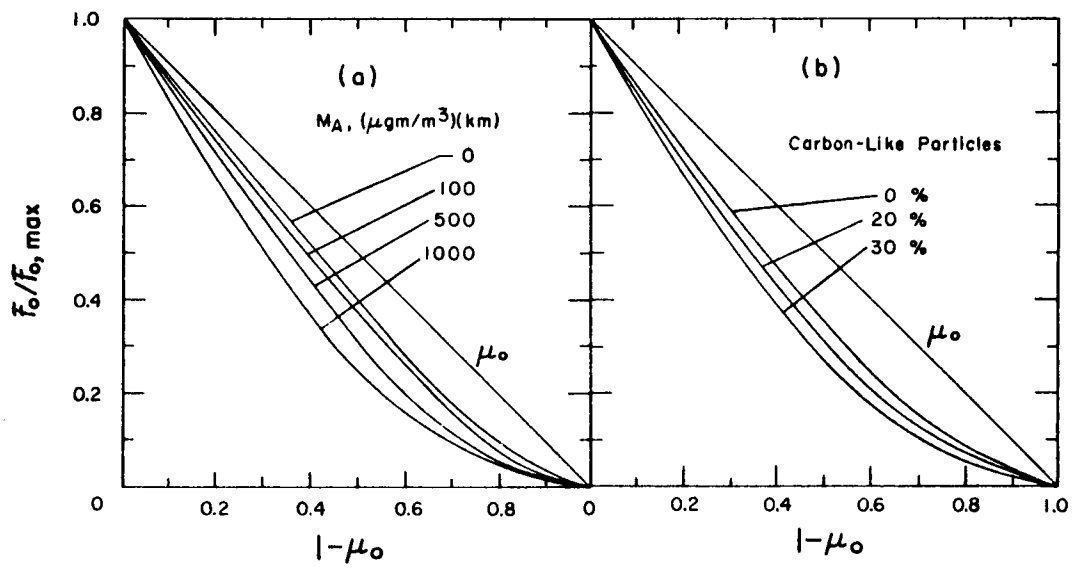

Fig. 7. Normalized surface solar flux for $z_\delta = 2$ km: (*a*) effect of M_A with 20 % by weight carbon aerosol, and (*b*) effect of aerosol composition with $M_A = 500$ ($\mu g/m^3$) (km).

studied and result in considerable saving of computational effort. This is important since the inclusion of accurate radiative transfer calculations in detailed circulation or climatic models is limited by computer time. It has been found that a small amount of absorbing particles in the pollution aerosol can cause large heating rates, particularly if the vertical concentration distribution is highly peaked as in a Gaussian plume model. It was also shown that in order to correctly determine the radiative flux at the earth surface it is necessary to prescribe both the total amount of particles and their absorption and scattering characteristics.

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ПРОГНОЗ ПОТОКА СОЛНЕЧНОЙ РАДИАЦИИ И СКОРОСТЕЙ НАГРЕВА В ЗАГРЯЗНЕННОЙ АТМОСФЕРЕ

Представлен прямой метод решения уравнения переноса радиации в приближении сферических гармоник для неоднородной, поглощающей, излучающей неизотропно рассеивающей атмосферы. Приближения P_1 и P_3 метода сферических гармоник срьвниваются с другими более детальными методами для определения потока солнечной радиации и его дивергенции в поглощающей и рассеивающей атмосфере с произвольной индикатрисой. Для модельной атмосферы были вычислены скорости нагрева благодаря поглощению аэрозолями загрязненнй как функции аэрозольного состава, распределения концентрации и угла возвышения солнца. Определены также скорости нагревания солнечной радиацией для двуокиси азота, озона и двуокиси серы. Найдено, что небольшое количество поглощающих частиц может существенно влиять на перенос радиации в атмосфере.