On the effect of radiative exchange on the growth by condensation of a cloud or fog droplet

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(Received 27 March 1975; revised 10 November 1975)

SUMMARY

The effect of radiative heat transfer on droplet growth is assessed. It is estimated that radiative cooling is the principal agent of droplet growth in radiation fog, and that supersaturations are very small - slight undersaturation may even occur - and may be relatively unimportant for droplet growth in this environment. Gravitational settling is shown to be the principal limiting factor on droplet growth in radiation fog. It is also suggested that radiative transfer may have some relevance to the unexpectedly large numbers of 5μm radius droplets observed in cumulus clouds by Warner (1969).

1. INTRODUCTION

The kinetics of the condensation and evaporation of a droplet in stationary, continuous vapour and temperature fields was first discussed by Maxwell (1890), and his equations (with the boundary conditions sometimes modified to take account of the effect of droplet curvature (the Kelvin effect) and of dissolved salts on the vapour pressure at the droplet surface) have been widely used in the microphysical modelling of cloud growth (e.g. Mason 1971).

It is also known that the surface boundary conditions must be corrected for the rapid increases in temperature and vapour concentration gradients that occur within about one mean-free-path of the droplet surface. The effect of this correction is discussed by Fuchs (1959) and Fukuta and Walter (1970) who conclude that while the effect of the temperature 'jump' is probably negligible for droplets of radius > 1μm, the effect of the vapour jump has some influence on droplet growth up to about 10μm radius. The effect of this is discussed briefly in the appendix but is not included in the main theory in this paper, nor are the possible effects of surfactants on droplet growth considered.

The effect of net radiative exchange of heat between the droplet and its environment has usually been assumed to be negligible. Fuchs considers the radiative exchange arising solely from the temperature difference (of order 10⁻² degC) between the droplet and its environment (both assumed to radiate as black bodies) and not surprisingly found it to be negligible. However, there may be commonly occurring situations in the atmosphere (in the upper parts of stratiform cloud and fog) where radiation can become significant and would be expected to affect the growth of individual droplets. The purpose of this note is to consider this possibility.

2. THEORY

The addition of a radiative transfer term into Maxwell's heat transfer equation for a droplet yields

\[ L \rho_L \frac{dr}{dt} = (K/r)(T_r - T) - \mathcal{R} \]

(1)

where

- \( L \) = latent heat of condensation
- \( r \) = radius of droplet
- \( \rho_L \) = density of droplet
- \( K \) = thermal conductivity of air
The corresponding equation for the water budget of the drop is given as

$$\rho_d \frac{dr}{dt} = \left( D_w \frac{W}{r} \right) \left[ \rho - \rho_s(T_r) \right]$$

where

- $\rho$ = vapour density away from droplet
- $\rho_s(T_r)$ = saturation vapour density at surface determined by its temperature
- $D_w$ = diffusion coefficient of water vapour in air.

In a cloud interior, where $\mathcal{R} \approx 0$, Eqs. (1) and (2) show that a growing droplet must be warmer than its environment, and the water vapour and temperature gradients are therefore always opposed. It follows that the atmosphere must be supersaturated with respect to the droplet temperature and therefore to the air temperature (since $\rho_s(T_r) > \rho(T)$).

This is the situation prevailing where the droplet environment is being cooled by ascent or by radiation. In a radiation fog, however, there is evidence (Roach et al. 1976) that radiative cooling due to water vapour and carbon dioxide soon becomes small compared with that due to the droplets themselves. Thus the principal heat sink is transferred from the air to the droplets, and the droplets become colder than their environment, but (as seen from Eq. (1)) still grow. For a droplet in equilibrium with its environment while being radiatively cooled, i.e. $\frac{dr}{dt} = 0$, $\mathcal{R} = (K/r)(T_r - T)$. Also $\rho = \rho_s(T_r) < \rho_s(T)$.

In section 3, we show that a typical value of $\mathcal{R}$ on a radiation night is $-30 \text{W m}^{-2}$; then if $r \approx 10 \mu m$ we find $(T_r - T) \approx -10^{-2} \text{K}$. Hence the atmosphere is undersaturated (by about $7 \times 10^{-4}$ in this example), and droplet growth can occur in an atmosphere of zero supersaturation if it is being radiatively cooled.

