Reports

Venus Clouds: A Dirty Hydrochloric Acid Model

Abstract. *The spectral and polarization data for Venus are consistent with micrometer-sized aerosol cloud particles of hydrochloric acid with soluble and insoluble iron compounds, whose source could be volcanic or crustal dust. The yellow color of the clouds could be due to absorption bands in the near ultraviolet involving ferric iron and chlorine complexes. The ultraviolet features could arise from variations in the concentrations of iron and hydrochloric acid in the cloud particles.*

In spite of four successful penetrations of the atmosphere of Venus by Venera entry probes and two flybys by Mariner spacecraft, the question of the composition of the clouds of Venus remains controversial. Among the many materials that have been suggested as the dominant component of the clouds are H_2O liquid or ice, partially hydrated ferrous chloride, ammonium chloride, mercury halides, carbon suboxide, and dust (1–5).

Any proposed substance must be consistent with the following observations. Theoretical analyses by Hansen and Arking (6) of the polarization data of Coffeen and Gehrels (7) have provided extremely strong evidence concerning the physical nature of the cloud particles. The particles are spherical and, thus, are almost certainly liquid. They have indices of refraction of $1.45 \pm$ 0.02, and they exist in a narrow range of sizes, with radii close to 1 μm. Because of the detailed matching of the theoretical model to the observed curves for polarization as a function of phase angle at different wavelengths, it is unlikely that Hansen and Arking's conclusions are seriously in error.

The spectrum of Venus is shown in Figs. 1 and 3. The cloud particles absorb strongly in the near infrared in the region between 3 and 5 μm and also have absorption features in the visible from 0.4 to 0.6 μm and in the ultraviolet below 0.4 μm. For wavelengths longer than about 5 μm the radiation is primarily due to thermal emission, and it is difficult to know whether the spectral features are to be interpreted as absorption or emissivity bands. For wavelengths shorter than about 0.3 μm the radiation can be accounted for almost entirely by Rayleigh scattering from gas above the clouds, and little information can be obtained about the absorbing properties of the cloud particles in this spectral region (8). Several strong absorption bands of atmospheric CO_2 between 1 and 3 μm make it difficult to ascertain the exact spectrum of the clouds in this region of the near infrared.

The only gases in the atmosphere of Venus that have been detected spectroscopically from the earth are CO_2, H_2O, CO, HCl, and HF. The mixing ratios of the latter four gases, at the level in the atmosphere of Venus at which the CO_2 pressure is approximately 0.2 atm and the temperature is of the order of 250°K (altitude approximately 60 km), are of the order of 10^{-4} to 10^{-6}, 10^{-4}, 10^{-6}, and 10^{-8}, respectively (3, 9). By contrast, the Venera probes measured mixing ratios of 10^{-3} to 10^{-2} for H_2O. It is not clear how these conflicting sets of data are to be reconciled, except that the earth-based determinations of H_2O and CO_2 refer to different positions in the atmosphere and to different times.

