Extinction efficiency in the infrared (2–18 μm) of laboratory ice clouds: observations of scattering minima in the Christiansen bands of ice

W. Patrick Arnott, Ya Y. Dong, and John Hallett

Extinction measurements with a laser diode 0.685 μm and a Fourier transform infrared spectrometer 2–18 μm were performed on laboratory ice clouds 5 μm ≤ D ≤ 70 μm grown at a variety of temperatures, and thus at a variety of crystal habits and average projected crystal area. Ice clouds were grown by nucleation of a supercooled water droplet cloud with a rod cooled with liquid nitrogen. The ice crystals observed were mainly plates and dendrites at the coldest temperatures (−15 °C) and were mainly columns and needles at warmer temperatures (−5 °C). The crystals were imaged with both a novel microscope equipped with a video camera and a heated glass slide and a continuously running Formvar replicator. The IR spectral optical-depth measurements reveal a narrow 0.5-μm-width extinction minimum at 2.84 μm and a wider 3-μm-width minimum at 10.5 μm. These partial windows are associated with wavelengths where the real part of the index of refraction for bulk ice has a relative minimum so that extinction is primarily due to absorption rather than scattering, i.e., the Christiansen effect.

Introduction

Understanding radiative transfer in a cloudy atmosphere calls for knowledge of ice-crystal single-scattering properties. Applications of this knowledge include climate modeling; passive2–5 and active remote sensing of cirrus clouds with ground and satellite observation of cloud emission, extinction, and backscatter; and target detection when cirrus clouds are present.6 Extinction of light by small ice crystals D < 50 μm, for example in cirrus clouds is related mainly to crystal projected area in the visible but deviates from this relation in the IR. However, snow crystals and snowflakes are at the very large end of the particle size spectrum, where D = 100–10000 μm, so that extinction is determined mainly by the average projected area of the crystal.7–9 Theoretical results give us clue to the numbers of small ice crystals that are necessary to influence the radiative properties of cirrus clouds.10 In situ cirrus sampling with research aircraft equipped with a Formvar ice-crystal replicator indicates that the number of naturally occurring small ice crystals is sufficiently high that these crystals can contribute significantly to cloud radiative properties11 such as solar extinction and IR emission. Ray-tracing algorithms, which are most useful in the visible, have been developed for modeling of single-scattering properties of ice crystals when the size parameter $x = \pi D/\lambda$, D is the crystal maximum dimension, λ is the vacuum wavelength is >30.1,12,13 An algorithm for size parameter ≤10 is the discrete dipole approximation.14 Laboratory measurements of well-characterized ice clouds are useful for understanding the single-scattering properties of cirrus clouds that contain small ice crystals and for providing data against which algorithms for IR single-scattering properties can be tested.

Phase function measurements in laboratory ice clouds have been performed previously for the 0.633-μm and 10.6-μm CO₂ laser wavelengths15 see also Refs. 12 and 16 for additional 0.633-μm and
0.550-µm measurements, respectively. Measurements at the same size parameter showed that the phase functions were similar in the near-forward-scattering direction on account of the diffraction component but that the phase function in the IR was significantly less than that in the visible at scattering angles $>90^\circ$.\textsuperscript{15} For the purposes of furthering the discussion of previous studies and introducing the topic of this paper, we introduce $\sigma_{10.6}$ and $\sigma_{0.633}$ as the laboratory cloud-extinction coefficients in units of inverse meters for $\lambda = 10.6$ and 0.633 µm wavelengths, respectively. If we assume single scattering, we may define a size-distribution-averaged IR extinction efficiency as $Q_{\text{ext,IR}}(\lambda = 10.6) = 2\sigma_{10.6}/\sigma_{0.633}$, where $\sigma_{0.633}$ is a measure of the projected area of crystals in the cloud. The limiting value for a very large size parameter is $Q_{\text{ext,IR}}(\lambda = 10.6) = 2$, with half this value from refraction and half from the combination of refraction and absorption. Measurements\textsuperscript{15} indicated that $Q_{\text{ext,IR}}(\lambda = 10.6) = 0.94$ for "mature laboratory ice clouds." Measurements of $Q_{\text{ext,IR}}(\lambda)$ for $\lambda = 2$−18 µm for laboratory ice clouds grown at a variety of cloud temperatures and also at a variety of crystal concentrations and habits are reported in this paper.