If Eqs. (1) and (2) are combined with the Clausius-Clapeyron relationship and the gas law for water vapour, we obtain

$$\frac{dr}{dt} = (1/A)(s/r - D\mathcal{R})$$

where

$$A = \frac{L}{KT} \left( \frac{LM}{RT} - 1 \right) + \frac{RT}{D_w M e_s(T)}.$$ 

$$D = \frac{1}{KT} \left( \frac{LM}{RT} - 1 \right);$$

and

- $s = \text{supersaturation}$
- $M = \text{molecular weight of water (18g mole}^{-1})$
- $R = \text{gas constant (8.33 mole}^{-1}\text{deg}^{-1})$
- $\rho_d = \text{density of droplet fluid (10}^6\text{g m}^{-3})$
- $D_w = \text{molecular diffusion coefficient for water vapour (2.3} \times 10^{-5}\text{m}^2\text{s}^{-1})$
- $e_s(T) = \text{saturation vapour pressure at temperature } T$
- $K = \text{molecular conductivity of air (2.4} \times 10^{-2}\text{W m}^{-1}\text{deg}^{-1})$.

The effects of droplet curvature on vapour pressure result in the addition of a term proportional to $r^{-2}$ whilst that of the dissolved solute in a term proportional to $r^{-4}$ to the right-hand side of Eq. (3). In general, the effects of these terms become small as $\mathcal{R}$ becomes large (i.e. with increasing droplet radius) as discussed in section 2(b).

(a) The radiation term

The interception of radiation by a spherical homogeneous droplet of radius $r$, refractive
index \( m = n - in' \) will result in the absorption by the droplet of radiation at a rate, \( F_a \), given by

\[
F_a = \pi r^2 \int \int Q_a(r, m, \lambda) I(s, \lambda) \, d\lambda d\Omega
\]

(4a)

where \( Q_a(r, m, \lambda) \) is an efficiency factor for absorption; \( I(s, \lambda) \) is the intensity of radiation within a wavelength interval \( \lambda \) to \( \lambda + d\lambda \) incident from directions within a solid angle \( d\Omega \). Since \( m \) is only a function of wavelength, we can write \( Q_a = Q_a(r, \lambda) \).

The droplet will also emit radiation at its own temperature, \( T \), at a rate which is given from Kirchoff's Law as

\[
F_e = \pi r^2 \int \int Q_e(r, \lambda) B(T, \lambda) \, d\lambda d\Omega
\]

where \( B(T, \lambda) \) is the black-body emission given by the well-known Planck function, and is independent of \( s \), so that

\[
F_e = 4\pi r^2 Q_e(r)\int B(T, \lambda) \, d\lambda
\]

(4b)

It follows that

\[
F_a - F_e = 4\pi r^2 \mathcal{R}
\]

(5)

For a large absorbing sphere for which \( R \gg \frac{\lambda}{\lambda n} \) \( Q_a \rightarrow 1 \) and \( F_e = 4\pi r^2 \sigma T^4 \) which is the familiar Stefan–Boltzmann radiation law and \( \sigma \) is the Stefan–Boltzmann constant.

If the radiating droplet is in a plane, stratified atmosphere, we may write \( d\Omega = \sin \theta d\theta d\phi \), where \( \theta \) is the zenith angle and \( \phi \) the azimuth of the vector \( s \). Since \( I \) will be independent of \( \phi \), we can write Eq. 4(a) as

\[
F_a = 2\pi r^2 \int_0^\pi \int_0^\pi Q_a(r, \lambda) I(\theta, \lambda) \sin \theta \, d\theta d\lambda
\]

(6)

We may define a mean efficiency factor, \( Q_a(r) \), averaged over wavelength by

\[
Q_a(r) = \frac{\int_0^\pi \int_0^\pi Q_a(r, \lambda) I(\theta, \lambda) \sin \theta d\theta d\lambda}{\int_0^\pi \int_0^\pi I(\theta, \lambda) \sin \theta d\theta d\lambda}
\]

(7)

which applies if \( Q_a(r) \) is relatively insensitive to the typical range of distribution of \( I(\theta, \lambda) \) occurring in the atmosphere. \( Q_a(r) \) was evaluated for two regimes of \( I(\theta, \lambda) \):

(i) a black body enclosure (approximating to upward radiation at low levels)

(ii) a clear sky radiation field typical of that found at low levels.

