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On the cloud absorption anomaly

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SUMMARY

This paper provides an overview of the subject of absorption of solar radiation by water clouds in the earth's atmosphere. The paper summarizes the available evidence which points to disagreements between theoretical and observed values of both cloud absorption and reflection. The importance of these discrepancies, particularly to remote sensing of clouds as well as to studies of cloud physics and earth radiation budgets, is emphasized. Existing cloud absorption and reflection measurements are reviewed and the persistent differences that exist between calculated and measured near-infrared cloud albedos are highlighted. Various explanations for these reflection and absorption discrepancies are discussed and a simple outline of the theory of cloud absorption is provided. This outline is used to examine the large-droplet hypothesis as well as the effects of absorbing aerosol and enhanced water vapour continuum absorption. A further hypothesis regarding the effects of cloud inhomogeneities is also examined. While the theory of cloud absorption is not completely understood, especially with regard to inhomogeneous clouds, the underlying conclusion of this paper points to the need for better measurements of solar radiation in clouds, water vapour absorption and microphysics properties of clouds.

1. INTRODUCTION

The flow of radiant energy to and from the atmosphere is fundamental to the maintenance of the earth-atmosphere climate system. Basic to our understanding of such a system is the interaction of radiation with the atmosphere and the subsequent transformation of radiation into other forms of energy. The importance of these interactions to the climate system is evident by reference to the global and annual average energy balance. Representations of this budget have varied in detail over the years since the original version of Dines (1917) which is reproduced in Fig. 1 (a) in modified form. For instance, the planetary albedo determined by Dines to be about 50% has been considerably revised in the more modern depictions of the energy balance (compare Fig. 1 (a) and Fig. 1 (b)).

Despite the fundamental nature of the topic and the period of time that has elapsed since the initial estimate of Dines, a number of major uncertainties in the magnitude of some of the energy balance components still remain. For instance, the energy transfer at the surface of the earth remains uncertain on the global scale and the global monitoring of this energy flow represents a major obstacle confronting the climate research community. The purpose of this paper is partly to provide a review of the research that has exposed yet another uncertainty in the global energy balance, namely the absorption of solar radiation in clouds. The nature of the uncertainty has been stated in a number of different ways by several investigators. For example, Fritz (1951) commented that 'Preliminary results from these [McDonald's] measurements indicate that the absorption by these deep widespread systems averages about 20% of the solar radiation incident on the cloud. . . . This amount of absorption is much higher than the maximum of 6% which Hewson's theoretical calculations indicate.'

Thus a paradox is said to have emerged (Wiscombe *et al.* 1984) that measurements of cloud solar absorption tended to exceed theoretical estimates. Obviously Fritz's statement indicates that anomalous cloud absorption has loomed as a concern in atmospheric radiation for more than three decades. Aircraft observations such as those of Robinson (1958), Drummond and Hickey (1971), Reynolds *et al.* (1975), Rozenberg *et al.* (1974), Herman (1977), Stephens *et al.* (1978), Twomey and Cocks (1982), Stephens

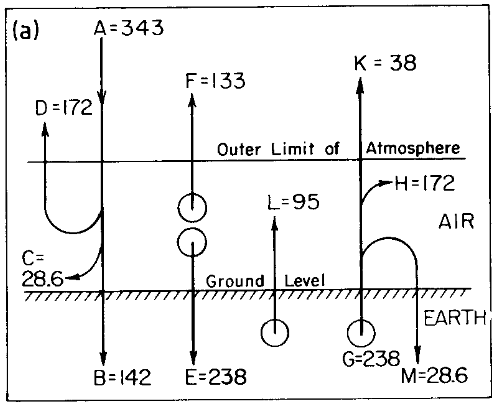

Figure 1 (a). The early schematic representation of the annual heat balance of the atmosphere. The original numerical values assigned to the various components are included in units of $W m^{-2}$ for comparison with Fig. 1 (b). The components are A: solar insolation, B: solar energy absorbed at the surface, C: solar absorption in the atmosphere, D: reflected solar radiation, E and F: emitted longwave to earth and space from the atmosphere, G: emitted from surface, H: absorbed longwave in the atmosphere, K: transmitted longwave to space, L: flux to atmosphere other than radiation, and M: reflected longwave back to surface (after Dines 1917).

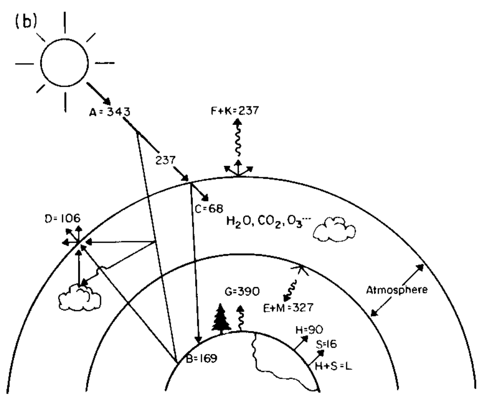

Figure 1 (b). A modern representation of the global annually averaged energy balance. The estimated solar absorption by the atmosphere is $68 W m^{-2}$ of which slightly in excess of $30 W m^{-2}$ might be attributed to absorption in cloud (after Ramanathan 1987).

and Platt (1987), Hignett (1987), Foot (1988) and the more recent results of the First ISCCP Regional Experiment (FIRE) reported by Nakajima *et al.* (1990) have continued to fuel this issue with reports of measurements at variance with theoretical expectation. This paper provides an overview of these observations, attempts to emphasize any consistent trend in the observations and aims to provide a critique of the various explanations proposed to account for the discrepancies.

Before doing so, a relevant and obvious question to ask is, why be so concerned with large errors in a relatively small component of the global energy cycle? One compelling reason for the interest in the cloud absorption problem is apparent from the consideration of the information contained within Fig. 2. Shown are the latitudinal variations of the two components of the earth radiation budget (ERB). The difference between these two large components, emphasized as the shaded region of Fig. 2, provides the fundamental drive of the atmospheric circulation. This drive, however, is a small

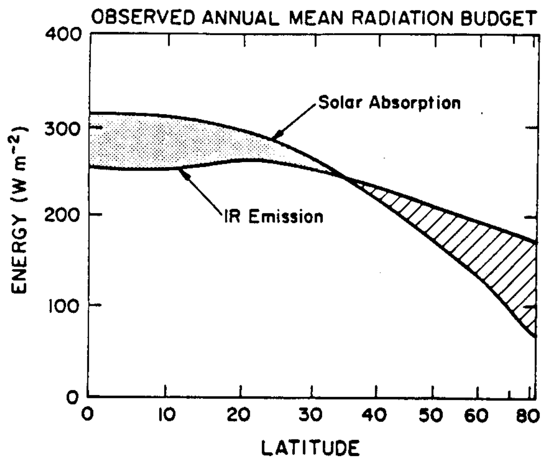

Figure 2. Annual zonal mean estimates of absorbed solar radiation and outgoing longwave (IR emission) obtained by satellites. Shaded regions denote net heating and dashed regions are net cooling (after Ramanathan 1987).

residual that results from the balance between the absorbed solar and emitted infrared radiation. For the example presented, the maximum net annual heating of 50–60 W m^{-2} in the tropics is actually only 20% of the total absorbed solar radiation in these regions. This implies that roughly 80% of the solar radiation absorbed by the earth-atmosphere system is used to heat the surface and drive oceanic circulations, whereas the remaining 20% is available to drive the atmospheric circulation. Consequently, small uncertainties in the absorbed solar radiation translate into a much larger uncertainty in the drive of the atmospheric and oceanic circulations.

Another reason to be concerned about the quoted lack of agreement between theory and observation lies in the uncertainty introduced both from our understanding of the transfer of solar radiation through clouds and, subsequently, from our ability to calculate this energy transfer. As a specific example, the uncertainty in cloud absorption has a direct and significant bearing on studies that attempt to use solar reflection measurements to estimate mean size of cloud droplets, among other properties. Measurements of near-infrared reflection are often exploited in such studies since water vapour absorption is considered to be small compared to liquid and solid absorption (i.e., in the near-infrared windows). Such an approach has been outlined by Twomey (1971), applied to the measurements of Blau *et al.* (1966) by Hansen and Pollack (1970), to the clouds of Venus by Pollack *et al.* (1978) and to the measurements collected over marine stratocumulus clouds by Nakajima *et al.* (1990) among other studies.

Quite apart from these issues, absorption of solar radiation in clouds is also important for the energy balance of the cloud itself. Herman and Goody (1976) demonstrated the delicate balance between longwave cooling and shortwave heating in Arctic stratus, Stephens *et al.* (1978) reported that on occasions solar heating was observed to exceed longwave cooling as did Twomey (1983) in his studies of Californian stratus. If absorption is appreciably greater than currently predicted by theory, then solar heating would be even more important and may perhaps even dominate the longwave cooling under certain circumstances.

Current explanations for the discrepancies between theoretical and observed cloud absorptions can be divided into two broad categories. One of these categories focuses attention on uncertainties in cloud optical properties which arise, for example, either from uncertainties in the numbers and sizes of cloud droplets or from uncertainties in the composition of the cloud volume. For instance, it has been proposed that absorption by foreign solid aerosol particles, either imbedded in or interstitial to cloud droplets, might provide an additional source of absorption. The second category, and one much less developed, queries the relevance of the plane parallel theory when applied to clouds which are in reality spatially variable. The explanation proposes that heterogeneities, when properly accounted for in the theory, might explain the observed discrepancy.

The plan of the paper is as follows. The next section discusses the mounting observations that have isolated the anomaly. Section 3 offers a simple theoretical description of absorption in clouds and attempts to isolate the key optical properties of the clouds that determine the absorption. This description is then followed by a discussion of the relationship between these optical properties and the cloud microphysical, aerosol and water vapour properties. The discussions of Sections 3 and 4 are brought together in Section 5 where it is shown how large droplets and the presence of aerosol influence cloud absorption. The hypothesis that both cloud absorption and reflection at near-infrared wavelengths can be significantly influenced by water vapour continuum absorption is also explored in this section. Section 6 reviews the effects of spatial heterogeneities on cloud absorption and a summary of the discussion is provided in the final section of the paper.

2. Observational evidence

(*a*) *Broadband measurements of shortwave absorption in cloud*

Subsequent to the work of Fritz, aircraft measurements by Robinson (1958), Griggs (1968), Drummond and Hickey (1971) and Reynolds *et al.* (1975) added to the cloud absorption controversy with estimates based on broadband flux measurements in the range 20–40%. Table 1 which is adapted from Rawlins (1989) summarizes various estimates of solar absorption based on a variety of aircraft measurements conducted over the past thirty or more years. This list of values shown is not meant to be complete but

TABLE 1. Previous aircraft measurements of solar absorption within layer cloud, including the standard error or range of measurements (adapted from Rawlins 1989)

Author	Mean absorptance (± error) and/or range	Mean cloud thickness or range	Comments
Neiburger (1949)	0·07 ± 0·02	250m	75 cases, some negative
Fritz & Macdonald (1951)	0·27 ± 0·03	5–7km	1 case
	0·15 ± 0·05		average of 7 cases
Chel'tsov (1952)	0·035 ± 0·002	360m	
	0·072 ± 0·018	530m	
Robinson (1958)	0·22		but 0·19 even in thinnest
	0·13 to 0·29		clouds
Koptev and Voskresenskii (1962)	0·02–0·10	200–500m	
Griggs (1968)	0·04	300m	36 cases
	−0·30 to 0·28		
Goisa & Shoshin (1969)	0·072		
Paltridge (1971)	<0·02	180–560m	3 cases
Reynolds *et al.* (1975)	0·12 to 0·36	1–5km	3 cases, albedo 0·37 to 0·46
Herman (1977)	0·07 ± 0·08	100–800m	6 cases, albedo 0·60 to 0·75
Stephens *et al.* (1978)	0·087	400m	8 cases, albedo 0·50 to 0·75
	0·0 to 0·2		
Slingo *et al.* (1982)	0·068 ± 0·026	435m	albedo 0·68 ± 0·02
Foot (1988)	0·10 to 0·15	1000m	albedo 0·82 ± 0·02
Rawlins (1989)	0·02 to 0·12		Broken cloudiness

is merely introduced to emphasize the wide range of values obtained. On the basis of these results various authors have suggested that the absorption within clouds is anomalously large relative to equivalent theoretical values (which are typically 0·05–0·1 for the cloud types under consideration). It must also be noted that not all measurements disagree with observations as indicated by the observations reported by Slingo *et al.* (1982) and Rawlins (1989).

