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The Case for Ice Clouds on Venus

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Abstract. A paper by Rea and O'Leary, claiming that ice clouds are inconsistent with the near infrared spectra of Venus, is examined. Ice crystals with diameters of a few microns are found to be consistent with polarimetric observations and with all ground-based and high-altitude observations of Venus near 1.5-, 2-, and 3- μ wavelengths. However, it does not seem possible reliably to extract cloud parameters from the spectral details, because uncertainties of 20% or more in absolute reflectivity occur in existing spectra. To explain the low 3- μ reflectivity with particles of dimensions compatible with polarization data requires a 3- μ absorption coefficient \geq several hundred cm^{-1} . Ice crystals satisfy this requirement without contradicting data at other wavelengths; various common carbonates, silicates, and oxides, as well as dry ice, do not. Adsorbed water also seems unlikely. The Venera 4 water vapor observations, if reliable, independently demonstrate the existence of ice clouds on Venus, and with optical depths consistent with those obtained from analysis of the near infrared spectra. Some dust may nevertheless be mixed with the ice.

In a recent paper Rea and O'Leary [1968] have reexamined the infrared continuum albedo of Venus and have deduced that the aerosols scattering infrared radiation are almost certainly not composed of H₂O ice crystals. Opposite conclusions were reached by Bottema et al. [1965] and Sagan and Pollack [1967], who made use of similar data. In this note we present a critique of the Rea and O'Leary paper, as well as a reassessment of the evidence for ice clouds on Venus.

Since some of the arguments presented below are based on the two stream reflectivity equations given in our earlier paper, we first consider an apparent discrepancy between the theoretical predictions and laboratory measurements. Rea and O'Leary point out that we derive an average particle size for a synthetic ice cloud, generated by Zander [1966], that is an order of magnitude larger than obtained experimentally by direct sampling of the cloud; they suggest several reasons for the disagreement, including the possible invalidity of our formulas. In an effort to further test our theoretical results, we have derived an average particle dimension for a dense terrestrial cirrus cloud, observed by Blau et al. [1966]. The reflectivity measurements are summarized in Figure 2 of the paper by Rea and O'Leary. Using the reflectivity minimum at 2.0 μ , we obtain an average diameter of 40 μ . An essentially identical result is found from the 1.5- μ reflectivity minimum. We note that while Rea and O'Leary arbitrarily normalized the cloud reflectivity to agree with the laboratory curve at 1.2 μ , reasonable variations in the reflectivity at this wavelength do not affect our results; substantial changes in optical depth are needed to change the reflectivity at 1.2 μ from the assigned value, which, moreover, is typical of dense clouds. The derived particle size agrees well with typical dimensions for cirrus ice crystals, which have an average base of tens of microns and an average length of hundreds of microns [Blau et al., 1966]. We are certainly not a factor of ten too large in our estimates.

This leads us to believe that some of the difference between theory and laboratory results may be attributed to Zander's sampling of only the top layer of his cloud, where small particles can be expected because of sedimentation of the larger particles toward the bottom of the apparatus, as well

as the greater exposure of particles near the top to ambient conditions, which would tend to evaporate them. In addition, Zander states that a fraction of the aerosol crystals were bound together into larger aggregates. Since the scattering contribution of a crystal of a given size is weighted by the extinction cross section (which varies roughly as the square of the linear dimension), these large aggregates may well have dominated the scattering events. The optical constants used in our two-stream calculations were obtained from an extensive literature survey by Irvine and Pollack [1968]. The general validity of our two-stream approximation, in the present context, has been demonstrated by Irvine [1968] in a comparison with exact solutions of the equation of radiative transfer.

Rea and O'Leary argue that all the decrease in the reflectivity of Venus near 2- μ wavelength is attributable to carbon dioxide, and hence no absorption remains to attribute to the aerosol. This conclusion is based on a comparison of the absorptivity exhibited by Venus and by laboratory samples of CO₂ at wavelengths of 1.58, 1.60, and 1.98 μ . Although the carbon dioxide absorption on Venus is influenced by multiple scattering whereas the laboratory result is a direct transmission measurement, they claim that the similar values of the absorptivity at the three wavelengths permit such a comparison and cite an analogous procedure employed by Connes et al. [1967].

