

Microwave Boundary Conditions on the Atmosphere and Clouds of Venus¹

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ABSTRACT

The dielectric properties of H₂O and H₂SO₄ at microwave frequencies have been calculated from the Debye equations. The derived frequency and temperature dependence agrees well with existing data. The dielectric properties of H₂O/H₂SO₄ mixtures are deduced and, for a well-mixed atmosphere, the structure of H₂O and H₂O/H₂SO₄ clouds is calculated. With the COSPAR model atmosphere and the calculated cloud models, the microwave properties of the atmosphere and clouds are determined. The 3.8 cm radar reflectivity of the planet, the Mariner 5 S-band occultation profile, and the passive microwave emission spectrum of the planet together set an upper limit on the mixing ratio by number of H₂O of $\sim 10^{-3}$ in the lower Venus atmosphere, and of H₂SO₄ of $\sim 10^{-5}$. The polarization value of the real part of the refractive index of the clouds, the spectroscopic limits on the abundance of water vapor above the clouds, and the microwave data together set corresponding upper limits on H₂O of $\sim 2 \times 10^{-4}$ and on H₂SO₄ of $\sim 9 \times 10^{-6}$. Upper limits on the surface density of total cloud constituents and of cloud liquid water are, respectively, $\sim 0.1 \text{ g cm}^{-2}$ and $\sim 0.01 \text{ g cm}^{-2}$. The infrared opacities of 90 bars of CO₂, together with the derived upper limits to the amounts of water vapor and liquid H₂O/H₂SO₄, may be sufficient to explain the high surface temperatures through the greenhouse effect.

1. Introduction

Water vapor is a spectroscopically detected constituent of the Venus atmosphere above the clouds [for a review, see L. D. G. Young (1972)] and water is an effective absorber of microwave radiation in both its vapor and liquid phases. Because of the temperature structure of the atmosphere and the presence of sulfuric acid in the clouds, the water vapor abundance must be greater below than above the Venus clouds. Microwave observations constitute the only remote method for investigating the water content of the deep atmosphere of Venus. Direct spacecraft observations of water have so far been restricted to the middle atmosphere. Microwave observations also set an upper limit on the integrated abundance of liquid water and other absorbers. The first such calculation set an upper limit of $\sim 0.1 \text{ g cm}^{-2}$ to the liquid water in the Venus clouds (Sagan and Giver, 1961). Previous attempts to place an upper limit on the total abundance of water on Venus from the microwave spectrum have assumed either the presence of uniformly mixed water vapor alone (Pollack and Morrison, 1970; Janssen *et al.*, 1973) or the presence of uniformly mixed water vapor and a relatively thin, isothermal, liquid water cloud (Sagan and Giver, 1961; Barrett and Staelin, 1964). In the first case, the derived upper limit on the water vapor mixing ratio by number, a value between 2×10^{-3} and 5×10^{-3} , was below the minimum amount of water needed to condense liquid water clouds, assuming no impurities; it was therefore

concluded that liquid water could be ignored as a source of microwave opacity. In the second case, details of cloud structure and the relation of vapor pressure to cloud thickness were ignored since the calculations were performed at a time when the atmospheric structure was poorly known; the authors concluded that high water vapor abundances and relatively thick liquid water clouds might be compatible with the microwave data.

Recently, Hansen and Hovenier (1974) have deduced two important properties of the upper cloud particles from an analysis of the polarization of light reflected from Venus:

- 1) The particles are spherical with a mean radius of $1 \mu\text{m}$. Therefore, the cloud particles are liquid down to temperatures between 230 and 250 K.
- 2) The real part of the refractive index for visible light is 1.44.

The only material so far proposed that can remain liquid at these temperatures and that has this refractive index is sulfuric acid solutions with concentration by mass of about 75%. This composition, suggested by A. T. Young (1973), explains the previously puzzling $11.2 \mu\text{m}$ band (A. T. Young, 1973), and is consistent, while many other materials are not, with the $3 \mu\text{m}$ absorption feature (Pollack *et al.*, 1974).

Since there is strong evidence supporting a cloud composed of sulfuric acid solution and since the composition and structure of the noncondensing part of the Venus atmosphere is better known, detailed cloud models can now usefully be compared to the microwave

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data in order to set more stringent upper limits on the abundances of cloud constituents. Radar and microwave occultation data will be used as well as the passive microwave emission from the planet. In Section 2, the available microwave data are enumerated and the general calculation scheme outlined. In Section 3 the optical properties at these wavelengths of plausible clouds are set forth; and in Section 4 the relevant physics is described for the calculation of more realistic cloud models than have previously been employed. Results on abundances are exhibited in the final sections.

2. Microwave data

Fig. 1 displays the microwave brightness temperature of Venus between 0.1 and 10 cm as compiled by Pollack and Morrison (1970) with the addition of more recent measurements by Rather *et al.* (1974) at 0.14 cm, Ulich *et al.* (1973) at 0.31 and 0.86 cm, Ulich (1974) at 0.21 cm, Janssen *et al.* (1973) at 0.85, 0.98, 1.18, 1.33, 1.40 and 1.46 cm, and Warnock and Dickel (1973) at 7.89 and 9.26 cm. In the absence of a significant microwave phase effect, no correction need be made for the phase of the observations. Janssen *et al.* claim that the calibration of their data is consistent with the flux standards used by Pollack and Morrison in compiling the older data; yet these two data sets near 1 cm are clearly inconsistent. The data of Rather *et al.*, Ulich *et al.*, and Ulich are not referenced to the same flux standards used by Pollack and Morrison which may explain the difference between these two data sets; however, in their discussion of calibration Ulich *et al.* (1973) seem to favor a higher flux standard and, correspondingly, brightness temperatures some 10–30 K higher than previously reported. These inconsistencies will hopefully be resolved in the near future. For the time being, they limit the precision with which conclusions can be drawn from the microwave spectrum.

The small differences in atmospheric structure between the COSPAR model (Marov, 1972) based primarily on the Venera entry measurements and the NASA standard model based primarily on the Mariner 5 and Mariner 10 radio occultation results can produce small differences in calculated brightness temperatures; however, larger differences are engendered by differing assumptions on the CO₂ abundance. For example, the data of Rather *et al.* (1974), Ulich *et al.* (1973), Ulich (1974) and Janssen *et al.* (1973) are consistent with the brightness temperature spectrum calculated for an atmosphere composed of 90% CO₂ and 10% N₂ with no other opacity source, but are only barely consistent with a 97% CO₂ atmosphere. On the other hand, the data reported by Pollack and Morrison (1970) are consistent only with a spectrum calculated assuming some additional opacity source besides CO₂, and adding a microwave-transparent gas such as N₂ to the model Venus atmosphere only compounds the problem.

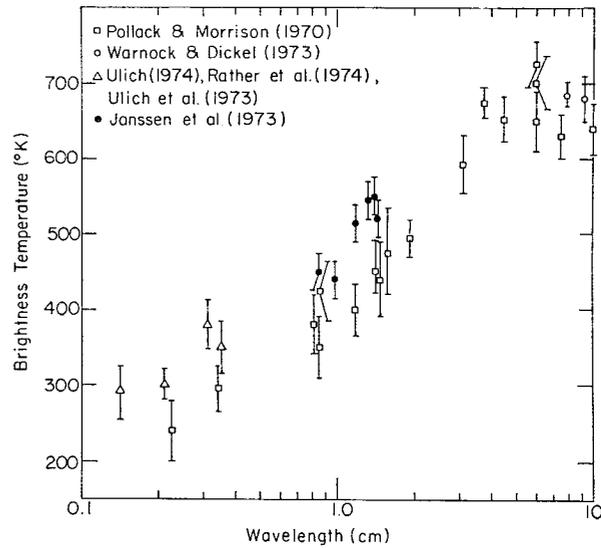


FIG. 1. Microwave brightness temperature spectrum of Venus.

