# On the Possible Absorption of Visible Light by Clouds

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ABSTRACT

Danielson *et al.* have compared theoretical and experimental data on shortwave cloud albedos and have suggested that absorption by the cloud nuclei is responsible for the tendency of measured albedos to be lower than those based on theoretical predictions for a non-absorbing atmosphere. That suggestion is examined in the light of what is known about the size and composition of natural cloud nuclei but cannot be supported on these grounds.

## 1. Introduction

It is traditionally assumed that clouds are black (albedo, zero) for terrestrial radiation and white (albedo, unity) for solar radiation. However, with the advent of numerical computations of multiple scattering some element of uncertainty has appeared—computed albedos for a conservative scattering cloud become greater than 0.80 for optical thicknesses $\tau \gtrsim 50$ and exceed 0.90 when $\tau$ exceeds a hundred or so. These figures are not at all sensitive to the scattering diagram (phase function) of the individual droplets. Measured albedos, however, do not exceed $\sim$0.80, yet an optical thickness of 100 does not imply a great linear depth and should occur frequently; under typical conditions a stratocumulus cloud layer 1 km thick will have an optical thickness of 30–100. It is therefore surprising that some measured albedos have not exceeded 0.8.

Quite a small degree of absorption at each scattering event would suffice to bring the albedo down to $\leqslant 0.8$ for all layer thicknesses, because of the large number of scatterings involved—the absorption of one-thousandth of the scattered light, i.e., a single scattering albedo $\tilde{\omega}$ of 0.999, would be enough. Danielson *et al.* (1969) have proposed that the nuclei upon which the cloud droplets are formed give rise to this degree of absorption with a loss per scattering $(1-\tilde{\omega})$ of $\sim 10^{-3}$, and therefore a limit of about 0.8 to the cloud albedo even for the deepest layers.

The consequences of an absorption by optically deep clouds of 20% of the incident solar radiation could be quite far-reaching (especially since only quite moderate physical thicknesses correspond to large optical depths). A cloud with a typical extinction coefficient of $10^{-3}$ cm$^{-1}$ and a single scattering albedo of 0.999 would have an absorption coefficient of $10^{-6}$ cm$^{-1}$; it would therefore absorb $2\times10^{-6}$ cal cm$^{-3}$ min$^{-1}$ from an incident flux of 2 cal cm$^{-2}$ min$^{-1}$. For comparison, it may be noted that the latent heat stored in the cloud liquid water would be of the order of $2\times10^{-4}$ cal cm$^{-3}$. In terms of heating, $2\times10^{-6}$ cal cm$^{-3}$ min$^{-1}$ would be equivalent to several degrees per day.

In view of these consequences it seems desirable to examine further the suggestion that the nuclei can give rise to significant absorption.

## 2. Size and composition of cloud nuclei

To initiate condensation at supersaturations of a few tenths of one per cent, such as exist in natural clouds, a particle must, from nucleation theory (see, for example, Fletcher, 1962), be either 1) water-soluble and a few hundredths of a micron in radius, or 2) insoluble and wettable and a few tenths of a micron in radius.

The sort of nucleus invisaged by Danielson *et al.*—black, of the order of 0.5$\mu$ radius—would fall in the latter category; however, there is considerable experimental support for the contention that the vast majority of natural cloud nuclei are in the former category, i.e., water-soluble particles a few hundredths of a micron in radius. Diffusion coefficient measurements (Twomey, 1965) suggest a nucleus radius of the order of $3\times10^{-6}$ cm, while experiments on the volatility of the nuclei at elevated temperatures (Twomey, 1968) show that they are relatively volatile, most probably ammonium sulfate or a similar material. Furthermore, when droplets are grown on natural nuclei at cloud-like supersaturation in a diffusion cloud chamber, it is noteworthy that the nuclei themselves, before deoplets form on them, are not detectable by the eye or by photography, either at 90° or at 20–30°, in the intense illuminating beam ($\sim$500 lm cm$^{-2}$) used to make the droplets visible and photographically recordable. From the known parameters of the illuminating system, combined with Mie theory and tests on artificial point sources, the minimum detectable size can be estimated quite closely at 0.6–0.7$\mu$ for 90° viewing and 0.2$\mu$ for 20–30° viewing; one can therefore set an upper limit of 0.2$\mu$ for nucleus radius from these considerations alone. If the nuclei were insoluble, it would require a rather implausible combination of circumstances to reconcile a radius $\leqslant 0.2\mu$ with the ability to nucleate at fractional supersaturations