We disagree. Such a method is valid only when the monochromatic absorption coefficient, k_{ν} , varies in the same manner for all the wavelength regions over a frequency interval equal to the resolution of the spectrometer. This is not to be expected for the cited wavelength intervals. Even for single rotational lines, as employed by Connes et al. [1967], this procedure is totally valid only when the lines have the same half widths. The 1.58- and 1.60- μ domains lie within the centers of carbon dioxide absorption bands, where k_{ν} will vary greatly from the center of a rotational line to a wavelength intermediate between two such lines. However, the 1.98- μ wavelength domain lies between two strong absorption features; here k_{ν} tends to be more nearly constant.

To pursue this line of argument further, we now present a hypothetical situation in which there is little CO_2 absorption at 1.98μ , but the observed strong absorption at 1.58 and 1.60μ . We approximate the variation of k_v over a resolution element at 1.58μ with two absorption coefficients, $k_v(\ell)$ (1.58) and $k_v(s)$ (1.58), where $k_v(\ell) \gg k_v(s)$. A similar approximation is used at 1.60μ , and k_v near 1.98μ is taken to have a single value, $k_v(1.98)$. So that the laboratory transmission results be similar at the three wavelengths, we require $k_v(1.98)$ to have a value intermediate between $k_v(\ell)$ and $k_v(s)$. We further suppose that the density of aerosols on Venus is such that little CO_2 absorption takes place between successive scattering events when k_v is $k_v(s)$ or $k_v(1.98)$, but significant absorption takes place when k_v is $k_v(\ell)$. In this event, Venus will look black at wavelengths appropriate to $k_v(\ell)$ and white at $k_v(s)$ and $k_v(1.98)$. Smearing by the finite spectrometer resolving power will lead to a substantial CO_2 absorption at 1.58μ and 1.60μ with little CO_2 absorption at 1.98μ . The above example illustrates how the observed absorptivity of Venus at 1.98μ could be smaller than at the other two wavelengths, and the reverse be true for the laboratory measurements, such as the ones preferred by Rea and O'Leary. Such a reversal would not be expected if the procedure employed by Rea and O'Leary were generally valid. Our hypothetical example, of course, does not demonstrate significant cloud absorption at 1.98μ .

Perhaps the most potent argument advanced against the presence of ice clouds is the apparent absence of the broad-band ice feature at 1.5μ , which would appear as a local reflectivity minimum. Rea and O'Leary considered Zander's laboratory ice clouds to be characterized by a size of 1μ and the cirrus cloud of Blau *et al.* to be characterized by sizes one to two orders of magnitude larger. Since spectra of both clouds exhibited a strong absorption feature at 1.5μ , they deduced that submicron-sized particles would be needed to reduce the ice band to an acceptable level of insignificance. But such particles are unlikely, by terrestrial analogy.

For reasons given earlier, we do not believe Zander's spectra are characterized by a particle size as small as 1μ and have accordingly used the reflectivity formulas given in our previous paper, as well as an optical depth of 30 derived there, to calculate the strength of the $1.5\text{-}\mu$ band as a

function of particle size. Table 1 summarizes the results of these calculations, with the symbol $R(x)$ denoting the reflectivity at a wavelength of x microns. Since the 1.5- μ band is fairly broad, it is necessary in order to detect it to compare the reflectivity at 1.5 μ with that at a wavelength a few tenths of a micron away. We see that for acceptably small particle sizes the 1.5- μ feature is quite weak. The values for $R(1.5)$ and its ratios can also be used for the center of the 2- μ ice band, if the particle diameters are halved.

In order to estimate the constraints placed by the observations on the value of the reflectivity ratios, we first estimate their error. Because the two wavelengths at which R must be measured are considerably farther apart than for gaseous absorption features, the observation is more difficult to perform. The search for the "limonite band" on Mars illustrates this point. One observer [Tull, 1966] found a weak broad-band feature near the appropriate wavelength, whereas two other observers [Younkin, 1966; Sinton, 1967] did not [cf. Pollack and Sagan, 1967].