2. Extinction Measurements

A. Cloud Chamber and Optical Arrangement

The experimental arrangement is shown in Fig. 1. The cloud region is the interior of a cold box measuring 1 m high $\times$ 1.2 m $\times$ 1.2 m. A precooling chamber, a vertical tube with antifreeze circulation around the outside jacket, with a diameter of 15 cm and a height of 92 cm sits above the cold box to precool the water droplets (D = 9 µm $\pm$ 3) from two ultrasonic nebulizers. The mist, which was precooled to $-3^\circ$ C in the precooling chamber and gradually reached the cold-box air temperature, is warmer than the interior, becomes stably stratified, and occupies the space at the chamber top. It is noted that the ice cloud grew in a region of temperature gradient, with the temperature at the cold-box top $\approx -3^\circ$ C and a lower temperature 15 cm below the top. Cloud temperatures in this paper refer to a measurement taken 15 cm below the cold-box top. The cold-box temperature within 10 cm of the bottom was typically $2^\circ$ C colder than the reported cloud temperature. Ice-cloud growth begins when the cloud is homogeneously nucleated with a wire rod [length 1 m, diameter 3 mm] cooled with liquid nitrogen. Crystals grow at the expense of water vapor supplied by the supercooled drops and fall through the Fourier transform infrared spectrometer (FTIR) and laser beams. Measurements were performed between cold-box midpoint temperatures of $-21$ to $-5^\circ$ C so that a range of crystal habits and average crystal dimensions were covered.

A thin sheet of plastic wrap (generic supermarket brand had less absorption than both more expensive brands and a Mylar sheet serves as a window to the cold-box interior for both IR and visible, and although the wrap was a source of heat transfer, it was needed to keep the cold air from falling out of the box. The window was heated from the outside with a warm air stream to prevent condensation. A curved mirror of focal length 152.4 cm and diameter 15.2 cm reflected enough visible and IR energy. The IR source and detector and the laser diode were all at distances from the mirror that were approximately equal to twice the focal length so that the IR beam diameter was approximately the same at the source and the detector. The source and detector input-optics areas are nearly equal so that the entire unscattered beam is largely collected by the detector. A resistance heater was placed on the back of the mirror to prevent condensation. On reflection by the mirror, IR and laser beams exited the cold box and traversed a distance (specified below) to their detectors. The angles between the incoming and the outgoing beams for laser and IR beams were 2.2°, and 2.8°, respectively, so that actual in-cloud path lengths for both the IR and the visible were well approximated as 228 cm. A somewhat similar arrangement was used to view the halomeration in artificial ice clouds.\textsuperscript{17}

The FTIR Bomem manufacture consists of a hot source followed by a scanning Michelson interferometer and a ZnSe output window. The modulated beam has an 80-mr full maximum angle divergence and a maximum beam diameter of 4.5 cm at the output window. The IR detector (mercury cadmium telluride type) was mounted off the right side and 12 cm in front of the IR source. Operation of the spectrometer is controlled by computer that receives the detector signal and performs the necessary Fourier transform of the spatial interferogram into the IR wavelength spectrum. The mirror-to-FTIR source-window distance was 302 cm. Typically 30 scans were averaged over a period of 42 s, and the resolution in wave numbers was 4 cm$^{-1}$.

The visible beam of wavelength 0.685 µm had an estimated net output power of 5 mW. It was from a laser diode collimated with a spherical ball lens to have a beam divergence of 3 mrad and a minimum beam width of 3.2 mm. The output power was monitored with the photodiode supplied in the laser diode package. The laser detector consisted of an integrating sphere with an input port diameter of 3.8 cm located a
distance of 396 cm from the mirror. The entire unscattered beam is collected by the integrating sphere. A tube with a blackened inner portion was mounted perpendicular to the integrating-sphere port to prevent stray light from entering the sphere. A phototransistor was placed on another port of the integrating sphere. Signals from both the phototransistor and the photodiode on the laser diode were measured with a strip chart recorder. The space between the laser diode and the mirror was 290 cm.

The typical procedure for extinction measurements was to take a reference measurement in the IR and the visible just before adding water drops to the precooling chamber. After the visible optical depth started increasing when the ice cloud grew, simultaneous transmission measurements, equal to the cloud spectrum divided by the reference spectrum, were taken in the visible and the IR. The basic assumption is that the observed extinction is entirely due to the presence of ice crystals between the radiation sources and detectors. The purpose of the reference spectra is to remove the effects of source-frequency response, gaseous absorption, transmission losses in the entrance window, and reflection losses at the mirror. It is assumed that the relative humidity of ambient air at the mirror level is at ice saturation both before and during the typical 7-min cloud life cycle.

Denote by $T_{\text{vis}}$ and $T_{\text{IR}}$ the transmission measurements at 0.685 µm and IR, respectively. The optical depth was estimated from the relation

$$\tau = -\ln(T).$$

The fractional error in optical-depth measurements because of light scattered into the finite collection angle of the detector was estimated to be less than 1% for the 0.685-µm wavelength and less yet for the IR. Although snow is well known to scatter by diffraction a nonnegligible amount of energy into typical detectors, the fractional amount of light scattered into typical detectors is much less for the comparatively much smaller particle laboratory ice-cloud scatter. The fractional error in underestimation of extinction measurements scales as the product of single-scatter albedo $D^{-2}\lambda^{-2}$ so that IR measurements are less error prone than are visible measurements for comparable source and detector dimensions.