The details of this computation are outlined in an appendix and the result shown in Fig. 1. Differences between the curves are small, and it is reasonable to represent them by a simple universal curve of the form

\[
Q_a(r) = Q_a(r)_{\text{max}} \left[ 1 - \exp(-\beta r) \right]
\]

(8)

In practice, \( Q_a(r)_{\text{max}} = 1.18; \beta = 0.28 \mu m^{-1} \).

Eqs. (4)–(7) can now be combined to give

\[
\mathcal{R} = \pi Q_a(r) \int_0^\pi \int_0^\pi \frac{1}{2} I(\theta, \lambda) \sin \theta d\theta - B(T, \lambda) \, d\lambda
\]

(9)

Eqs. (4)–(9) define radiative transfer across a spherical surface. In any model of the interaction of droplet growth with radiation in a plane, stratified atmosphere, upward and
downward radiative fluxes \((F_\uparrow\) and \(F_\downarrow\)) are resolved across a plane, horizontal surface. We may define:

\[
\begin{align*}
F_\uparrow &= \pi \int_{\pi/2}^{\pi} I \sin \theta \sin \theta \, d\theta \, d\lambda \\
F_\downarrow &= \pi \int_{0}^{\pi/2} I \sin \theta \cos \theta \, d\theta \, d\lambda \\
F_\uparrow &= 2\pi \int_{\pi/2}^{\pi} I \sin \theta \cos \theta \, d\theta \, d\lambda \\
F_\downarrow &= 2\pi \int_{0}^{\pi/2} I \sin \theta \cos \theta \, d\theta \, d\lambda \\
\sigma T^4 &= \pi \int B \, d\lambda 
\end{align*}
\]

Thus \(\mathcal{R} = Q_a(r)[\frac{1}{2}(F_\uparrow + F_\downarrow) - \sigma T^4] = Q_a(r)F\).

In the appendix, it is shown that \(F_\downarrow = (1.015 \pm 0.01)F_\downarrow; F_\uparrow = -F_\uparrow\). Therefore

\[
\mathcal{R} = Q_a(r)[\frac{1}{2}F_T - \sigma T^4], \hspace{1em} \text{very nearly}
\]
where \( F_T = F \downarrow - F \uparrow = \text{total} \)
\( F_N = F \downarrow + F \uparrow = \text{net} \)

Typically, at low levels under a clear sky \( F \downarrow \sim -0.8F \uparrow \) and \( F \uparrow \sim -0.6T^4 \); whence
\( \mathcal{R} \sim -0.1Q_s\sigma T^4, \sim -30Q_sW \text{ m}^{-2} \) for \( T \sim 275K \).

This is probably a typical radiation environment for a fog droplet growing near the top of a fog. Near the top of layer cloud in the lower troposphere, the ratio \( F \downarrow/F \uparrow \) may have decreased to about 0.7, thus tending to increase \( \mathcal{R} \) but this will be offset by a rather lower value of \( T \).

Near the top of convective cloud in the middle, or upper troposphere, \( F \downarrow/F \uparrow \) may have decreased to about 0.6 and \( T \) to about 255K and \( \mathcal{R} \) may be as high as \(-50Q_s(r)W \text{ m}^{-2}\), but the effect of this on growth rate will be offset by the decrease in \( D/A \) (Eq.(3)) with temperature. Conversely a droplet near the base of a small cumulus drifting over a (mainly) sunlit surface could experience a value of \( \mathcal{R} \) of 20-30\( Q_s(r)W \text{ m}^{-2}\).

\[(b) \text{ The general properties of the droplet growth equation} \]

Eq. (3) in its full form may be written
\[
dr/dt = (1/A)[s/r - B/r^2 + Cm/r^4 - DFQ_s(r)] \tag{12}
\]
where \( A, B, C, D \), are temperature dependent coefficients; \( F \) and \( Q_s(r) \) are defined by Eqs. (8) and (11); and \( m \) is mass of solute.

The general properties of Eq. (12) have been evaluated for ammonium sulphate, as a commonly occurring hydroscopic aerosol in inland areas. The growth parameters for ammonium sulphate have been given by Garland (1969), and the corresponding values of the coefficients are listed in Table 1.