Why is there such a range in estimates of cloud absorption? Part of the explanation lies in the type of measurements made and the approach used to estimate the absorption. Typically the absorption estimates were derived from broadband measurements of solar fluxes. The general approach used in these studies is to average the individual measurements of solar flux along the flight path at cloud base and top. Net fluxes at cloud top and base are then obtained from the difference of these averaged fluxes and the absorption subsequently derived as a residual. This residual is typically much smaller than the individual measured fluxes themselves and is often smaller than the experimental error associated with the procedure. For example, Twomey (1983) reported measurements of shortwave fluxes above and below stratus cloud off the Californian coast that on analysis provided negative absorptions. Bonnel *et al.* (1983) analysed radiation measurements made in both mid-latitude and near-equatorial stratocumulus cloud fields that were short lived and highly inhomogeneous. They reported several cases of large absorptions but dismissed these because of the extreme cloud variability along the flight levels. The effects of the heterogeneities both on the method of analysis and on cloud absorption itself have been proposed as a possible source of discrepancy. In recognition of this possible effect, refinements of the experimental procedures have been developed (Ackerman and Cox 1981; Rawlins 1989) and these are further described in section 6 of this paper.

(*b*) *Measurements of cloud albedo*

Reliable measurements of broadband cloud absorption are difficult to make since they rely on averaging spatially and temporarily varying data. The concerns about the credibility of the experimental procedure, the obvious sampling problem and experimental accuracy are all legitimate and figure as important reasons as to why the analysis of broadband flux measurements, while suggestive of an absorption anomaly, remained unconvincing for so long. By contrast, the albedo of the cloud is an intrinisically more accurate and reliable measurement to make, although the problem of defining the cloud properties at the time of measurement remains, especially when quantitative comparisons with theory are sought. Despite these difficulties a few quantitative comparisons between theory and observations of broadband albedos have been attempted (e.g., Herman and Curry (1984); Hignett (1987); Stephens *et al.* (1978); Foot (1988)) with the general conclusions that the theory and observations broadly agree. An example of this type of agreement is demonstrated in the work of Hignett (1987) as shown in Figs. 3(a) and 3(b). The measurements are compared against the spread of albedo values derived from calculations based on the observed range of integrated liquid water paths. This agreement is misleading, however, as further consideration of spectral flux measurements demonstrates. For example, the visible and near-infrared flux measurements of Hignett show systematic departures from theory that are not evident in the broadband comparisons. These differences are shown in Figs. 4(a) and 4(b) in which the ratios of the near-infrared to visible albedo are presented. These ratios indicate that the measured values of near-infrared cloud albedo are significantly less than those predicted by theory. This same type of discrepancy also seems to be apparent in comparisons of even finely resolved spectral radiances.

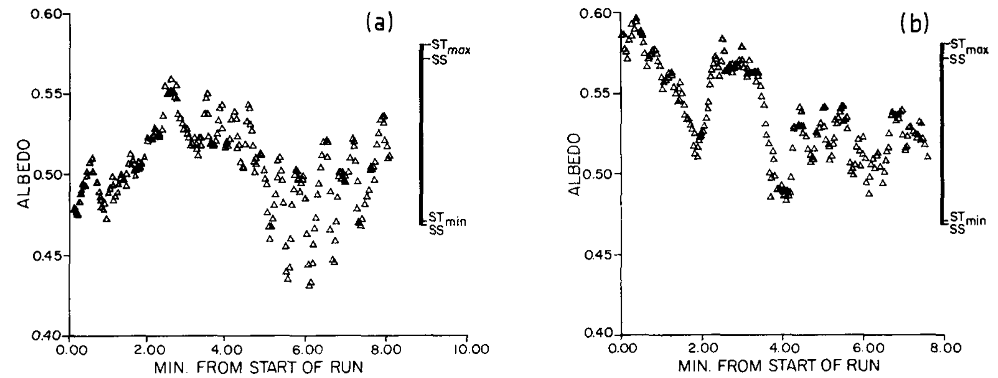

Figure 3 (a). Total albedo against time from an aircraft run just above cloud top; airspeed was $100 \mathrm{m\,s^{-1}}$. ST and SS refer to two different model values for maximum and minimum liquid water path clouds. The vertical bar emphasizes the range expected from theoretical calculation (after Hignett 1987).

Figure 3 (b). As Fig. 3 (a) except for a second aircraft run (after Hignett 1986).

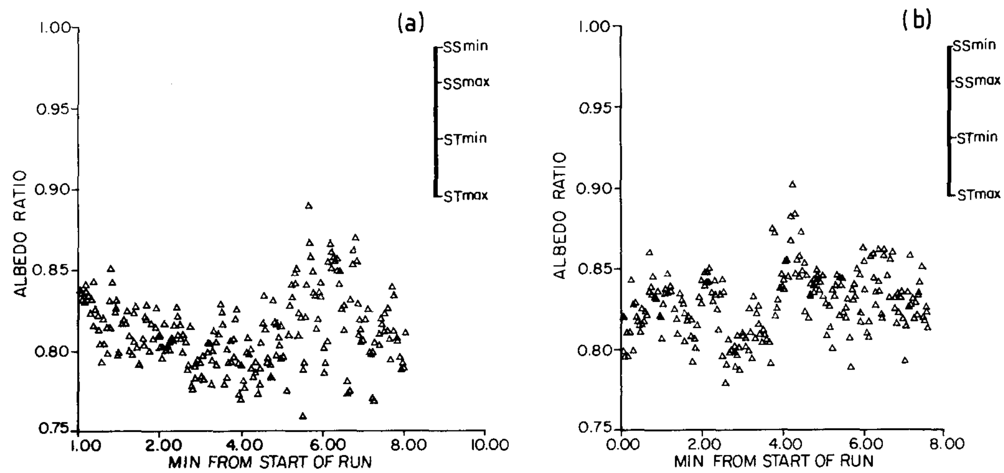

Figure 4 (a). Ratio of near-infrared to visible albedo against time for the run in Fig. 3 (a); notation also the same.

Figure 4 (b). As Fig. 4 (a) but for the run in Fig. 3 (b) (after Hignett 1987).

(c) *Measurements of spectral reflection by clouds*

Although measurements of broadband albedo are more reliable than broadband measurements of absorption, the comparisons with calculations still do not lead to definitive conclusions about discrepancies between theory and observations, and it has been evident for some time that even more sophisticated radiometric techniques are needed to resolve the cloud absorption question. Certainly the desire to reconcile the cloud absorption issue was the principal motivation for the spectral measurements reported by Stephens and Platt (1987) and by King *et al.* (1986) and it seems to have been a significant issue in the observational studies of Twomey and Cocks (1982).

Unfortunately, very few spectral measurements of cloud reflectance are reported in the literature at a resolution sufficient to resolve even gross spectral features, and there are even fewer cases for which supplemental cloud microphysics data are available for a comprehensive comparison with theory. Early observations of cloud reflectance in the near-infrared were published by Blau *et al.* (1966) mainly over ice clouds. These data were subsequently compared with theoretical simulations by Hansen and Pollack (1970) but discrepancies between the measurements and these calculations are difficult to establish. The near-infrared cloud reflectance measurements obtained by radiometers on Soviet Cosmos satellites, however, revealed evidence of a near-infrared absorption anomaly. In fact, Rozenberg *et al.* (1974) concluded that: 'Terrestrial clouds contain matter whose specific absorption averages two orders of magnitude higher than absorption of liquid-drop or crystalline water. . . . The spectral trend of the specific absorption of this matter is the same as for pure water or ice, i.e. the absorption coefficients of pure water are enhanced in clouds'.

More recent spectral reflectance and transmittance measurements from a series of aircraft flights were published by Doherty and Houghton (1984). As we discuss below, cloud optical thickness and some measure of the mean droplet size are two parameters that can be juggled in the calculations in an attempt to match the observations. According to the results of both Doherty and Houghton and those introduced in the following examples, the reflections at all wavelengths generally cannot be consistently matched with a theory that uses a single unique set of microphysical parameters. The parameters that provide best agreement at the shorter wavelengths invariably lead to an overestimate of reflection in the near-infrared (for example at 1·6 μm). This general result was also a conclusion of Georgiyevskiy and Shukurov (1985) based on analyses of spectral transmittance measurements of clouds.

Three studies in which both spectral radiation measurements and cloud microphysics data were collected are those of Twomey and Cocks (1982), Stephens and Platt (1987) and Foot (1988). The results of Twomey and Cocks are reproduced in Fig. 5 for discussion. The upper panel provides the time-varying reflectances measured from a four-channel radiometer, the second panel superimposes the theoretically predicted reflectances for these four wavelengths using values of optical thickness and mean droplet size derived from *in situ* cloud physics measurements, whereas the lower panel includes the theoretical results obtained using optical thickness and mean droplet size which were optimized to match the observations. The optimum droplet sizes determined in this way tend to be significantly larger than those measured. Twomey and Cocks also estimated the range of values of the bulk-water absorption required to bring the observations and theory into closer agreement. Their results, reproduced in Table 2 together with accepted values of the bulk absorption coefficient of water, suggest that a pronounced increase in cloud absorption is needed to account for the differences between observations and theory. Using the multichannel radiometer described in Doherty and Houghton (1984), Foot (1988) also reached similar conclusions by demonstrating that the measured near-infrared reflections could be matched with calculated reflectances if the droplet size was increased to values above those recorded by *in situ* measurements. Stephens and Platt (1987) also provided comparisons between observed and simulated reflection spectra based on data collected from three flights above and within stratocumulus cloud layers. Two of these comparisons are reproduced in Fig. 6. Incorporating the measured cloud droplet size distributions in their calculations, Stephens and Platt (1987) concluded that theory tended to overestimate the near-infrared reflection, especially in the windows.

The recent FIRE marine stratocumulus experiment (e.g. Albrecht *et al.* 1988) also provides the opportunity of making use of *in situ* spectral radiation and microphysics measurements using low-flying aircraft, coincident remote radiometric observations from the higher-flying NASA ER-2 aircraft and coincident data from various meteorological satellites. King *et al.* (1990) report on the analyses of spectral radiance data and droplet-size information obtained from measurements made on the University of Washington C-131A research aircraft which flew in the middle of thick clouds. Based on arguments of

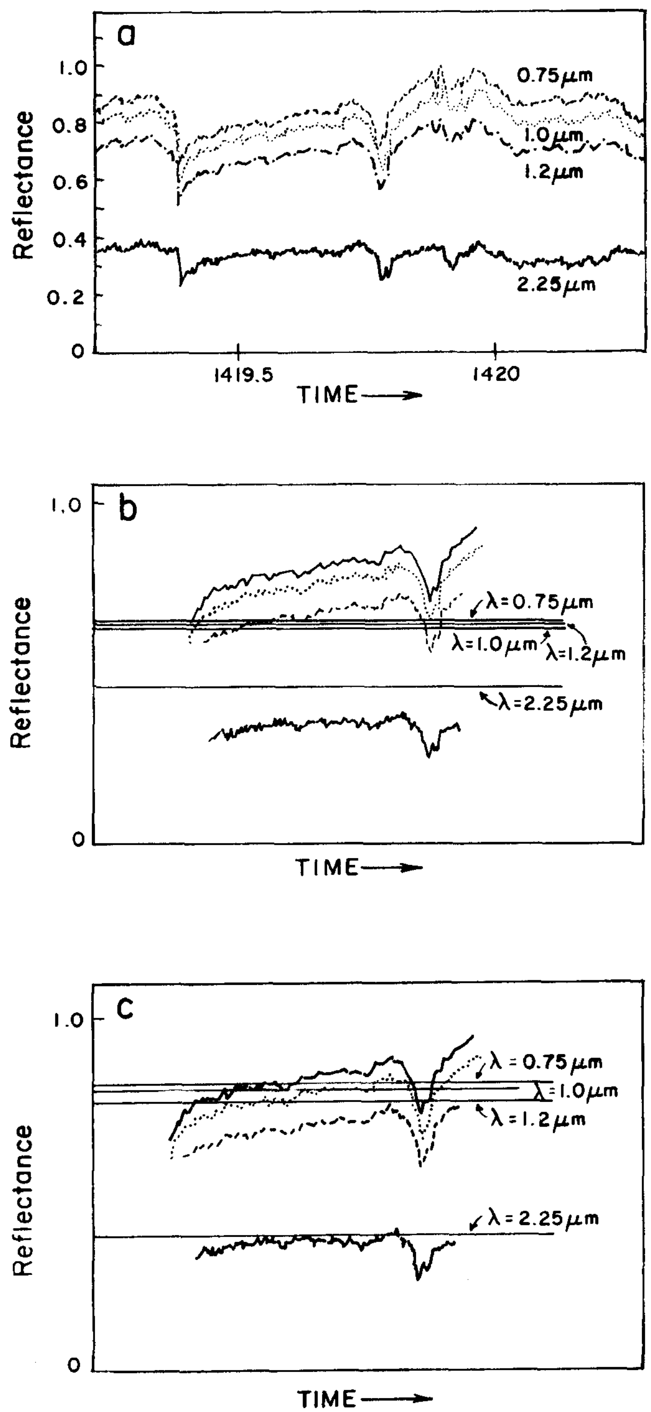

Figure 5. Time-varying cloud reflectances from a four-channel spectrometer (a). Portions of the data are shown in (b) along with theoretical values calculated with optical thickness τ and droplet sizes $\bar{r}$ given by measured cloud microphysics (horizontal lines). Lower panel is similar to (b) except that τ and $\bar{r}$ were optimized to provide the best fit to data; $\tau = 32$ and $\bar{r} = 12\ \mu$m (after Twomey and Cocks 1982).

