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Chemical composition of Venus atmosphere and clouds: Some unsolved problems

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Abstract

In spite of many spacecrafts that visited Venus, chemical composition of the Venus atmosphere and clouds present many challenging problems in observation and theory. The following problems are briefly discussed below: (1) molecular oxygen above the clouds, (2) lightning, (3) the blue absorption in the clouds, (4) mode 3 particle controversy and the Vega X-ray fluorescent observations, (5) search for new chlorine and sulfur species, and (6) vertical and spatial variations of water vapor and CO.

Keywords: Venus, Atmosphere; Venus, Clouds; Venus, Spectroscopy; Venus, Chemical composition

1. Introduction

Despite numerous spacecraft that visited Venus, chemical composition of the Venus atmosphere and clouds present many challenging problems. Some of these problems will be discussed below in arbitrary order. Our discussions are actually brief indications of the problems, and their more detail and thorough considerations are given in the original papers which are reviewed in *Venus* (1983), *Venus II* (1997), and Krasnopolsky (1986).

2. Molecular oxygen above the clouds

Each solar photon at $\lambda < 200$ nm and 200–218 nm impacting Venus is absorbed by CO_2 and SO_2, respectively. The solar photon fluxes are 2×10^{13} and 8.5×10^{13} ph cm^{-2} s^{-1} at the Venus orbit, respectively, and the global-mean column photolysis rates of CO_2 and SO_2 are quarters of these values. Almost all oxygen atoms formed are initially converted into O_2 by the three-body association and some catalytic reactions. The total column rate of the O_2 production is $\sim 10^{13}$ cm^{-2} s^{-1}, and one might expect significant quantities of O_2 in the stratosphere of Venus.

However, O_2 has not been found on Venus, and the most restrictive observations were made by Trauger and Lunine (1983). (O_2 cannot exist in the hot reducing lower atmosphere of Venus, and some data on the presence of O_2 there are erroneous.) They used a triple pressure-scanned Fabry–Perot interferometer to search for a Doppler-shifted line of the O_2 band at 762 nm. The instrument had a resolving power $\lambda/\delta\lambda = 2.4 \times 10^5$. No absorption was found, and a 2-sigma upper limit to the equivalent width of the P9 line was 2×10^{-6} nm, that is, 3.5×10^{-5} cm^{-1}. Then Trauger and Lunine (1983) applied a radiative transfer code to this line assuming the uniformly mixed O_2 and derived an upper limit of 0.3 parts per million (ppm) to the O_2 mixing ratio.

Though this upper limit was calculated for the uniformly mixed oxygen, it is actually weighted to a so-called level of line formation which is in the clouds at $\tau(1-g) = 1$. Here τ is the cloud optical depth and $g \approx 0.75$ is the scattering asymmetry factor. This level is at 62 km in the atmosphere. All photochemical models (Krasnopolsky and Parshev, 1981, 1983; Yung and DeMore, 1982; Mills, 1998) predict

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highly variable vertical distribution of O_2 with the bulk oxygen above 62 km.

Therefore, the model with the mixed oxygen is not the best presentation of the upper limit to O_2 on Venus. A simple reflection model is more adequate. The solar light undergoes a two-way absorption in the atmosphere above a reflecting surface at 62 km in this model. The phase dependence of Venus near 762 nm (Irvine, 1968) shows that the Lambert reflection is a reasonable approximation for Venus at this wavelength. (Brightness is proportional to cosine of solar zenith angle and does not depend on viewing angle for the Lambert reflection.) The observations of Trauger and Lunine (1983) were made for the light from the whole disk at phase (Sun–planet–Earth) angle of 90°. Then integration over the sunlit hemisphere of Venus results in a total airmass factor of $3\pi/2 \approx 4.7$. According to the spectroscopic database HITRAN 2004, the strength of the P9 line is 9×10^{-24} cm at 200–250 K. Therefore the 2-sigma upper limit of $3.5 \times 10^{-5}\,\text{cm}^{-1}$ to the line equivalent width results in the O_2 column abundance of $3.5 \times 10^{-5}/(1.5\pi \times 9 \times 10^{-24}) \approx 8 \times 10^{17}\,\text{cm}^{-2}$.