<table>
<thead>
<tr>
<th>( T ) (K)</th>
<th>( A )</th>
<th>( B )</th>
<th>( C )</th>
<th>( D )</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>265</td>
<td>2.50</td>
<td>2.25</td>
<td>6.00</td>
<td>3.23</td>
<td>( 10^{-2} \mu\text{m}^{-2}\text{s}^{-1} ) for ( r ) in ( \mu\text{m} )</td>
</tr>
<tr>
<td>280</td>
<td>1.24</td>
<td>1.15</td>
<td>6.00</td>
<td>2.48</td>
<td>( 10^{-2} \mu\text{m}^{-2} ) in ( g )</td>
</tr>
</tbody>
</table>

The effect of the radiation term on equilibrium values of radius as a function of \( s \) is shown in Fig. 2. Radiative cooling has the general effect of depressing all the equilibrium curves to lower values of \( s \); for values of \( m \) greater than about 2\( \times 10^{-13} \)g, \( s \) remains negative for quite moderate radiative cooling. This reflects the suggestion above that a radiatively cooled droplet can continue to grow in an unsaturated environment, providing its \( s/r \) co-ordinates lie above the equilibrium curve corresponding to the appropriate values of \( F \) and \( m \).

The effect of a temperature increase from 265K to 280K (not shown) on the equilibrium curves is to increase the equilibrium values of \( r \) for a given \( s \) by about 20\% for the largest values of \( |F| \).

The growth equation has also been integrated to investigate the effects of variations in relevant parameters on the growth of a single droplet (Fig. 3). All droplets were started with a radius of 4\( \mu\text{m} \). This value was chosen for convenience of presentation; a different starting value shifts the curves laterally without changing their shape.

The first group of curves shows the effect of radiation on the growth of a droplet in a constant supersaturation of \( 5\times10^{-4} \), and shows that radiative cooling could grow a
droplet of given solute mass to about 25μm radius in the same time (30–40min) that it will take the droplet to reach 15μm in the absence of radiative cooling.

The implication of this result for the growth of droplets in the cumulus model due to Mason and Jonas (1974) has recently been investigated by Brown (unpublished), but he finds that radiative cooling, even if applied (unrealistically) throughout the cloud, produces a relatively minor effect. This is probably because supersaturations of order 0.3% probably occur in the updraught of growing cumulus, and would grow the droplets much faster than radiative cooling.

Curves 4–6 show growth curves in zero supersaturation for various values of $F$. The dominant effect of radiative cooling is shown by curve 4. Curve 7 shows that the effect of radiative cooling is offset by an under-saturation of $5 \times 10^{-4}$, while curves 5 to 7 suggest there is a range of conditions in which droplets of about 5μm radius do not change much in size. One is reminded of Warner’s (1969) observations of unexpectedly large numbers of droplets of this size in cumulus, and it may be that a varying balance between radiative cooling and slight undersaturation may occur in convective cloud debris away from the main cloud development regions and would favour the presence of droplets about this size.

Curves 8–10 demonstrate that droplet growth rate becomes largely dependent on radiative cooling for droplet radii greater than about 10μm – a decrease in solute mass of a factor of 30 simply delays the time taken to reach a given radius in a given supersaturation by a few minutes.

Finally, curves 11 and 12 show that while temperature variations had little effect on the equilibrium $s/r$ curves, (2), they had a large effect on droplet growth rates. For a droplet with $m = 10^{-12}$g, an increase in temperature of 15K (curve 11 → curve 1) appears to have a similar effect to the introduction of radiative cooling (curve 2 → curve 1).
The influence of radiative transfer on droplet growth is likely to be most significant in fog (particularly radiation fog), layer cloud, and ice cloud in the upper troposphere. It has been observed (e.g. Garland 1971; Roach, Brown, Caughey, Garland, Readings 1976) that most of the liquid-water content of fog is contributed to by droplets in the 5-10μm radius range which of course contribute mainly to the radiative cooling of the fog, but are also subject to the gravitational settling which Brown and Roach (1976) found to reduce the liquid water content and therefore the radiative cooling.

Two aspects of radiation fog will be briefly looked at here:

(a) An estimate of the characteristic level of supersaturation likely to be found in radiation fog.

(b) The effect of gravitational settling in limiting the growth of a given droplet.
Supersaturation in fog

Neglecting the effects of turbulence, the one-dimensional heat balance equation may be written

$$\frac{dT}{dt} = -\frac{1}{\rho_a} \frac{dF_n}{dz} - \frac{Ldx}{c_p dt}$$

(13)

where $\rho_a$ is air density and $x$ is vapour mixing ratio.