TABLE 2. RANGE OF VALUES OF ABSORPTION COEFFICIENT (cm^{-1}) NEEDED TO BRING MEASUREMENTS AND CALCULATIONS INTO AGREEMENT (AFTER TWOMEY AND COCKS 1982). COMPARE THE LISTED 0·75 μm REFLECTANCES WITH THOSE PROVIDED IN FIG. 5(a).

Reflectance at 0·75 μm	κ (1·0 μm)	κ (1·2 μm)	κ (2·25 μm)
1·0 ± 0·002	0·5–0·9	1·0– 1·9	25–35
0·825 ± 0·01	1·3–1·7	5·0– 6·7	35–55
0·63 ± 0·01	1·8–3·0	6·0–10·3	29–51
0·5 ± 0·01	2·0–3·4	8·4–12·0	29–46
Accepted bulk values	0·35–0·36	1·02–1·03	17–22

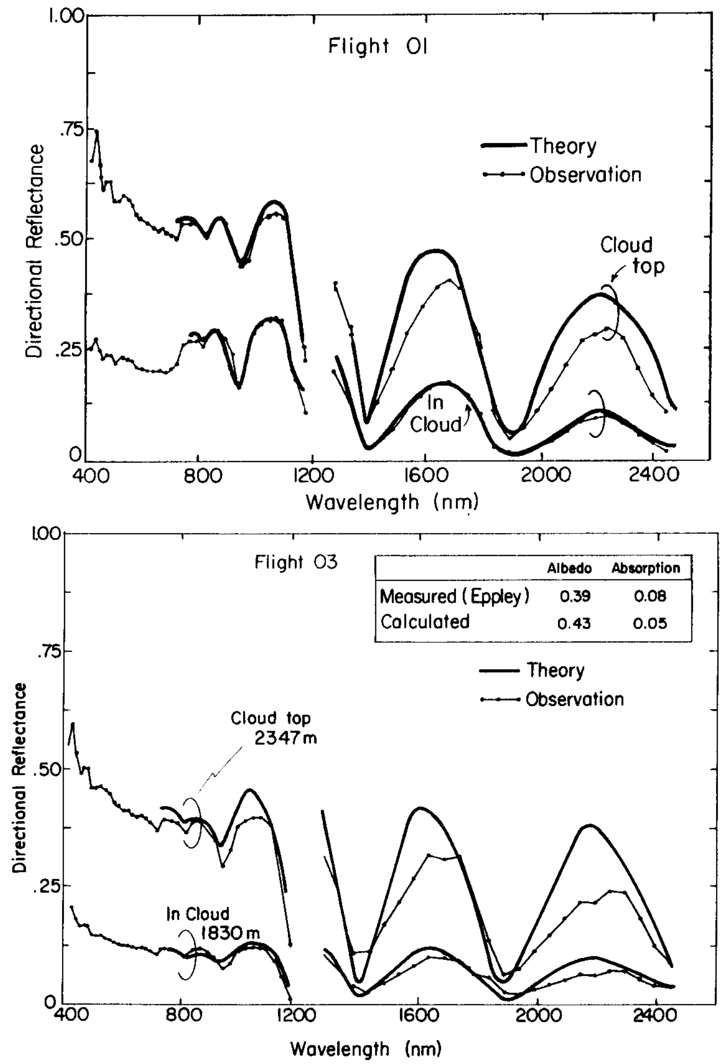

Figure 6. Comparison of the calculated (heavy curves) and measured (light curves) spectral reflectance for two of the three Sc studied by Stephens and Platt (1987). The comparisons are shown for cloud top and some level approximately in the middle of the cloud. Also included are the measured and theoretically derived broadband albedos and shortwave absorption estimates (after Stephens and Platt 1987).

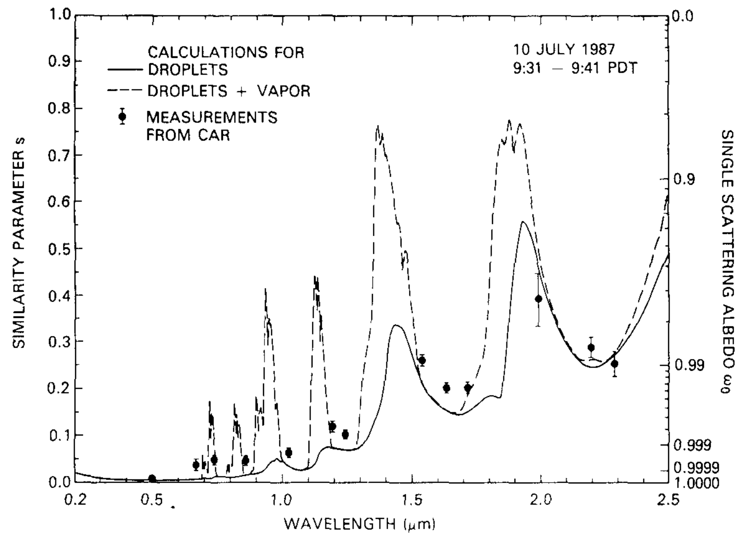

Figure 7. Calculations of the single-scatter albedo (right scale) as a function of wavelength for water droplets alone (solid) and droplets plus vapour (dashed) for the cloud droplet size distribution and water vapour conditions of a marine stratocumulus cloud sampled during the FIRE. The measurements are averages derived using the analysis scheme of King (1981). The left-hand scale applies to the similarity parameter which is a scaling parameter uniquely related to single albedo (after King *et al.* 1989).

diffuse transport, King (1981) earlier demonstrated that the ratio of nadir to zenith intensity, deep in thick clouds, reduces to a simple function of the single-scatter albedo ($\tilde{\omega}_0$) and asymmetry parameter. (The meaning of these parameters is provided below). With an estimate of the asymmetry parameter provided from the measured droplet size distributions by Mie theory together with the measured intensity ratio, King *et al.* were able to deduce the single-scatter albedo at thirteen different wavelengths corresponding to the channels of the radiometer. The resultant spectrum derived from their analyses is shown in Fig. 7 for the cloud case sampled on 10 July 1987. The comparisons do show some discrepancies between theory and observations. For instance, it appears that the value of droplet absorption $(1 - \tilde{\omega}_0)$ measured in channel 8 (1·57 μm) is 0·0115 versus the value of 0·0075 predicted from theory, thus suggesting a substantially larger absorption at this wavelength. Despite this discrepancy, a general interpretation of these results in terms of both the broadband cloud absorption and reflection are difficult to make. So far only one case has been analyzed by King *et al.* Certainly more comparisons are required and some inferences about the broadband properties need to be drawn from future analyses.

Another important result from the FIRE in which spectral measurements of reflection were compared with theoretical values has been reported by Nakajima *et al.* (1990). In that study, spectral reflection measurements at 0·754, 1·65 and 2·16 μm corresponding to the channels of a multichannel radiometer which was flown on the NASA ET-2 aircraft are used to retrieve the optical thickness and effective particle radius. Their results are shown in Fig. 8 in the form of a scatter diagram that compares remotely-sensed values of droplet size to the *in situ* values. The results show that the remotely-sensed values are systematically larger than the *in situ* values. This tendency to overestimate the particle size is also consistent with the previous findings of Twomey and Cocks (1982), Foot (1988) and Stephens and Platt (1987).

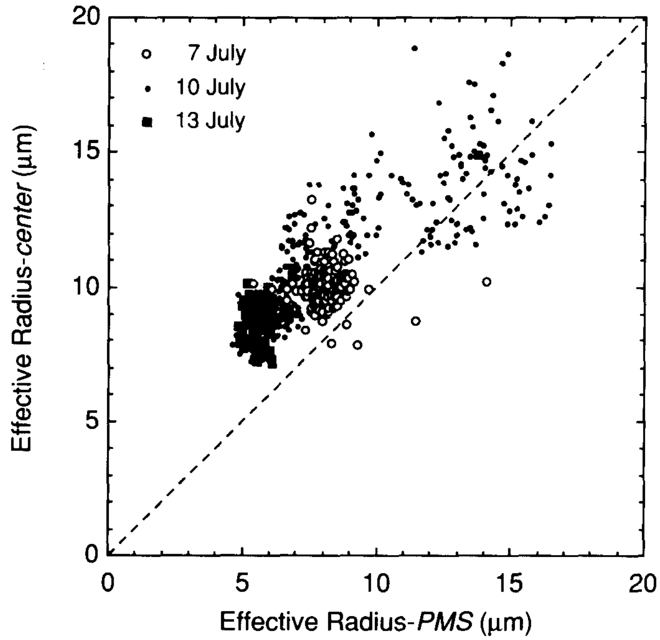

Figure 8. Scatter diagram comprising *in situ* and remotely-sensed effective radius. The effective radius centre is weighted to the centre of the cloud (after Nakajima *et al.* 1990).

The results described above, based on analyses of spectral measurements, together with broadband data like those of Hignett, tend to point to a consistent difference between measured and calculated cloud reflection and, by inference, cloud absorption in the near-infrared and show that perhaps these discrepancies are largest in the windows. While anomalous absorptions in the visible cannot strictly be ruled out, the large values of visible reflectance reported by Twomey and Cocks and by Stephens and Platt do not generally support this hypothesis.

3. Theoretical limits of solar absorption in clouds

A brief and simple theoretical account of solar radiative transfer in clouds is now presented. The aim is to establish the connections between the absorption and cloud albedo and the various cloud optical properties that are defined by the microphysical properties of the cloud. Two-stream methods are generally accurate and have been widely used to study many types of transfer problems (for instance radiation in plant canopies, the insulating properties of fibreglass, the spectral properties of certain paint pigments, the turbidity of oceans and properties of planetary atmospheres among many others) and several reviews of the subject can readily be found (Welch *et al.* 1980; Meador and Weavor 1980; King and Harshvardhan 1986).

The two-stream version of the radiative transfer equation follows by consideration of the energy balance of a small but finite region of the cloud and by further separating the flux in the upward (+) and downward (−) directions in these regions. The two-stream equations can be written in the form

$$\mp \frac{dF^{\pm}}{d\tau} = -[D(1-\tilde{\omega}_0)+\tilde{\omega}_0 b]F^{\pm} + \tilde{\omega}_0 b F^{\mp} + F_0 e^{-\tau/\mu_0}\tilde{\omega}_0 \begin{pmatrix} b_0 \\ f_0 \end{pmatrix} \tag{1}$$

where D is a measure of the diffuseness of the radiation. The quantity τ is the optical depth in the cloud and an increment of optical depth is defined as

$$\Delta\tau = (\alpha_{\text{abs}} + \alpha_{\text{sca}})\Delta z, \tag{2}$$

where Δz is the geometric thickness of the layer and α_{abs} and α_{sca} are the volume absorption and scattering coefficients respectively. The single-scatter albedo $\tilde{\omega}_0$ is

$$\tilde{\omega}_0 = \frac{\alpha_{\text{sca}}}{\alpha_{\text{abs}} + \alpha_{\text{sca}}}. \tag{3}$$

The other quantities in Eq. (1) are expained in the Appendix. Within the context of (1), F_0 is the flux through a surface at cloud top normal to the collimated flow, and $\mu_0 F_0$ is the value of this flux through a horizontal surface at cloud top. The angle formed between the surface normal to the collimated flow and the horizontal is the solar zenith angle θ_0 and we set $\mu_0 = \cos\theta_0$. The factor $F_0 e^{-\tau/\mu_0}$ in (1) represents the amount of this collimated flux transmitted to the τ level within the cloud.