A more recent search for O_2 on Venus (Mills, 1999a) gave a less restrictive upper limit of 3 ppm to O_2 uniformly mixed above 58 km. The limit to the O_2 line equivalent width was 1.2×10^{-5} nm, above the limit of Trauger and Lunine (1983) by a factor of 6, although the instrument resolving power was better by a factor of 2.5. This is because the entrance aperture was 2.2 arcsec^2 while the Fabry-Perot interferometer collected the light from the whole sunlit part of Venus, which was 220 arcsec^2 in the observations by Trauger and Lunine (1983).

Another side of the O_2 problem is that the photochemical models generally disagree with the upper limit to O_2 (Table 1). The latest models by Mills (1998) with the updated kinetic data are still above the observed upper limit even for some deviations from the standard rate coefficients (one-sigma model, see also Pernice et al. (2004)). Mills (1998) also adopted eddy diffusion higher by an order of magnitude than that recommended by von Zahn et al. (1980), Gerard et al. (1981), and Krasnopolsky (1986). The high eddy diffusion facilitates transport of O_2 to a region near 70 km where O_2 is effectively destroyed. Therefore, the high eddy diffusion helps to reduce the column O_2 abundance. Thus, both detection of O_2 and its photochemical interpretation present challenging problems.

Table 1
Column abundance of O_2 in photochemical models

Models	O_2, cm^{-2}	O_2
Krasnopolsky and Parshev (1981)	4×10^{18}	5
Yung and DeMore (1982), model C	1.5×10^{19}	19
Mills (1998), Nominal model	8×10^{18}	10
Pernice et al. (2004), One sigma model	2×10^{18}	2.5

The last column gives ratios of the calculated O_2 to the measured O_2. The measured O_2 is the upper limit of $8 \times 10^{17}\,\text{cm}^{-2}$ derived in this work from the observation by Trauger and Lunine (1983).

3. Lightning

Electric signals detected by the Venera 11–14 landing probes (hereafter V11–14), the Pioneer Venus (PV) orbiter, and during the Galileo flyby of Venus have been interpreted as originating from lightning (see Russel (1991), Grebowsky et al., 1997 and references therein). Optical data on lightning are more controversial.

The V9 and 10 optical spectrometers revealed one flashing region (Krasnopolsky, 1983); therefore, the flashing rate and lightning energy deposition based on these data are highly uncertain. The mean flash duration is 0.25 s (Fig. 1), similar to that on the Earth. However, no strokes were detected within the flashes. Maybe, the number of strokes is significantly larger than 4 on the Earth and individual strokes could not be resolved by the instrument. The mean light energy of a flash in the visible range is 2×10^7 J, not very different from the flashes on the Earth. There is no significant depletion in the spectrum of lightning below 550 nm by the Rayleigh extinction, and this rules out lightning in the lowest 20 km from volcanic eruptions etc. The observed spectrum agrees with the simulated spectrum (Borucki et al., 1996) which consists mostly of a strong continuum and a few lines. An increase in the simulated spectrum below 450 nm because of the CN bands is compensated in the observed spectrum by the blue absorption in the upper cloud layer. The O 777 nm line is not prominent in the observed spectrum because of the low sensitivity (2% of the peak value) and random flashing.

A search for lightning by the PV star sensor (Borucki et al., 1991) had some severe restrictions, and the observations were possible just for 83 s in two sessions. No flashes were observed with an upper limit of $3\,\text{km}^{-2}\,\text{y}^{-1}$. Similarly, a search by the photometers on the Vega 1 and 2 balloons (Sagdeev et al., 1986) was negative despite the total observing time of 60 h in the nighttime conditions.

Ground-based search for lightning on the Venus night side (Hansell et al., 1995) revealed seven flashes per ~4 h of the total exposure. Based on the early photographic spectra of simulated lightning, the authors adopted that 40% of the total light energy is emitted in the O 777 nm line which they used for the detection of lightning. Later photoelectric spectra (Borucki et al., 1996) showed a dominant contribution of the continuum which reduces the O 777 nm line to ~3%. Therefore, the detected flashes had a light energy exceeding 1.2×10^9 J, two orders of magnitude above the mean light energy, and it is difficult to compare the observed very low flashing rate with that from the other sources. Anyway, this experiment also confirms lightning on Venus.

