A Theoretical Investigation of the Effect of Aerosol Pollutants on Shortwave Flux Divergence in the Lower Troposphere

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ABSTRACT

This study is aimed at evaluating shortwave effects of aerosol pollutants in the lower troposphere (surface to 2 km). We have solved the radiative transfer equation for a plane-parallel atmosphere of infinite extent in the horizontal but inhomogeneous and of finite extent in the vertical. The vertical inhomogeneity is due to the presence of a non-uniform aerosol distribution, and a non-uniform concentration distribution of absorbing gases. The shortwave spectrum is divided into a sufficient number of intervals to simulate absorption characteristics of various atmospheric gases. The equation of radiative transfer is solved for these spectral intervals taking into account all orders of scattering.

Simulations have been made, using several representative vertical distributions of aerosols, typical aerosol values of refractive indices, and various size distribution functions. The analyses of these results indicate that the spectrally integrated flux divergence in the lower troposphere is highly dependent on solar zenith angle, the imaginary part of the refractive index, and the height and size distributions of the aerosol. The effect of gaseous absorption, however, has a smaller influence on the flux divergence.

1. Introduction

Aerosols found in the earth’s atmosphere are either natural or anthropogenic in origin. Recent observations (e.g., by Kondratyev et al., 1972) show that natural aerosols may be significant for absorption of shortwave radiation in the troposphere. Such measurements indicate that absorption of shortwave radiation by aerosols is at least comparable to that by water vapor.

The anthropogenic aerosol is estimated to be about 10% of the total global aerosol component (Williamson, 1973). Due to emissions from process industries and fossil fuel consumption, anthropogenic aerosol concentrations are known to increase dramatically in urban atmospheres. Several observational studies (e.g., Meetham, 1945; Roach, 1961; Monteith, 1966; Peterson and Flowers, 1974) indicate that the shortwave flux is significantly altered in urban atmospheres with high concentrations of aerosols.

In their theoretical investigations Atwater (1970) and, more recently, Bergstrom and Viskanta (1973) have included aerosols in tropospheric modeling of radiative transfer. These authors found significant alterations of the vertical temperature structure in the lower troposphere when aerosols were included in their atmospheric models.

The purpose of this paper is to describe some preliminary results of a theoretical study which is aimed at evaluating the effect of various aerosol characteristics, e.g., size distribution, refractive index, and the vertical distribution of aerosol concentrations, on the vertical profile of the shortwave flux divergence in the lower troposphere.

2. Atmospheric model

The radiation boundary layer model, taken to extend through the lower 2 km of the atmosphere, is for average mid-latitude summer conditions. The variations of pressure, temperature, water vapor and ozone with height are those given by McElroy et al. (1970). The atmosphere was assumed to be cloudless and homogeneous in the horizontal direction, and radiation reflected from the surface was neglected. The total unscaled water vapor and ozone amounts were 1.826 g cm\(^{-2}\) and 0.005 atm-cm, respectively. To simulate aerosol contribution from anthropogenic sources, the total number of particles in 1 cm\(^3\) column in the radiation boundary layer was increased from 1.646\times10\(^9\) [in Model C of Braslau and Dave (1973)] to 1.886\times10\(^4\). By the use of this number of particles, three typical aerosol concentration profiles were generated for the
boundary radiation layer. These three profiles, designated as distributions 1, 2, and 3, are shown in Fig. 1. In distribution 1, the aerosol concentration is independent of pressure, while in distribution 2 there is a strong decrease with decreasing pressure; in distribution 3 there is a strong increase with decreasing pressure.