Details of the general solution to (1) are provided in the Appendix. It is particularly useful to consider the theoretical behaviour of the absorption and albedo in the limits as the optical thickness of the cloud $\tau^* \rightarrow 0$ and $\tau^* \rightarrow \infty$. For optically thick clouds with $\tau^* \rightarrow \infty$, it follows directly from (A9 a) that

$$\mathcal{R}_\infty = \frac{1}{\mu_0}\left[Z_+(\mu_0) - Z_-(\mu_0)\frac{h_-}{h_+}\right] \tag{4a}$$

$$\mathcal{A}_\infty = 1 - \mathcal{R}_\infty \tag{4b}$$

where $\mathcal{R}_\infty$ and $\mathcal{A}_\infty$ are respectively the albedo and absorption of this 'semi-infinite' cloud. According to these simple relationships both the albedo and absorption approach fixed asymptotic limits as τ^* increases. These upper (theoretical) limits are defined solely by the optical properties (constants) of the cloud, namely $\tilde{\omega}_0$, b_0, f_0, D and b as well as by the solar geometry as expressed by μ_0. The invariant nature of $\mathcal{R}$ and $\mathcal{A}$ with increasing optical thickness is well known and forms the basis for other well-established radiative transfer principles (such as the principles of invariance discussed in Chandrasekhar 1960). Figure 9 illustrates the basic relationship between $\mathcal{A}_\infty$ and the droplet absorption factor

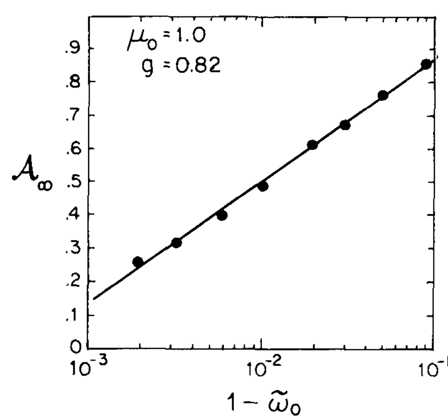

Figure 9. Illustration of the relation between cloud absorption and $1 - \tilde{\omega}_0$ (the single-scatter co-albedo) for a semi-infinite cloud with $\theta_0 = 0°$ and $g = 0{\cdot}82$.

$1 - \tilde{\omega}_0$ based on (4 a) and (4 b) for $\theta_0 = 0$ and an asymmetry factor $g = 0{\cdot}82$. We introduce the expression

$$\mathcal{A}_\infty \approx constant \times (1 - \tilde{\omega}_0)^{0{\cdot}4} \tag{5}$$

which is depicted as the solid curve in Fig. 9 for the convenience of later discussion.

For optically thin clouds with $\tau^* \to 0$, it again follows from (A9 a) and (A9 b) that

$$\mathcal{R}_0 = \frac{\tau^*}{\mu_0} \tilde{\omega}_0 b_0 \tag{6a}$$

$$\mathcal{T}_0 = 1 - \frac{\tau^*}{\mu_0}(1 - \tilde{\omega}_0 f_0) \tag{6b}$$

and

$$\mathcal{A}_0 = \frac{\tau^*}{\mu_0}(1 - \tilde{\omega}_0). \tag{6c}$$

Therefore in the thin limit, both the albedo and absorption of the cloud vary linearly with optical thickness and, as expected, respectively depend on the backscatter and absorption properties of the individual cloud droplets. Furthermore, the dependence of absorption on solar zenith angle varies between the thin and thick limits. For thin cloud, the absorption increases as solar zenith angle increases (i.e. as μ_0 decreases in (6*c*)) whereas the absorption in the semi-infinite limit can be shown to decrease with increasing solar zenith angle. These results are consistent with those reported by others (e.g. Davies *et al.* 1984; Stephens 1978 among others).

4. Cloud optical properties

Two parameters that appear to have a major bearing on the amount of solar radiation absorbed and reflected by a cloud layer are the extinction coefficient α_{ext} (through its presence in τ^*) and the single-scattering albedo $\tilde{\omega}_0$. A third parameter, the amount of backscattered radiation, is also important and is related in the manner discussed by King and Harshvardhan (1986) to the asymmetry factor which is derived from the scattering phase function. The value of the asymmetry factor is much less variable in water droplet clouds and perhaps better known than either α or $\tilde{\omega}_0$. The latter properties are hereafter referred to as cloud optical properties and combine in a complex way to influence the gross absorption by cloud. For instance, suppose a cloud is composed of droplets that have associated with them the value $\tilde{\omega}_0 = 0{\cdot}999$ and that these droplets are sufficiently numerous that $\alpha_{ext} = 40\,\text{km}^{-1}$. Thus while a single droplet absorbs only 0·1% of the radiation incident on it, a 1 km thick cloud of such droplets absorbs approximately 10% of the incident radiation that illuminates the cloud. In this simple illustration, multiple scattering between cloud droplets acts to enhance the absorption in the cloud over the absorption by a single particle almost one hundredfold.

Given the combined importance of these properties to cloud absorption, it is reasonable to suspect, and thus not surprising to find, that the incorrect specification of either $\tilde{\omega}_0$ or α_{ext} or both, in the theory, figure as possible sources for the apparent discrepancy between theory and observation. As we will show, these properties not only depend on the number concentration of cloud droplets but also on the size and composition of the droplets, the amount of water vapour in the volume, the strength of this vapour absorption, as well as on the existence of aerosol in the volume. That is,

$$\alpha_{ext} = \alpha_d + \alpha_v + \alpha_a, \tag{7}$$

where the three terms on the right-hand side of (7) respectively represent the extinction by droplets, water vapour and aerosol. In a similar way, we write

$$1 - \tilde{\omega}_0 = [\alpha_{\text{abs,d}} + \alpha_{\text{abs,v}} + \alpha_{\text{abs,a}}]/\alpha_{\text{ext}} \tag{8}$$

for total absorption. We now aim to provide some assessment of how the above-mentioned factors influence both $\tilde{\omega}_0$ and α_{ext} and, by (4*a*), (4*b*), (6*a*–6*c*), the bulk absorption by clouds.

(*a*) *Cloud droplet effects*

In dealing with the effects of droplet microphysics on the radiative transfer through clouds, its proves convenient to characterize such a polydispersion in terms of a general form of gamma distribution

$$\eta(r) = \frac{N_0}{\Gamma(j) r_m} \left(\frac{r}{r_m}\right)^{j-1} \exp\left(-\frac{r}{r_m}\right) \tag{9}$$

where N_0 is the total (volume) concentration of droplets, r_{m} is a characteristic radius of the distribution and j is a constant which is often an integer. The characteristic radius r_{m} and j are closely related to the mode, mean and effective (r_{e}) radii

$$\begin{cases} r_{\text{mode}} = (j-1) r_{\text{m}} \\ r_{\text{mean}} = j r_{\text{m}} \\ r_{\text{e}} = (j+2) r_{\text{m}} \end{cases} \tag{10}$$

In the discussion to follow, examples of calculations will be presented for the Deirmendjian (1969) C1-cloud model which is specified by (9) with $j = 7$ and $r_{\text{m}} = 2/3\ \mu\text{m}$.

The liquid water content w associated with such a distribution of droplets simply follows as

$$w = \frac{4}{3}\pi\rho_{\text{water}} \int_0^\infty \eta(r) r^3 dr = \frac{4}{3}\pi\rho_{\text{water}} N_0 r_{\text{m}}^3 f(3) \tag{11}$$

where ρ_{water} is the density of water and

$$f(l) = \frac{\Gamma(j+l)}{\Gamma(j)}. \tag{12}$$

Similarly, the cross-sectional area per unit volume of the distribution is

$$A = \pi \int_0^\infty \eta(r) r^2 dr = \pi N_0 r_{\text{m}}^2 f(2). \tag{13}$$

The volume extinction and absorption coefficients are given by

$$\alpha_{\text{ext}} = \pi \int_0^\infty \eta(r) Q_{\text{ext}} r^2 dr \tag{14a}$$

and

$$\alpha_{\text{abs}} = \pi \int_0^\infty \eta(r) Q_{\text{abs}} r^2\, dr, \tag{14b}$$

where Q_{ext} and Q_{abs} denote respectively the extinction and absorption efficiencies for drops of radius r. We now seek explicit relationships between these coefficients and some measure of the droplet size. If we consider the large x_m limit ($x_m = 2\pi r_m/\lambda$, λ for wavelength), then $Q_{ext} \approx 2$ and it follows that

$$\alpha_{ext} \approx 2A = \frac{3w}{2\rho_{water} r_e}. \tag{15}$$

Therefore according to this relationship, and under conditions of fixed liquid water content, the extinction α_{ext} scales as the inverse of r_e. Stephens (1978) derived (15) and further assumed r_e to be an implicit function of w to arrive at a relationship between α_{ext} and cloud liquid water content and thus express the optical thickness of the cloud solely in terms of the vertically integrated liquid water content (referred to as the cloud liquid water path, LWP).

Ackerman and Stephens (1987) invoked a simple and conceptually approximate model of droplet scattering (van de Hulst's 1957 anomalous diffraction theory) to examine the relationship between single scatter albedo and r_e. This relationship can be expressed as

$$\tilde{\omega}_0 \approx \frac{1}{2} - \frac{1}{(j+1)j}\left[\frac{j}{v_m(v_m+1)^{j+1}} + \frac{1}{v_m^2(v_m+1)^j} - \frac{1}{v_m^2}\right] \tag{16}$$

where the integrations (14a) and (14b) were carried out analytically using the size distribution (9) together with the analytic expressions for Q_{abs} and Q_{ext} provided by the approximate scattering theory. In (16), v_m is an absorption scaling parameter defined as

$$v_m = 2\kappa r_m \tag{17}$$

where $\kappa = 4\pi n'/\lambda$ is the bulk absorption coefficient of water at the wavelength λ. One interpretation of v_m is that it is a single parameter that combines the effects of both droplet size and absorption properties of water at a single specified wavelength. Over the wavelength range of interest, the complex part of the refractive index for water $n' < 0{\cdot}3$ and $10^{-7} < v_m < 10$ for $r_m < 1{\cdot}0$ μm as in Deirmendjian's C1-, C2- and C3-cloud models.

Equation (16) provides us with a single parametric relationship between the cloud optical properties and cloud droplet size (through r_m) and composition (through n') based on a simple scattering theory. However, this relationship is still somewhat complex. To simplify matters, Ackerman and Stephens (1987) approximated (16) by

$$1 - \tilde{\omega}_0 \approx \mathit{constant} \times \kappa r_e^p \tag{18}$$

and further established three broad absorption regimes which they characterized in terms of both κ and p as:

- 'weak absorption', $\kappa < 5\ \mathrm{cm}^{-1}$ and $p = 1$,
- 'moderate absorption', $5\ \mathrm{cm}^{-1} < \kappa < 100\ \mathrm{cm}^{-1}$ and $1 > p > 0{\cdot}7$, and
- 'strong absorption', $\kappa > 100\ \mathrm{cm}^{-1}$ and $p < 0{\cdot}7$.

Examples of the relationship (18) are depicted in Fig. 10 as solid curves and these are compared to those (open circles) from the modified anomalous diffraction theory (Ackerman and Stephens 1987). Also shown in the inset is the variation in p with the absorption coefficient κ. The essential point of this diagram is that it demonstrates that the absorption by cloud particles increases with increasing values of r_e but the particular rate of increase varies according to the strength of liquid water absorption (i.e. κ) and, as we show below, this has a profound effect on the sensitivity of cloud absorption to changes in droplet size.

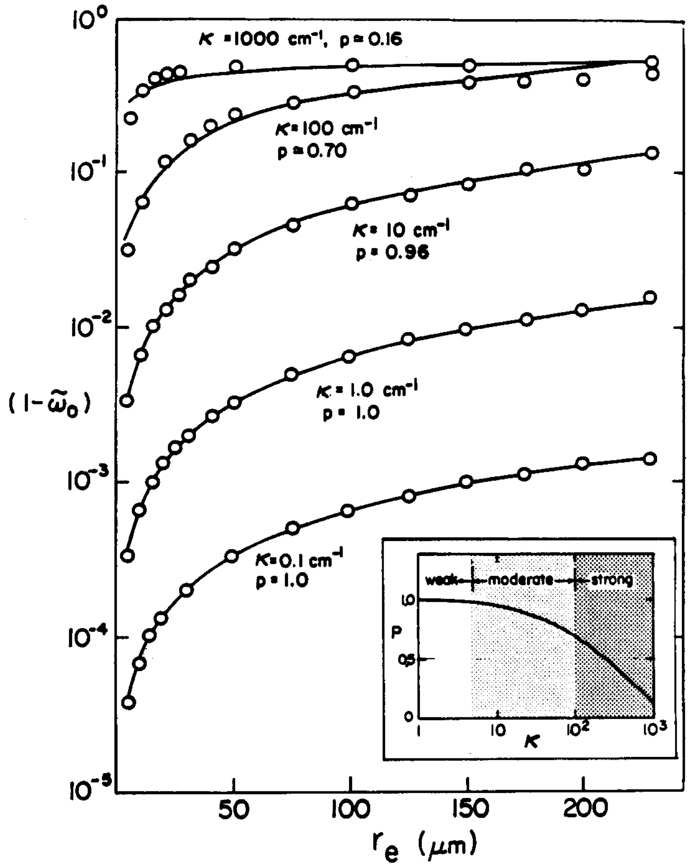

Figure 10. The single-scatter co-albedo as a function of effective radius for selected values of κ. The solid curves represent the relationship described by Eq. 18 for the values of p indicated and the open circles apply to van de Hulst's anomalous diffraction theory. The inset depicts the breakdown of the weak, moderate and strong absorption regimes.