To estimate the magnitude of the error that may be present for such observations of Venus, we compared different measurements of the same reflectivity ratio. Without much effort we found three examples of substantial differences. Kuiper [1964] estimates $R(2.4)/R(2.3)$ as 1.1, while Bottema et al.'s [1965] measurement implies a value of 0.85. Taking account of the cruder resolving power of Bottema et al. only aggravates the problem. Moreover, the earth's atmosphere is very transparent at these two wavelengths. Two spectra taken by Kuiper [1964] only half an hour apart appear to show a difference in the value of $R(1.98)/R(2.3)$ of 20%. Finally, Moroz [1963] finds $R(1.5)/R(1.7)$ to be 1.11, while his spectra in a later paper [Moroz, 1965] imply a value of 1.47. Thus it seems that errors of 20% may easily be present in measurements of reflectivity ratios.

Moroz finds $R(1.5)/R(1.2)$ to be 0.89, while Kuiper's spectra imply a value of about 1 for $R(1.5)/R(1.7)$. Allowing for a 20% uncertainty, an average size less than about 7.5 μ would be compatible with these observations, according to Table 1. Such sizes mean that the 2- μ band, while

somewhat stronger, would also be fairly weak and that Kuiper's [1964] rejection of ice clouds from the value of the ratio $R(1.93)/R(2.3)$ can be faulted on the same grounds as in the discussion of the $1.5\text{-}\mu$ feature above.

Table 1 gives the single-scattering albedo, $\tilde{\omega}_0$, for ice crystals at the three wavelengths of interest. We see that $\tilde{\omega}_0$ is quite close to 1 for small particle sizes, so that other sources of absorption in the Venus atmosphere, such as possible condensation nuclei and the far wings of gaseous absorption features, may lead to a distortion of the actual reflectivity from that of a pure ice cloud.

We have now discussed the major reasons advanced by Rea and O'Leary against the presence of ice clouds on Venus and have found them far from conclusive. We now consider some of the data that we viewed as favorable for ice clouds and that Rea and O'Leary consider to be ambiguous. In principle, if we have a sufficient number of reflectivity points, even if these are not at absorption maxima, we can still make a reliable identification of composition. The variation of reflectivity with wavelength depends on the variation of the absorption coefficient with wavelength, and even substances as closely related as water and ice differ significantly in this respect. In our earlier paper, we did not think that Bottema et al. [1965] could readily make the CO_2 correction at $2.0\ \mu$, but we concluded that the remainder of their corrected curve was valid, since the water-vapor correction at $1.8\ \mu$ implied a ratio $R(1.75)/R(2.3)$ in agreement with a value of the same ratio found by Moroz. We note that at $1.75\ \mu$ there is no Venus water-vapor correction. In addition, the particle size deduced from the curve of Bottema et al. agreed well with that deduced from $R(2.5)$ and $R(1.75)/R(1.2)$. At that time, we believed reflectivities could be measured more accurately than the above discussion suggests they can. Accordingly, we no longer consider the region between 1 and $3\ \mu$ sufficiently well defined to permit a definitive compositional analysis. We hope that, in the future, high-quality determinations of R will be made in this region at places far removed from absorption lines.

The one unambiguous feature remaining in the Venus spectra, which offers positive evidence for ice clouds, is the low reflectivity between 3.0μ and 3.3μ . The low reflectivity between 2.7μ and 3.0μ is strongly influenced by the CO_2 intercombination bands near 2.75μ , while the low reflectivity beyond 3.3μ may be influenced by the HCl fundamental at 3.45μ (and at longer wavelengths by the CO_2 fundamental at 4.3μ). It would seem that the CO_2 bands near 2.75μ have little influence at wavelengths longer than 3.0μ since the reflectivity is more than a factor 10 higher at 2.5μ than at 3.0μ , although both are equally distant from 2.75μ . Because of the large decrease in reflectivity, there can be little doubt as to the reality of this feature. It has also been observed both by Bottema et al. [1965] and by Moroz [1965]. While Rea and O'Leary admit the reality of this feature, they claim that many substances absorb strongly beyond 3μ and that even if H_2O is responsible it may be present only as an adsorbed coating on a particle of dust.