For the calculations presented here the structure of the atmosphere has been taken to be that presented by Marov (1972) and the abundance of CO₂ has been taken to be 97% as reported by Vinogradov *et al.* (1971). The resulting calculated brightness temperature spectra will be compared to the large, self-consistent data set presented by Pollack and Morrison (1970) in order to set upper limits on the cloud opacity. In Section 5 variations in these derived upper limits due to a relaxation of assumptions will also be discussed.

Once all opacity sources have been accounted for and a relation between physical temperature *T* and the optical depth τ at a particular level of the atmosphere is found, the disk-integrated brightness temperature at each wavelength can be calculated using

$$T_B = 2eT_s E_3(\tau_s) + 2 \int_{\tau_0}^{\tau_s} T(\tau) E_2(\tau) d\tau + T_0 [1 - 2E_3(\tau_0)]. \quad (1)$$

The first term is the emission from the surface, at temperature *T_s*, with emissivity *e*, attenuated by the whole of the atmosphere and clouds which together have a total optical depth τ_s . *E_n*(τ) is given by

$$E_n(\tau) = \int_0^1 \mu^{n-2} e^{-\tau/\mu} d\mu,$$

where $\cos^{-1}\mu = \theta$, the viewing angle. The second term is the total emission of the atmosphere, composed of contributions from each atmospheric layer attenuated by the layers above. The third term is the emission of that fraction of the atmosphere, assumed isothermal, which is above the cloud layers. Since τ_0 , the optical depth of the high atmosphere, is taken to be 0.01, the

contribution of the third term is only a few degrees Kelvin.

Determining the value of ϵ in (1) presents problems similar to those encountered when using radar data to measure atmospheric opacity; both require knowledge of the surface properties at the appropriate wavelengths. Observations of the brightness temperature and radar reflectivity at longer wavelengths, where atmospheric effects are negligibly small (see, e.g., Pollack and Sagan, 1965; Pollack and Morrison, 1970), place constraints on the dielectric properties of the surface material and show that the emissivity and reflectivity are approximately constant down to a wavelength of 20 cm. However, the uncertainties in the data as well as in the composition and structure of the surface materials make extrapolation to shorter wavelengths uncertain. The surface reflectivity becomes especially sensitive to surface structure which has a size comparable to the wavelength.

Actual measurements of rocks and rock powders (Campbell and Ulrichs, 1969) and experience with the radar properties of the Moon and Mars (see, for example, Olhoeft and Strangway, 1974) probably limit the uncertainty of the extrapolated reflectivity to less than 50% as long as no "gravel" is present; this corresponds to an uncertainty in the emissivity probably less than 10%. Since the extrapolation of the short-wavelength emissivity contains less uncertainty and since the part of the microwave spectrum most sensitive to surface properties is also the part least sensitive to atmospheric and cloud properties, these difficulties will not affect significantly the comparison of models with the data. A value of ϵ between 0.8 and 0.9 seems consistent with the long-wavelength data (Pollack and Sagan, 1965; Pollack and Morrison, 1970).

The atmospheric attenuation in excess of that provided by CO₂ needed to match the spectral data decreases the calculated radar reflectivity by an amount smaller than the uncertainty in the surface reflectivity. Since it is the intent of this paper to set limits on the atmospheric properties and not to study the surface properties in detail, the limit on the total, two-way atmospheric attenuation, A_{radar} , at 3.8 cm of 9 dB estimated by Ingalls and Evans (1969), who do consider surface properties, will be adopted here as a check on the results derived from the microwave spectrum.

The radio occultation results from Mariner 5 (Fjeldbo *et al.*, 1971) are also considered. Since, at S-band wavelength the opacity of all plausible cloud constituents is much smaller than for the X-band radar measurements, these data are much less sensitive to the presence of the cloud. The temperature dependence of the cloud opacity exhibited in Section 3 and the cloud structure in Section 4 together place the bulk of the effective cloud absorption at or above 50 km. The measured attenuation of the atmosphere at these levels, A_{occ} , is effectively zero with an uncertainty of about 0.5 dB; there-

fore, 0.5 dB will be taken as an upper limit to the cloud and atmospheric opacity above 50 km.

3. Absorption coefficient of cloud and atmospheric constituents

In order to calculate the optical thickness of each layer of the atmosphere, three sources of opacity must be accounted for: the opacity of the main constituent gases of the atmosphere (assumed here to be 97% CO₂ and 3% N₂); the opacity of any minor constituent gases (in this case H₂O); and the opacity of the cloud itself.

The optical depth τ of an atmospheric layer is given by the total absorption coefficient of the layer, k , times the thickness of the layer, z . For CO₂, the absorption coefficient is given by an expression derived from one given by Barrett and Staelin (1964) based on measurements by Thaddeus and Ho (cf. Ho *et al.*, 1966):

$$k_{\text{CO}_2} = 1.35 P^2 T^{-4.6} \lambda^{-2} f \text{ [km}^{-1}\text{]}, \quad (2)$$

where P is the total atmospheric pressure in bars, T the temperature in units of 100 K, λ the wavelength of radiation in centimeters, and f a function of the ratio of CO₂ to total pressure. For a CO₂ mixing ratio of 97% by number, $f = 0.94$.

For H₂O vapor the absorption coefficient for radiation near the resonant absorption feature at 1.35 cm is derived from expressions given by Chung (see Barrett and Staelin, 1964):

$$k_{\text{H}_2\text{O}} = k_{\text{res}} + k_{\text{nonres}},$$

$$k_{\text{nonres}} = 4.97 P V T^{-2.5} \lambda^{-2} \alpha_{\text{H}_2\text{O}} \text{ [km}^{-1}\text{]}, \quad (3)$$

$$k_{\text{res}} = (3.6 \times 10^4) (1 + 3.14 \alpha_{\text{H}_2\text{O}}) P^2 F \lambda^{-2} G \alpha_{\text{H}_2\text{O}} \text{ [km}^{-1}\text{]}, \quad (4)$$

where

$$V = 5.38 (1 + 3.14 \alpha_{\text{H}_2\text{O}}) P T^{-0.625},$$

$$F = [(900 \lambda^{-2} + 22.237)^2 + V^2]^{-1} + [(900 \lambda^{-2} - 22.237)^2 + V^2]^{-1},$$

$$G = T^{-4.125} \exp(-6.44/T).$$

In these expressions $\alpha_{\text{H}_2\text{O}}$ is the number mixing ratio of water vapor, P is in bars, T in 100 K and λ in cm; k_{res} is the absorption coefficient due to the rotational transition line at 1.35 cm.

Calculating the absorption coefficient of the cloud particles is greatly simplified since the cloud particles are much smaller than the wavelengths considered here. Analysis of polarization of light reflected from Venus leads to a mean particle size in the cloud tops of 1 μm (Hansen and Hovenier, 1974). The formation of droplets large enough to fall out of the cloud will not only affect the structure of the cloud but also the calculation of the absorption coefficient in the small-particle limit. The effects of precipitation will be neglected and it will be shown later that this is permissible.