(*b*) *Aerosol effects*

Danielson *et al.* (1969) provided one of the earliest investigations of the effects of aerosol on cloud absorption. This study was motivated by early observations of stratocumulus cloud albedo which were reported to be between 0·7 and 0·8. Assuming these clouds to be optically thick, one finds from the semi-infinite approximation that values of $1 - \omega_0$ of the order of 10^{-3} are required to match these observations. This is a factor of 10^4 larger than the co-albedo derived for pure water droplets at visible wavelengths. These arguments were used both by Danielson *et al.* (1969) and later by Chýlek *et al.* (1984) in their studies of aerosol effects on cloud albedo. Such arguments, however, are based on two false assumptions. The assumption that the clouds are semi-infinite is not correct as observations reported for similar clouds found the optical depths to be of the order of a few tens rather than a few hundreds. That the semi-infinite assumption is inappropriate is also apparent from the cloud transmission measurements also reported for these clouds. For example, values between 5% and 10% were reported by Foot (1988) and even larger values of transmission were measured by Stephens *et al.* (1978). The second limitation is that the clouds are assumed to be horizontally homogeneous. It is shown below that proper account of both finite vertical thickness and horizontal heterogeneity directly impacts on the hypothetical semi-infinite limits of cloud albedo and absorption.

Nevertheless, aerosol particles mixed in cloud can affect the cloud absorption in a number of different ways. For example, absorbing aerosol particles which do not serve as condensation nuclei but which remain interstitial to the cloud droplets can further add to droplet absorption. This mixture of aerosol and cloud droplets is referred to as an external mixture. Even though the contribution by these particles to the total volume extinction and volume scattering is small, the relative contribution to absorption can be large particularly in the visible spectral region where droplet absorption is normally negligible. This point is emphasized in Fig. 11 in which the ratio of absorption derived with and without aerosol is presented as a function of wavelength. These results were

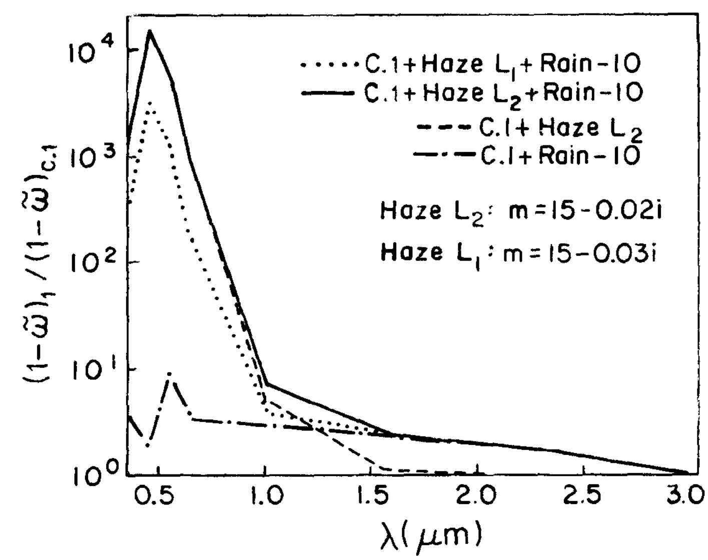

Figure 11. The ratio of particle absorption to droplet absorption, determined for four external mixtures of aerosols and cloud droplets (modelled by Haze-L_1 and Haze-L_2 distributions in combination with C1- and Rain-10 cloud distributions, after Newiger and Bähnke 1981).

taken from the work of Newiger and Bähnke (1981) who assumed two different values of refractive index for the aerosol and based these values on measurements of Fischer (1973) and others. The calculations were carried out for the size distributions indicated on the diagram. Details of these distributions can be found in Newiger and Bähnke (1981). These results point to the potential of aerosol particles for increasing the absorption of cloud predominantly at visible wavelengths.

The aerosol particles that might be contained within the cloud droplet itself can also exert an influence on both cloud albedo and absorption. In describing these effects, we consider two types of mixtures: aerosols that are totally soluble and therefore mix throughout the drop (volume mixture) and wettable but insoluble aerosols that are either in the form of a solid core surrounded by a shell of pure water (as considered for instance by Danielson *et al.* 1969) or aerosols that are distributed throughout the droplet (Chýlek *et al.* 1984). Both cases are referred to here as an internal mixture and are depicted in Figs. 12(a) and 12(b).

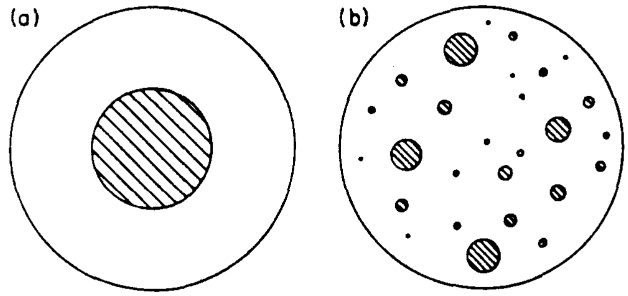

Figure 12. Two schematic examples of internal mixtures of water and graphitic carbon. In (a) the carbon forms a strongly absorbing solid core whereas in (b) the soot particles are randomly distributed throughout the volume of the drop (after Chýlek *et al.* 1984).

Internal mixtures of graphitic carbon in water droplets and the effects of these mixtures on cloud optical properties have been the subject of a number of studies. Danielson *et al.* (1969), for instance, conclude that for mixtures of the type illustrated in Fig. 12 (a), carbon nuclei that occupy about 10% of the droplet volume are required to obtain the single-scattering co-albedo of 10^{-3} which they claim is required to explain the observed cloud albedos. Chýlek *et al.* (1984), by contrast, proposed a mixed model of the type illustrated in Fig. 12 (b) in which carbon particles were considered to be randomly distributed throughout the droplet volume and obtained values of the visible co-albedo similar to those of Danielson *et al.*, but for two orders of magnitude less carbon. The specific absorption of a water–carbon internal mixture of the type modelled by Chýlek *et al.* is shown in Fig. 13 as a function of the graphitic carbon volume fraction V of the mixture. The calculations apply to a 5 μm radius water droplet in which either 0·3 μm or 0·5 μm radii carbon particles are randomly distributed within the droplet. The calculated particle absorptions are compared to the absorption of free carbon in air (solid, horizontal lines) and to mixed particles containing either a solid carbon core or shell. The results show that the absorption efficiency of randomly mixed carbon–water droplets is more than twice that of the same carbon particles in air (at least for $V < 10^{-3}$) and further that the random internal mixture is mostly more absorbing than either the shell or core models. The latter results however must be considered with some caution as the optical properties of a mixture of the type proposed by Chýlek *et al.* are determined by treating the particle as a homogeneous droplet possessing some single 'effective' refractive index which is some combination of the carbon and liquid water values. The results of this type of study therefore depend largely on how this refractive index is determined and there are significant problems with this approach (Bohren 1986).

Another study of the effects of irregularities in cloud droplets on the absorption can be found in the work of Twomey (1987). In that study, Twomey demonstrated that internal scattering within the droplet due to the presence of air bubbles acts to enhance the absorption by the droplet. However, no direct experimental confirmation or refutation of the predicted effects of internal scattering is presently available and no data are available which might indicate even the magnitude of internal scattering in atmospheric water clouds.

(c) *Water vapour absorption*

In addition to water droplet and aerosol absorption, the water vapour in the cloud volume also contributes to the total solar absorption. In fact, many of the early studies of cloud absorption assumed that the droplet contributions were negligibly small compared to the vapour absorption. However, this assumption has no foundation. Stephens

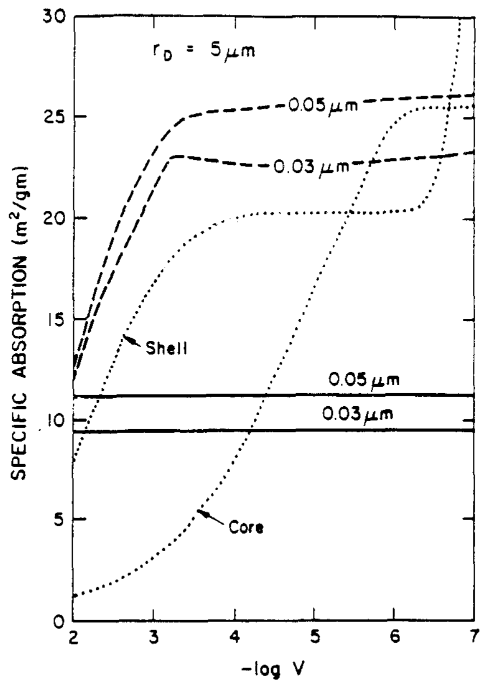

Figure 13. Specific absorption of water–graphitic carbon internal mixture with carbon randomly distributed throughout the volume of a droplet is more than a factor of two higher than the specific absorption of carbon in external mixture (solid lines). The radius of the water droplet is 5 μm. Two different radii of carbon particles (0·03 and 0·05 μm) are considerd. For comparison, the specific absorption of a water–graphitic carbon layered sphere is also shown (shell and core refers to the location of carbon). *V* is the graphitic carbon volume fraction of the mixture (after Chýlek *et al.* 1984).

(1978) showed that the contributions by droplets and vapour were typically of equal importance in defining the total cloud absorption. Estimating the relative contributions of water and vapour is, however, complicated since the absorption bands of liquid water largely overlap the molecular water vapour absorption bands. This feature complicates the calculations of both the total solar absorption in cloud and the relative contributions by the different water phases as it is necessary to determine the amount of solar radiation absorbed by water vapour in the clear sky above (and below) the cloud in order to account properly for the radiation available for absorption in the cloud. The complexity of cloud absorption is well illustrated in Fig. 14 from the work of Davies *et al.* (1984). The diagram shows the spectral distribution of vapour absorption within the cloud (solid curve), cloud droplet absorption (dashed curve) and the vapour absorption in the atmospheric column above the cloud (dotted curve). Vapour absorption is small at low wavenumbers (longer wavelengths) owing to the depletion of solar radiation above the cloud by the strong column vapour absorption. At larger wavenumbers where vapour absorption is weaker, this depletion is considerably reduced and the vapour absorption in the cloud is more comparable to the column absorption. Note also that the cloud droplet absorption is generally similar in magnitude to the vapour absorption although the relative contributions vary significantly with the type of atmosphere in which the cloud is imbedded (Slingo and Schrecker 1982).

One of the difficulties in treating the overlap between liquid and molecular absorption lies in the need to deal with the very different frequency dependences of liquid water and vapour absorption. The contribution by molecular absorption to the cloud optical

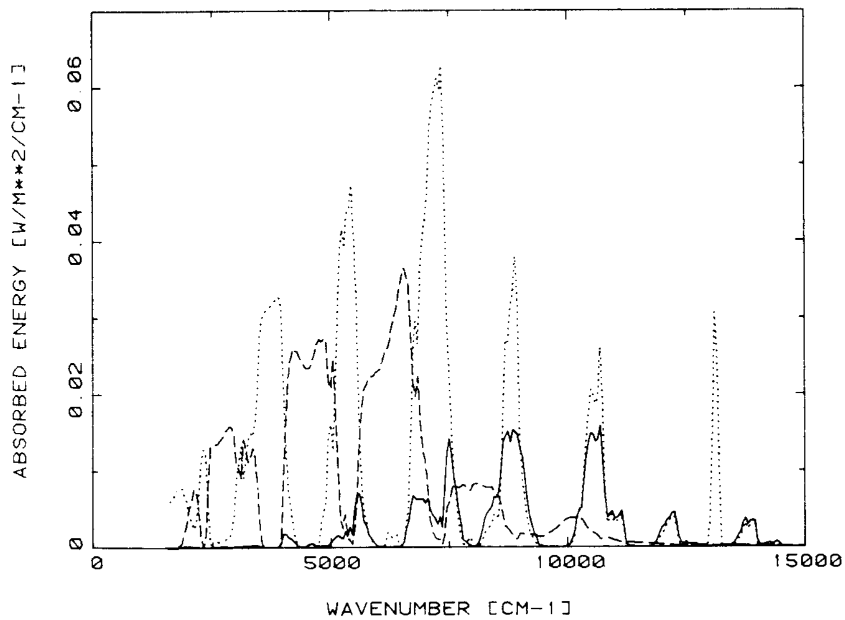

Figure 14. Spectral absorption at $50\,\text{cm}^{-1}$ resolution by cloud water vapour (solid), cloud droplets (dashed) and column vapour (dotted) typical of a 1 km stratus cloud with cloud top at 2 km above the ground illuminated by an overhead sun (after Davies *et al.* 1984).

thickness is a highly variable function of wavelength. The water vapour spectrum provided in this diagram is therefore highly smoothed and is by necessity an overly simple depiction of this wavelength structure. The actual way that molecular line absorption and its overlap with water absorption is treated in calculations is a complicated topic and has been reviewed by Stephens (1984).