In an effort to investigate the variety of compositions that can cause such low reflectivity near 3μ , we estimate the value of the absorption coefficient, k , at these wavelengths. When particles become sufficiently small compared to a wavelength, the absorption cross section will always dominate over the scattering cross section, as long as k is not identically zero. Thus k could be quite small and yet there could be a very low reflectivity beyond $3\text{-}\mu$ wavelength. If k is less than 100 cm^{-1} , formulas given by van de Hulst [1957], when coupled with our reflectivity equations, imply that almost all particles must be smaller than 0.085μ in diameter for $R(3 \mu) < 5\%$. Such particle sizes seem quite incompatible with the polarization estimates of several microns [van de Hulst, 1952]. The exact value implied by the polarization/phase-angle curve depends on the index of refraction of the particle, but all reasonable indices of refraction lead to sizes in excess of 1μ .

In addition, such small particles could not account for the value of 0.35 for $R(3.2)/R(2.5)$ found by Sinton [1963] near inferior conjunction. As inferior conjunction is approached, the reflectivity value will increase from

the low values found by Moroz and Bottema et al. and asymptotically approach a value of $[1 - \tilde{\omega}_0(3.2)]/[1 - \tilde{\omega}_0(2.5)]$ expected when single scattering dominates (see, e. g., formulas given by Chamberlain [1965]). Using our reflectivity equations and an appropriate choice of l for the anisotropy parameter, 2β , we find $\tilde{\omega}_0(2.5) \approx 0.7$ and $\tilde{\omega}_0(3.2) \approx 0.1$ are needed for small particles to match the observations at the smaller phase angles. This implies an upper limit of 0.15 to the reflectivity ratio at large phase angles, in contradiction to Sinton's result. By contrast, large-particle scattering results in an upper limit of 0.5.

If the particles are comparable to or larger than a wavelength near 3μ , they must possess a sufficient thickness, d , and absorption coefficient, k , so as to absorb a significant fraction of the radiation passing through the particle. This would imply a sufficiently low value of the single-scattering albedo for a very low reflectivity to result. Sample calculations with our two-stream formulas indicate that kd must be greater than 0.5 if the reflectivity is to be below 5%. The polarization size data, cited above, and a consideration of the small fall times of large particles suggest that the average particle size of the aerosols is probably below 50μ ; Ryan [1964] gives values of the terminal velocities of various sized particles in the terrestrial atmosphere that are approximately equal to values appropriate for the level of interest in the Venus atmosphere. Particles of diameter 1μ , 10μ , 50μ , and 500μ will fall 10 km in 1200, 12, 0.58, and 0.023 days, respectively. Such an upper bound on d implies a lower bound on k of 100 cm^{-1} throughout the 3.0- to 3.3- μ wavelength region. At the center of the band causing the absorption, k may be 10 times larger or more. Accordingly, a lower bound on the value of k at the band center is about 1000 cm^{-1} . Furthermore, this band must be located within or very close to the 3.0- to 3.3- μ domain. Since the largest value of k at the center of a band, other than a fundamental, for ice and most other materials is 100 cm^{-1} , the lower limit on k derived above suggests that a fundamental is responsible for the low 3- μ reflectivity on Venus.