With the particle size much smaller than the wavelength, the scattering cross section is much smaller than

the absorption cross section which is just the geometric cross section. (For the small-particle approximation, the scattering cross section is the Rayleigh value which for 10 μm droplets and 0.1 cm radiation is $\sim 10^{-12} \text{ cm}^{-2}$ which is much smaller than the geometric cross section for absorption, $\sim 10^{-6} \text{ cm}^{-2}$.) Neglecting scattering then, the absorption coefficient, in the small-particle limit, becomes linearly proportional to the line-of-sight bulk of the absorber:

$$k_e = 0.3\pi c_1 \lambda^{-1} (\rho_c / \rho_l) \text{ [km}^{-1}\text{]}, \quad (5)$$

where λ is the wavelength (cm), ρ_l the density of individual droplets (g cm^{-3}), ρ_c the bulk density (g m^{-3}), and c_1 depends on the real and imaginary parts of the dielectric constant, ϵ_1 and ϵ_2 respectively:

$$c_1 = 6\epsilon_2 [(\epsilon_1 + 2)^2 + \epsilon_2^2]^{-1}. \quad (6)$$

ϵ_1 and ϵ_2 depend on the real and imaginary parts of the refractive index, η and χ respectively, as

$$\left. \begin{aligned} \epsilon_1 &= \eta^2 - \chi^2 \\ \epsilon_2 &= 2\eta\chi \end{aligned} \right\}$$

The theoretical description of the dielectric behavior of non-interacting dipoles dissolved in a non-polar fluid, given by Debye (see Fröhlich, 1958), postulates that the position of the dipole relative to its nearest neighbors is maintained by viscous forces. Energy from the oscillating electric field of radiation can be absorbed to excite oscillations in the position of the dipole which relaxes back to its initial state by viscous damping. Since the strength of the damping is proportional to the amplitude of the oscillations, the relaxation is exponential in time with a decay constant proportional to the viscosity of the fluid. This description is valid as long as the interaction of the dipoles with each other can be neglected.

For pure polar liquids, the densities are sufficiently high that the interaction between dipoles cannot be neglected and the Debye theory should not generally apply. However, the experimental work of Collie *et al.* (1948), Saxton (1952), and Chamberlain *et al.* (1966) has verified that the complex dielectric constant of water does obey the Debye equations in its frequency and temperature dependence. Specifically,

$$\epsilon_1 = \epsilon_\infty + (\epsilon_s - \epsilon_\infty) / (1 + y^2), \quad (7)$$

$$\epsilon_2 = (\epsilon_s - \epsilon_\infty) y / (1 + y^2), \quad (8)$$

where ϵ_∞ is the high frequency value of the dielectric constant, ϵ_s the static dielectric constant, and $y = \omega t_r$, where ω is the frequency and t_r the relaxation time. Fig. 2 shows theoretical values of the real and imaginary parts of the refractive index for water calculated from the Debye equations and the experimentally measured values of Lowan (1949), Goldstein (1951) and Gunn and East (1954) as functions of frequency and temperature.

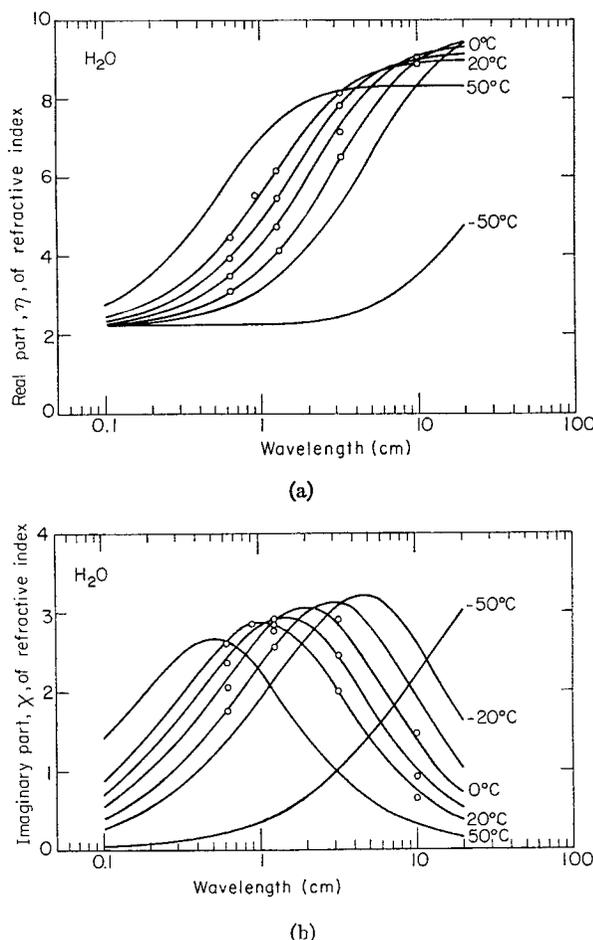


FIG. 2. Real part η (a) and imaginary part χ (b) of refractive index of pure water as a function of wavelength and temperature. Curves, calculated from Debye theory, are compared with experimental points.

The excellent agreement between the experimental values of the dielectric constant and the values calculated from the Debye equations might be due to the short-order structure created in water by the molecular interaction. X-ray studies have shown that, despite the fluid nature of water at room temperature, the molecular interaction—hydrogen bonding—creates a relatively fixed relationship between each molecule and its few nearest neighbors. Therefore, like the description of a dilute solution of dipoles above, each molecule has a fairly fixed position relative to its neighbors and the restoring force is of such a nature as to produce an exponential relaxation of any perturbation on this position. The decay constant is proportional to the viscosity since the viscosity of water is dominated by this same interaction. The variation from exact regularity of the molecular positions introduces a dispersion in relaxation times (Fröhlich, 1958) which can be accounted for in the above equations by introducing a dispersion parameter β such that y becomes $y^{1-\beta}$ (Cole and Cole, 1941); however, in the case of water this

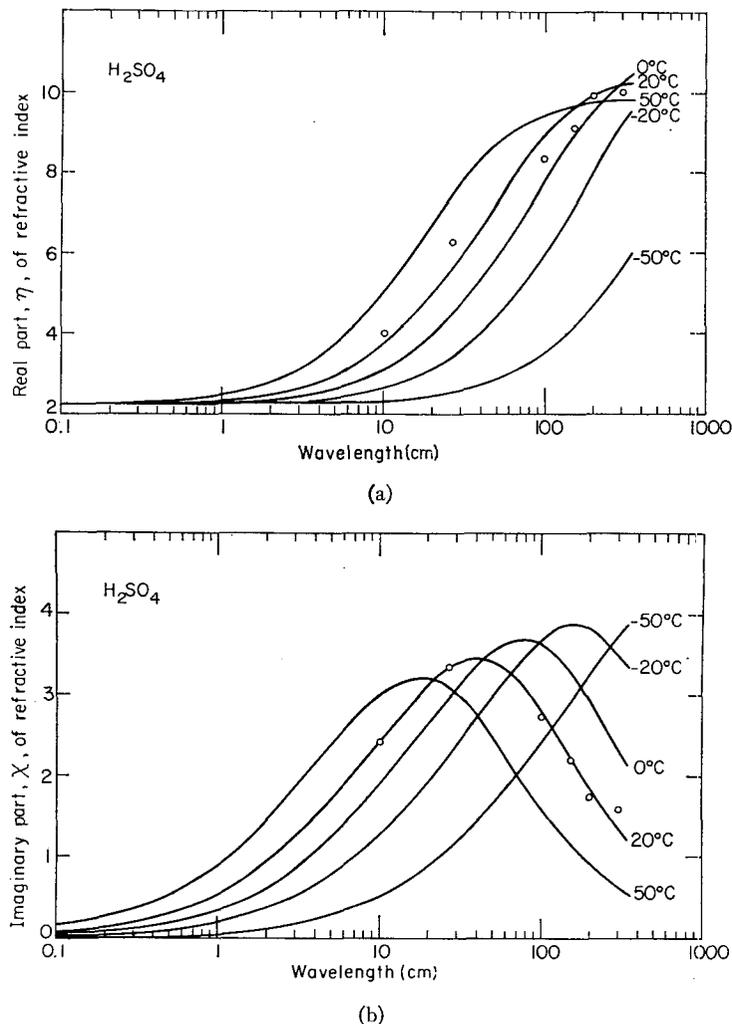


FIG. 3. As in Fig. 2 except for pure sulfuric acid.

correction has proved unnecessary to the precision required here.