5. Discussion

(*a*) *Cloud drop microphysics*

Wiscombe *et al.* (1984) argue that the existing microphysics probes flown on current research aircraft fail to detect the presence of large droplets which they claim are often present in real clouds. Based on earlier sensitivity studies of Welch *et al.* (1980), they postulated that a relatively small number of large drizzle sized droplets would enhance cloud absorption enough to account for the noted anomaly. After a more careful theoretical consideration, however, they were forced to conclude that the effect of large droplets on absorption was small. This conclusion was in direct contrast to the earlier results of Welch *et al.* (1980) and to the work of Twomey and Bohren (1980) which had demonstrated a much greater sensitivity of cloud absorption to droplet size.

We now attempt to clarify these results using the theoretical developments outlined above. For the sake of discussion, consider the case of a homogeneous cloud layer with a fixed liquid water content. The importance of the volume extinction α_{ext} and single scattering albedo $\tilde{\omega}_0$ to both cloud albedo and absorption was emphasized above. It was also shown using (15), under conditions of fixed liquid water content, that α_{ext} scales as the inverse of r_e whereas the relationship between single scattering albedo and r_e, according to the simplified expression (18), scales as some power of r_e, which in turn depends on the strength of absorption. The ultimate relationship between cloud albedo, absorption and r_e results from a combination of these two effects. Figures 15 (a) and (b)

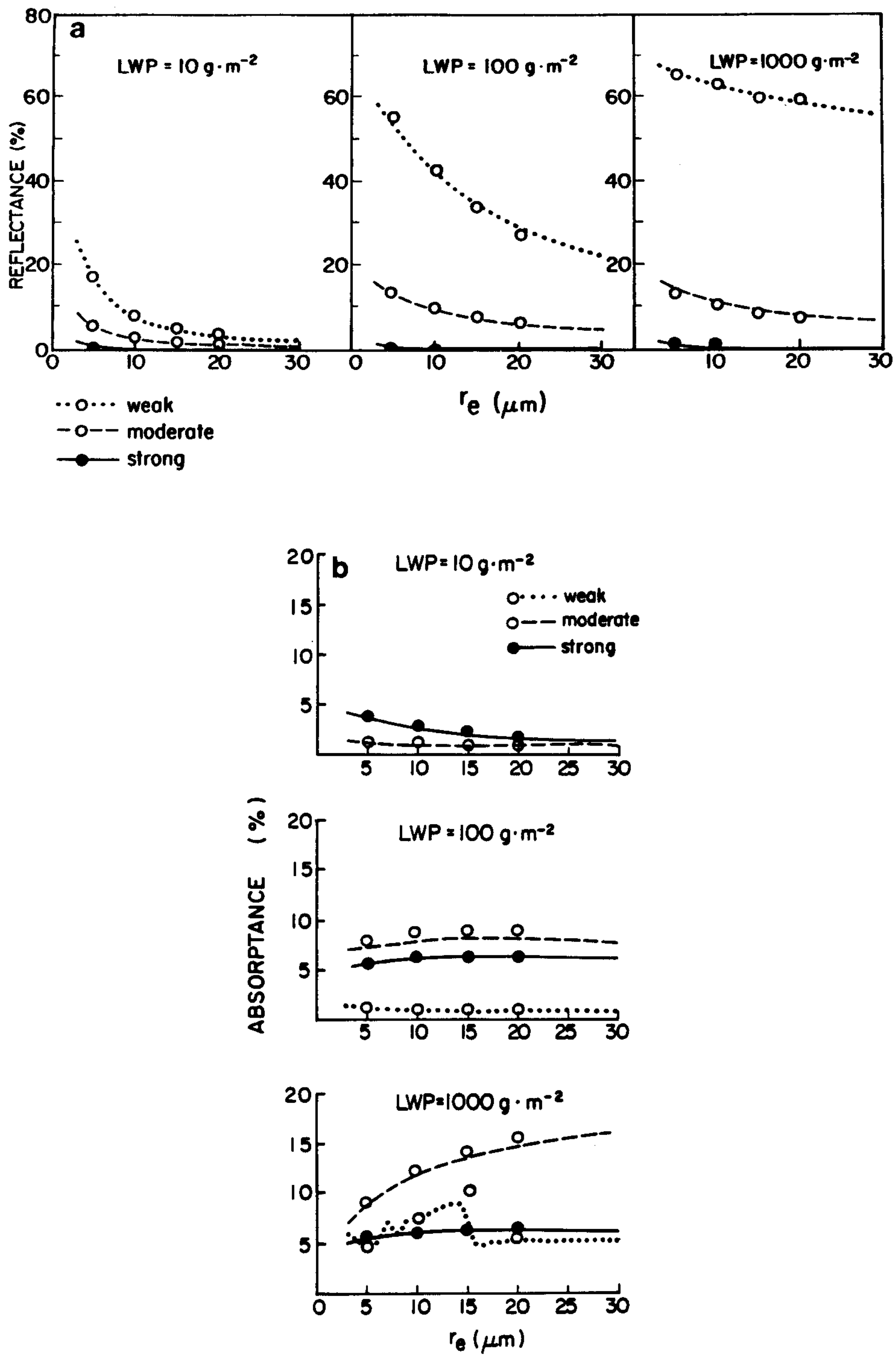

Figure 15. Contributions to (a) reflectance and (b) absorptance by the three absorption regimes as a function of r_e for cloud liquid water paths specified. The symbols refer to calculations performed using single-scattering properties determined from Mie theory (no significance should be attached to the difference between symbols) and the curves apply to calculations using the diffraction approximation to particle scattering assuming an overhead sun (after Ackerman and Stephens 1987).

provide an illustration of such relationships broken down into the three broadband regions determined according to the absorption criteria defined above. Consider the absorption relationships shown in Fig. 15(b) for discussion. According to (6*c*), the theoretical absorption in the thin limit (i.e. small liquid water paths such as indicated by LWP = 10 g m^{-2} case) is given directly by the product of α_{ext} and $1 - \tilde{\omega}_0$. Exploiting the r_e dependences of these parameters previously given by (15) and (18), it follows then, that the thin limit absorption scales as r_e^{p-1}. Since the absorption in this case is dominated by the strong regime for which $p < 0{\cdot}7$, it follows that cloud absorption is overall a *decreasing* function of *increasing* r_e. By contrast, the absorption in the semi-infinite limit represented here by the LWP = 1000 g m^{-2} example, varies more or less as $(1 - \tilde{\omega}_0)^{0{\cdot}4}$ (cf. Fig. 9) and therefore scales as r_e^l. The power $l \approx 0{\cdot}4 \approx 0{\cdot}3$ since absorption in this limit is dominated by the moderate regimes with $p > 0{\cdot}7$. Thus, and in contrast to the thin limit case, absorption in the semi-infinite limit increases with increasing r_e. This is similar to the result of Twomey and Bohren (1980) who used the more elegant theory of Chandrasekhar (1960) for isotropic scattering in a semi-infinite atmosphere. For intermediate ranges of LWP, and for ranges more typical of the clouds in which many of the absorption measurements were made, the relationship between droplet size and absorption lies somewhere between these two extremes and only very weakly depends on r_e.

The relative proportions of the strongly absorbed wavelengths to the moderately absorbed and weakly absorbed wavelengths varies according to the solar zenith angle. However, the results of Wiscombe and Welch (1986) imply that these proportions only change the arguments presented here significantly at sun angles much lower than that typical of the measurements reported. Based on the analysis given here, it thus seems that the existence of large droplets is unlikely to explain the majority of the observed anomalies. However, this conclusion neither diminishes the importance of large drops to cloud reflection (Fig. 15(a)) nor lessens the need for more comprehensive measurements of droplet size distributions.

(*b*) *The influence of aerosol on cloud absorption*

One of the first to recognize the potential effect of interstitial aerosol on cloud absorption was Twomey (1972). Using considerations of cloud physics, he argued that the bulk of the aerosol in cloud would be found between the droplets. Twomey then estimated the effects of this aerosol on the bulk cloud absorption by imbedding a small amount of absorbing aerosol of a specified optical thickness (τ_{abs}) in a hypothetical cloud layer composed of non-absorbing cloud droplets. His results are reproduced in Fig. 16 in which absorption is shown as a function of the cosine of the solar zenith angle. The dashed curve is the absorption of solar radiation by an aerosol layer in the absence of cloud, which increases as the solar zenith angle increases. With the addition of more cloud (i.e. increasing cloud optical thickness), the dependence of the cloud–aerosol layer absorption on solar zenith angle changes. This figure shows that the absorption in thick clouds ($\tau^* = 64$) increases with decreasing zenith angle in contrast to the aerosol-only case and the thin cloud behaviour lies somewhere between these two cases. For overhead sun, photons penetrate deeply into the cloud before undergoing an interaction with cloud droplets and the probability that they then undergo absorption before escaping back through the cloud top is also high. When the sun is near the horizon, the depth of penetration is not as great and photons can escape from the cloud top before being absorbed. Thus the absorption in thick cloud is maximum for high sun and smallest for low sun. For the thin cloud, the probability that vertically incident photons penetrate directly through the cloud before undergoing an absorption interaction is high, whereas

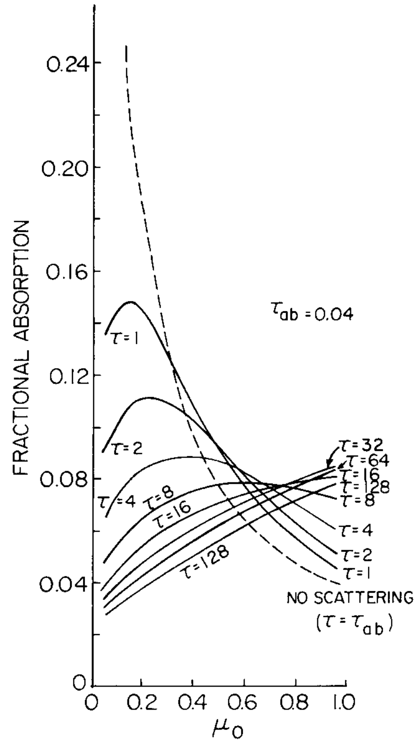

Figure 16. Absorption versus cosine of the solar zenith angle for an absorbing layer of optical thickness 0·04 externally mixed with non-absorbing scatterers of varying optical thickness τ. Curves are shown for τ = 0, 1, 2, 4, 8, 16, 32, 64 and 128 (after Twomey 1972).

the probability of absorption is greater for those photons that enter the cloud along an oblique path simply because the path through the cloud is longer. The end result, as shown in Fig. 16, is for an enhancement of the absorption by aerosol by, almost, a factor of two for thick cloud irradiated by sunlight at zenith angles less than about 60°. Twomey's results apply to the visible wavelengths at which water droplets are non-absorbing and highlight how the scattering between non-absorbing cloud droplets enhances absorption by interstitial absorbing aerosol.

Others such as Grassl (1975), Ackerman and Baker (1977) and Newiger and Bähnke (1981) also demonstrate how the presence of absorbing aerosol in cloud increases the bulk absorption of the cloud as a whole. However, significant increases only result when copious amounts of aerosol are assumed and many of the reported measurements were taken from clouds far from pollution sources. Furthermore, the spectral measurements of Twomey and Cocks (1982) and Stephens and Platt (1987) showed no indication of foreign absorption at shorter wavelengths and that the anomaly seemed to be mainly associated with the near-infrared wavelengths.

As mentioned previously, Chýlek *et al.* (1984) using refractive index mixing rules, demonstrated that significant absorptions could be produced by smaller amounts of internally mixed graphitic carbon compared to the external mixing studies described above. An example of their results for a C1-cloud model type are reproduced in Fig. 17 which presents reflection spectra for clouds composed of carbon–droplet mixtures for different fractional volume amounts of carbon. As expected, the most dramatic effects

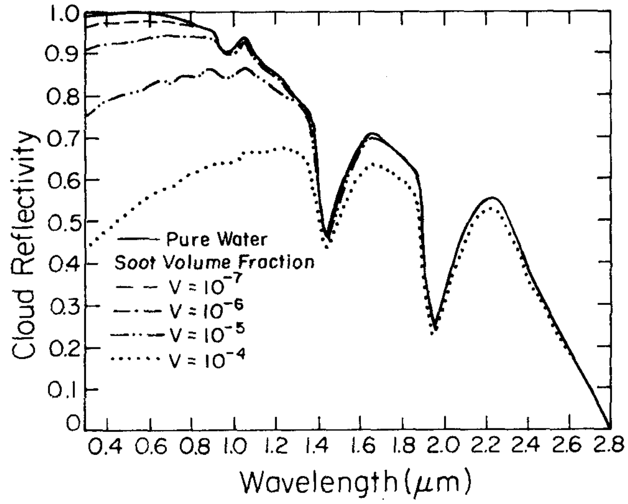

Figure 17. Spectral dependence of cloud reflectivity for different soot volume fractions internally mixed in C1-cloud drops. The effects of soot are most pronounced in the visible wavelengths. Because of the increasing absorption of water in the near-infrared, soot has negligible influence on the radiative properties in the near-infrared spectral region (after Chýlek *et al.* 1984).

of carbon on cloud reflection occur at visible wavelengths. Only relatively large volume fractions of carbon produce any significant reductions at the near-infrared wavelengths.