The preceding observations argue against most of the proposed substances as the major constituent of the clouds. The requirement that the major constituent must be a liquid eliminates ice, ferrous chloride, ammonium chloride, and dust. The narrow size distribution implies that the substance is condensable, which argues against the chlorides and dust. Ferrous chloride has a prominent Fe^{2+} absorption band at 1.0 μm, which is not observed in the spectrum of Venus (10). Although H_2O liquid
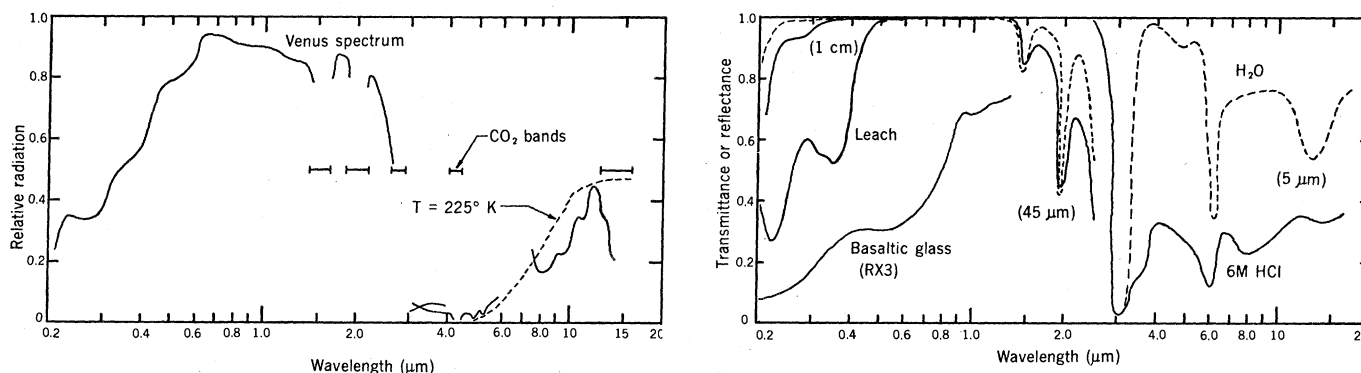

Fig. 1 (left). Relative reflectivity and thermal emission spectrum of Venus. Data from the following sources have been used to construct this spectrum: below 0.3 μm (8), 0.3 to 1.0 μm (30), 0.8 to 1.2 μm (10), 1.2 to 3.5 μm (2, 32), 3.0 to 14 μm (33). The near-ultraviolet and near-infrared values have been normalized to the Bond albedo values of Irvine *et al.* (30); the far-infrared curve has been arbitrarily joined to the near-infrared curve at 3.2 μm. The bars indicate the positions of strong CO_2 bands in the atmosphere of Venus. A strong H_2O band in Earth's atmosphere occurs at 6 to 7.5 μm, and an ozone band occurs at 9.8 μm. Fig. 2 (right). Transmission spectra of H_2O and $6M$ HCl. The absorption cell thicknesses are, for 0.2 to 0.7 μm, 10 mm; for 0.7 to 2.5 μm, 0.04 mm; for 2.5 to 16 μm, 0.005 mm. The data for 2.5 to 16 μm were adapted from (13); the data for 0.2 to 2.5 μm were measured on a Cary 14 spectrophotometer. Also shown are the reflection spectrum of a powder of an artificial basaltic glass of composition approximating that of Apollo 11 lunar samples (this spectrum has been multiplied by 3) and the transmission spectrum of a solution resulting from leaching this glass with $6M$ HCl for 60 hours (cell thickness, 1.0 mm).

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and ice, ferrous chloride, and ammonium chloride absorb strongly at 3.0 μm, they become translucent again past 4.0 μm, and clouds consisting of micrometer-sized particles of these substances would be bright at long wavelengths. The spectra of C_3O_2 and mercury halides do not match the detailed spectrum of Venus (3, 5). Of all the proposed substances, only C_3O_2 has a refractive index close to 1.45. The albedos of volcanic dusts are too low for the clouds to consist primarily of this material.

The pressure of H_2O vapor in the atmosphere, as determined from observations from the earth, is too low by significantly more than an order of magnitude for it to be in equilibrium with H_2O liquid or ice at 250°K. However, if the Venera mixing ratios are correct, water clouds would have no difficulty in forming. Recent analyses of the Mariner 5 radio occultation data (11) indicate that the temperature profile of the atmosphere of Venus is consistent with the presence of a condensing component at altitudes of 60 to 75 km. With a mixing ratio of 10^{-4} for H_2O, water clouds could exist at an altitude of 75 km (temperature about 210°K and pressure about 10^{-2} atm). The haze or cloud layer apparently extends to 120 km (12); between 75 and 120 km the temperature may be well below 200°K (11, 12), and water clouds would be stable for mixing ratios as low as 10^{-5}.