As a result of the temperature dependence of the refractive index shown in Fig. 2, the cloud opacity at longer wavelengths is dominated by the colder water near the top of the cloud even though, as shown in Section 4, the mass of the cloud is dominated by the warmer water. In fact water warmer than about 373 K ceases to be an effective absorber of microwave radiation. Thus the contribution to the emergent microwave flux of a liquid water cloud is a sensitive function of the freezing temperature of the droplets. Since the wavelength of the χ maximum varies as a function of temperature and, therefore, as a function of position in the cloud, the already broad frequency dependence of the cloud opacity is even further broadened. In the case when the liquid water in the cloud dominates the microwave spectrum near 1 cm, no "shoulder" appears in the spectrum due to water vapor (see, e.g., Pollack and Morrison, 1970). That is, the absence of the 1.35 cm

feature in the Venus microwave spectrum is not so sharp a constraint on the abundance of water if extensive liquid water clouds are also present.

The dielectric properties of sulfuric acid have also been shown to obey the Debye equations with the introduction of a dispersion in the relaxation time; Fig. 3 shows the calculated and measured (Brand *et al.*, 1953) values of η and χ . Since the measured values were obtained at only one temperature, the temperature dependence was assumed to be that predicted by Debye's theory as is true for water. Note that the values of η and χ shown in Fig. 3 give a value of c_1 in Eq. (6) at 1 m wavelength that is as large as that for water at 1 cm; however, the value of k is two orders of magnitude less for sulfuric acid because of the λ^{-1} dependence in Eq. (5). Because the wavelength dependence of c_1 for both water and sulfuric acid is weaker than a linear dependence, the maximum values of k occur for the 0.1 cm wavelength. Sulfuric acid becomes an effective absorber only at wavelengths < 20 cm.

Because pure H_2SO_4 is capable of dissociating itself, there are always some ions in solution which can change the nature of the restoring force; however, since the temperatures considered here are much less than the dissociation temperature of sulfuric acid, the concentration of such ions, and their effect on the dielectric properties, is probably small. It should be noted that because of the wavelength dependence of ϵ_1 and ϵ_2 in Eq. (6) it is the warmer sulfuric acid which dominates the opacity of a pure sulfuric acid cloud in contrast to the case for water. However, the opacity of sulfuric acid warmer than about 400 K can be neglected.

The theoretical description of the dielectric properties of solutions of sulfuric acid in water is complicated not only by the lack of theoretical and experimental knowledge concerning pure polar fluids but also by the complicated interaction between water and sulfuric acid molecules in mixtures including a large concentration of ions as solutes. However a plausible estimate of the dielectric properties of these mixtures can nevertheless be performed.

The small dispersion in relaxation time observed in both pure water and pure sulfuric acid indicates that the arrangement of molecules in these fluids must be very nearly uniform such that every molecular dipole is in the same electric field. Because of the uniformity, the dipoles all absorb at the same frequency producing a much stronger absorption than if they absorbed at different frequencies. Therefore, the addition of a small amount of H_2SO_4 to water should weaken the absorption by water by disrupting the uniform distribution of the water dipoles. Likewise, the addition of a small amount of H_2O to sulfuric acid should weaken its absorption.

The absorption by stronger solutions, for which the ratio of the number of H_2O molecules to the number of H_2SO_4 molecules is near unity, is complicated by the increasing number of ions present as solutes. The concentration of ions becomes large enough that ionic interactions can dominate the motion of the molecules; but, even more important, the increase of ion concentration means that the dominant molecular species is changing from H_2O and H_2SO_4 to H_3O^+ and HSO_4^- . Since the strength of the ionic interaction is much greater than the strength of hydrogen bonding between like molecules, either the new constituents, H_3O^+ and HSO_4^- , become harder to move or they form larger associations which tend to move as a unit. Either of these effects would serve to shift the relaxation time to a smaller value producing absorption at a frequency different from that of H_2O or H_2SO_4 . These associations would also decrease the uniformity of the arrangement of any remaining molecules, spreading their absorption over a broader frequency interval with a consequent reduction of absorption at any one frequency.

Therefore, the effect of mixing these two strongly interacting molecules is to weaken the absorption due

to each species by decreasing their number and the uniformity of their environment and possibly to introduce absorption at some other frequency. This sort of behavior is suggested by measurements of absorption band strengths in the infrared region of the spectrum by Giguère and Savoie (1960). They observe that bands which are strong in dilute solutions (8.5% H_2SO_4 by mass) weaken and disappear for concentrations higher than 78.5%, while strong bands in 100% H_2SO_4 weaken and disappear for concentrations less than 78.5%. The same behavior is also suggested by variations in infrared spectra with concentration as measured by Remsberg (1973) and Palmer and Williams (1975) for sulfuric acid solutions, by Draegert and Williams (1968) for other acid solutions, and by Gurikov *et al.* (1969) for aqueous solutions of acetonitrile.

For the purposes of this paper two extreme assumptions have been adopted to simulate the range of possibilities for the dielectric properties of mixtures of water and sulfuric acid:

1) To simulate the case discussed above, we assume that the absorption of the droplets is simply a superposition of the absorption of pure water and pure sulfuric acid—in proportion to their respective number concentrations in the solution. This assumption gives a broad and strong absorption feature which smoothly changes its properties from those of pure water to those of pure H_2SO_4 , mimicking the behavior suggested qualitatively by the spectra of Remsberg (1973), Giguère and Savoie (1960) and Palmer and Williams (1975). This assumption will be referred to as the “full absorption” model and may closely represent the actual properties of H_2O – H_2SO_4 mixtures.

2) For completeness and to simulate the case in which the new ionic species can only absorb effectively at much higher frequencies, we assume that H_2O and H_2SO_4 form pairs which cannot absorb at all in the microwave region. This assumption implies absorption only by excess water or excess sulfuric acid; i.e., concentrations of sulfuric acid by mass less than the monohydrate concentration of 80.5% absorb as an excess amount of water, while concentrations greater than 80.5% absorb as an excess amount of sulfuric acid. This is certainly a minimum assumption; it makes the water or sulfuric acid molecules which are chemically inactive in the solution (i.e., not dissociated) the only absorbers in the droplet. This assumption will be referred to as the “effective absorption” model. These two assumptions should bracket the actual behavior of H_2O – H_2SO_4 mixtures.

4. Cloud structure

As shown in Section 3, the microwave opacity of the cloud depends only on the bulk content as long as the droplets remain small compared to the wavelength of the radiation. Two quantities must be known before the vertical distribution of bulk content can be calcu-

TABLE 1. Values of $\alpha_{\text{H}_2\text{O}}$ and $\alpha_{\text{H}_2\text{SO}_4}$ for models used in microwave calculations.

Model name	$\alpha_{\text{H}_2\text{O}}$ (%)	$\alpha_{\text{H}_2\text{SO}_4}$ (%)
Venera	2.0 - 3.0	0
Pure water	1.4 - 2.0	0
Sulfuric acid	0.001-1.0	0.00045-0.045
Young "wet"	0.39	0.22
Young "dry"	0.001-0.01	0.00014
Prinn	0.01	(~0.0009)

lated: first, the ratio of the amount of material in vapor form to the amount in condensed form at equilibrium, and second, the amount of condensable material available at a given level in the atmosphere. The former corresponds to a statement about the equilibrium vapor pressure over the condensate at a given temperature and depends on whether the vapor is maintained in a supersaturated, saturated or undersaturated state. Supersaturation or undersaturation can only occur when the cooling or heating of an air parcel occurs faster than the diffusion time of the vapor onto or off a droplet. The vapor diffusion time is roughly proportional to the pressure of the background gas; the diffusion time at the 10 bar level on Venus is roughly 100 s. Significant variations from vapor equilibrium would require sustained vertical velocities much greater than 10 m s^{-1} throughout most of the atmosphere; therefore the vapor is everywhere in equilibrium with the condensate (Gierasch, 1975).