Let us now investigate what other plausible cloud constituents might meet the conditions given above. First consider dust particles. According to the American Institute of Physics Handbook [Grey, 1963], calcite, quartz, magnesium oxide, and sapphire (aluminum oxide) have values of k at 3.0μ of 2 cm^{-1} , 0.75 cm^{-1} , $<0.05 \text{ cm}^{-1}$, and 2 cm^{-1} , respectively. Such materials are unlikely to cause the low Venus reflectivity. Put another way, laboratory slabs of some of these substances will be opaque, but involve dimensions far larger than seem admissible for Venus aerosols. Measurements by Clark [1957] on common terrestrial silicates indicate values of $k(3.0 \mu)$ approaching 30 cm^{-1} for some specimens; however, such specimens exhibit even larger values of $k(1.5 \mu)$, leading to even lower reflectivities at 1.5μ , contrary to observation. Reflectivity measurements by Hovis [1965] of the iron oxides, Fe_2O_3 and Fe_3O_4 , show no enhancement of the absorption at 3μ over that at 2.5μ , except for an absorption that is clearly attributable to water of hydration.

Hovis' results lead to a consideration of another possibility — that water of crystallization or adsorbed water present in or on dust particles can cause the low $3\text{-}\mu$ reflectivity. But water of crystallization is unlikely to be present, since the dust particles would have lost any such water when at the high temperature of the lower atmosphere and surface [cf. Sagan et al., 1965] and would not have had time to regain it in the relatively short period a given particle would spend in the higher, colder portions of the atmosphere. In addition, for adsorbed water (and/or adsorbed HCl and HF) to cause such a low reflectivity, thicknesses of more than 1μ (i. e., more than 10^4 molecules deep) would be required so that $kd \gtrsim 1/2$; this seems very unlikely. Large amounts of other molecules of the general form H-X, which absorb near 3μ , are not to be expected on Venus on thermodynamic equilibrium grounds [cf. Lippincott et al., 1967]. Finally, CO_2 condensation clouds have strong intercombination bands at 2.7μ , but the wings do not extend beyond 2.8μ [Osberg and Hornig, 1952].

Above we have examined some of the most likely cloud components and have found them unlikely to produce the low reflectivity observed for Venus near 3μ . We now consider the ability of H_2O ice clouds to match the observations. Because of the low infrared cloud temperatures, we consider it likely that the upper portions of a Venus water cloud would be ice and not liquid water [cf. Sagan, 1960]. Throughout the 3.0 - to $3.3\text{-}\mu$ region, ice has values of k in excess of 100 cm^{-1} and has a fundamental with a value of $1.4 \times 10^4 \text{ cm}^{-1}$, centered at 3.1μ . Thus, ice clouds meet all the requirements given above. We now consider in more detail the particle sizes required for ice crystals to produce the low reflectivity and inquire how compatible such particles are with the requirements for a weak $1.5\text{-}\mu$ band. To some extent, it is not meaningful to speak of an average particle size, since both sufficiently large particles ($d \gtrsim 4 \mu$) and sufficiently small particles ($d \lesssim 0.6 \mu$) are capable of meeting the requirement that the reflectivity be less than 5% throughout the 3 - to $3.3\text{-}\mu$ domain. We can certainly say that average particle sizes in excess of 4μ are permitted, and, depending on the particle-size distribution function, smaller average sizes may be allowed. Thus, there is overlap with the requirement from the $1.5\text{-}\mu$ band that $d < 7.5 \mu$. In addition, we require that $d > 1 \mu$ for the meteorological reasons advanced by Rea and O'Leary; we find that ice crystals with mean diameters between 4μ and 7.5μ (and perhaps 1 to 7.5μ) are compatible with the observations. Dollfus [1961] has deduced a value for water-particle sizes of 2.5μ from the polarization data. While Dollfus' work strictly applies only to water droplets, the same result should apply to ice crystals, since the two materials have almost identical visual indices of refraction [van de Hulst, 1952]. Finally, for reasons given in our earlier paper, we note that the polarization and spectral sizes need not exactly agree and that the latter will probably be somewhat larger.