It can also be shown (using the Clausius-Clapeyron relationship) that

$$\frac{dx}{dt} = \frac{LM}{RT^2} \frac{dT}{dt} + \frac{x e}{d} = -\frac{dw}{dt}$$

(14)

where $x_e$ is saturation vapour mixing ratio; $s$ is supersaturation; and $w$ is liquid water content. (g/g dry air).

Let us suppose that the fog droplets are monodisperse with a concentration of $N m^{-3}$.

Then

$$\frac{dw}{dt} = 4\pi r^2 N (\rho_L/\rho_a) (dr/dt)$$

(15)

where $dr/dt$ is given by Eq. (3).

Assuming:

(i) The effects of droplet curvature and solute mass on droplet growth may be neglected ($r > \sim 4 \mu m$).

(ii) Supersaturation has reached equilibrium ($ds/dt = 0$).

(iii) Radiative flux divergence is entirely due to radiative loss from droplets.

Then

$$\frac{dF_n}{dz} = -4\pi r^2 N \mathcal{R}$$

(16)

Eqs. (13)-(17) can be combined to give

$$s/r = \mathcal{R}(D - AX)$$

where $1/X = \rho_L [L + (RT^2 c_p LM x_e)]$

(17)

Since $D$ and $AX$ are of similar magnitude, Eqs. (3) and (17) show that the supersaturation appears to be unimportant compared with radiative cooling for droplet growth in the radiation fog situation. Inserting characteristic values: $(D-AX) \sim 0.2 m W^{-1}$; $\mathcal{R} \sim -30 W m^{-2}$; and $r \sim 8 \mu m$; then $s \sim -5 \times 10^{-5}$, i.e. slight undersaturation. If an additional flux divergence, $G$, due to gaseous absorption, is introduced into Eq. (3) then Eq. (17) becomes

$$s/r = \mathcal{R}(D - A)X - GAX$$

(18)

A gaseous contribution to cooling rate as small as $0.2 K h^{-1}$ is sufficient to offset this slight undersaturation. Thus a cooling rate of $2 K h^{-1}$ due to water vapour and carbon dioxide, such as might occur in the early stages of fog formation near the ground, may induce supersaturation of a few units of $10^{-4}$. This initial increase in $\sigma$ is to be expected as the main cooling has not yet been transferred from minor gaseous constituents of the air to the growing droplets.

Gravitational settling

Combining Eqs. (3) and (17) gives $dr/dt = -\mathcal{R}X$, from which it would appear that droplet growth (initially about $15 \mu m h^{-1}$ for $\mathcal{R} \sim -30 W m^{-2}$) is only limited by temperature dependence. This equation is identical to the equation derived by Stewart (1955) in a brief section on the interaction of radiative cooling and fog development. Stewart defined the radiative heat loss by the droplet simply as $\pi r^2 F_n$ which differs from our definition (Eqs. (5) and (11)) in some important respects. Stewart's definition results in a under-
estimate of cooling rates of some $30\text{-}50\%$, but this is in favour of his contention that radiative cooling is responsible for the rapid decrease in visibility often observed.

As the droplet grows, so it begins to settle through the (developing) fog and becomes progressively radiatively shielded by the fog above. Thus we may write

$$\frac{dr}{dt} = V \frac{dr}{dz} \propto r^2 \frac{dr}{dz} \propto \frac{dm_d}{dz}$$

(19)

![Graph](image)

Figure 4. Some droplet growth curves as a function of depth while settling through fogs of different (but uniform) opacities. Small figures on curves denote time in minutes after leaving top of fog.

<table>
<thead>
<tr>
<th>Curve</th>
<th>$r_0(\mu m)$</th>
<th>$\alpha (m^{-1})$</th>
<th>$G (K \cdot h^{-1})$</th>
<th>$F (W \cdot m^{-2})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3</td>
<td>0.1</td>
<td>0</td>
<td>-30</td>
</tr>
<tr>
<td>2</td>
<td>6</td>
<td>0.1</td>
<td>0</td>
<td>-10</td>
</tr>
<tr>
<td>3</td>
<td>9</td>
<td>0.06</td>
<td>-3</td>
<td>-30</td>
</tr>
<tr>
<td>4</td>
<td>3</td>
<td>0.03</td>
<td>0</td>
<td>-10</td>
</tr>
<tr>
<td>5</td>
<td>3</td>
<td>0.06</td>
<td>-3</td>
<td>-30</td>
</tr>
</tbody>
</table>