The second quantity required corresponds to a statement on the assumed total abundance of a condensable constituent and its vertical distribution. If the mixing time is much smaller than the fallout time, then the cloud will be well-mixed; i.e., the ratio of cloud constituent to CO_2 will be independent of altitude. Since the vapor diffusion time is much shorter than the mixing time, the particular distribution of cloud constituent between vapor and condensed phases will depend only on "local" conditions. The horizontal homogeneity of the Venus clouds as indicated by Mariner 10 terminator photographs (Hapke, 1974), by measurements of the amount of CO_2 above the clouds (A. T. Young, 1975); and by infrared temperature measurements (Murray *et al.*, 1963; Ingersoll and Orton, 1974), and the presence of high-velocity flows throughout the clouds as indicated by the Venera 8 Doppler measurements (Marov *et al.*, 1973), by cloud motions observed in ground-based UV photographs (Dollfus, 1975) and Mariner 10 UV photographs (Murray *et al.*, 1974), and by measurements of the differential Doppler shift of CO_2 absorption lines across the disk of Venus (Traub and Carleton, 1975), all together imply that sufficient motions are present to mix the Venus clouds thoroughly.

The Venera 8 photometer results (Marov *et al.*, 1973) suggest that the lower portions of the Venus clouds, most likely to be affected by precipitation, extend down

to approximately 35 km altitude, well below the level at which both water and sulfuric acid cease to be effective absorbers of microwave radiation. Therefore, in order to simplify calculations, the effects of precipitation on cloud structure will be neglected and a constant mixing ratio for all cloud constituents assumed. The validity of this assumption will be discussed later in relation to those cloud models found to be consistent with the microwave data.

Since the total amount of any condensable substance is constant, the amount in the condensed phase is just the difference between the total amount and the equilibrium vapor pressure over the condensate; i.e.

$$\alpha_c = \alpha_T - \alpha_v, \quad (9)$$

where the number mixing ratio of the condensed phase is represented by α_c , the vapor phase by α_v , and the total by α_T .

Two types of water clouds are considered here. In the first, water condenses onto an aerosol which does not affect the vapor pressure in equilibrium with the droplet; i.e., α_v is a function of temperature only. Type I occurs when the condensation nuclei are not easily soluble in water (e.g., rock dust) or when the mass of the condensation nuclei is negligible compared to the mass of the droplet and forms only a very dilute solution. In the cloud Type II, water condenses on sulfuric acid particles of sufficient mass to alter significantly the equilibrium vapor pressure of water over the droplet; α_v is then a function of the concentration of water in the droplet as well as the temperature. For Type II, Eq. (9) is rewritten as

$$\alpha_v(x, T) = \alpha_{\text{H}_2\text{O}} - m(1-x)\alpha_{\text{H}_2\text{SO}_4}/x, \quad (10)$$

where

$$m \equiv \mu_{\text{CO}_2}^2 / (\mu_{\text{H}_2\text{O}} \mu_{\text{H}_2\text{SO}_4}),$$

μ_{CO_2} , $\mu_{\text{H}_2\text{O}}$ and $\mu_{\text{H}_2\text{SO}_4}$ are the molecular weights of CO_2 , H_2O and H_2SO_4 , respectively, and x is the concentration by mass of sulfuric acid in the droplets. $\alpha_v(x, T)$ is the number mixing ratio of water vapor, $\alpha_{\text{H}_2\text{O}}$ the mixing ratio of the total quantity of water in all phases (assumed constant), and $\alpha_{\text{H}_2\text{SO}_4}$ the number mixing ratio of sulfuric acid which is assumed to be entirely in the condensed phase. This is approximately the case for temperatures $\lesssim 400 \text{ K}$ and $\alpha_{\text{H}_2\text{O}} \gtrsim 10^{-5}$ (Wofsy, 1974). Since at higher temperatures the cloud is an ineffective absorber of microwave radiation, overestimating the bulk of the lower cloud does not constitute a serious error.

Table 1 exhibits the range of compositions assumed for these models. The pure water models span a range of values of $\alpha_{\text{H}_2\text{O}}$ which includes the values measured by Venera spacecraft as reported by Vinogradov *et al.* (1971) down to the minimum value for which Type I liquid water clouds can condense. The sulfuric acid models include two models proposed by A. T. Young (1973) as well as one proposed by Prinn (1973). Em-

ploying Marov's (1972) model atmosphere and the values of α_v as a function of temperature for pure water from the *International Critical Tables* (Washburn, 1928; Keyes, 1928), Eq. (9) gives the bulk liquid water content at each level of the cloud. The results are displayed in Fig. 4 where they are compared with a profile proposed by Vinogradov *et al.* (1971) on the basis of Venera measurements of the water vapor abundance.

Calculation of the vertical structure of sulfuric acid cloud models requires the solution of Eq. (10) which is performed graphically in Fig. 5. The family of curves which slope from the upper left to the lower right and which are labelled by temperature represent α_v as a function of the droplet concentration x (Blake and Greenwalt, 1928; Timmermans, 1960). Each family of curves originating from the right side of the graph and sloping toward the lower left represents the value of the right side of Eq. (10) for different values of α_{H_2O} and $\alpha_{H_2SO_4}$. The intersection of two of these curves, one from each family, gives the values of α_v and x which satisfy Eq. (10) for a given temperature, α_{H_2O} and $\alpha_{H_2SO_4}$. The bulk liquid water content at each level of the cloud is then $\alpha_{H_2O} - \alpha_v$. Fig. 6 displays the result as well as the specific model proposed by Prinn (1973).

Prinn's model is unrealistic since it requires α_v and x to be constant with altitude. Fig. 5 shows that if the value of $\alpha_v = 10^{-4}$ is to remain constant with altitude throughout the cloud, then $\alpha_{H_2SO_4} < 2 \times 10^{-6}$. This cloud is less dense than Prinn's model by an order of magnitude; it also has a sulfuric acid concentration which never reaches 75% unless the temperature at the top is less than 235 K. If, on the other hand, the concentration is required to remain at 75% throughout a large part of the cloud, then Fig. 5 suggests that the values of both α_{H_2O} and $\alpha_{H_2SO_4}$ would have to be several orders

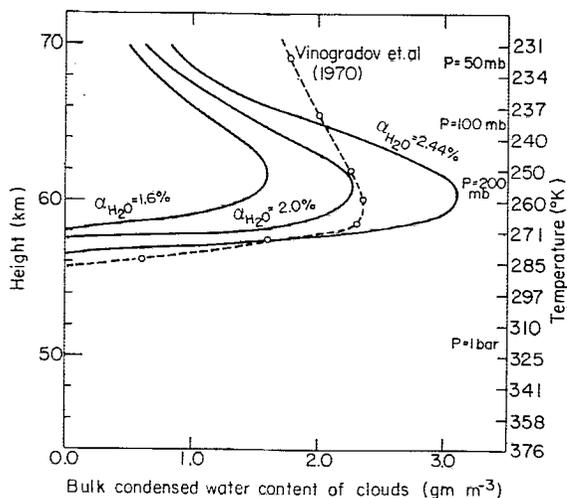


FIG. 4. Bulk condensed water content of pure water clouds for various α_{H_2O} as a function of altitude or temperature.