The visible optical depth $\tau_{\text{vis}}$ during a typical 7-min cloud lifetime is shown in Fig. 2. The vertical bars in Fig. 2 are separated by the time interval necessary for one complete set of 30 FTIR scans. Typically $\tau_{\text{vis}}$ increased to a maximum value and decayed more slowly. Single-scattering contributions were sufficient to explain the main features of the extinction measurements in snow for optical depths less than unity. Reference 8 also shows the calculated effect of multiple scattering on a 0.633-µm laser beam captured completely by the detector. Figure 13 of Ref. 8 shows that the apparent extinction derived from the laser beam transmittance is only approximately 3% less than the single-scattered extinction (i.e., the true extinction coefficient), a result that is valid for optical depths up to approximately 6, taking into account all orders of multiple scattering. The contribution of multiple scattering to the measurements reported here at a similar wavelength of 0.685 µm should be even less than 3% because the laboratory ice crystals are much smaller than snowflakes and thus have wider average scattering angles.

B. Ice-Crystal Observation

Two cloud probes were used to size and count cloud crystals, a continuous-running formvar replicator and a newly developed cloud scope. The formvar replicator has a sampling arm that protrudes 40 cm into the cold box. It is usually deployed on a wing-mounted pod of a research aircraft for in situ sampling of cloud particles and is a descendant of other replicators developed at the Desert Research Institute. The arm tip was typically 10 cm to the side of, and 10 cm below, the path of IR and laser beams in the cold box. Film (16-mm width) rolls out onto the arm and is coated with a liquid Formvar–chloroform solution at a distance $L = 4$ cm before the arm tip. Ice crystals fall onto the liquid plastic, which envelopes them. When the crystals evaporate, a cast or crystal replica is left behind on the now-hardened plastic film (Fig. 3). The replicator was used only to obtain an estimate of the number of crystals per volume. Denote by $D$ the diameter of a circle that just fits around the hexagonal perimeter of an ice crystal, and denote by $H$ the crystal length parallel to the $c$ axis. The number of crystals per unit volume in a particular size interval, denoted by $N(D)$, can be estimated from replicator data:

$$N(D) = \text{film speed} \frac{1}{\nu(D)} \frac{1}{L} \frac{\#\text{particles}}{\text{area of film}},$$

where the film speed is 1 cm/s, $L$ is the length of replicator arm where the Formvar solution is exposed.
to crystals and \( v_c(D) = 2.96 \times 10^{-2}D^{0.824} \) cgs units is a computed crystal fall speed for plate-type crystals.\(^{19}\) We estimated the number of particles per area of film by taking microscope photos and projecting the negatives with an enlarger onto a white surface, where counts and sizing were performed. In Eq. 2 it is apparent that larger crystals have a greater sample volume than the smaller crystals. The replicator-size-dependent collection efficiency was estimated to cut off at approximately 7 µm (see Fig. 4) and could have been increased by the use of an air stream to carry crystals to the Formvar more effectively than does the natural falling motion. Spikes in the data shown in Fig. 4 are likely due to undersampling, as only 132 crystals were counted for the distribution.

If we assume single scattering, the visible optical depth for randomly oriented hexagonal crystals is\(^{3}\)

\[
\tau_{\text{vis}} = 2z \int_{D_{\text{min}}}^{D_{\text{max}}} \frac{3}{4} \left( \frac{\sqrt{3}}{4} D + H(D) \right) \frac{N(D) dD}{D^2},
\]

where the factor of 2 is the extinction efficiency in the geometric optics limit (i.e., particle dimensions are much greater than the wavelength); \( z = 228 \) cm is the total beam path length in the cloud; \( D_{\text{min}} \) and \( D_{\text{max}} \) are the smallest and largest crystals in the cloud, respectively; and the first term in the integral is the average projected area of a randomly oriented hexagonal crystal. The integral can be converted to a sum for use with experimental data. Using

\[
H(D) = 1.41 \times 10^{-2}D^{0.474}
\]

as a relation for crystal geometry\(^{20}\) for \( 10 \mu m \leq D \leq 3000 \mu m \), and the experimental data shown in Fig. 4, gives us \( \tau_{\text{vis}} = 0.99 \). This value of \( \tau_{\text{vis}} \) corresponds to time near the peak optical depth in Fig. 2, which is the region of replicator data studied. The average crystal D was \( D = 29.5 \) µm, and the total number of crystals per unit volume summed over all sizes was \( N_{\text{total}} = 420 \) crystals per cm\(^3\) or \( \approx 1 \) crystal per 2 mm\(^3\).