The recent detection of NO (Krasnopolsky, 2006) in the lower atmosphere of Venus is a convincing and independent proof of lightning. The measured NO absorption

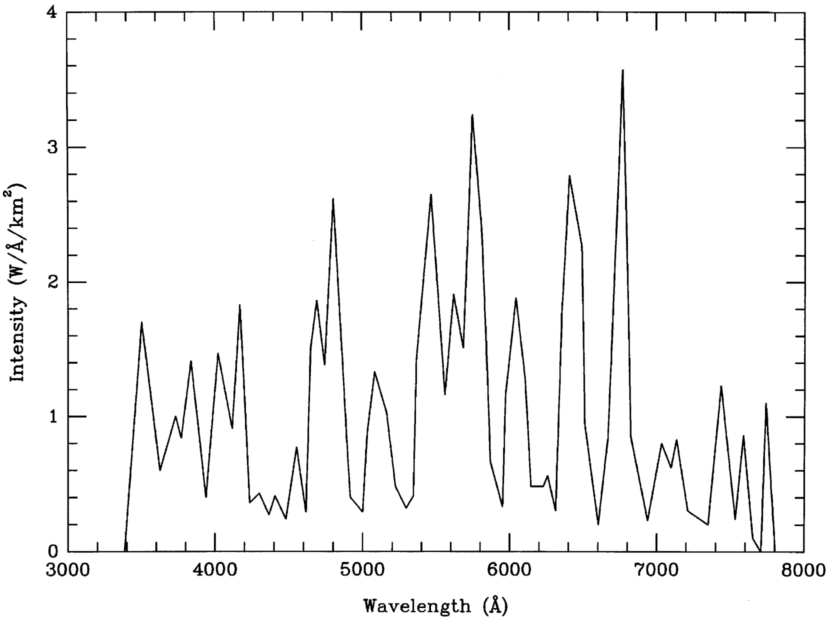

Fig. 1. One of seven spectra of lightning observed by V9. Four hundred and eighty points of the spectrum from 3000 to 8000 Å were consecutively measured in 10 s; therefore, the spectrum also reflects a temporal behavior of the thunderstorm region. A mean duration of the flashes is 0.25 s and their light energy is 2×10^{7} J (Krasnopolsky, 1983).

corresponds to a mixing ratio of 5.5 + 1.5 ppb below 65 km. Lightning is the only source of NO in the lower atmosphere, and the appropriate lightning energy deposition of $0.19\pm0.06\,\mathrm{erg\,cm^{-2}\,s^{-1}}$ results in a global flashing rate of $\sim 90\,\mathrm{s^{-1}}$ for a mean flash energy of 10^{9} J. This rate is in a reasonable agreement with the other optical data.

The measured NO abundance is smaller by a factor of 5 than that suggested by Yung and DeMore (1982) in their model B. NO in that model was a basic catalyst which provided the balance of the major photochemical products on Venus. Despite the observed smaller NO abundance, the photochemical impact of NO is significant in the atmosphere above the clouds.

Lightning may be searched on Venus from an orbiter using a light and cheap device. This device could be designed to measure the basic properties of lightning and to close some existing uncertainties in the problem of lightning on Venus.

4. Blue absorption in the clouds

The blue absorption in the Venus clouds (Fig. 2) is known for a long time, and it is responsible for the yellowish color of Venus. The detection of SO_2 (Esposito et al., 1979; Barker, 1979) explained the absorption below 320 nm while an absorber at 320–500 nm is a subject of discussion here. Near UV marking and contrasts observed by cameras from the Venus orbiters and ground-based telescopes are due to this absorber. The observed correlation between the absorber and SO_2 may be explained (Esposito et al., 1983) by either dynamics (downward and upward flows in the clouds) or chemistry. The latter could mean that the absorber is a product of the SO_2 chemistry.

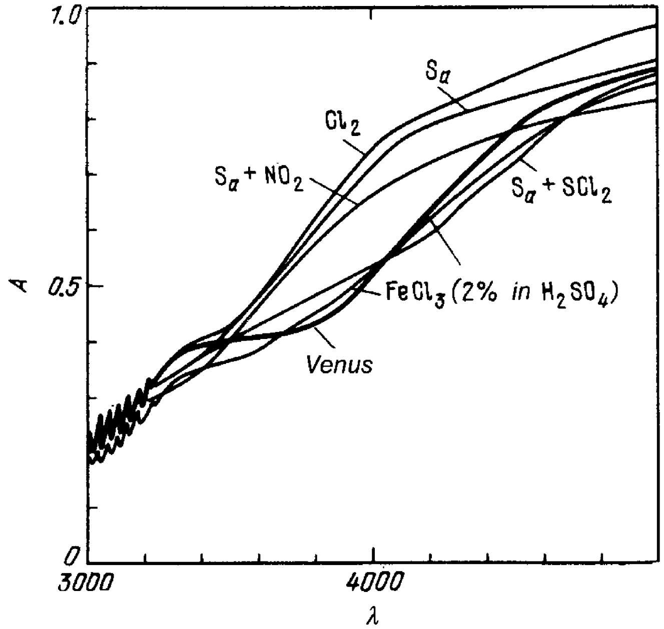

Fig. 2. Venus albedo at 3000–5000 Å (Barker et al., 1975, solid line) is compared with calculated albedo for various absorbers. S_a is the aerosol sulfur. From Zasova et al. (1981).