The spectra of water and 6M hydrochloric acid are shown in Fig. 2. Except for minor differences the spectrum of ice is similar to that of liquid H_2O. The changes caused by the addition of HCl are due mainly to the hydronium ion OH_3^+ in the acid (13), so that the addition of any strong acid to water results in a similar spectrum. Strong bases, but not salts, have a somewhat similar, but less pronounced, effect. The most striking change is the series of wide, overlapping OH_3^+ bands, which keep the transmissivity of the acid solution low over the entire range of wavelengths from 3 μm to beyond 16 μm. Thus, the low reflectivity of Venus in the range from 3 to 5 μm would be accounted for if OH_3^+ ions are abundant in the clouds. Lewis (4) has shown that the relative abundances of HCl and H_2O vapor observed in the atmosphere of Venus require that any liquid in equilibrium with these gases must be a hydrochloric acid solution of the order of 25 percent by weight (about 5 to 7M). Terrestrial volcanic exhalations contain abundant SO_2. Therefore, pos-
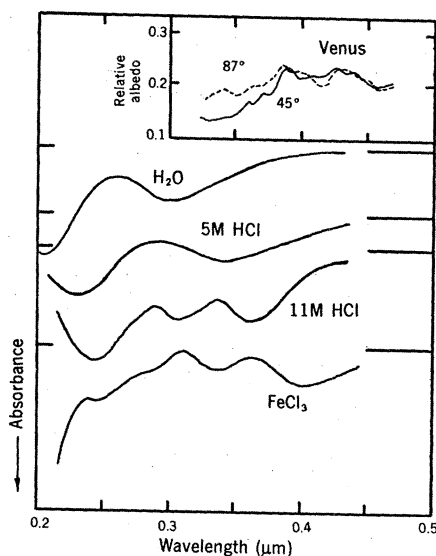
Fig. 3. Near-ultraviolet spectrum of Venus and absorption spectra of ferric ions. (Top) Relative reflectivity spectrum of Venus by Glushneva (22), as reduced by Cruikshank and Thompson (10). (Bottom) Absorption spectra of Fe^{3+} ions in various hydrochloric acid solutions (34) and of $FeCl_3$. (The absorbance is proportional to the negative logarithm of the transmittance. To avoid confusion, the absorbance spectra have been displaced in the vertical direction by arbitrary amounts.) The spectrum of Fe^{3+} in H_2SO_4 solutions is similar to the H_2O curve.

sible alternatives to HCl could be H_2SO_3 or H_2SO_4. However, SO_2 vapor has not been detected spectroscopically on Venus. Thus, the most probable additive to the clouds on Venus is HCl.

Rea and O'Leary (14) have objected to ice and water clouds on Venus on the grounds that the bands at 1.5 and 1.9 μm are not visible in the spectrum of the planet. This objection does not apply to the present model because the bands would be quite weak for clouds composed of particles as small as 1 μm and also because the addition of HCl decreases and widens the bands.

At 8 μm the acid spectrum has a broad band that is not present in the water spectrum that corresponds to a minimum in the thermal spectrum of Venus.

Although a detailed scattering calculation remains to be done, the infrared spectrum of hydrochloric acid, in contrast to that of pure H_2O liquid or ice, appears to be compatible with the spectrum of Venus. If this identification is correct, ice clouds are probably much rarer on Venus than on the earth, since the addition of HCl to water depresses the freezing temperature to below 200°K for 6M hydrochloric acid (4).

Hydrochloric acid has an extremely

weak absorption band at 0.28 μm. However, none of the observed constituents of the atmosphere of Venus absorb appreciably in the region from 0.3 to 0.6 μm. Therefore, it is necessary to resort to indirect arguments to account for the yellowish color of the clouds. Terrestrial clouds and aerosols form by nucleation onto both soluble and insoluble substances and thus are not composed of pure H_2O. The compositions of the nuclei of terrestrial clouds are not well known, but analyses (15–17) of aerosol residues reveal three types of substances: NaCl from ocean spray, ammonium and sulfate ions and other materials primarily from industrial sources, and natural dust from volcanoes and the crust. Because of unfavorable surface conditions, the first two sources are presumably not present on Venus. However, because of the high temperature of the surface, volcanoes should be even more prevalent on Venus than on the earth. Volcanic dust has been estimated to reach altitudes greater than 60 km on the earth (18, 19). Other finely divided solids from the crust could be carried into the stratosphere by turbulence.