The instrument shown schematically in Fig. 5 is called the cloud scope and is a video microscope system, with the small microscope plate heated to sublime or melt incoming ice crystals after their images have been recorded. Crystals are drawn into the funnel (Fig. 5b) by the suction blower, and some portion of the crystals are collected in the area on the microscope slide imaged by the objective. A heater wire is used to prevent condensation on the objective and to heat the microscope slide so that the rate of

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**Fig. 3.** Photographs of ice crystals collected with the Formvar technique.

**Fig. 4.** Number concentration computed from the replicator data, plate crystals, for a cold-box midpoint temperature of \(-15^\circ\)C.

**Fig. 5.** Cloud-scope schematic: (a) overall, (b) detailed.
Crystals grew in a region of strong temperature gradient where, for example, the temperature at the top of the cold box in Fig. 1 was $-3^\circ C$ and 15 cm below the cold box top the temperature was $-21^\circ C$. Sublimation can be used to estimate crystal mass. The dimensions of the microscope slide are $\sim 2.5$ cm $\times$ 1 cm. A thermistor is attached to the slide to monitor temperature so that a slide temperature of $\sim -0.5^\circ C$ was attained. The image from the objective is reflected by the mirror into the direction of the eyepiece. A lensless video camera (not shown) is in close contact with the eyepiece for recording of crystal images. The flow velocity at the funnel entrance was estimated to be $\sim 2$ cm s$^{-1}$. The operator pulses the blower until a number of ice crystals have landed on the slide. With the video recorder running, crystal images are stored during the $\sim 2$ s less for crystals smaller than $\sim 20$ µm necessary for sublimation. This process is continued during the cloud life cycle, usually yielding 100–300 crystal images. Figure 6 shows a dendrite, which was one of the largest crystals imaged during the measurements, at a cold-box midpoint air temperature of $-21^\circ C$, and other crystals in varying evaporating and melting stages. We measure D for platelike crystals and estimate H from the power-law relation, Eq. 4, but can measure both D and H for columnlike crystals because they usually land with the crystal c axis parallel to the microscope plate. All number concentrations reported here (except for Fig. 4) were obtained with manual frame-by-frame analysis of cloud-scope data.

The dimensions of the microscope slide are $1$ cm $\times$ 1 cm. A thermistor is attached to the slide. With the video recorder running, crystal images during the measurements, at a cold-box midpoint air temperature of $2-0.5^\circ C$ was attained. The image from the objective is reflected by the mirror into the direction of the eyepiece. A lensless video camera in close contact with the eyepiece for recording of crystal images. The flow velocity at the funnel entrance was estimated to be $\sim 2$ cm s$^{-1}$. The operator pulses the blower until a number of ice crystals have landed on the slide. With the video recorder running, crystal images are stored during the $\sim 2$ s less for crystals smaller than $\sim 20$ µm necessary for sublimation. This process is continued during the cloud life cycle, usually yielding 100–300 crystal images. Figure 6 shows a dendrite, which was one of the largest crystals imaged during the measurements, at a cold-box midpoint air temperature of $-21^\circ C$, and other crystals in varying evaporating and melting stages. We measure D for platelike crystals and estimate H from the power-law relation, Eq. 4, but can measure both D and H for columnlike crystals because they usually land with the crystal c axis parallel to the microscope plate. All number concentrations reported here (except for Fig. 4) were obtained with manual frame-by-frame analysis of cloud-scope data.

Denote by $\langle P \rangle$, $\langle D \rangle$, $\langle H \rangle$, and $\langle $aspect ratio$ \rangle$ the average ice-cloud single-particle properties, where, for example,

$$\langle P \rangle = \frac{\int_{D_{\min}}^{D_{\max}} P|D| H|D| N|D|dD}{N_T}.$$

The small crystals studied were likely to have no preferred orientation, as the Reynolds number was estimated to be $<1$ for all crystals. These properties are a measure of cloud-particle size and aspect ratio and are calculated from measured number concentrations.

