Shortwave Radiative Effects of Unactivated Aerosol Particles in Clouds

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ABSTRACT

Clouds in some polluted areas may contain high concentrations of anthropogenic aerosol particles. The possible role of these particles in perturbing the optical and dynamical properties of the clouds is an important question for climate studies. The direct radiative effects of unactivated aerosol particles in stable stratus clouds have been calculated at $\lambda=0.5$ μm. Several simplifying assumptions have been made relating the behavior of such particles in the high humidity environment within the cloud to their physicochemical make-up. It is shown that the energy absorbed by particles within the clouds may be, for realistic concentrations, comparable to the latent heat released and thus may play a significant role in cloud dynamics in some areas. These results are shown to be relatively insensitive to the assumptions about the particle properties within the cloud.

1. Introduction

There is increasing evidence, both theoretical and experimental, that man-made aerosols can significantly alter the radiative characteristics of the lower atmosphere and thus may play an important role in determining global and regional climate (see, e.g., Mitchell, 1971; Hodge, 1971; Rasool and Schneider, 1971; SMIC, 1971; Bolin and Charlson, 1976). The most important single atmospheric parameter for the determination of the radiative properties of the troposphere is cloud cover. It is of interest, therefore, to estimate the extent to which pollutant particles can perturb the radiative characteristics of clouds. Such perturbations can arise both through modification by aerosols of the cloud droplet spectrum (Easter and Hobbs, 1974) and through direct interaction of the particles with the radiation passing through the cloud. In this paper we present a simple model of the latter phenomenon.

The possibility that the shortwave radiative properties of clouds may be significantly modified by atmospheric pollutants was implied by Twomey (1972) who considered the effects of adding absorbing layers to nonabsorbing clouds. Rozenberg (1973) showed that a particle layer of absorption coefficient $b_{abs}\approx 0.03$ km^{-1} in an infinitely thick, nonabsorbing cloud can decrease the albedo about 20%. Braslau and Dave (1974) have shown that absorbing particles may play an important role in determining atmospheric heating rates. These previous studies have not considered the effect of the high humidity environment on the properties of the particles in the cloud.

Typical atmospheric aerosols are mixtures of particles with varying chemical composition and as such may exhibit a variety of responses to increased humidity. Some of the aerosol particles are hygrophobic, while others [H_2SO_4 and $(NH_4)_2SO_4$, for example] are hygroscopic (Covert *et al.*, 1972). Depending on the type of particles present we can expect the following kinds of radiative effects in water clouds due to the presence of nonactivated aerosol particles:

1) Hygrophobic particles such as flyash, organic tars and other carbonaceous compounds, which are strong light absorbers, will tend to lower the albedos of thick clouds and contribute to local heating.

2) Nonactivated hygroscopic or deliquescent particles, which may or may not be strongly light absorbing, may increase cloud albedos by increasing droplet concentrations and also may contribute to heating due to their light absorption properties.

3) Activation nuclei, which are usually nonabsorbing (Twomey, 1970), increase droplet concentrations and thereby increase cloud albedos.

In this paper we shall confine ourselves to a preliminary investigation of the direct radiative effects at $\lambda=0.5$ μm of unactivated particles in and below stratus clouds and shall not include effects of deformation of the cloud droplet spectrum by different types of particles.[2] In Section 2 we present the models used for the

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[2] The actual proportion of particles captured (or activated) depends on particle chemistry, size distribution and the thermo-

TABLE 1. Model parameters. The refractive index $n_r - in_i$, single scattering albedo $\tilde{\omega}_0$, asymmetry factor g, extinction coefficient b_{ext}, mean radius and standard deviation r_g and $\ln\sigma_g$, are given for two log-normal distributions (models 1 and 2), and for a Junge-type particle distribution, model 3, $n(r)\sim r^{-4}$, $r_{min} < r < r_{max}$.