Residues of terrestrial stratospheric aerosols include Si, S, and Fe (15); no Ni is detected, which indicates that the source of the Fe is not meteoritic. Any volcanic dust finding its way into the stratosphere of Venus would be attacked by the hydrochloric acid clouds. Volcanic glass is especially susceptible to acid leaching because of its high density of solid-state defects. It has been shown that glass in lunar soil is strongly attacked by hydrochloric acid and that all the cations that are abundant in the lunar soil are also present in the solute (20). Thus, if the crust of Venus is similar to the crusts of the earth and the moon, ions of the following elements should be found as solid nuclei and in solution in the aerosols of Venus: Si, Al, Ca, Fe, Mg, Na, K, and Ti and smaller amounts of Cr, Mn, and V. Because of the much greater chemical activity of hydrochloric acid compared with water, the concentrations of these ions should be much greater in Venusian than in terrestrial aerosols.

The coloration of terrestrial and lunar rocks and minerals is due almost entirely to absorption bands associated with the transition elements Fe, Ti, Cr, Mn, and V, with Fe being by far the most important because of its geochemical abundance. The Fe^{3+} ion has strong absorption bands in the spectral region from 0.2 to 0.4 μm, and the yel-

low or red color of most iron compounds is due to these bands. The spectra of several iron-bearing materials are shown in Figs. 2 and 3.

Although the atmosphere of Venus is slightly reducing, some Fe^{3+} should be present, either because of oxidation by the oxygen formed in the photodissociation of CO_2 or from thermal oxidation reactions of the type $FeO + CO_2 \rightleftharpoons Fe_2O_3 + CO$, which would take place at the surface (21). Using a simple two-stream scattering model for an optically thick cloud layer and the absorbances of ferric salt solutions measured on a Cary 14 spectrophotometer, I estimate that Fe^{3+} concentrations of the order of $0.1M$ should account for the edge at $0.4 \mu m$ in the spectrum of Venus and for the yellow color of the clouds of Venus. The ferric band at $0.24 \mu m$, shown in Fig. 3, is probably unobservable because of atmospheric Rayleigh scattering. Ferrous iron, Fe^{2+}, should also be abundant in the clouds of Venus. However, this ion has only a weak band at $1.0 \mu m$, and much more ferrous than ferric iron could be present without causing a detectable band.

The position of the ferric bands in the near ultraviolet depends on the type and normality of the solvent. In water or in weak hydrochloric acid the ferric ion is complexed by H_2O molecules, which results in a band at $0.30 \mu m$. As the HCl concentration increases, complexes of $FeCl^{2+}$ and $FeCl_2^+$ become significant and the band shifts to $0.34 \mu m$. At concentrations above about $6M$ HCl the dominant complex is $FeCl_4^+$, and the band splits into two components centered at 0.31 and 0.36 μm. In $FeCl_3$ salt the band is at $0.40 \mu m$. In H_2SO_4 the ferric band is at $0.30 \mu m$.

The reflection spectrum of Venus is poorly known in the ultraviolet. However, a spectrum obtained by Glushneva at a phase angle of $45°$ (22), shown in Fig. 3, is suggestive of a minimum at $0.34 \mu m$. More high-precision observations of Venus in the near ultraviolet are necessary. Such observations could confirm the presence of Fe, HCl, and liquid H_2O in the clouds of Venus and indicate their abundances and the type and normality of the acid.

When they are surrounded by anions, the cations of all the transition elements have absorption bands in the visible and near infrared due to d-electron transitions (23). For instance, Ti^{3+} has a band at $0.50 \mu m$. However, these bands are relatively weak, and it is doubtful

that any of them could account for the minimum at $0.5 \mu m$ in the spectrum of Venus, without a prominent Fe^{2+} band at $1.0 \mu m$. Much stronger electron-transfer bands can occur in the visible when two multivalent ions of transition elements are sufficiently close together that their d orbitals overlap (24). The intensity of this type of band depends on the product of the concentrations of the ions; the wavelength depends on the degree of overlap and on the nature and structure of the surrounding ions.