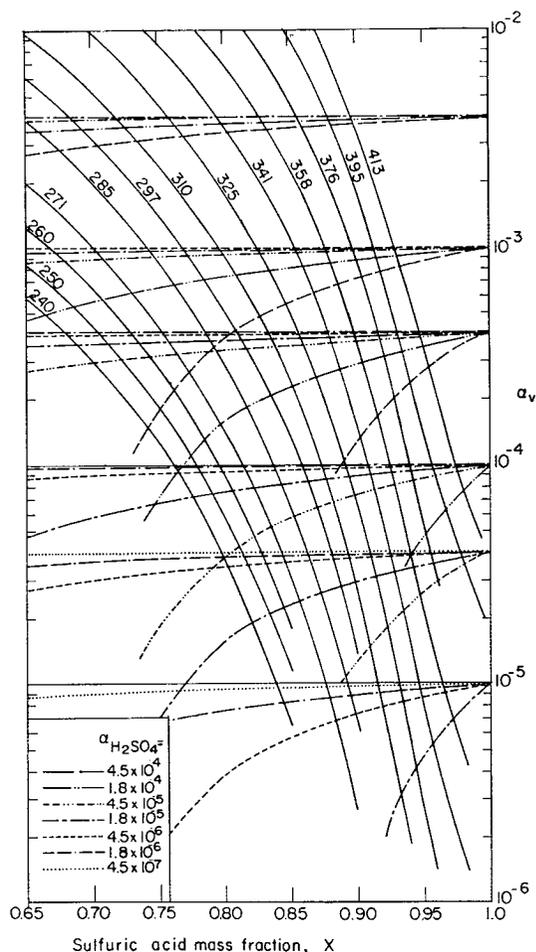


FIG. 5. Graphical solution of Eq. (10). The family of curves originating from upper left represent α_v , the equilibrium vapor pressure of water, as a function of temperature, and the sulfuric acid mass fraction x of the cloud droplet. The family of curves originating from the right edge represent the right-hand side of Eq. (10); each family corresponds to a different value of α_{H_2O} and each curve in a family a different value of $\alpha_{H_2SO_4}$.

of magnitude greater than assumed. Considerations of this sort lead to two general conclusions:

- 1) The concentration of the cloud droplets varies significantly over the vertical extent of the cloud.
- 2) If $\alpha_{H_2O} / \alpha_{H_2SO_4} \lesssim 10$, α_v in the deep atmosphere can be much greater than it is above the clouds.

5. Results

The conclusions of Sections 3 and 4 are used to calculate the opacity of the cloud and atmosphere system in occultation and radar geometry and to calculate the microwave spectrum of the system; a comparison of these calculations with observation sets upper limits on the abundance of water, α_{H_2O} , and sulfuric acid, $\alpha_{H_2SO_4}$, in the atmosphere of Venus. Table 2 is a summary of these results.

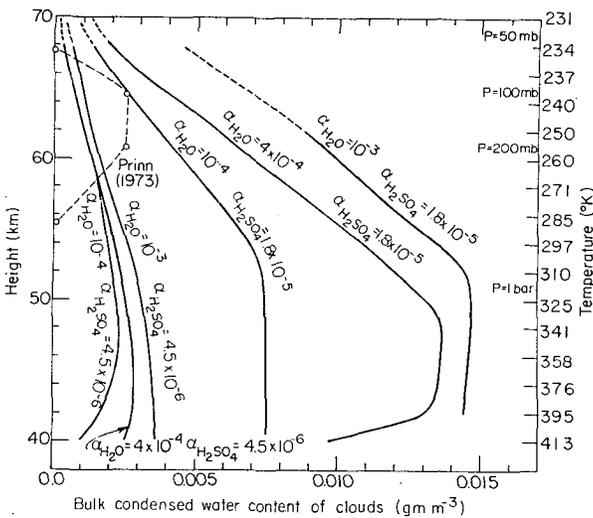


FIG. 6. Bulk condensed water content of sulfuric acid clouds with different α_{H_2O} and $\alpha_{H_2SO_4}$ as a function of altitude or temperature.

Certain general features of these calculations should be noted since they place constraints on any similar calculations, set upper limits on water abundance which are nearly independent of the model used, and indicate possible future measurements that would better define the upper limits. The lack of a feature at 1.35 cm sets an upper limit on α_{H_2O} of order 5.0×10^{-3} , as Pollack and Morrison (1970) and Janssen *et al.* (1973) have already concluded. Massive clouds,

TABLE 2. Summary of upper limits on α_{H_2O} and $\alpha_{H_2SO_4}$ for various combinations of microwave data and various assumptions. "Full Abs" and "Eff Abs" refer to the two assumptions made concerning the dielectric properties of $H_2O-H_2SO_4$ mixtures.

Assumptions used	Upper limit on α_{H_2O}	Upper limit on $\alpha_{H_2SO_4}$
1.35 cm H_2O feature	$\sim 5 \times 10^{-3}$	—
Occultation data		
Full Abs	$4 \times 10^{-4} - 4 \times 10^{-3}$	$4.5 \times 10^{-6} - 9 \times 10^{-6}$
Eff Abs	4×10^{-3}	9×10^{-6}
Radar data: $CO_2 \sim 97\%$		
Full Abs	4×10^{-4}	9×10^{-6}
Eff Abs	4×10^{-4}	1.8×10^{-5}
Janssen <i>et al.</i> , Rather <i>et al.</i> , Ulich and Ulich <i>et al.</i> data: $CO_2 \sim 90\%$		
Full Abs	10^{-4}	9×10^{-6}
Eff Abs	10^{-4}	4.5×10^{-5}
Pollack and Morrison data: $CO_2 \sim 90\%$		
Full Abs	10^{-2}	4.5×10^{-4}
Eff Abs	10^{-2}	10^{-3}
Pollack and Morrison data: $CO_2 \sim 97\%$		
Full Abs	2×10^{-4}	9×10^{-6}
Eff Abs	2×10^{-4}	4.5×10^{-5}

however, would change this result in two ways: (i) the opacity of a very massive cloud would dominate the spectrum, becoming optically thick before the vapor, and thereby obscuring the water vapor feature; and (ii) if the ratio of sulfuric acid to water is sufficiently large, most of the water is in the condensate form rather than the vapor form (see Fig. 5) thereby removing the vapor from the atmosphere. However, even in the extreme case of a pure N_2 atmosphere, clouds more massive than 0.2% of the atmosphere by mass would be optically thick at sufficiently cold temperatures making them inconsistent with the microwave brightness temperature data. Even these clouds are not sufficiently massive to change this result; thus 0.5% can be taken as a firm upper limit on the water abundance on Venus. This result clearly excludes the pure water models of Vinogradov *et al.* (1971) and others (cf. Fig. 4).

The radio occultation measurements at $\lambda > 10$ cm are insensitive to the CO_2 abundance, but can, in principle, serve as a good direct measure of the cloud mass. Since the water abundance is $\leq 0.5\%$, only the cloud can absorb (weakly) the radio transmission; and since the cloud bulk density above 50 km scales roughly as the assumed abundance of water and sulfuric acid (see Fig. 6), the radio occultation amplitude data in this range serve to measure the mass of the cloud. Thus, in Table 2, the upper limit set by these data alone is of order 10^{-3} for α_{H_2O} and 10^{-5} for $\alpha_{H_2SO_4}$.

Sulfuric acid does not contribute significantly to the opacity of the clouds unless $\alpha_{H_2O}/\alpha_{H_2SO_4} \lesssim 1$, in which case the cloud is essentially pure sulfuric acid. If this ratio is $\gtrsim 1$, then the combination of the temperature and frequency dependence of the dielectric constant of $H_2O-H_2SO_4$ mixtures with the concentration and bulk density structure makes the liquid water in the upper part of the cloud the dominant source of opacity. If the concentration in these upper regions is as low as 75%, as suggested by the index of refraction (A. T. Young, 1973), then the assumption of water-like dielectric properties is a good one. [See Remsberg (1973) and Giguère and Savoie (1960) for comparison of the infrared spectra of 8.5 and 75% solutions of sulfuric acid.]