Parameter	Cloud droplets	Aerosol particles Model 1	Model 2	Model 3
			1.55	1.55
$n_r - in_i$	$1.33-0.0i$	$1.55-0.0i$	$-0.01i$	-0.046
$\tilde{\omega}_0$	1.00	1.00	0.94	0.78
g	0.86	0.6	0.6	0.6
b_{ext} (km^{-1})	70.5	0.1	0.1	0.1
r_g (μm)	6.0	0.02	0.02	
$\ln\sigma_g$	0.26	0.80	0.80	
r_{min} (μm)				0.04
r_{max} (μm)				10.

cloud and particle parameters, in Section 3 the calculational method, and in Section 4 the results.

2. The models

We consider a two-layer system consisting of a cloud layer, containing both cloud and unactivated aerosol particles, and a subcloud layer, bounded by the cloud bottom and the ground. We assume that both layers are plane-parallel, horizontally homogeneous and of infinite horizontal extent. The cloud base is at 1.5 km and its thickness varies from 0.2 to 1 km. The aerosol particles are assumed to be well mixed within each layer. The only differences between the properties of the particles in the cloud and those below the cloud are due to the high relative humidity within the cloud.

The cloud-droplet size distribution used in the calculation is log-normal, i.e.,

$$\frac{dN}{d(\ln r)} = \frac{N_0}{(2\pi)^{\frac{1}{2}} \ln\sigma_g} \exp\{-[\ln^2(r/r_g)]/2\ln^2\sigma_g\}, \quad (1)$$

where $dN/d(\ln r)$ is the number of droplets per cubic centimeter of radius r in the interval $\ln r$, $\ln r + d(\ln r)$; r_g is the number mean radius and is taken to be 6.0 μm while σ_g is the geometrical standard deviation and is taken to be 1.3; and N_0, the total number of droplets per cubic centimeter, is computed by fixing the liquid water content of the cloud at 0.3 gm m^{-3}. This model of the cloud is similar to model 3 used by Twomey *et al.*, (1966).

In order to cover a realistic range of urban aerosol types we chose three different distributions for the subcloud particles. Model 1 consists entirely of nonabsorbing particles. Models 2 and 3 consist of moderately and heavily absorbing particles, respectively. The size distribution parameters for models 1 and 2 were found by Whitby *et al.* (1971) in Los Angeles smog. Model 3 uses a power-law size distribution of the form $dN/dr = Cr^{-4}$, $r \leqslant 10$ μm, and corresponds to that reported by Hänel (1972) for Mainz, Germany. The values of the various parameters are given in Table 1.

The effect on the particles caused by introducing them into the high humidity environment of a cloud will depend on the chemical composition of the particles. In order to cover a range of possible behaviors we consider the following cases.

a. Nonabsorbing particles

In general, particles which do not absorb light in the visible tend to be electrolytic solution droplets. We assume that the growth of these particles can be described by the Köhler curves for salt solution droplets (see, e.g., Mason, 1957, p. 25) so that the mean radius of the aerosol sample changes by a factor of about 5 in the transition from relative humidity 80% below the cloud to slightly above 100% in the cloud. The width of the distribution is assumed constant during this process. More accurate calculations, including the widening of the distribution at high relative humidities, as found by Hänel (1972), will not substantially affect the approximate results derived here, since scattering by the droplet distribution depends to a good approximation on the volume mean radius and the maximum variation in the absorption cross section with increasing relative humidity is less than a factor of 2 (Prishivalko and Astafyeva, 1974). The refractive index of the wet particles in the cloud is computed by taking real and imaginary parts of the approximate relation

$$(n^2-1)_{droplets} = \frac{(n^2-1)_{solute}V_{solute} + (n^2-1)_{H_2O}V_{H_2O}}{V_{solute}+V_{H_2O}}, \quad (2)$$

where V_{solute} and V_{H_2O} are the volumes of the solute and accreted water. This type of particle is used in model 1 with the parameter values given in Table 1.