Two different size-frequency distributions were assumed for the aerosols. The first of these was the Haze L distribution of Deirmendjian (1969), and the second was Junge's distribution (1955) with the slope parameter \(a = 3\). The mass loading of particulate material was taken to be the same for the two models, an assumption which necessitated increasing the total number of particles from \(1.886 \times 10^8\) for the Haze L model to \(1.368 \times 10^9\) for the Junge model. The normal optical thickness of the radiation boundary layer for aerosol scattering and absorption is shown as a function of wavelength for the two models in Fig. 2. This case is for aerosol distribution 1 and for an index of refraction of \(n = 1.50 - 0.01i\). For this purpose, scattering and absorption efficiency factors were calculated for the values of the size parameter \(x\) of the particle given by \(x = 0.02 (0.02) 40.0, 40.0 (0.1) 135.0\), and for the value of refractive index under investigation by using a procedure outlined by Dave (1968). From Fig. 2 it can be seen that for wavelengths of the solar spectrum which are significant in the scattering and absorbing process, the Haze L size distribution is a more efficient scatterer and absorber of the radiation than the Junge distribution.

Computations of aerosol absorption and scattering were made for the refractive indices shown in Table 1. These have been found to be representative of aerosols over highly populated areas (e.g., Twitty and Weinman, 1971).

### Table 1. Refractive index of aerosols, and a qualitative indication of the strength of their absorption, for the aerosol models considered.

<table>
<thead>
<tr>
<th>Refractive index</th>
<th>Aerosol absorption</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.50-0.01i</td>
<td>none</td>
</tr>
<tr>
<td>1.50-0.01i</td>
<td>moderate</td>
</tr>
<tr>
<td>1.50-0.10i</td>
<td>strong</td>
</tr>
<tr>
<td>1.66-0.06i</td>
<td>none</td>
</tr>
<tr>
<td>1.66-0.25i</td>
<td>very strong</td>
</tr>
<tr>
<td>1.80-0.50i</td>
<td>very strong</td>
</tr>
</tbody>
</table>

3. Computational procedure

In designing the computer programs for determining radiative effects of aerosols in the atmospheric boundary layer, the following factors were considered for reducing the computational requirements:

1) Since aerosol changes take place primarily within the boundary layer, changes at higher levels can be reasonably neglected for present purposes.

2) Because of the relatively small magnitude of the downward radiation at the top of the boundary layer resulting from backscattering of light directed upward through that layer, it is reasonable to consider the radiation incident on the top of the layer to be independent of the optical properties of the layer itself. This backscattered component is normally only a very few percent of the total incident flux and is thus of minor significance in the overall energy budget.

By use of the results of Braslau and Dave (1973), the direct solar flux and the azimuth-independent component of the diffuse sky radiation at the top of the boundary layer were computed. These quantities were evaluated at 2° intervals of angle in each of 80
wavelength intervals through the 300-2500 nm spectral region (Braslau and Dave, 1972) for two extreme solar zenith angles (0°, 80°). Transfer calculations for both direct and diffuse components of radiation incident at the top of the radiation boundary layer were carried out, taking into account all orders of scattering by aerosols and gaseous molecules of the atmospheric model. The computational procedure is based on a Fourier representation of the normalized scattering phase function and an iterative technique for solution of the equation of transfer (Braslau and Dave, 1973). Since the objective was to compute flux and the resultant divergence, the scalar form of the equation was utilized.

The accuracy of the flux divergence values computed using this method depends upon the quadrature taken for the integration over the azimuth angle and the number of layers into which the atmosphere is divided. Test calculations by Braslau and Dave (1973) with somewhat similar procedures showed net fluxes to be consistent to ±2%. Since the azimuth and optical thickness intervals used here did not exceed those of the test calculations, we estimate our flux divergence values to be of the same or higher relative accuracy.

The upward and downward quasi-monochromatic diffuse flux were evaluated at 20 equally spaced pressure levels within the 200 mb thick boundary layer, from which the resultant spectrally integrated upward and downward flux at these levels were computed. These data were then used to obtain the vertical profile of the flux divergence for each radiation boundary layer model.