Since in a steady-state cloud the net downward flux of particles is a constant throughout the cloud, the conversion of cloud droplets to precipitation and their removal by fallout implies that the number density of droplets is constant in altitude because the number density is the flux divided by the fall velocity which is nearly constant in the deep atmosphere. Therefore, in one scale height the ratio between the precipitation-controlled mixing ratio and a constant mixing ratio is $\sim \frac{1}{3}$. Since the contribution of the lower scale height of these tenuous sulfuric acid clouds to the total opacity and total microwave emission is small compared to that of the upper part of the cloud, the neglect of precipita-

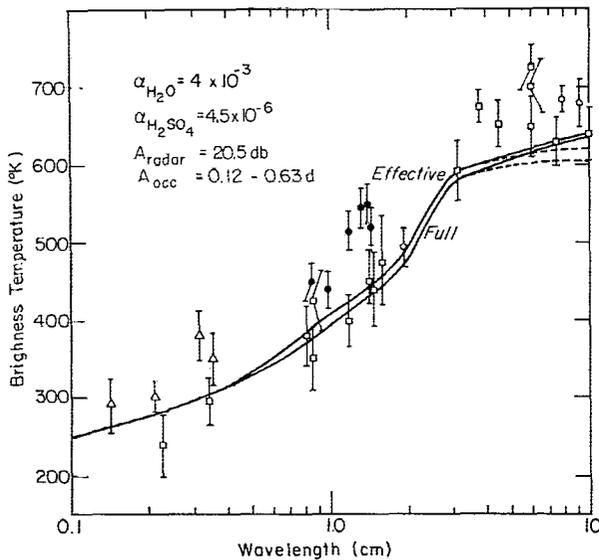


FIG. 7. Calculated microwave spectrum for cloud model with $\alpha_{H_2O} = 4 \times 10^{-3}$ and $\alpha_{H_2SO_4} = 4.5 \times 10^{-6}$.

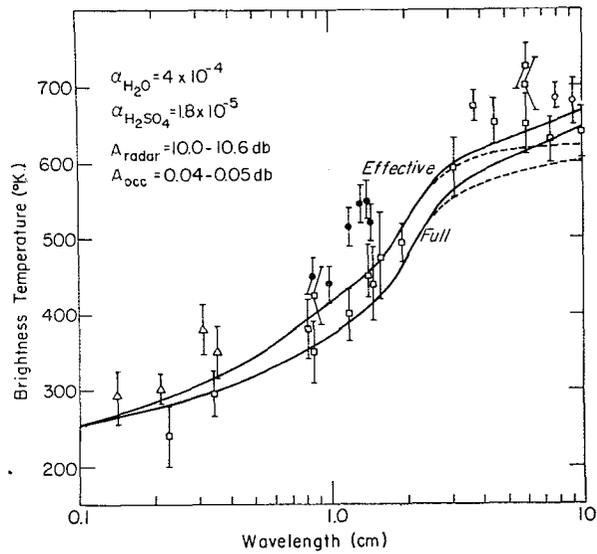


FIG. 9. Calculated microwave spectrum for cloud model with $\alpha_{H_2O} = 4 \times 10^{-4}$ and $\alpha_{H_2SO_4} = 1.8 \times 10^{-5}$.

tion effects does not produce a significant change in the conclusions drawn.

Also shown in Table 2 are the limits set by the radar reflectivities and the microwave spectrum. The calculated microwave spectra for 97% CO₂ and the values of α_{H_2O} and $\alpha_{H_2SO_4}$ indicated in Table 2 are displayed in Figs. 7-12 where the two solid curves labelled "Full" and "Effective" represent the calculated spectra for the two assumptions about the dielectric properties of H₂O-H₂SO₄ mixtures, and the dotted lines indicate the effect of changing the surface emissivity from 0.9 to 0.8. All of these results are consistent with an upper limit on $\alpha_{H_2O} \approx 10^{-3}$ and on $\alpha_{H_2SO_4} \approx 10^{-5}$. (The radar

results set an upper limit on $\alpha_{H_2O} \approx 4 \times 10^{-4}$; but because of the uncertainty in surface properties, this result has been given less weight.) These limits definitely rule out all pure water models such as that proposed by Vinogradov *et al.* (1971) and the "wet" model proposed by A. T. Young (1973).

6. Discussion

In this Section, the characteristics of those cloud models found to be consistent with observations will be discussed and compared to other observations of the Venus clouds.

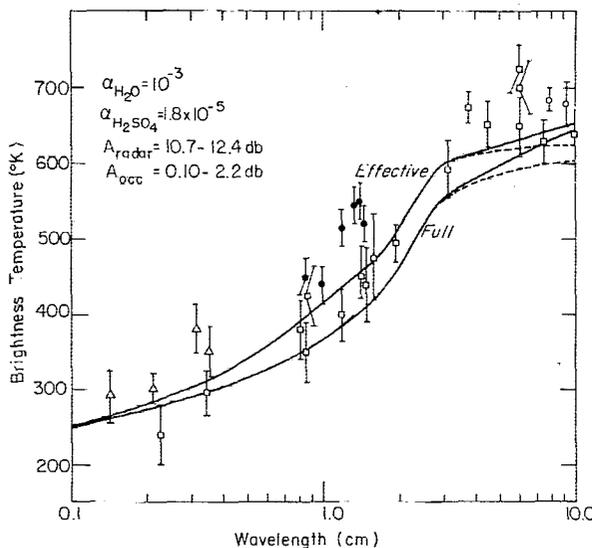


FIG. 8. Calculated microwave spectrum for cloud model with $\alpha_{H_2O} = 10^{-3}$ and $\alpha_{H_2SO_4} = 1.8 \times 10^{-5}$.

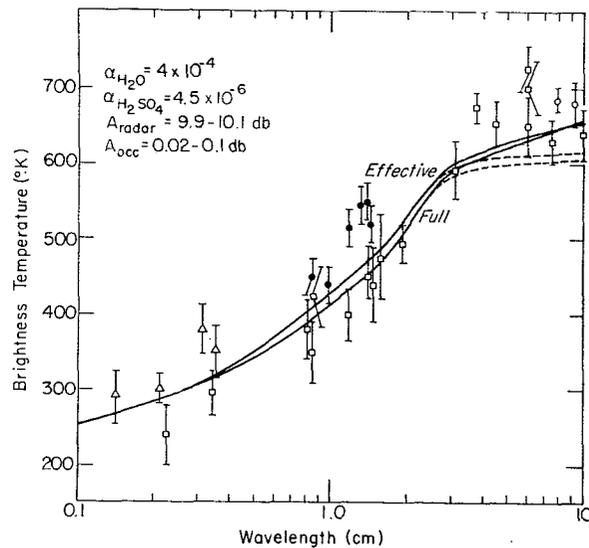


FIG. 10. Calculated microwave spectrum for cloud model with $\alpha_{H_2O} = 4 \times 10^{-4}$ and $\alpha_{H_2SO_4} = 4.5 \times 10^{-6}$.

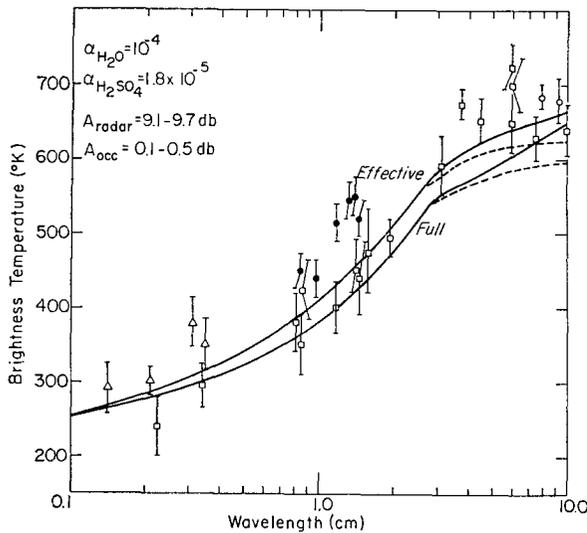


FIG. 11. Calculated microwave spectrum for cloud model with $\alpha_{H_2O} = 10^{-4}$ and $\alpha_{H_2SO_4} = 1.8 \times 10^{-5}$.