Assuming no scattered contributions, we take the optical depth per unit length of cloud at $\lambda = 0.685$ µm, also known as the extinction coefficient, to be

$$\frac{\tau_{\text{vis}}}{\text{length}} = \frac{\sigma_{\text{ext}}}{\langle P \rangle} = \int_{D_{\min}}^{D_{\max}} \sigma_{\text{ext}}|D| N|D|dD = 2N_T^{-1}\langle P \rangle.$$

The IR extinction coefficient is given by

$$\frac{\tau_{\text{IR}}(\lambda)}{\text{length}} = \int_{D_{\min}}^{D_{\max}} Q_{\text{extIR}}(\lambda; D) P|D| H|D| N|D|dD,$$

where $Q_{\text{extIR}}(\lambda; D)$ is the size- and wavelength-dependent extinction efficiency of a single crystal and averaging over random crystal orientations is assumed. The average cloud-extinction efficiency is taken to be

$$\langle Q_{\text{extIR}}(\lambda) \rangle = \frac{1}{N_{T}^{-1}\langle P \rangle} \int_{D_{\min}}^{D_{\max}} Q_{\text{extIR}}(\lambda; D) P|D| H|D| N|D|dD,$$

which in terms of optical depths is

$$\langle Q_{\text{extIR}}(\lambda) \rangle = 2 \frac{\tau_{\text{IR}}(\lambda)}{\tau_{\text{vis}}}.$$

An IR transmission measurement yields the quantity

$$NT = \int_{D_{\min}}^{D_{\max}} NDdD.$$

C. Measured IR Cloud-Extinction Efficiency

The circle diameter that just touches the outer hexagonal perimeter of a pristine ice crystal is denoted by $D$, $H[D]$ is the crystal length parallel to the c axis, $P[D]$ and $H[D]$ is the average over all orientations of crystal projected area, N$[D]$ is the number of crystals per volume per size interval, and $D'/H'$ is the aspect ratio so that an aspect ratio of $\geq 1$ corresponds to plates and an aspect ratio of $<1$ corresponds to columns. The total number of crystals per volume is

$$\langle P \rangle = \frac{\int_{D_{\min}}^{D_{\max}} P|D| H|D| N|D|dD}{N_T}.$$

The small crystals studied were likely to have no preferred orientation, as the Reynolds number was estimated to be $<1$ for all crystals. These properties are a measure of cloud-particle size and aspect ratio and are calculated from measured number concentrations.

Assuming no scattered contributions, we take the optical depth per unit length of cloud at $\lambda = 0.685$ µm, also known as the extinction coefficient, to be

$$\frac{\tau_{\text{vis}}}{\text{length}} = \frac{\sigma_{\text{ext}}}{\langle P \rangle} = \int_{D_{\min}}^{D_{\max}} \sigma_{\text{ext}}|D| N|D|dD = 2N_T^{-1}\langle P \rangle.$$

The IR extinction coefficient is given by

$$\frac{\tau_{\text{IR}}(\lambda)}{\text{length}} = \int_{D_{\min}}^{D_{\max}} Q_{\text{extIR}}(\lambda; D) P|D| H|D| N|D|dD,$$

where $Q_{\text{extIR}}(\lambda; D)$ is the size- and wavelength-dependent extinction efficiency of a single crystal and averaging over random crystal orientations is assumed. The average cloud-extinction efficiency is taken to be

$$\langle Q_{\text{extIR}}(\lambda) \rangle = \frac{1}{N_{T}^{-1}\langle P \rangle} \int_{D_{\min}}^{D_{\max}} Q_{\text{extIR}}(\lambda; D) P|D| H|D| N|D|dD,$$

which in terms of optical depths is

$$\langle Q_{\text{extIR}}(\lambda) \rangle = 2 \frac{\tau_{\text{IR}}(\lambda)}{\tau_{\text{vis}}}.$$

An IR transmission measurement yields the quantity

$$NT = \int_{D_{\min}}^{D_{\max}} NDdD.$$
The measured average cloud-extinction efficiency is thus

\[ \langle Q_{\text{extIR}}(\lambda) \rangle = 2 \frac{\ln T_{\text{IR}}}{\ln T_{\text{vis}}} \].

In the limiting case in which \( D \gg l, l_{\text{IR}} = l_{\text{vis}} \), and we obtain \( Q_{\text{extIR}}(\lambda) \approx 2 \) as expected. The extinction measurements discussed below are all presented in the normalized form given by Eq. (11). The size parameter is estimated by

\[ x = \frac{\pi (D/H)^{1.2}}{\lambda} \],

where \( \lambda \) is the wavelength in air.

Figure 7a shows measured \( Q_{\text{extIR}}(\lambda) \), and Fig. 7b shows the corresponding number concentration for a cloud composed mainly of plates and a cold-box temperature of \(-21^\circ C\). The vertical bars in Fig. 7a (and in Figs. 8a, 9a, 10a, and 11a) indicate wavelengths for which gaseous or cold-box window absorption was too high for sufficient signal to noise.