b. Light absorbing particles

These tend to be at least partially insoluble in water but their growth under conditions of high humidity is as yet poorly understood. We therefore suggest three possible types:

(a) Totally hygrophobic particles, whose optical properties are independent of relative humidity.

dynamic parameters within the cloud. There is little theoretical or experimental information on which to base estimates of the effectiveness of these processes. We have assumed, in accordance with Twomey (1972) and estimates of in-cloud scavenging efficiencies (see, e.g., Slinn and Hales, 1971) that a large fraction of the absorbing aerosol particles entering a stable stratus cloud remain outside the cloud droplets. This would be the case, for example, if CCN and light absorbing particles have different origins. Comparison of our results to those of Grassl (1975) show that in the visible the direct radiative effects of aerosols in clouds are not sensitive to this assumption.

(b) Totally water soluble particles, whose optical properties are derived as functions of relative humidity in a manner analogous to the nonabsorbing particles using Eq. (2).

(c) Partially water soluble particles which consist of an inner nonsoluble core surrounded by a sheath of aqueous salt solution. Winkler (1974) has found that in Mainz about 60% by mass of the aerosol fraction in the radius range $0.1 \leqslant r \leqslant 1.0$ μm is water soluble, and the water soluble fraction consists largely of nonabsorbing electrolytes. Therefore, we have assumed the inner core is of mass $m=0.4\ m_0$, where m_0 is the initial mass of the particle (in the low-humidity environment below the cloud). The outer radius of the compound particle is calculated by assuming the sheath growth follows the Köhler curves and the equilibrium radius at saturation is that corresponding to a salt solution droplet of solute mass $0.6\ m_0$. The index of refraction of the inner core is calculated from that of the original particle by Eq. (2), assuming the real parts of the index of refraction of the soluble salts are the same as the real part of the insoluble core and the imaginary part of the index of refraction of the soluble salts is zero. The index of refraction of the sheath is taken to be that of water.

Of the three particle types, the third is probably the most commonly found, while the other two may be considered limiting cases. We incorporate these possible behaviors into our models by assuming that the particles in model 2 are either hygrophobic or water soluble (a and b above), while the particles in model 3 may exhibit any of the three kinds of behavior. The size distribution parameters and the indices of refraction of the various kinds of particles are given in Table 1. Included in the table are the calculated values of the single-scattering albedo ω_0 and the asymmetry factor g. The total number of particles in the subcloud layer is determined by setting the extinction coefficient b_{ext} to 0.1 km^{-1} in this region. The values of the optical properties thus obtained are in the range of values measured (Weiss *et al.*, 1976) in a variety of locations and thus may be taken to be representative of actual particle distributions. The value of b_{ext} for particles in the cloud is found by assuming the same number density of particles in the cloud as in the subcloud layer and then computing the extinction of the larger particles within the cloud, taking into account the change of refractive index given by Eq. (2).

Urban aerosol distributions are in fact composites of all the types of particles described above, although in many cases they are dominated by particles exhibiting one of the types of behavior we have discussed. By treating the idealized cases in which the growth behaviors are separated we can estimate the order of magnitude of the effects and of the variability due to chemical composition of the particles.

3. Calculational method

The general equations of radiative transfer in an azimuthally symmetric medium can be written as

$$\frac{dI_i}{d\tau} = M_{ij}(\tau) I_j(\tau) - F_i(\tau), \tag{3}$$

where $I_i(\tau)=I(\mu_i,\tau)$ is the intensity of the diffuse radiation at an angle $\cos^{-1}(\mu_i)$ from the vertical at an optical depth τ and $F_i(\tau)$ is a source term. For solar radiation $F_i(\tau)$ represents the diffuse radiation scattered from the direct beam into the direction μ_i. Repetition of an index implies summation over all possible values of the index.