The water vapor mixing ratio above the clouds of Venus has been measured many times [see L. D. G. Young (1972) for a review of these measurements]. The range of number mixing ratios found for water is between 10^{-6} and 10^{-4} ; however, Fig. 5 shows that, for certain values of $\alpha_{H_2O}/\alpha_{H_2SO_4}$, the vapor mixing ratio of water may not be uniform with altitude because of the changing concentration of the droplets. Therefore a better measure of the water vapor abundance is the number of precipitable microns of water above a certain level. This has been shown to be time-variable about a mean value of $\sim 60 \mu m$ by Schorn *et al.* (1969). The major uncertainty left in comparing this number to a

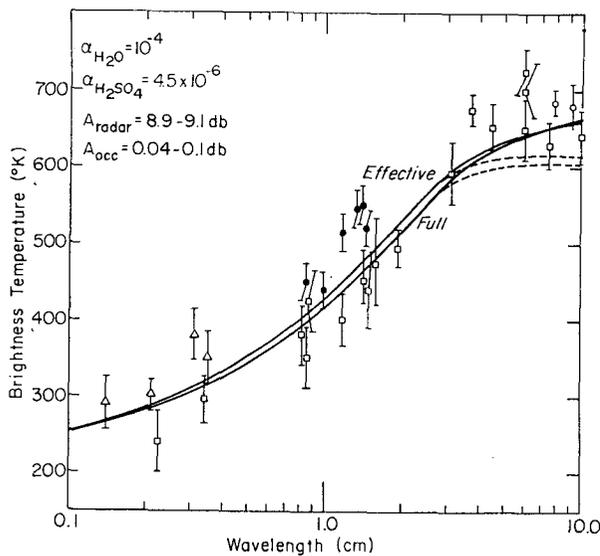


FIG. 12. Calculated microwave spectrum for cloud model with $\alpha_{H_2O} = 10^{-4}$ and $\alpha_{H_2SO_4} = 4.5 \times 10^{-6}$.

TABLE 3. Range of cloud models consistent with microwave data and derived values of α_v and x at reference level where $T = 250$ K.

Concentration x of H_2SO_4 by mass (%)	$\alpha_{H_2SO_4}/\alpha_{H_2O}$	Water mixing ratio α_v
~ 50 (maximum H_2O)	5×10^{-4}	4×10^{-3}
> 87 (maximum H_2SO_4)	2	3×10^{-6}
75	5×10^{-2}	2×10^{-4}
70	2×10^{-2}	4×10^{-4}
90	4	$< 10^{-6}$
95	10	$< 10^{-7}$

specific model is the uncertainty of the position of the reference level. For example, L. D. G. Young (1972) reports a 20–30 K spread in temperature for different CO_2 bands depending on the band strength.

The same problem occurs for the polarization measurements and their analysis by Hansen and Hovenier (1974) who conclude that the best fit to the data is provided by a 76% sulfuric acid solution at the 50 mb level of the atmosphere. This uncertainty in the position of the reference level also applies to an analysis of infrared data by Samuelson *et al.* (1974) who derive a number density of cloud droplets of $\sim 1000 \text{ cm}^{-3}$ and a number mixing ratio of $\sim 2 \times 10^{-6}$ for some level in the cloud.

These three quantities—water vapor abundance above the clouds, concentration of sulfuric acid in cloud droplets, and bulk content of the cloud (all referred to the same level in the atmosphere)—together with the microwave observations can be used to select consistent cloud models. Table 2 summarizes the upper limits on α_{H_2O} and $\alpha_{H_2SO_4}$ derived from the comparison of calculations and observations. The results should be compared to Fig. 6. The cloud with asymptotic water content 0.0075 gm m^{-3} is consistent with all data and thus any combination of α_{H_2O} and $\alpha_{H_2SO_4}$ which results in a cloud of approximately the same water content and bulk content will be consistent with the data as well. Since α_{H_2O} is constrained by the absence of a feature at 1.35 cm in the microwave spectrum, the maximum water cloud has $\alpha_{H_2O} = 4 \times 10^{-3}$ and $\alpha_{H_2SO_4} = 1.8 \times 10^{-6}$ (about half the mass of the cloud model

TABLE 4. Range of cloud models consistent with microwave data with H_2SO_4 concentration in droplets between 75–85% by mass and derived values of water vapor abundance above given reference level.

T reference (°K)	x (%) H_2SO_4 by mass	α_{H_2O}	$\alpha_{H_2SO_4}$	Vapor abundance (precipitable μm)
260	75	3×10^{-4}	9×10^{-6}	~ 370
260	85	4×10^{-5}	2×10^{-5}	~ 25
250	75	2×10^{-4}	9×10^{-6}	~ 180
250	85	3×10^{-5}	2×10^{-5}	~ 14
240	75	2×10^{-4}	5×10^{-6}	~ 90
240	85	2×10^{-5}	4×10^{-5}	~ 5
235	75	10^{-4}	2×10^{-6}	~ 30
235	85	2×10^{-5}	4×10^{-5}	~ 3

shown in Fig. 7). This model is equivalent to water vapor and an extremely tenuous cloud. The maximum sulfuric acid cloud has $\alpha_{\text{H}_2\text{O}} \leq 10^{-5}$ and $\alpha_{\text{H}_2\text{SO}_4} = 1.8 \times 10^{-5}$ (equivalent to the model shown in Fig. 11). This model has a pure sulfuric acid cloud as the major opacity source.

For a reference level temperature of 250 K, Table 3 shows the H_2SO_4 concentration in cloud droplets, the value of $\alpha_{\text{H}_2\text{SO}_4}/\alpha_{\text{H}_2\text{O}}$, and the water vapor mixing ratio α , for these two cloud models as well as for cloud models with differing H_2SO_4 concentrations. All of these cloud models have an asymptotic bulk water content of 0.0075 g m^{-3} , a total surface density of $\sim 0.1\text{--}0.3 \text{ g cm}^{-2}$, a liquid water surface density of $\sim 0.01\text{--}0.02 \text{ g cm}^{-2}$, and a droplet number density at 250 K (assuming $1 \mu\text{m}$ droplets) of $\sim 1000 \text{ cm}^{-3}$. This table illustrates the range of cloud models which is compatible with the observed mass and microwave properties of the clouds.

Table 4, on the other hand, illustrates the range of models compatible with the observed sulfuric acid concentration and microwave properties for different reference level temperatures and shows the derived abundance of water above the reference level in precipitable microns. The data of Schorn *et al.* (1969) imply that the sulfuric acid concentration in the droplets is probably higher than 75% and that the reference level temperature is $\lesssim 250 \text{ K}$.

All of these data together sharply restrict the possible values of $\alpha_{\text{H}_2\text{O}}$ and $\alpha_{\text{H}_2\text{SO}_4}$. If the abundance of CO_2 is 97% and the "full absorption" model approximates the dielectric properties of $\text{H}_2\text{O}\text{--}\text{H}_2\text{SO}_4$ mixtures, then the microwave data together with the observations discussed above imply $\alpha_{\text{H}_2\text{O}} = 2 \times 10^{-4}$ and $\alpha_{\text{H}_2\text{SO}_4} = 9 \times 10^{-6}$ for the clouds of Venus.

Sagan (1960, 1969) has suggested that the high surface temperatures on Venus are due to a $\text{CO}_2\text{--}\text{H}_2\text{O}$ greenhouse effect. Pollack (1969) calculates the required value of $\alpha_{\text{H}_2\text{O}}$, assuming water to be entirely in the vapor phase, to be $\sim 5 \times 10^{-3}$, a factor 20 above values derived here. However L. D. G. Young (private communication, 1974) finds that the opacity of CO_2 hot bands may have been underestimated in previous greenhouse calculations, lowering the required water content. In addition, liquid $\text{H}_2\text{O}\text{--}\text{H}_2\text{SO}_4$ mixtures are strong absorbers of infrared radiation in the 5 to 20 μm region (Palmer and Williams, 1975), and the total column density of these clouds, $\sim 0.1\text{--}0.3 \text{ g cm}^{-2}$ may be sufficient to provide the additional opacity needed for the greenhouse effect.

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