4. Results of computations

a. Effect of aerosol refractive index

The vertical profiles of the flux divergence due to the combined absorption by gases and aerosols are shown in Fig. 3 for two different zenith angles of the sun (0°, 80°) and for all the values of the refractive indices listed in Table 1. The aerosol vertical distribution 1 and size distribution Haze L were selected for these computations. For the case of gaseous absorption only (m = 1.50–0.0i and m = 1.66–0.0i) the flux divergence is almost constant within the radiation boundary layer. The gaseous absorption is due primarily to water vapor.

Using a moderately absorbing aerosol (m = 1.50–0.01i) the magnitude of the vertical profile of the flux divergence (at 0° solar zenith angle) increases throughout the radiation boundary layer, the increase being a factor of 3.6 at the top and 3.1 at ground level. The larger absorption at the top is due to the presence of greater incident flux at these levels. Using a strong aerosol absorber (m = 1.50–0.10i) further enlarges the flux divergence to 11 times at the top and 7.6 times at the ground level, compared to the case for m = 1.50–0.00i. Of the aerosols listed in Table 1, the carbonaceous aerosol (m = 1.80–0.50i) suggested by Twitty and Weinman (1971) causes the maximum values of flux divergence at both 0° and 80° solar zenith angles.

The effect of multiple scattering on flux divergence calculations for non-absorbing aerosols (m = 1.50–0.0i

![Graph showing vertical profiles of shortwave flux divergence](image-url)
and $m=1.66-0.00i$) can also be evaluated from Fig. 3. Except for the near-ultraviolet, these non-absorbing aerosols have a scattering optical thickness which increases rapidly as the real part of their refractive index increases (Brassiau and Dave, 1973). This causes additional multiple scattering for the case $m=1.66-0.0i$, resulting in a longer traverse path and supplemental gaseous absorption. This appears to be the reason for the slight increase of flux divergence in the simulation using the aerosol with $m=1.66-0.0i$.

The total flux divergence in the radiation boundary layer as a function of solar zenith angles $0^\circ$ and $80^\circ$ is presented in Table 2 for three aerosol models of refractive index. The refractive indices all have constant values for their real parts but increasing values for their imaginary parts. Therefore, each aerosol model has a successively larger absorbing optical thickness, and its use results in a commensurate growth in absorption. The total flux divergence at $0^\circ$ resulting from embedding a moderately absorbing aerosol ($m=1.50-0.01i$) in the radiation boundary layer is 3.5 times greater than that for a non-absorbing aerosol ($m=1.50-0.00i$). At a solar zenith angle of $80^\circ$, this value becomes 7.3. For the strong absorbing aerosol ($m=1.50-0.10i$), the flux divergence grows to 9.5 and 22.7 times at $0^\circ$ and $80^\circ$, respectively. From these data it appears that increasing the imaginary part of the refractive index by an order of magnitude (i.e., 0.01i to 0.10i increases the total absorption in the radiation boundary layer by a factor of 3 for both $0^\circ$ and $80^\circ$ solar zenith angles).

b. Effects of aerosol size distribution

The normal scattering optical thicknesses for the Haze L and Junge size distributions are plotted as a function of wavelength in Fig. 2. The values of scattering optical thickness for the Haze L distribution increase between the wavelength interval 300–600 nm then decrease for the remaining wavelength values. The normal scattering optical thickness for the Junge distribution has decreasing values throughout the spectral region 300–2300 nm. Both distributions have equal values at a wavelength of 450 nm. At wavelengths less than this, Junge’s distribution has larger optical thickness values than the Haze L distribution. However, at wavelengths >450 nm the reverse is true. The Haze L distribution has a larger normal absorption optical thickness than the Junge distribution throughout the spectral interval 300–2300 nm.