$r_0 =$ initial droplet radius
where $V$ = fall velocity of droplet $\propto r^2$ by Stoke's Law and $m_d = \text{droplet mass}$. Hence the total increase in mass is given by

$$\Delta m_d \propto - \int R \, dz \quad . \quad . \quad . \quad (20)$$

$R$ is changing with time in a Lagrangian sense (following the droplet deeper into the fog) and an Eulerian sense due to the feedback of the developing fog on the radiation field as a whole although a steady state of fog is assumed in this particular calculation.

If we take the 'artificial' case of a stable, uniform fog with $F(z) = F(0) \exp(-az)$, then by combining with $\frac{dr}{dt} = -RX$ and Eqs. (8), (11) and (19) we obtain

$$\frac{dr}{dz} = - \frac{X \exp(-az)}{kr^2} F(0)Q_d(r)_{max}(1 - \exp(-br)) \quad . \quad (21)$$

Eq. (21) has been used to evaluate some growth curves (Fig. 4) as a function of depth in a stable fog of uniform opacity. The curve parameters are listed in Table 3.

All curves tend to a limiting value after about 15–20m fall, which takes about 20min for 9µm droplets (curve 3) to about 1h for 3µm droplets. The effect of reducing the initial radiative cooling by a factor of three results (curve 4) in the reduction of the increase in droplet radius by a factor of about two. An additional radiative cooling due to gaseous constituents of about 3K h$^{-1}$ increases (curve 5) the ultimate radius by only about 15%. A reduction in the opacity ($\alpha$) of the fog results in increased growth and considerably increased fall before the droplet approaches a limiting radius.

In this over-simplified model it is claimed to demonstrate no more than the interaction of radiative cooling and gravitational settling on droplet growth in fog, and to suggest that it is difficult to grow a fog droplet much beyond 10µm radius, and this is borne out by observations of drop size distribution in fog.

A more comprehensive model must take account of an initial size distribution of condensation nuclei, and of the subsequent feedback of fog development on the radiation field. It is intended that this paper and the 'macrophysical' fog model of Brown and Roach (1976) will form the basis for such a 'microphysical' model of fog development.

### REFERENCES

Fuchs, N. A. 1959 Evaporation and droplet growth in gaseous media, Pergamon Press.
Herman, B. M. 1952 A statistical model for water vapour absorption, Ibid., 78, pp. 165-169.
GROWTH OF DROPLETS


APPENDIX

The values of $Q_a(r, \lambda)$ and $I(\theta, \lambda)$ are fairly well defined by theory (e.g. Herman 1962, Goody 1964) but their precise evaluation is tedious.

Evaluation of $Q_a(r, \lambda)$. Herman gives curves of absorption efficiencies as a function of $2\pi r/\lambda$ for wavelengths of 4 to 24$\mu$m which covers most of the spectrum of thermal importance in the atmosphere. The curves for $Q_a$ all resemble an exponential curve of the form $Q_a = (Q_{a \text{max}}[1 – \exp(-2\pi r/\lambda)]$. Best fits to this function gave values of the empirical constants $(Q_{a \text{max}}$ and $\gamma$ for each wavelength interval (Table 1) from which a two-dimensional matrix $Q_{ik}$ was obtained

$$Q_{ik} = [(Q_{a \text{max}})[1 – \exp(-2\pi r/\lambda)]$$

Evaluation of $I(\theta, \lambda)$. In order to give a rough approximation to a typical distribution of clear-sky intensity near the ground, it was assumed that the atmosphere was isothermal – equivalent to assuming that all the downward radiation originates in the lowest layers, and with emissivity depending on total precipitable water vapour and zenith angle expressed and form suggested by Goody (1952). This led to the matrix $E_{ij}$:

$$E_{ij} = 1 – \exp \left[ -2.54 \frac{W}{\zeta_j w_i} \left( 1 + \frac{12.45 W}{\zeta_j w_i} \right)^{-\frac{1}{2}} \right]$$

where $W = \text{total precipitable water vapour}$

$$\zeta_j = 0.01(1 + 2j) = (\cos \theta)_j$$

$$w_i = \text{precipitable water vapour required to produce an emissivity of } 0.5 \text{ (from Cowling 1950).}$$