In the vertically homogeneous case $M_{ij}(\tau)=M_{ij}$ independent of τ. The general solution to Eq. (3) in this case is

$$I_j(\tau) = W_{ji}[L_i' \exp(k_i\tau) - G_i(\tau)], \tag{4}$$

where

$$G_i(\tau) \equiv \int_0^\tau \exp[k_i(\tau-\tau')] W_k^{-1} F_k(\tau') d\tau' \tag{5}$$

and W_{ji} is the jth component of the ith eigenvector of M_{ij} corresponding to the ith eigenvalue k_i, i.e.,

$$M_{ij}W_{jk} = k_i W_{ik}. \tag{6}$$

The L_i' are constants which are evaluated from the boundary conditions for the problem.

The individual term M_{ij} of the matrix $\mathbf{M}$ represents the contribution to the intensity of diffuse radiation scattered from H_j to H_i by particles. In the vertically homogeneous case Eq. (3) reduces, of course, to the familiar equations (Chandrasekhar, 1970) which have been solved by a variety of approximations. We have written them in more general notation because the ordinary methods of perturbation theory can be applied to Eqs. (3)–(6) in this form to yield closed expressions for the radiative fluxes if $M_{ij}(\tau)=M_{ij}^{(0)}+M_{ij}^{(1)}(\tau)$, where $M_{ij}^{(1)}(\tau)/M_{ij}^{(0)}<1$, as is often true for atmospheric applications.

Since our present purpose is merely to estimate the radiative effects of representative aerosol samples, the parameters of which are not precisely known, it is appropriate to choose a quick and approximate solution. Consequently, we let $\mathbf{M}$ be a 2×2 matrix, which is the simplest possible case and calculate the values of the M_{ij} using the discrete ordinate method of Chandrasekhar (Liou, 1973, 1974). In order to check the accuracy of this two-stream method, we have compared calculated results for one homogeneous layer with those obtained by the doubling method (Van de Hulst, 1968; Twomey, 1972; Sagan and Pollack, 1967). In the conservative case ($\omega_0=1.000$) for $\tau \leqslant 1.0$ with no direct beam and for $\tau \geqslant 8.0$, the two-stream results are good to within 2%. The accuracy increases with op-

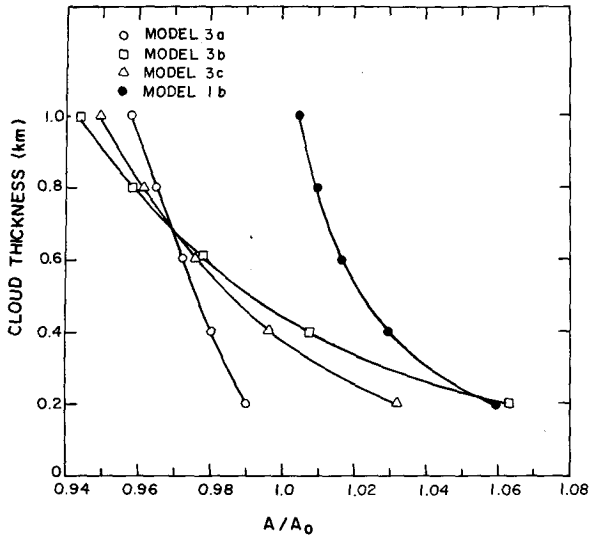

FIG. 1. Fractional albedo change due to unactivated particles as a function of cloud thickness. Fractional albedo change is defined as the ratio A/A_0, where A_0 is the system albedo with no unactivated particles in the cloud and A is the albedo of the same cloud when unactivated particles are added to it.

tical depth for large τ ($\tau>8.0$). For absorbing media ($\omega_0<1.000$) this same degree of accuracy requires $\tau\geqslant 32.0$ or $\tau<0.5$.

We let $I_1^{(c)}(\tau)$ and $I_2^{(c)}(\tau)$ be the downward and upward intensities in the cloud and define $I_1^{(s)}(\tau)$ and $I_2^{(s)}(\tau)$ analogously for the subcloud layer.