Because of the abundance of Fe, the band at $0.5 \mu m$ in the spectrum of Venus is probably a $Fe^{2+}-Fe^{3+}$ electron-transfer band, although if Ti is as abundant in the crust of Venus as it is in the lunar maria, a $Fe^{2+}-Ti^{4+}$ band may also be involved. In the Venusian aerosols, conditions appropriate for electron-transfer bands probably would not occur in the aqueous state because the solutions would have to be too concentrated. More favorable conditions would occur in precipitates involving iron complexed with oxygen, ammonia, chloride, carbonate, or other ions, and in the insoluble portion of the aerosol nuclei. An example of a solid-state, electron-transfer band in the visible spectrum of an artificial basaltic glass containing Fe^{2+}, Fe^{3+}, and Ti^{4+} is shown in Fig. 2. In this spectrum the tails of strong bands of Fe^{3+} and Ti^{4+} in the far ultraviolet are also prominent, but the Fe^{2+} band at $1.0 \mu m$ is extremely weak, and no band is present at $0.34 \mu m$.

The index of refraction of the upper Venusian clouds is close to 1.45 (6), whereas the index of refraction of a $6M$ HCl solution is approximately 1.39 and is only weakly dependent on temperature (25). However, most solutes raise the index of refraction of an aqueous solution by 0.01 to 0.03 for each mole of the dissolved material per liter of water (26). For instance, a $6M$ HCl solution containing 1.7 moles of dissolved $FeCl_3$ per liter of solution has an index of refraction of 1.45. Thus, if the Venusian aerosols contain dissolved salts in concentrations of a few moles per liter, their index of refraction would have the required value. Exactly the same situation occurs in terrestrial water aerosols. Water has an index of refraction of 1.33; however, the scattering properties of the aerosol particles can be accounted for by Mie theory if a value of 1.50 rather than 1.33 is used (16). The large refractivity is due to both dissolved salts and the insoluble portion of the nuclei. Thus, a

value of 1.45 for Venusian hydrochloric acid aerosols is eminently reasonable.

The partial pressure of H_2O vapor in a gas containing suspended aerosols will in general not be the same as the tabulated value of the vapor pressure, which refers to the equilibrium partial pressure over a flat, infinite surface of pure water. The growth of aerosol particles and their equilibrium vapor pressure depend on a number of factors, including the size, shape, and wettability of the nuclei, the surface tension, and the concentration of dissolved salts (15, 17). In particular, the partial pressure of water in a solution is reduced in proportion to the mole fraction of solute, in accordance with Raoult's law, so that it is possible for aerosols to grow under such conditions that the partial pressure of water is less than half the vapor pressure at that temperature. However, this effect is probably not sufficient to reconcile the Venera and earth-based measurements, nor would it allow the formation of water aerosols if H_2O mixing ratios of 10^{-6} are typical.

To the eye the disk of Venus is featureless, but when the planet is observed at wavelengths below $0.4 \mu m$ large diffuse markings, which persist for several days or weeks, become visible. Since the apparent period of rotation of the ultraviolet features is of the order of 5 days, while the solid body of the planet rotates with a period of 243 days, the markings are obviously due to atmospheric rather than surface features. Dollfus (27) and Kuiper et al. (28) have published a large number of photographs of these clouds. A natural explanation in terms of the model discussed here is that the markings reflect variations in the intensity of the ferric band.

Several lines of evidence indicate that the ultraviolet features are associated with a partially transparent or incomplete cloud or layer of haze, which scatters light but absorbs only weakly in the ultraviolet and which overlies a cloud deck that absorbs more strongly in the ultraviolet.

1) The ultraviolet features are seen more often near the center of the disk than near the polar or equatorial limbs.

2) The markings occur more frequently at small phase angles than at large ones (29).