condensation must have been of that size. The weaknesses in this argument are several; it would be relevant only if the atmospheric aerosol was uniform in composition, and it furthermore assumes that the Frankfurt surface air would, if it took part in cloud condensation, give only a hundred or so droplets per cubic centimeter.

## 3. Discussion

droplets.

The apparent disparity between theory and absorption still remains to be explained. It may not, however, be a real disparity, since the measurements are few and can be criticized, especially as broad-band detectors were used. It would seem most desirable for further and more precise measurements of cloud albedo to be undertaken to find out if natural cloud albedos ever approach unity, as theory would demand in the conservative case.

—a perfectly wetted particle or even a water droplet 0.2$\mu$ in radius cannot nucleate condensation below about one-half of one percent supersaturation, so the particles would need to be wettable to a high degree and just below 0.2$\mu$. Soluble particles, on the other hand, can be considerably smaller and still nucleate condensation at a few tenths of one percent (because of vapor pressure lowering by the dissolved nucleus).

There is, therefore, little support for the assumption by Danielson *et al.* of absorbing nuclei of the order of 0.5$\mu$ radius. Their reference in support of this size was Byers (1965), whose sole grounds for the suggested size was the fact that the atmospheric aerosol (sampled by Junge at Frankfurt) contained a hundred or so particles per cubic centimeter greater than about half a micron; since this number is similar to cloud droplet concentrations it was suggested that the nuclei of cloud condensation must have been of that size. The weaknesses in this argument are several; it would be relevant only if the atmospheric aerosol was uniform in composition, and it furthermore assumes that the Frankfurt surface air would, if it took part in cloud condensation, give only a hundred or so droplets per cubic centimeter.

## 3. Discussion

If an absorption of 20% of the incident flux does, in fact, take place, an explanation other than that proposed by Danielson *et al.* seems to be required. If the absorption took place within the liquid droplet, the single-scattering albedo of 0.999 would imply an absorption coefficient for the liquid of around 1 $cm^{-1}$ for 5$\mu$ radius droplets. The presence of dissolved material at a concentration level of perhaps one part per million would certainly not give such a degree of absorption.

Apart from the soluble nucleus upon which cloud condensation occurs one would expect the droplets to contain other aerosol particles which coagulate either with the nucleus or with the droplets after cloud formation. However, using Fuch's (1964) coagulation coefficient values, an aerosol with $10^4$ particles $cm^{-3}$ following a Junge distribution down to $10^{-6}$ cm was calculated to diffuse to a 5$\mu$ radius cloud drop only at the rate $3\times10^{-20}$ $cm^3$ $sec^{-1}$; to collect material equivalent to a single 0.5$\mu$ radius particle a time of the order of a few months would therefore be required. The coagulation rate with the dry nucleus would, of course, be much smaller.

The possibility that "diffusiophoresis" in the vapor pressure gradients about growing cloud droplets causes a sufficient accumulation of particles must be discounted on the basis of the conclusions by Goldsmith *et al.* (1963) as to the magnitude of this effect.

## 4. Conclusions

Present knowledge of atmospheric particles and cloud condensation does not lend support to the suggestion by Danielson *et al.* that up to some 20% absorption of visible light is caused by the nuclei within the cloud droplets.

The apparent disparity between theory and absorption still remains to be explained. It may not, however, be a real disparity, since the measurements are few and can be criticized, especially as broad-band detectors were used. It would seem most desirable for further and more precise measurements of cloud albedo to be undertaken to find out if natural cloud albedos ever approach unity, as theory would demand in the conservative case.

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