The cloud properties were \( D = 31.3 \mu m, H = 9.2 \mu m \) (estimated from Eq. (4)), and aspect ratio = 3.2. The size parameter ranged from \( x = 27 \) for \( \lambda = 2 \mu m \) to \( x = 3 \) for \( \lambda = 18 \mu m \). Near 2 \( \mu m \), \( Q_{\text{extIR}}(\lambda) \approx 2 \), which is the large size parameter limit for all particles. A narrow extinction window \((0.5 \mu m) \) wide occurs near \( \lambda = 2.84 \mu m \), where \( Q_{\text{extIR}}(\lambda) < 1 \) occurs. Beyond 4 \( \mu m \) the curves in Fig. 7a, which correspond to measurements during different parts of the cloud life cycle, become separated, probably on account of differences in the ice-crystal-number distribution. The ordering for \( Q_{\text{extIR}}(\lambda) \) from highest to lowest at \( \lambda = 16 \mu m \) is \( \tau_{\text{vis}} = 0.55, 0.72, 0.89, 1.21 \), where the measurement during \( \tau_{\text{vis}} = 1.21 \) occurred near the maximum cloud optical depth during the cloud life cycle (see Fig. 2), and the others occurred after this time. Note that there is another extinction window near \( \lambda = 10.5 \mu m \). This window is deeper and broader than the window at 2.84 \( \mu m \). The wavelength of lowest \( Q_{\text{extIR}}(\lambda) \) near 10.5 \( \mu m \) is not precisely the same for different optical depths.
there is a 0.07-µm spread between the point when the cloud \( \tau_{\text{vis}} = 1.21 \) and 150 s later, when \( \tau_{\text{vis}} = 0.55 \), and this is an indication that the form of the number distribution changes. The curve with the highest value of \( Q_{\text{extIR}}(\lambda) = 10.5 \) µm is the last measurement made during the cloud life cycle and likely had larger \( D_1 \); longer crystal growth time than the other measurements. The spectral shift to a slightly longer wavelength for \( Q_{\text{extIR}}(\lambda) = 10.5 \) µm of larger particles is discussed in Section 3, below. Unfortunately not enough crystals were collected during the cloud life cycle to resolve differences in number concentrations only \( \approx 150 \) crystals were measured for the distribution in Fig. 7b). The windows are associated with Christiansen bands of ice as discussed in Section 3, below. The small rapid variations near 6 and 15 µm are associated with regions in which air density changes, resulting from slight temperature changes, causing gaseous absorption to vary.

Figure 8a is superficially similar to Fig. 7a; however, note that in Fig. 8b there are fewer larger particles than in Fig. 7b. The size parameter ranged from \( x = 22 \) at \( \lambda = 2 \) µm to \( x = 2.4 \) at \( \lambda = 18 \) µm. Note that two nearly coincident curves with \( \tau_{\text{vis}} = 1.60, 1.20 \) are overlaid. These measurements occurred at and slightly later than the time of maximum cloud optical depth. \( Q_{\text{extIR}}(\lambda) = 2 \) to 6 µm does not even reach 2, probably on account of an extinction minimum for \( x \) in this range.

Crystals were only collected during the time interval near the cloud maximum optical depth for a midpoint cold-box temperature of \(-10^{\circ} \text{C}\). \( Q_{\text{extIR}}(\lambda) \) in Fig. 9a corresponds to a measured \( \tau_{\text{vis}} = 2.46 \). Note that the number concentration in Fig. 9b is referenced to crystal H rather than D as are those in Figs. 10b and 11b) because the dominant crystal habit at this temperature was hollow columns. The cloud properties were \( D_1 = 15.6 \) µm, \( H_1 = 21.5 \) µm, and aspect ratio \( 0.8 \pm 0.2 \), yielding size parameters \( x = 29 \) for \( \lambda = 2 \) µm and \( x = 3.2 \) for \( \lambda = 18 \) µm.

Columns were again the dominant habit for the measurements shown in Fig. 10 although approximately 10% were thin plates. Note the shift to smaller sizes in Fig. 10b compared with Figs. 7b), 8b), and 9b). Cloud properties were \( D_1 = 9.8 \) µm, \( H_1 = 14.8 \) µm, and aspect ratio \( 0.7 \pm 0.3 \), yielding size parameters \( x = 19 \) for \( \lambda = 2 \) µm and \( x = 2.1 \) for \( \lambda = 18 \) µm. The extinction windows are more open for this case than any of the previous cases on
account of the lower size parameters. \( Q_{\text{extIR}}(\lambda = 4 \, \mu m) = 2.3 \) rises to this maximum value, generally indicating the presence of an extinction relative maximum for this size parameter. The visible optical depth was \( \tau_{\text{vis}} = 0.47 \) during the IR measurement. Note that the rapid roll-off of the size distribution in Fig. 10 likely indicates that the cloud-scope collection is reduced for crystals with a maximum dimension of less than 8 \( \mu m \).