The 15$\mu$m CO$_2$ band and the water vapour band at wavelengths greater than 22.5$\mu$m were assumed to be opaque. The following quantities were then evaluated:

$$I_k' = 0.02 \sum_{i=1}^{14} \sum_{j=1}^{49} \pi Q_{ik} B_i E_{ij}$$

$$F_r' = 0.02 \sum_{i=1}^{14} \sum_{j=1}^{49} \pi B_i E_{ij}$$

where dashes indicate normalization to black body radiation at 275K

$$Q_{ik} = I_k'/F_r'$$

$$Q_{Bk} = \sum_{i=1}^{14} Q_{ik} B_i$$

These expressions are the finite difference forms of Eq. (7) with $\Delta(\cos \theta) = 0.02$. The quantity:

$$F_{14}' = 0.04 \sum_{i=1}^{14} \sum_{j=1}^{49} \pi B_i E_{ij} \zeta_j$$

is the finite difference form of Eq. (9) with $\Delta(\cos \theta) = 0.02$. The...
was also evaluated and compared with $F_{TJ}$. The values of the parameters used in Eqs. (A1) to (A6) are shown in Table 2. $B_i$ = fraction of the total black body flux for $T = 275K$ within wavelength interval at $i$/th wavelength.

| Table 2. Parameters for evaluation of $Q_\omega(r, \lambda)$ and $I(\theta, \lambda)$ |
|---|---|---|---|---|
| $i$ | $\lambda$ (μm) | $(Q_\omega)_{max}$ | $\gamma$ | $B$ | $w$ (g cm$^{-2}$) |
| 1 | 4 | 1·00 | 0·007 | 0·0023 | 0 |
| 2 | 5 | 1·00 | 0·04 | 0·0100 | 0·03 |
| 3 | 6 | 1·24 | 0·4 | 0·0207 | 0 |
| 4 | 7 | 1·00 | 0·08 | 0·037 | 0 |
| 5 | 8 | 1·05 | 0·1 | 0·051 | 0·5 |
| 6 | 9 | 1·10 | 0·08 | 0·053 | 5 |
| 7 | 10 | 1·15 | 0·08 | 0·091 | 10 |
| 8 | 12 | 1·23 | 0·7 | 0·115 | 5 |
| 9 | 14 | | | | |
| 10 | 16 | | | | |
| 11 | 18 | | | | |
| 12 | 20 | 1·25 | 1·1 | 0·075 | 0·1 |
| 13 | 25 | | | | |
| 14 | 30 | | | | |

Results

- $W =$ 0·5 g cm$^{-2}$
- $F_{TJ} =$ 0·661
- $F_{J} =$ 0·644
- $\Delta =$ 0·017
- $w =$ 0·010
- $\Delta =$ 0·005

Thus conversion from spherical to plane geometry makes little difference ~ about 1% of black body flux to $F_{TJ}$ and hence about 0·5% to $F_T$.

The variation of $Q_\omega$ (for $W = 0·5g cm^{-2}$) and $Q_\omega$ with $r$ is shown in Fig. 1. The curves are very similar for rather different distributions of incident radiation, and it is suggested that the exponential curve (shown dotted) can be used for $Q_\omega(r)$ for the range of atmospheric infrared spectra likely to be found at low levels.

**Correction for ‘Kinetic Theory’ effect.** Fukuta and Walter (1970) suggest that the effect of the water vapour ‘jump’ within one mean-free-path of the droplet surface can be allowed for in the Maxwell equations by multiplying the vapour diffusion coefficient, $D_\omega$, by a factor $r/(r + 1)$ where $r$ is droplet radius and $I = (2\pi M/RT)^4(D_\omega/\beta)$, where $M$, $R$, $T$ have already been defined, and $\beta$ is the condensation coefficient – taken to be 0·0415 by Fukuta and Walter. Substituting this correction factor in Eq. (12) results, at a temperature of 280K, in the reduction of growth rate by about a factor of 3 at $r = 1μm$; by 30% at $r = 4μm$; by 15% at $r = 10μm$. Thus this is another effect which in common with the effect of droplet curvature and solute mass on the vapour pressure near the droplet surface, diminishes with increase in droplet radius. This correction has no effect on the equilibrium curves (Fig. 2) since it is only the factor $A$ in Eq. (12) which is modified.