We can then write the boundary condition equations as

$$\left.\begin{aligned} &I_1^c(0)=0\\ &I_1^c(\tau_1)=I_1^s(\tau_1)\\ &I_2^s(\tau_2)=A_e\left[I_1^s(\tau_2)+\frac{\mu_0 F_0}{2\mu}\exp(-\tau_2/\mu_0)\right]\end{aligned}\right\}, \tag{7}$$

where τ_1 is the optical depth of the cloud layer, τ_2 that of the cloud plus subcloud two-layer system, μ_0 the cosine of the solar zenith angle and A_e the albedo of the earth's surface.

Eq. (3) and the analogous equations for the $I_i^s(\tau)$, with the boundary conditions (7), can be solved to find the upward and downward intensities at any optical depth τ. The albedo of the total system is

$$A=\frac{2\mu I_2^c(0)}{F_0\mu_0}. \tag{8}$$

The fractional amount of energy transmitted to the ground is

$$T=[I_1^s(\tau_2)-I_2^s(\tau_2)](2\mu/F_0\mu_0)+(1-A_e)\exp(-\tau_2/\mu_0),$$

while the fractional amount of energy absorbed by the two-layer atmosphere is

$$F_{\text{abs}}=1-A-T. \tag{9}$$

We can also define the albedo of the cloud alone as $A_{\text{cloud}}=A-[2\mu I_2^s(\tau_1)]/F_0\mu_0$ and the fraction of incident energy transmitted from the cloud to the subcloud layer as $T_{\text{cloud}}=I_1^s(\tau_1)(2\mu/F_0\mu_0)+\exp(-\tau_1/\mu_0)$. Then the fraction of incident energy absorbed by the cloud alone is

$$F_{\text{abs}}^{\text{cloud}}=1-A_{\text{cloud}}-T_{\text{cloud}}. \tag{10}$$

4. Results

We have calculated the following quantities for $\lambda=0.5$ μm as functions of cloud thickness, surface albedo and angle of incidence of the solar beam:

- System albedo A defined in Eq. (8).
- Energy absorption F_{abs} by the two-layer system as defined in Eq. (9) and energy absorbed by the cloud alone $F_{\text{abs}}^{\text{cloud}}$ as defined by Eq. (10).
- Directed flux intensities $F_\uparrow(z)\equiv 2\mu I_2^c(z)$, $F_\downarrow(z)\equiv 2\mu I_1^c(z)$ and the flux divergence $(d/dz)[F_\uparrow(z)-F_\downarrow(z)]$.

In this section we shall discuss each of these in turn.

a. System albedo

Fig. 1 shows the ratio of system albedo with particles in the clouds to that with no unactivated particles in the clouds. The effect of adding absorbing particles can be to increase the system albedo for thin clouds as a result of increased backscatter by the particles, or to

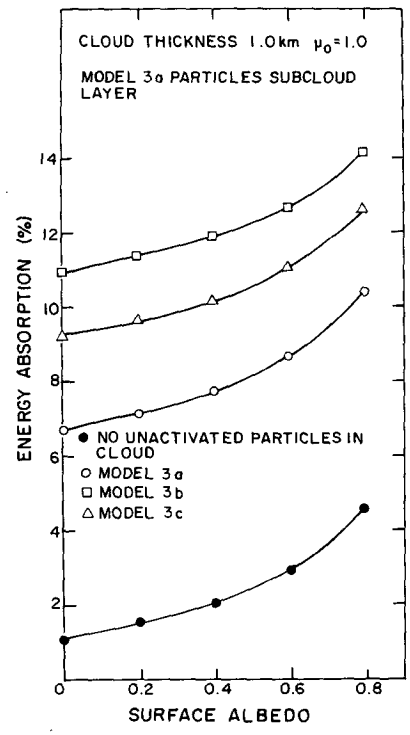

FIG. 2. Fractional absorption of incident energy at $\lambda=0.5$ μm by two-layer system as a function of underlying surface albedo. Cloud thickness is 0.6 km, solar zenith angle 0°.