The extinction curves cross in numerous places in Fig. 11a). The number concentration in Fig. 11b refers to a cloud mainly composed of hollow columns with a few (estimated 10%) thin plates. At 2.5 and 4 \( \mu m \) the \( Q_{\text{extIR}} \) values from highest to lowest correspond to visible optical depths \( \tau_{\text{vis}} = 1.92, 0.66, 0.40, 3.40, 2.12 \) and at 12 \( \mu m \) they correspond to \( \tau_{\text{vis}} = 1.92, 3.40, 0.66, 2.12, 0.40 \). Only \( \tau_{\text{vis}} = 1.92 \) occurred before the maximum \( \tau_{\text{vis}} = 3.40 \). Mean cloud crystal properties were \( D = 14.0 \, \mu m, H = 16.6 \, \mu m, \) aspect ratio \( = 0.9 \pm 0.2 \). The size parameter was \( x = 24 \) for \( \lambda = 2 \, \mu m \) and \( x = 2.7 \) for \( \lambda = 18 \, \mu m \). As the cloud evolved during its life cycle, particle sizes were such that extinction rose to a relative maxima for wavelengths between 2 and 6 \( \mu m \). The extrema of the extinction curves are more dramatic in Fig. 11a, in which crystals of aspect ratio near unity were more common than in Fig. 7a, in which higher aspect ratio plates were the dominant habit. Note also that the particle size spectrum is much broader in Fig. 7b than in Fig. 11b. Broad particle size spectra have an averaging to effect on the extinction efficiency for sufficiently large particles and small wavelengths.

3. Discussion

Figures 12a) and 12b) show the real \( (n_r) \) and imaginary \( (n_i) \) parts of the index of refraction for ice, respectively, from data compiled by Warren. The strong absorption feature near \( \lambda = 3.077 \, \mu m \) is near the wavelength range 2.8 to 3 \( \mu m \), where \( n_r(\lambda) = 1 \) is a relative minimum. This wavelength range is known as a Christiansen band, and the lack of scattering on account of similar \( n_r \) for the host medium and particle is known as the Christiansen effect. This effect has been mentioned in the literature for over 100 years. In general, power is removed from a beam of light by particles through processes of diffraction, refraction, and absorption. For \( n_r = 1 \) the refraction component of extinction is weak. Now as the size parameter is reduced the diffraction component

![Figure 11](image1.png)

**Fig. 11.** a) IR extinction efficiency during different portions of the cloud life cycle for mainly column crystals and a cold-box temperature of \(-5^\circ C\). b) Number concentration based on crystal H and \( t_{\text{vis}} = 1 \), yielding \( D = 14.0 \, \mu m, H = 16.6 \, \mu m, \) aspect ratio \( = 0.9, P = 249 \, \mu m^2, \) and 921 crystals per cm

![Figure 12](image2.png)

**Fig. 12.** a) Real and b) imaginary parts of the refractive index for bulk ice.
also weakens, so extinction is due mainly to absorption. The single-scatter albedo gives us the fraction of extinction that is due to scattering and is defined in terms of the cross sections as \( \omega_0 = \sigma_{\text{scat}}/\sigma_{\text{ext}} \). Note that \( \omega_0 \rightarrow 0 \) for sufficiently small particles and wavelengths in a Christiansen band. The Christiansen effect does not simply turn off when \( n_r \neq 1 \), or when typical particle \( D \) reaches some threshold value. In fact measured IR windows at both \( \lambda = 2.84 \) \( \mu \)m and \( \lambda = 10.5 \mu \)m in Figs. 7a, 8a, 9a, 10a, and 11a are due to the Christiansen effect, even though particles are not that small and \( n_r (\lambda = 10.5 \mu \)m) does not quite reach unity.

Consider a beam of light that is incident upon one face of a rectangular particle with projected area \( P \) and length \( D \) along the propagation direction. Anomalous diffraction theory gives extinction by this particle as

\[
\sigma_{\text{ext}}(\lambda; D) = 2P \left[ 1 - \exp(-kD\cos kDn_r - 1) \right],
\]

where \( k = 2\pi/\lambda \) is the propagation number of the medium. The interference of forward light passing through and near the particle is given by the cos term, as modified by particle absorption. This model makes the undesirable prediction that \( \sigma_{\text{ext}} \) will vary between limits of 0 and 4 when \( n_r \rightarrow 0 \), and the model predicts that extinction departure away from the limit of very large particles is solely due to absorption when \( n_r \rightarrow 1 \):

\[
\frac{\sigma_{\text{ext}}(\lambda; D \rightarrow \infty) - \sigma_{\text{ext}}(\lambda; D)}{2P} = \exp(-kDn_r). \tag{14}
\]

Remnants of the Christiansen effect are noticeable when the phase angle \( kDn_r - 1 \) \( \approx 0 \).