Photometric observations from the V14 landing probe (Ekonomov et al., 1983) showed that the true absorption at 320–390 nm exceeds that at 490–730 nm by more than a factor of 30 in the clouds at 58 to 62 km. The absorber is therefore distributed in the upper cloud layer. The measured absorption at 320–390 nm below 58 km is explained by SO_2 (Ekonomov et al., 1983) and results in the SO_2 abundances of 10, 20, and 50 ppm at 57, 54, and

50 km, respectively (Krasnopolsky, 1986). There are some other evidences (see Esposito et al., 1983, 1997 and references therein) that the absorber is at the altitudes of the upper cloud layer (57–70 km).

Many candidates were suggested for the absorber (see Esposito et al., 1983, 1997; Krasnopolsky, 1986). Some of them (aerosol sulfur, Cl_2, SCl_2, S_2O) are photochemical products and should exist in the Venus atmosphere. However, these species (except SCl_2) poorly match the observed absorption (Fig. 2), and the proposed abundances of Cl_2, SCl_2, and S_2O exceed those in the photochemical models by two orders of magnitude. Sulfur allotropes S_3 and S_4 proposed as the required absorber are quickly removed by photolysis in the visible range. All these species may contribute to the absorption, though their contributions are minor. Difficulties associated with more exotic absorbers like ammonium pyrosulfite $(NH_4)_2S_2O_5$, nitrosylsulfuric acid $NOHSO_4$, and perchloric acid $HClO_4$ are discussed by Krasnopolsky (1986).

Solution of ~1% $FeCl_3$ in sulfuric acid (Zasova et al., 1981) is the best candidate to the absorber (Fig. 2). The surface reflectivity at the V9 and 10 landing sites (Pieters et al., 1986) and the color images of the V13 and 14 landing sites favor the presence of ferric rocks, though ferrous rocks are more thermodynamically stable on Venus. Analysis of the X-ray fluorescent spectroscopy from the V12 probe (Petryanov et al., 1981a, b) revealed both the Cl 2.62 keV and Fe 6.9 keV peaks with an intensity ratio that exactly matched $FeCl_3$. If a mixing ratio of Fe_2Cl_6 is equal to 15 ppbv in a deep atmosphere, then iron chloride condenses at 47.5 km (Krasnopolsky, 1986) and its properties exactly match (Fig. 3) those of the mode 1 particles in the lower and middle cloud layers (Knollenberg and Hunten, 1980). This mode is of different origin (probably sulfur and/or sulfuric acid) in the upper cloud layer.