(c) *Water vapour absorption: a near-infrared continuum hypothesis*

The cloud absorption problem is not the only reported case of anomalous absorption of radiation in the atmosphere. In fact, more firmly established disparities between calculated and measured absorptions of the cloud-free atmosphere have been reported over a wide spectral region extending from the mid-infrared to the millimetre wave region. Like the cloud absorption anomaly, these observations tend to show observed absorptions in excess of theoretical prediction. The absorption excess at these longer wavelengths seems to possess the following general characteristics:

- the excess absorption is related to water vapour and has the nature of a continuum; that is, it is a slowly varying function of changing wavenumber,
- the absorption excess decreases with increasing temperature,
- it is greater for absorption in pure H_2O vapour (self broadening) than for absorption in a water-vapour–gas mixture (such as foreign broadened H_2O lines), and
- the relative discrepancy is greater in regions of weak absorption (such as in windows) than in regions of medium and strong absorptions.

The source of the discrepancy seems to lie in a poorly understood absorption mechanism which is postulated as either due to extreme wings of absorption lines of the H_2O molecule or due to H_2O molecule aggregates such as a dimer that consists of two water molecules bound together in some way (see Burch and Gryvnak 1980, for a more general review of this topic).

Perhaps the most celebrated absorption anomaly, at least from the meteorological perspective, is that found in the 8–13 μm region of the atmospheric absorption spectrum. Evidence for the enhanced absorption in this regime came from many different sources such as the early laboratory work of Burch (1970) and the measurements in the atmosphere reported by Cox (1969). (The reader is referred to the surveys of Roberts *et al.* (1976) and Burch and Gryvnak (1980) for more extensive discussions of the absorption in this spectral region.)

Measurements in the spectral regions that lie between the atmospheric windows at 100 GHz and 1000 GHz (i.e. 3·3 cm^{-1} to 33 cm^{-1}) and especially below 10 cm^{-1} have also identified absorption in excess of that predicted from line parameters (e.g. De Cosmo *et al.* (1983); Gebbie 1984). Figure 18 is introduced as a synthesis of various millimetre wave attenuation measurements (shown as symbols) along with spectra calculated with a simple continuum (curve A) included. It appears that the addition of a simple, empirical continuum is required to account for the observations obtained over a wide spectral region.

Continuum absorption has also been studied in other spectral regions, notably in the regions 333 cm^{-1} to 825 cm^{-1}, 1250 cm^{-1} to 2200 cm^{-1}, 2400 cm^{-1} to 2800 cm^{-1} (Burch and Gryvnak 1980) but, to the authors' knowledge, there are no measurements of water vapour continuum absorption in the near-infrared spectral regions at those wavelengths where discrepancies between observed and calculated cloud reflectances are greatest. It is a most attractive proposition to hypothesize that whatever causes the absorption anomalies at the longer wavelengths might also play a role in establishing the cloud reflection anomalies at near-infrared wavelengths.

Certainly the existence of such a continuum absorption in the near-infrared is likely but its strength is uncertain. In order to provide some indication of how such a continuum might affect both cloud reflection and absorption, a simple empirical continuum was superimposed on the water vapour band absorption spectrum. The object of such an exercise is not to provide a convincing proof of continuum effects on cloud absorption but to highlight yet another piece of the absorption puzzle that warrants further attention. The continuum devised in this study was increased systematically from zero at 0·7 μm to a maximum at 3·5 μm. The optical thickness associated with the continuum was derived in the way used by Kneizys *et al.* (1980) for the 3·3–4·2 μm spectral region. The optical thickness thus takes the form

$$\tau_{\text{cont}} = C_s \bar{u}$$

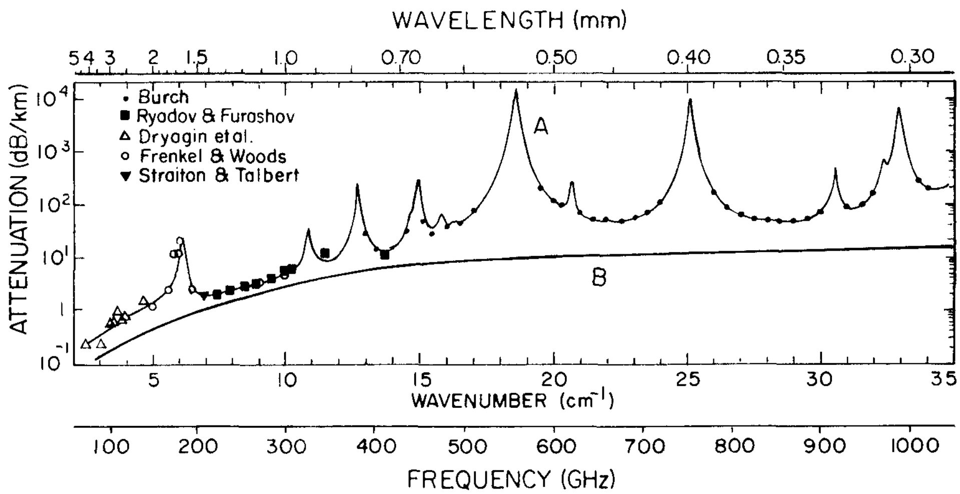

Figure 18. Spectral plots of the near-millimeter attenuation by atmospheric H_2O at sea level. H_2O content = 5·9 g m^{-3}. Curve A represents attenuation calculated by summing the theoretical contributions by all the lines and adding the continuum represented by curve B. The various points represent experimental data as reported by Burch and Gryvnak (1980).

where

$$\bar{u} = u\{e_{H_2O} + 0{\cdot}12(\bar{p} - e_{H_2O})\} \exp\left\{4{\cdot}56\left(\frac{296}{\bar{T}} - 1\right)\right\} \tag{19}$$

where u is the water vapour path (in g cm^{-2}), e_{H_2O} is the saturation vapour pressure associated with the temperature $\bar{T}$ and $\bar{p}$ is the total pressure. The spectral dependence of the absorption coefficient, C_s, was specified to be a linear function of wavelength for wavelengths greater than 0·7 μm. The form chosen for C_s is

$$C_s(\lambda) = 0{\cdot}04952\lambda - 0{\cdot}03467 \tag{20}$$

where C_s is in cm^2/(g atm) and λ is in μm. This formula corresponds to an average value of 0·156 cm^2/(g atm) at 3·85 μm which matches the values given by Kneizys *et al.* (1980) in LOWTRAN 5. The continuum absorption specified in this way also qualitatively agrees with the near-infrared continuum determined from overlapping the water vapour absorption lines compiled by AFGL (Geleyn, private communication).

Figure 19 illustrates the modification of the cloud reflection spectrum determined assuming only droplet absorption and scattering (bold line), the addition of water vapour line absorption to droplet effects (dashed line) and the addition of a continuum to water vapour line and droplet attenuation (fine line). The continuum absorption used in these calculations is increased tenfold from that given by (20) and the cloud parameters used in the calculations are indicated on the diagram. The added continuum has a significant impact on the spectral near-infrared reflectances in the windows. Table 3 lists values of the broadband near-infrared absorptions and albedos derived for clear-sky paths and for clouds with different combinations of droplet, line and continuum absorption. The

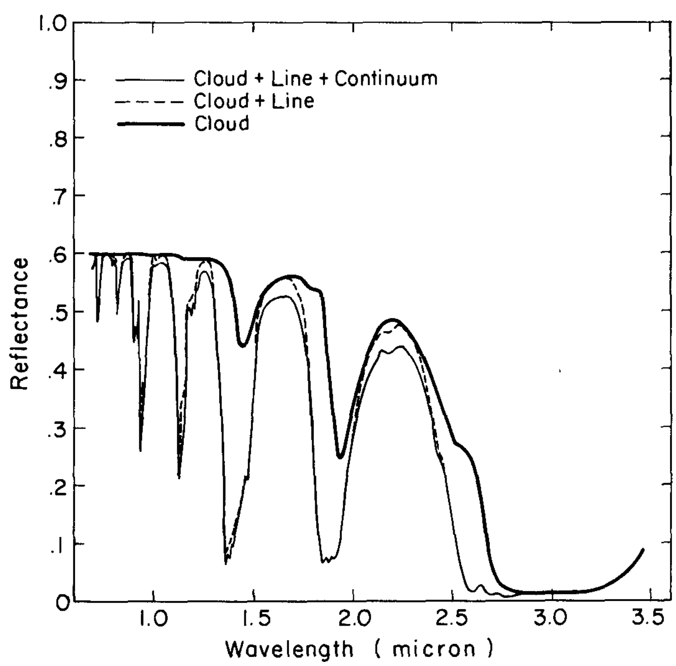

Figure 19. The near-infrared cloud reflection spectra derived for C1-cloud droplets and 1 km thick layer. Water vapour saturation is assumed for 273K. The spectra are shown for droplet scattering and absorption, droplet effects plus water vapour line absorption and for the addition of a continuum with an absorption strength increased tenfold over that given by equation (20). Note the effects in the windows centred at 1·6 μm and 2·2 μm.

TABLE 3(a). BROADBAND (0·7–5·0 μm) ABSORPTIONS OF FOUR HYPOTHETICAL TWO-LAYER ATMOSPHERES. THE BOTTOM LAYER IS A 1 Km THICK C1-CLOUD WITH VAPOUR SATURATION AT 273K (i.e. 0·485 g cm^{-2}). THE TOP LAYER CONTAINS WATER VAPOUR ONLY AND ABSORBER AMOUNTS ARE 0, 1, 2, AND 4 TIMES THE WATER VAPOUR IN CLOUD.

	F_c	Cloud	Cloud + Water Vapour	Cloud + Water Vapour + Continuum
Case 0	722 W m^{-2}	10·4%	20·2%	27·2%
Case 1	650 W m^{-2}	8·19%	15·3%	22·7%
Case 2	612 W m^{-2}	7·33%	13·1%	20·7%
Case 3	566 W m^{-2}	6·43%	10·8%	18·6%

Note F_c for fluxes at cloud top.

TABLE 3(b). AS IN TABLE 3(a), EXCEPT FOR REFLECTIVITIES.

	F_c	Cloud	Cloud + Water Vapour	Cloud + Water Vapour + Continuum
Case 0	722 W m^{-2}	54·0%	48·9%	45·3%
Case 1	650 W m^{-2}	55·3%	51·6%	47·8%
Case 2	612 W m^{-2}	55·7%	52·8%	48·9%
Case 3	566 W m^{-2}	56·2%	54·0%	50·0%

continuum used in this study produces a significant increase in the absorption (compare the values in the second and third columns of the Table) depending on the water vapour overburden. The four cases listed differ only in the vapour overburden assumed. The numbers listed to identify these cases correspond to 0, 1, 2 and 4 times the water vapour in the cloud layer. The effects of continuum absorption on the broadband near-infrared cloud albedo is also small despite the significant effects on spectral reflection in the windows as shown in Fig. 19.

6. THE INFLUENCE OF CLOUD HETEROGENEITIES ON CLOUD ABSORPTION

It was already mentioned above that one of the problems associated with the interpretation of the measurements of cloud absorption is the need to average data which contain large variabilities due to both the spatial and temporal fluctuations of clouds. Small biases introduced by inadequate sampling lead to spurious absorption estimates. As a result, it has been proposed that the discrepancies noted above occur largely either through inadequate sampling or by the overly simple treatment of heterogeneities in radiative transfer theory.

(a) *Cloud edge effects*

An obvious effect of cloud geometry on the transfer of solar radiation is the escape of solar energy through the sides of horizontally finite clouds in addition to the escape through the cloud base and cloud top. As a consequence, it is extremely difficult to characterize the total solar radiation budget of clouds with finite sides. In such cases, the absorption cannot be determined from the usual practice of measuring the solar fluxes at cloud top and base and taking differences of these fluxes. Absorptions derived in this way tend to be overestimated. In this case, the radiation that 'leaks' through the sides of the cloud is incorrectly interpreted as absorption within the cloud. Side effects were considered in the calculations of Welch *et al.* (1980) and by Newiger and Bähnke (1981) and in the analysis of the 1979 Summer Monsoon Experiment aircraft flux data by Ackerman and Cox (1981). Side effects were also dealt with in the analysis of Rawlins (1989) by considering the energy balance of the cloud in the form

$$\mathcal{A} + \mathcal{R} + \mathcal{T} + \mathcal{E} = 1 \tag{21}$$

where the $\mathscr{E}$ term is introduced to describe the net energy gain or loss through the cloud sides. If we define an apparent absorption as $\mathscr{A}' = 1 - \mathscr{R} - \mathscr{T}$, then the real absorption follows as

$$\mathscr{A} = \mathscr{A}' - \mathscr{E}. \tag{22}$$

Thus it only remains to estimate $\mathscr{E}$. One approach is to determine the magnitude of $\mathscr{E}$ for visible radiation assuming that the actual absorption at these wavelengths is negligible. The residual $\mathscr{A}'$ of visible radiation must then equal the side term $\mathscr{E}$. If it is assumed that this side term is the same in the near-infrared, then the cloud absorption follows from (22).