Although ice crystals are generally not spheres, the effects of Christiansen bands on cross sections can be evaluated through the analysis of scattering by such particles, also known as Mie scattering.\(^{24} \) Figures 13 through 16 show the extinction efficiency and the single-scattering albedo for ice spheres with diameters between 2 and 50 \( \mu \)m and wavelengths between 2 and 18 \( \mu \)m. Extinction efficiency generally oscillates, as can be seen in Fig. 13 by looking at fixed \( \lambda = 2.4 \) \( \mu \)m and variable \( D \). Extinction efficiency is also generally smaller for small particles, as can be seen in Fig. 13 if one looks at fixed \( D = 2 \) \( \mu \)m and variable wavelength. The character of extinction changes noticeably in the vicinity of \( \lambda = 2.8 \) \( \mu \)m, where the real part of the index of refraction for bulk ice approaches unity, as can be seen by looking at fixed \( \lambda \) and variable \( D \). The supposition that scattering is suppressed near the Christiansen bands is confirmed in the single-scatter albedo shown in Fig. 14. Note that at fixed \( \lambda = 2.2 \) \( \mu \)m and variable \( D \) the single-scatter albedo is approximately unity, indicating the dominance of scattering over absorption in determining overall extinction. However, extinction is almost entirely due to absorption for small \( D \) and fixed \( \lambda = 2.85 \) \( \mu \)m.

Extinction efficiency for ice spheres is shown in Fig.
later. Figure 15 indicates that larger wavelengths for the extinction-efficiency minimum should correspond to larger particles. This observation can be understood as follows. For small particles such as $D < 2 \mu m$, the phase shift $\kappa D n_r - 1$ is generally small, so by going to smaller wavelengths near the Christiansen band we can attain a lower $n_r$ [see Fig. 12b and Eq. (13)], which will correspond to a minimal extinction efficiency. As the particle size increases the phase shift becomes more critical so that the wavelength of relative minimum extinction efficiency must increase to minimize the quantity $n_r - 1$, with a corresponding penalty of increased $n_r$.

Figure 16 shows the single-scattering albedo in this wavelength range and confirms that scattering is mainly due to absorption at a fixed value of $\lambda = 10.5 \mu m$ and variable $D$. It is worth noting that the ice refractive-index plot, based on data compiled in Ref. 25, is significantly different in Fig. 12 than in plots based on older data.\footnote{Even though the older data are flawed, the Christiansen effect of reduced scattering where $n_r \rightarrow 1$ is apparent in the single-scatter albedo plots shown in figure 4 of Ref. 29. Recent measurements of $n_r$ for ice in the 1.5–2.5-µm wavelength range\footnote{An assessment of the accuracy of current measured complex refractive-index values for ice in the IR under a variety of temperatures and growth conditions, including ice with air bubbles, defects, and strain, is necessary before confidence can be placed in modeling efforts for the single-scattering properties of ice crystals.} disagree by as much as 15% with data compiled by Warren.\footnote{An assessment of the accuracy of current measured complex refractive-index values for ice in the IR under a variety of temperatures and growth conditions, including ice with air bubbles, defects, and strain, is necessary before confidence can be placed in modeling efforts for the single-scattering properties of ice crystals.}}

4. Conclusion

Inasmuch as cirrus clouds can have high concentrations of small crystals, with $D < 50 \mu m$.\footnote{Development of an aircraft version of the cloud scope was funded by the University Corporation for Atmospheric Research, grant S9302. Insightful comments by an anonymous reviewer are appreciated.} the measurements reported here have direct application to IR transfer in the atmosphere. Two partial windows for IR transfer through clouds with small crystals were found: a narrow window near 2.84 µm with a width of $\approx 0.5 \mu m$ and a window near 10.5 µm with a width of $\approx 3 \mu m$. The extinction coefficient in the windows is approximately one-half the value at nearby wavelengths. These windows are a manifestation of the Christiansen effect: wavelengths where the real part of the index of refraction for cloud particles approaches unity have reduced scattering coefficients. Extinction is mainly due to absorption in the Christiansen bands. These measurements should have application to development of numerical models for single-scattering properties of ice crystals in the IR. The window at 2.84 µm can be applied to the imaging of warm objects through cirrus clouds and to the transfer of a miniscule part of the solar spectrum. The window at $\approx 10.5 \mu m$ is in the wavelength range for the atmospheric gas window and a fair amount of terrestrial IR and thus could play a role in the global radiative balance. Future measurements will be undertaken to measure extinction and emissivity simultaneously so that absorption efficiency can also be determined, and we intend to produce clouds with narrower size distributions.

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