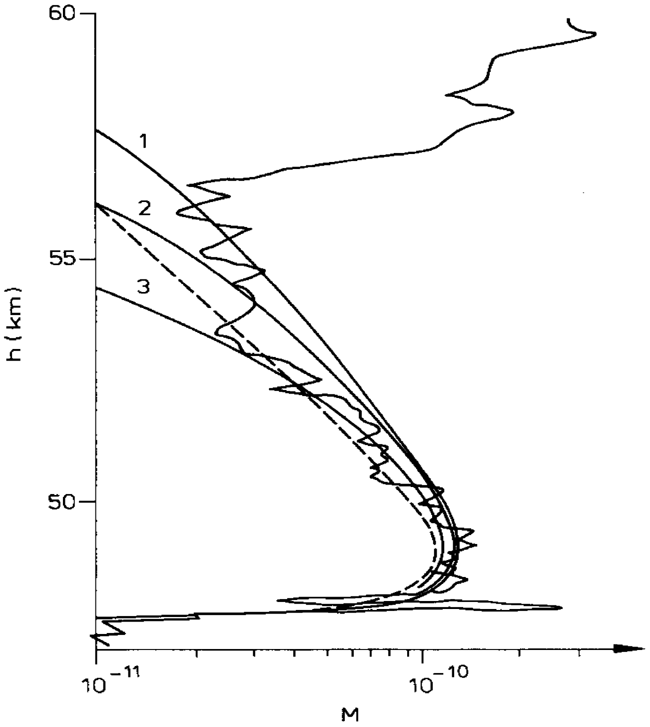

Fig. 3. Vertical profiles of the $FeCl_3$ aerosol mass loading M (in $g\,cm^{-3}$) calculated for the upward fluxes of the aerosol of (0.5, 1, and 2) $\times 10^{-12}\,g\,cm^{-2}\,s^{-1}$ (curves 1, 2, 3) are compared with the measured mass loading of mode 1 corrected for the higher density of $FeCl_3$. Profile for $K = 10^4\,cm^2\,s^{-1}$ and the zero flux is also shown (dashed curve). From Krasnopolsky (1986).

The iron chloride particles may be condensation centers for sulfuric acid which forms in the upper cloud layer. Then concentration of the $FeCl_3$ solution is ~ 1% in the mode 2 particles close to that required to explain the blue absorption. The reaction between $FeCl_3$ and concentrated H_2SO_4 is slow at temperatures of below 280 K typical of the clouds above 58 km, and the lifetime of the solution is close to the precipitation time of 1 month in the upper cloud layer. Colorless ferric sulfate replaces $FeCl_3$ near the bottom of the upper cloud layer, and ferric sulfate restores iron chloride in the reaction

$$Fe_2(SO_4)_3 + 6HCl + 3CO = Fe_2Cl_6 + 3H_2O + 3SO_2 + 3CO_2$$

in the lower atmosphere and on the surface (Krasnopolsky, 1989). This is the end of the atmospheric cycle of iron chloride.

5. Mode 3 particle controversy and Vega X-ray fluorescent observations

Mode 3 (Fig. 4) was observed by the PV cloud particle spectrometer as large particles with radii of ~3.5 μm in the middle and lower cloud layers (Knollenberg and Hunten,

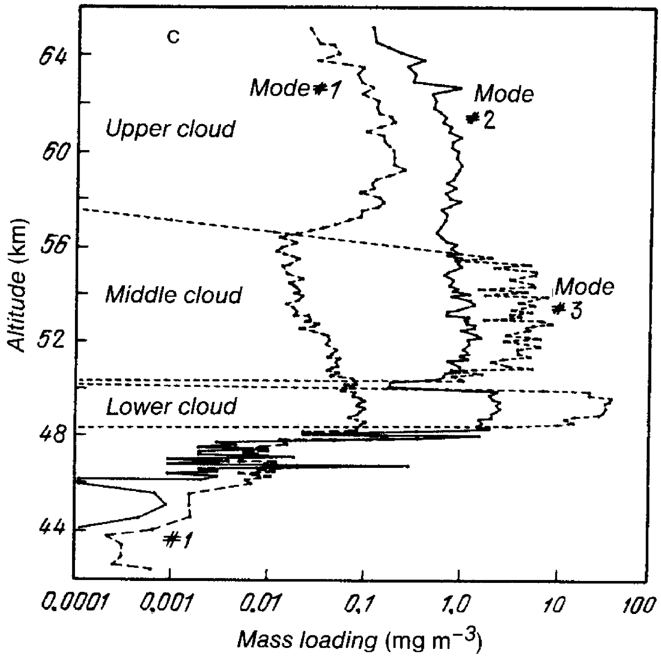

Fig. 4. Mass loading in the clouds of Venus as measured by the PV particle size spectrometer (Knollenberg and Hunten, 1980).

1980). These particles should be crystals with a high aspect ratio (Esposito et al., 1983) to agree with the simultaneous observations with the PV nephelometer (Ragent and Blamont, 1980) and solar flux radiometer (Tomasko et al., 1980). This interpretation requires the presence of an abundant condensable species other than sulfuric acid.

Toon et al. (1982) suggested that a discontinuity between mode 2 and mode 3 in the PV cloud particle spectrometer may be instrumental and mode 3 is a tail of mode 2. Then the mode 3 particles are spherical, their optical depth is larger by a factor of 3 and disagrees with the nephelometer and solar flux radiometer data. However, the particles consist of H_2SO_4 and other species are not required.

Three instruments on the Vega 1 and 2 landing probes collected and analyzed the cloud aerosol: the gas chromatograph (Porshnev et al., 1987), the mass spectrometer (Surkov et al., 1987), and the X-ray fluorescent multichannel radiometer (Andreychikov et al., 1987). The clouds were also studied by the particle size spectrometers and nephelometers (Gnedykh et al., 1987; Julanov et al., 1987). Unfortunately, data from the different experiments are sometimes contradictory and unexpected. Krasnopolsky (1989) made a semi-quantitative analysis and a summary of the observations.