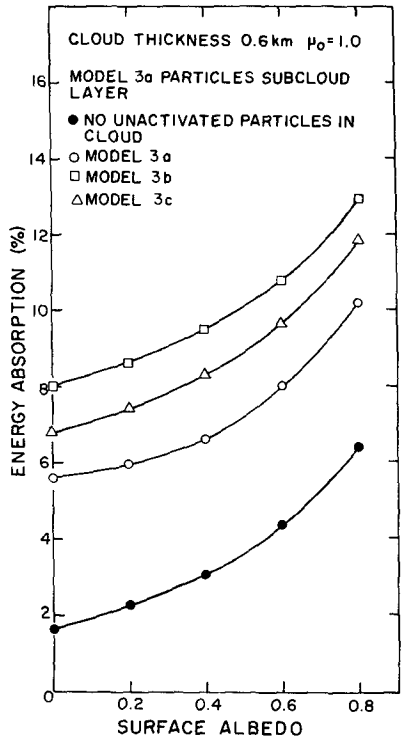

FIG. 3. As in Fig. 2 except for cloud thickness of 1.0 km.

decrease the albedo at larger optical thicknesses where absorption becomes important. For a given surface albedo, nonabsorbing unactivated particles increase the system albedo at all thicknesses. Note that because of the crudeness of the two-stream description at intermediate optical depths the results for cloud thicknesses <0.4 km may be in error by as much as 2%.[3]

b. *Energy absorption*

F_{abs} is shown for normal incidence in Figs. 2 and 3 for model 3 particles. The addition of water to a light-absorbing nucleus may change its light absorption efficiency. For the particles of model 3 the absorption coefficients are 0.027 km^{-1} at 80% RH, 0.052 km^{-1} for the solution droplets and 0.041 km^{-1} for the compound droplets at 100% RH. The particles in model 2, on the other hand, have absorption coefficient 0.006 km^{-1} at both 80% and 100% RH.

The large amount of absorption shown in Figs. 2 and 3 is a consequence of the fact that at vertical incidence absorption in a layer is enhanced by surrounding it by a multiple scattering medium (Twomey, 1972).

[3] *Note added in proof:* It is to be recalled that these approximations do not include possible modification of CCN spectra in polluted regions. Inclusion of this effect (as reported by Twomey) causes the calculated albedo change due to pollutants to be positive at greater cloud thicknesses.

The effects of surface albedo on F_{abs}^{cl} are slight. About 5–10% of the incident solar energy at $\lambda=0.5$ μm is absorbed by unactivated particles in thick clouds. If we assume the particle refractive index is independent of wavelength (Kondratyev, 1973) to obtain an estimate of total absorption, this corresponds to an average heating rate of about 0.2–0.5°C h^{-1}. In order to determine local heating rates due to particles it is necessary to examine the distribution of the energy absorption within the cloud.

c. *Directed flux intensities and flux divergence*

Fig. 4 shows upward and downward fluxes $F_\uparrow(z)$ and $F_\downarrow(z)$ and the "flux divergence" $[\Delta(F_\uparrow(z)-F_\downarrow(z))]/\Delta z$, where $\Delta z=0.05$ km, for surface albedo 0.0, $\lambda=0.5$ μm, model 3c. The assumption that both the subcloud and cloud layers are well-mixed causes the subcloud flux distributions to be more uniform than they should be but the main features of the graph are correct. The peak in both $F_\uparrow(z)$ and $F_\downarrow(z)$ at the top of the cloud is characteristic of thick clouds (Shettle and Weinman,