The resulting flux divergence in the radiation boundary layer for the simulations using the Haze L and Junge size distribution functions are shown in Fig. 4. The computations were made with the aerosol vertical distribution 1 and an aerosol refractive index of $m=1.50-0.01i$. At a solar zenith of $0^\circ$, the use of the Junge distribution resulted in a 15% reduction in flux divergence averaged throughout the radiation boundary layer. The decrease in flux divergence is somewhat larger at the top of the radiation boundary layer than at the bottom. At $80^\circ$ solar zenith angle the use of the Junge distribution caused slightly less flux divergence at levels above 863 mb. Below this level, the values were slightly larger than those resulting from the Haze L distribution.

The magnitude of the reversal of the flux divergence values is too small to be considered physically significant. It falls within the inaccuracy criteria of the numerical quadrature used.

c. Effects of vertical distribution of aerosols

In Fig. 5 the vertical variation of flux divergence for the three model aerosol vertical distributions are presented. The aerosol size distribution function and the refractive index employed for these computations are Haze L and $m=1.80-0.50i$, respectively. As expected, the flux divergence is highly dependent on the vertical distribution of aerosol number density. We chose the

![Diagram](image-url)
aerosol distribution 1 as a reference calculation, and discuss the flux divergence for the two remaining distributions in terms of their ratio to distribution 1. For the simulations with aerosol distribution 3 (at 0° solar zenith angle) the ratio of flux divergence at the top and bottom of the boundary layer is 1.68 and 0.30, respectively. At 80° this ratio is reduced to 1.50 at the top and 0.25 at the bottom of the boundary layer. The flux divergence ratio for the case using aerosol distribution 2 follows that discussed for distribution 3, except the location of the maximum and minimum ratios occur near the ground and at the top of the boundary layer, respectively. In the former case, at a solar zenith of 0°, this ratio is 1.80 near the ground and 0.32 near the top of the boundary layer, respectively. At 80° the ratio is 1.63 and 0.33, respectively.

The sensitivity of the flux divergence to the vertical gradient of the particle number density when the sun is at the vertical (0°) can also be seen in Fig. 5. At the 903 mb level there is a sharp change in the vertical slope of the flux divergence curve for the case of aerosol distribution 2. This pressure level (Fig. 1) coincides with a change in the vertical slope of the particle number density. However, at 80° solar zenith angle the effect is somewhat less.

The dependence of the flux divergence on the local particle number density as a function of solar zenith angle is presented in Fig. 6. In this figure we have plotted the ratio of the particle number density of aerosol distribution 3 to that of distribution 1, and the corresponding ratio of the flux divergence for the respective radiation boundary layer models. The size distribution and refractive index are Haze L and
$m=1.80-0.50i$, respectively. For the sun at the zenith, the ratio of the flux divergence is almost directly proportional to the ratio of the particle number density at that level. At 80° the dependency on local particle number density decreases, particularly at the upper levels of the radiation boundary layer. This is due to the longer path length at this solar angle, resulting in additional multiple scattering and absorption.

5. Discussion

The results presented in the preceding sections indicate that absorbing aerosols in the amounts associated with emissions from anthropogenic polluting sources do play a significant role in the absorption of shortwave radiation in the boundary layer. In fact, particular combinations of realistic aerosol parameters can cause much more shortwave absorption by the aerosols than by atmospheric gases.

The aerosol parameter which most significantly affects the magnitude and vertical profile of flux divergence is the imaginary part of the refractive index. The divergence is also sensitive to the vertical distribution of aerosol concentration, a parameter which is primarily controlled by the prevailing vertical variation of atmospheric stability.

A logical extension of the work reported here would be an inclusion of effects of ground reflection and of clouds. Unfortunately, the added computational burden necessary would have greatly exceeded that available for this project. It is hoped that the additional computation can be undertaken in a future study.

A major problem still remaining is the paucity of reliable observational information on both the imaginary part of the refractive index and the vertical distribution of aerosols. Our calculations show that these data are necessary if we are to exploit the full potential of accurate computational modeling of aerosol effects on shortwave flux divergence.

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**REFERENCES**