Some assessment of the magnitude of $\mathscr{E}$ is provided in Fig. 20 which is taken from the work of Newiger and Bähnke (1981) who performed radiative transfer calculations at the wavelength $\lambda = 0{\cdot}55\ \mu$m where droplet absorption is negligible. The results presented in this diagram shows the inferred absorption as a function of cloud horizontal extent for a 4 km thick cloud illuminated by a vertically incident solar source. As the horizontal extent of the cloud is systematically increased, then the inferred absorption decreases to the plane parallel value. Clearly the effect of radiative transfer through the sides of clouds, as emphasized by the dashed curve, assumes increasingly more importance with decreasing cloud horizontal extent. The earlier work of McKee and Cox (1974), Davies (1978) and the more recent work of Preisendorfer and Stephens (1984) demonstrated the theoretical importance of horizontal finiteness on radiative transfer through clouds.

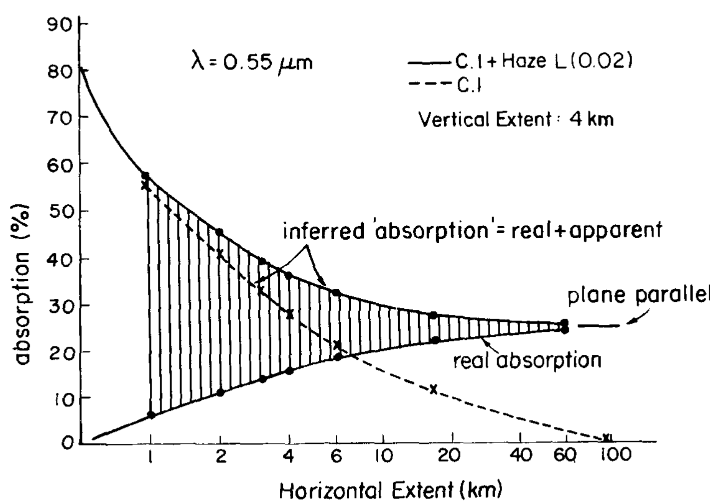

Figure 20. Real and apparent $0{\cdot}55\ \mu$m absorption as a function of the horizontal extent for two clouds of different composition and normal irradiance (after Newiger and Bähnke 1981).

(*b*) *The effects of spatial variations on absorption*

It has been suggested that the spatial and temporal variations of cloud influence the radiative transfer through them in such a way as to enhance the solar absorption in cloud. However, there is little evidence to support this suggestion when the hypothesis is tested using more sophisticated multidimensional radiative transfer models. Figure 21 is taken from Stephens (1988a) and shows the albedo and absorption averaged over a two-dimensional domain in which clouds were specified with geometries given by a simple Gaussian and harmonic functions. The calculated radiative properties for these hypothetical clouds are compared with the horizontally homogeneous cloud case. The results,

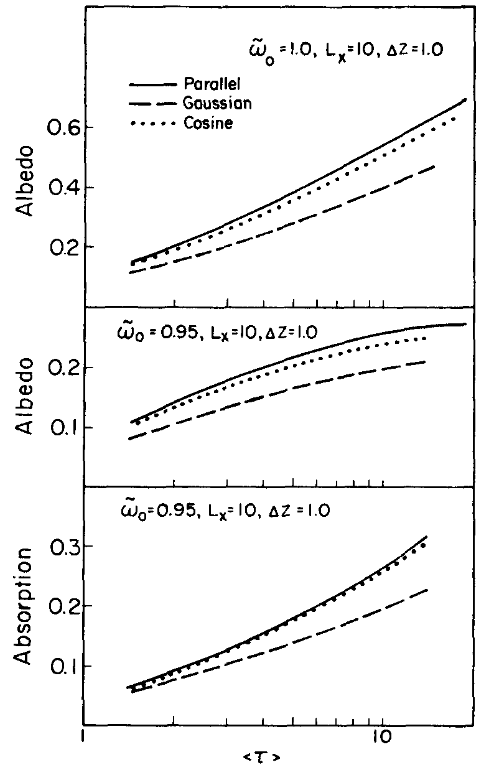

Figure 21. Albedo and absorptance as a function of area-averaged optical thickness, $\langle\tau\rangle$, for the plane parallel, cosine and Gaussian cloud distribution for the values of single-scatter albedo, horizontal scale of variability (L_x) and layer thickness Δz given (after Stephens 1988a).

presented as a function of the domain-averaged optical thickness, show that both the albedo and absorption of an inhomogeneous cloud distribution are systematically smaller than for the uniform cloud case for the same average optical thickness. The results further suggest that the departure from the homogeneous case depends not only on the averaged optical properties of the cloud but also on the nature of the cloud distribution. Taken at face value, these results do not support the contention that heterogeneities act in some way to enhance cloud absorption. This issue was taken up in the study of Stephens (1988b) who demonstrated theoretically the possibility of such an enhancement but was unable to establish clearly whether the properties of the heterogeneities needed to do so actually occur in real clouds. Nevertheless, the results do suggest that the heterogeneous clouds are less reflecting than equivalent plane parallel clouds. Whether this darkening effect is larger at absorbing near-infrared wavelengths than in the visible, which is explicit to the analyses decribed for edge effects, needs to be addressed in future research.

7. Summary and conclusions

This paper provides a review of the topic of solar radiation absorption in clouds. The far-reaching importance of the reported differences between theoretical and measured cloud absorptions to atmospheric research is discussed and the different explanations for the discrepancies are examined. The various types of observations which point to the anomaly are reviewed and it is concluded that the broadband absorption measurements, on the whole, remain inconclusive as they are difficult to make and suffer from large experimental uncertainty. Analyses of cloud reflection measurements, particularly near-infrared spectral reflectances, more convincingly point to discrepancies between theory and observations and we refer to this as a reflection darkening anomaly. Whether this darkening at these wavelengths is a result of enhanced cloud absorption or cloud transmission needs to be clarified.

A brief outline of the theory of solar radiative transfer in clouds is provided in order to establish both the importance of the attenuation coefficient and single-scatter albedo to this radiative transfer and to determine simple functional relationships between these cloud optical properties and the bulk absorption and reflection of clouds. Simple parametrizations of both the attenuation coefficient and single-scatter albedo are introduced in terms of cloud liquid water content, effective radius of the polydispersion and the bulk absorption coefficient of the droplets. In this way, the effect of droplet size and concentration on the cloud optical properties could be assessed as were the effects of both aerosol absorption and water vapour absorption.

Based on the theory outlined in this paper, it is demonstrated that the relationship between droplet size and cloud absorption is a complicated function of both absorption strength and optical thickness (or cloud liquid water path). This complex relationship is such that an increase in the size of cloud droplets in thin cloud actually reduces the cloud absorption whereas the reverse applies for optically thick (semi-infinite) clouds. Given the values of liquid water paths measured, it does not appear that the existence of undetected large droplets can explain the majority of the observed anomalies although the droplet size effects on cloud albedo might still be highly significant.

The effect of aerosol on cloud absorption and reflection is also reviewed and it is generally considered that the most dominant influence of these particles, whether externally or internally mixed with cloud droplets, is on the shorter visible wavelengths. While anomalous absorption in the visible cannot be strictly ruled out, the spectral reflectance measurements discussed in this paper generally do not seem to support this hypothesis. The possible effect of unaccounted-for water vapour absorption in cloud is also examined in the form of a near-infrared water vapour continuum. However, to explain the type of reflection and absorption anomalies described in the literature, a continuum effect substantially larger than that derived from simple line overlap would be required. The last of the explanations reviewed in this paper dealt with the effects of cloud heterogeneity on cloud absorption and reflection. The research which has focused on this topic, on the whole, seems to suggest that heterogeneous clouds are less reflecting than equivalent plane parallel clouds and that they are also less absorbing. Whether the reflection darkening effect by heterogeneities is larger for absorbing near-infrared wavelengths than for visible wavelengths has not been convincingly addressed.

At the time of writing this review article, no one explanation for the discrepancies between theory and measurement of solar radiative transfer in clouds has clearly emerged as more plausible than any other. Perhaps it is unlikely that there is one explanation for all the observed differences. The important issue here, however, is that the problem of the absorption anomaly has focused attention on the previous inadequacy of the observations and has highlighted the need for both more sophisticated instrumentation for measuring solar radiative transfer in clouds and an improved observing strategy.

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APPENDIX

For an absorbing cloud layer, $\alpha_{abs} > 0$ and $\tilde{\omega}_0 < 1$. Under these conditions, the general solution of (1) follows as

$$F^+(\tau) = C_+h_+e^{k\tau} + C_-h_-e^{-k\tau} + F_0Z_+e^{-\tau/\mu_0}$$
$$F^-(\tau) = C_+h_-e^{k\tau} + C_-h_+e^{-k\tau} + F_0Z_-e^{-\tau/\mu_0} \quad \text{(A1)}$$

where

$$Z_\pm(\mu_0) = \tilde{\omega}_0\left[\frac{\left(\frac{f_0}{b_0}\right)\tilde{\omega}_0 b + \left(\frac{b_0}{f_0}\right)\left\{D(1-\tilde{\omega}_0) + \tilde{\omega}_0 b \mp \frac{1}{\mu_0}\right\}}{k^2 - \left(\frac{1}{\mu_0}\right)^2}\right], \quad \text{(A2)}$$

$$k = [(1-\tilde{\omega}_0)D\{(1-\tilde{\omega}_0)D + 2\tilde{\omega}_0 b\}]^{1/2}, \quad \text{(A3)}$$

and

$$h_\pm = 1 \pm (1-\tilde{\omega}_0)D/k \quad \text{(A4)}$$

and where the $C_\pm$ represent the boundary conditions. The parameters b, b_0 and f_0 used here are functions of D, μ_0, g and $\tilde{\omega}_0$. The appropriate forms of these parameters generally depend on the problem at hand (cf. Meador and Weavor 1980; King and Harshvardhan 1986).

Consider an isolated cloud layer illuminated only by a collimated source of radiation and set the boundary fluxes as

$$F^-(z=0) = 0 \quad \text{(A5}a\text{)}$$
$$F^+(z=z^*) = 0 \quad \text{(A5}b\text{)}$$

where the level $z = z^*$ is used to denote the cloud base. With these boundary conditions in (A1), it follows that

$$C_- = \frac{F_0}{\Delta(\tau^*)}\{Z_+(\mu_0)h_-e^{-\tau^*/\mu_0} - Z_-(\mu_0)h_+e^{k\tau^*}\} \quad \text{(A6}a\text{)}$$

$$C_+ = \frac{-F_0}{\Delta(\tau^*)}\{Z_+(\mu_0)h_+e^{-\tau^*/\mu_0} - Z_-(\mu_0)h_-e^{-k\tau^*}\} \quad \text{(A6}b\text{)}$$

where we introduce

$$\Delta(\tau^*) = h_+^2e^{k\tau^*} - h_-^2e^{-k\tau^*} \quad \text{(A7)}$$

and use τ^* for the optical thickness of the cloud layer. For the example under consideration, the albedo of the cloud is

$$\mathcal{R} = \frac{F^+(0)}{\mu_0F_0} \quad \text{(A8}a\text{)}$$

and the transmission is

$$\mathcal{T} = \frac{F^-(z^*)}{\mu_0F_0} + \exp(-\tau^*/\mu_0). \quad \text{(A8}b\text{)}$$

These definitions together with (A2) and (A6) in (A1) give

$$\mathcal{R} = \frac{1}{\mu_0\Delta(\tau^*)}[Z_+(\mu_0)\{\Delta(\tau^*) - \Delta(0)e^{-\tau^*/\mu_0}\} - Z_-(\mu_0)h_+h_-(e^{k\tau^*} - e^{-k\tau^*})] \quad \text{(A9}a\text{)}$$

and

$$\mathcal{T} = \mathrm{e}^{-\tau^*/\mu_0} + \frac{1}{\mu_0\Delta(\tau^*)}[Z_-(\mu_0)\{\Delta(\tau^*)\mathrm{e}^{-\tau^*/\mu_0} - \Delta(0)\} - - Z_+(\mu_0)h_+h_-\mathrm{e}^{-\tau^*/\mu_0}(\mathrm{e}^{k\tau^*} - \mathrm{e}^{-k\tau^*})] \quad \text{(A9}\,b\text{)}$$

with cloud absorption defined as

$$\mathcal{A} = 1 - \mathcal{R} - \mathcal{T}. \quad \text{(A9}\,c\text{)}$$

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