3) The ratio of the reflectivity of Venus in the ultraviolet to that in the visible is considerably higher at large than at small phase angles (30).

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4) The minimum at 0.34 μm, which is apparent in Glushneva's spectrum at a phase angle of 45°, seems either to have disappeared or to have shifted to shorter wavelengths in the spectrum at 87°.

A decreased strength of the ferric band in the near ultraviolet could be due to lower concentrations of either Fe^{3+} or HCl, or both, in the upper layer of haze. Such a decrease may reflect more favorable conditions for the condensation of H_2O vapor at higher altitudes, due to the lower temperatures. Alternatively, larger Fe^{3+} concentrations at lower altitudes could be due to increases in the supply of silicate dust from volcanic eruptions on the surface. Physically, the ultraviolet features could represent holes; local deficiencies of iron in the upper clouds; or local concentrations of iron, possibly related to the surface distribution of active volcanoes, in the lower clouds.

In the far-infrared spectrum of Venus, three minima appear to be present at 8, 9.5, and 11 μm. The depression at 9.5 μm is probably due to a CO_2 band at 9.4 μm, although there is no sign of a minimum at 10.4 μm, where another CO_2 band is located. The 8-μm feature may be an absorption band of hydrochloric acid. The minimum at 11 μm may be associated with silicate aerosol nuclei. Silicates possess emissivity minima in the range from 9 to 11 μm due to the Si-O vibration band. The location of the band depends on the composition; it is shifted toward 9 μm for acidic rocks and toward 11 μm for ultrabasic silicates. Carbonates also have an emissivity minimum at 11 μm (31).

In order for silicate bands to be seen in emission, silicates would have to be abundant at the level from which the thermal radiation originates. However, the hydrochloric acid band must be seen in absorption, which requires a relatively lower silicate and higher HCl concentration at greater altitudes. Thus, the ultraviolet markings and the thermal infrared bands may be closely associated, and simultaneous monitoring of Venus in these two spectral regions may prove highly informative.

Finally, it must be stressed that these conclusions apply only to the visible clouds. Other substances may be important in the lower atmosphere.

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Photolysis of Formaldehyde as a Hydrogen Atom Source in the Lower Atmosphere

Abstract. *The rates of formaldehyde photodecomposition into hydrogen and formyl radicals and hydrogen and carbon monoxide molecules in sunlight-irradiated atmospheres have been estimated from extinction data and photochemical results. These data should prove useful in the development of models for the chemical changes that take place in the polluted atmosphere.*

Formaldehyde is a common product of the incomplete combustion of hydrocarbons. It is a significant minor product present in atmospheres polluted with auto exhaust where it amounts to some 40 to 50 percent of the total aldehydic compounds from this source (1). Formaldehyde may be introduced directly into the atmosphere or formed in the atmosphere by a variety of reactions; these include reactions of ozone with terminal olefins and reactions of oxygen with alkyl and alkoxyl radicals. The highest recorded total aldehyde concentration from data available up to 1959 was 1.9 parts per million (ppm) reported in Los Angeles (2). Typical CH_2O concentrations in Los Angeles in 1961 were in the range 2 to 19 parts per hundred million (pphm) (1). The natural processes for the removal of CH_2O in the atmosphere are of increasing concern. In particular, the rate of sunlight photolysis of CH_2O vapor in

Table. 1. Quantum yields ϕ for the primary photochemical processes in the photolysis of CH_2O as a function of wavelength and molar extinction coefficients ϵ of CH_2O vapor integrated over 100-Å bands centered at the given wavelengths.

Wavelength (Å)	$\bar{\phi}_1$	$\bar{\phi}_2$	$\bar{\epsilon}_\lambda$ (liter mole^{-1} cm^{-1}) for 100-Å band
2900	0.81	0.19	8.33
3000	.66	.34	8.51
3100	.52	.48	8.23
3200	.40	.60	6.13
3300	.29	.71	6.19
3400	.18	.82	5.17
3500	.09	.91	2.19
3600	.01	.99	0.46