We conclude this note with a brief discussion of the relevant results of the entry capsule of the Soviet Venera 4 spacecraft [Tass, 1967], news of which reached us after the above portion of this note was completed. Two independent measurements of the Cytherean atmosphere with phosphorous pentoxide gas analyzers yielded water-vapor mixing ratios (by mass) of between 0.1% and 1%. The measurements were made at the 0.5- and 1.5-bar

pressure levels; from the temperatures measured, the water vapor amounts are definitely below their saturation values, so the results pertain to the region below the cloud bottoms. The announced total pressure at the surface, as determined by Venera 4, is 20 ± 2 bars. On the other hand, the radar determination of the planetary diameter implies a surface pressure closer to 60 bars [Sagan, 1967; Pollack and Sagan, 1968]. If both values are carried, the 0.1% mixing ratio implies a loading density of water vapor above the surface of 20 to 70 g cm^{-2} ; 1% mixing ratio implies 200 to 700 g cm^{-2} . From these loading densities, the intersection of adiabats for uniform mixing with the saturation vapor-pressure curve gives the temperature at which saturation occurs. If appropriate homomorphic condensation (or crystallization) nuclei are present, condensation (or crystallization) will occur at saturation. Carrying out this procedure [cf. Sagan, 1960, Figure 4], we find that saturation occurs at temperatures between 240 and 270°K — quite plausible temperatures for the bottoms of water clouds whose tops are at 210° to 230°K. If the Venera 4 results are reliable, there must be water clouds on Venus.

We may next inquire whether there is enough water vapor in the atmosphere to yield clouds of the thickness — τ between 18 and 43 — that we have derived [Sagan and Pollack, 1967]. The loading density, L , of ice in the clouds is easily seen to be $\sim (4/3)r\rho\tau$, where r is the particle radius and ρ the particle density. For $1 \mu \leq r \leq 10 \mu$ and $18 \leq \tau \leq 43$, we find $2.4 \times 10^{-3} \text{ g cm}^{-2} \leq L \leq 5.7 \times 10^{-2} \text{ g cm}^{-2}$. The atmospheric structure determined by Mariner 5 near the Cytherean tropopause permits us to calculate an upper limit to the amount of water (in vapor or condensed form) above any level at which condensation is assumed to begin; we assume that the amount of water present in all phases equals that present as water vapor were no condensation to have occurred. For cloud-bottom temperatures between 235° and 273°K and water-vapor mixing ratios in the lower atmosphere between 0.1% and 1%, we find amounts of water entirely consistent with the values of L we derived from the visible and infrared data.

Finally, the discrepancy between the very low water-vapor mixing ratios determined spectroscopically, but applying principally to the region above the clouds, and the large amount of water vapor found by Venera 4, but applying principally to the region below the clouds, is in itself an argument for the condensation of water vapor — i. e. , for water clouds.

In summary, we have found that the low reflectivity at $3\ \mu$ is fully compatible with ice clouds with mean diameters of several microns, a particle size independently deduced from polarization studies. Other alternative materials, plausible on some grounds, are inconsistent with the $3\text{-}\mu$ observations. The molecule responsible for this feature must have an absorption coefficient $k \sim 1000\ \text{cm}^{-1}$ for a fundamental centered near $3.0\ \mu$. It does not seem likely that, apart from water, any cosmically abundant material exists that is stable in the Venus atmosphere and that possesses the required properties. It is also very unlikely that the water is present as adsorbed water or as water of crystallization. Quite independently, the Venera 4 results, if reliable, demonstrate that extensive ice clouds must exist on Venus. The probable presence of condensation and/or crystallization nuclei, as well as the yellow color index of Venus, suggests that some dust may be mixed with ice in the Cytherean clouds.

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TABLE 1. Strength of the 1.5- μ Ice Band as a Function of Particle Diameter, d

| d | R(1.5) | $\frac{R(1.5)}{R(1.7)}$ | $\frac{R(1.5)}{R(1.2)}$ | $\tilde{\omega}_0(1.2)$ | $\tilde{\omega}_0(1.5)$ | $\tilde{\omega}_0(1.7)$ |
|---------|--------|-------------------------|-------------------------|-------------------------|-------------------------|-------------------------|
| 1 μ | 0.78 | 0.94 | 0.92 | 0.99995 | 0.997 | 0.9993 |
| 10 | 0.45 | 0.70 | 0.57 | 0.9995 | 0.974 | 0.993 |
| 100 | 0.08 | 0.38 | 0.14 | 0.995 | 0.794 | 0.933 |