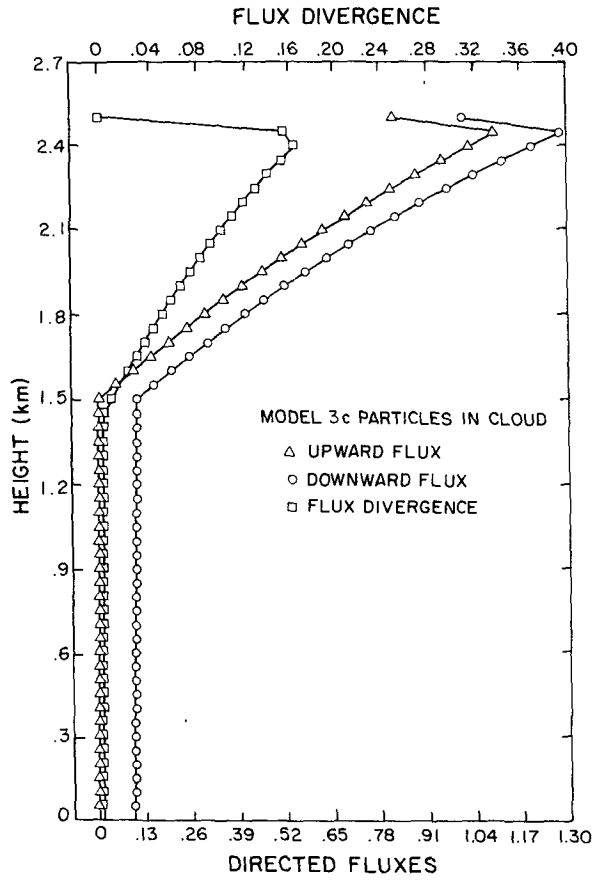

FIG. 4. Directed fluxes $F_\uparrow(z)$ and $F_\downarrow(z)$ and flux divergence $\Delta[F_\uparrow(z)-F_\downarrow(z)]/\Delta z$, $\Delta z=0.05$ km, for vertically incident unit flux. Model 3 particles in 1 km thick cloud.

1970). At $\lambda = 0.5$ μm there is no light absorption by "clean" cloud droplets and therefore the flux divergence within the cloud is entirely due to the unactivated particles. For particles in model 2 the maximum local heating rate is about 0.05°C-h^{-1} which is in qualitative agreement with the results of Braslau and Dave (1974).

5. Conclusions

The dominant processes in the energy balance of stratus clouds are the radiational cooling and the cooling of the rising air parcels modified by the latent heat release as cloud droplets form. The moist adiabatic lapse rate is on the order of 3°C km^{-1}. For the typical vertical velocities of a few centimeters per second associated with stratus clouds this suggests cooling rates due to lifting on the order of 0.04 to 0.4°C h^{-1}.

Estimates of radiational cooling in clouds from actual measurements are difficult to find. Paltridge (1974) presents flux profiles which give an average rate of cooling of 0.65°C h^{-1} through an 800 m thick cloud with a liquid water content of 0.1–0.2 g m^{-3}. The profiles suggest that the cooling rate is considerably larger at the top of the cloud, as might be expected, but the data presented are not sufficiently detailed to enable computation of local heating rates. In a paper on the formation of stratus clouds above mixed layers, Lilly (1968) computes a value of 0.055°C m s^{-1} for the average radiative heat loss from the top of the clouds. This figure is equivalent to an average cooling rate of 0.2°C-h for a 500 m thick cloud.

Comparing these figures with results of average excess heating on the order of 0.2–0.5°C h^{-1} suggests that absorbing particles may have a significant effect on the energetics of stratus clouds. The influence of this additional heating is difficult to state unequivocally. More complete calculations, incorporating spectral variation of the aerosol refractive index, a more realistic model of particle growth within the cloud, and possible effects of deformation of the cloud droplet spectrum by the addition of particles are needed in order to estimate the relative contributions of aerosols, water vapor and the liquid water in the radiation budget of clouds. It would be of interest to sample unactivated particles in clouds and to measure their physical and chemical properties, as well as their vertical distribution within clouds. Such measurements, although difficult to perform, may yield valuable information about possible anthropogenic influence on the radiative properties of stratus clouds.

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