Sulfur aerosol was detected for the first time by the gas chromatograph with a mass ratio $S_x/H_2SO_4 \approx 0.1$ (Porshnev et al., 1987). Previously, there were indirect indications of a possible presence of sulfur aerosol in the clouds of Venus. The measured ratio is close to the mass flux ratio of 0.04 in the model by Krasnopolsky and Pollack (1994) and the flux ratio of 0.1 in the nominal model by Mills (1998).

The X-ray fluorescent radiometer detected variable abundances of S, Cl, and P in the clouds and a column abundance of iron (Andreychikov et al., 1987). The data on chlorine may indicate (Krasnopolsky, 1989) the presence of aluminum chloride $AlCl_3$ in the middle cloud layer, and the abundant phosphorus in the lower cloud layer may exist as a partially hydrated phosphoric anhydride $P_2O_5 + H_3PO_4$. Though the X-ray fluorescent radiometer was switched off below 47 km, the phosphoric anhydride aerosol should extend to 25 km. This agrees with the particle size spectrometer and nephelometer data which show the abundant aerosol down to 33 km where these instruments were switched off. A similar extent was also observed by V8. Krasnopolsky (1989) suggested that a dense aerosol below the usual lower cloud boundary near 47 km may be an indication of phosphoric anhydride. Composition of the cloud aerosol based on the Vega 2 observations is given in Table 2. Some of these data and identifications are rather uncertain and need further study.

Table 2
Composition of the clouds at the Vega 2 site

Species	Altitude (km)	Abundance
H_2SO_4	52–62	$\sim 5\,mg\,m^{-3}$
S_8	52–62	$S_8/H_2SO_4 \approx 0.1$ by mass
$FeCl_3$	47–52	~1% of the total column mass loading at 47 km
$P_2O_5 + H_2O$	25–52	$\sim 5\,mg\,m^{-3}$
$AlCl_3$	53–58	$\sim 3\,mg\,m^{-3}$

From Krasnopolsky (1989).

6. Search for new chlorine and sulfur species

The photochemical model by Mills (1998) predicts significant abundances of Cl, Cl_2, S_2O_2, and $ClSO_2$ that are $\sim 10^{16}\,cm^{-2}$ above 58 km. The smaller abundances are predicted for SCl_2 ($\sim 10^{15}\,cm^{-2}$) and $COCl_2$ ($\sim 10^{14}\,cm^{-2}$). Some of these abundances may be detectable by ground-based and orbital observatories and by spacecraft missions to Venus. Any detection and even a restrictive upper limit would be of great interest for Venus' photochemistry. Recently $ClCO_3$, a key intermediate in the basic catalytic cycles in the Venus atmosphere, was synthesized in laboratory (Pernice et al., 2004).

7. Vertical and spatial variations of water vapor

Spectroscopy of the Venus nightside emissions in the windows at 2.3, 1.7, 1.3, and near 1 μm gave the H_2O mixing ratio of 30 ppm below the clouds (Pollack et al., 1993). A reanalysis of the observations from the V11–14 probes (Ignatiev et al., 1997) is based on high-temperature spectroscopic data and agrees with this value. Ignatiev et al. (1997) indicate a possible increase of the H_2O mixing ratio in the lowest 5 km. Meadows and Crisp (1996) derived H_2O increasing from 20 ppm at 45 km to 45 ppm below 30 km. The PV mass spectrometer measurements resulted in 60 ppm at 50–10 km decreasing to 10 ppm near the surface (Donahue and Hodges, 1992).

Variations of water vapor in the upper cloud layer were studied by Ignatiev et al. (1999) using the infrared spectra from the V15 orbiter. The mean water abundance varies from ~15 ppm at 60 km to ~5 ppm at 63 km. The value at 63 km is close to the mean abundance from the ground-based spectroscopy by Barker (1975) which also refers to 63 km. Local (place-to-place) variations of H_2O are of an order of magnitude in both observations. Koukouli et al. (2005) analyzed the H_2O bands in both the V15 infrared spectra and the PV Orbiter Infrared Radiometer obsevations. They confirmed the basic results of the V15 observations in Ignatiev et al. (1999). However, while the PV OIR nighttime and morningside observations agree with the data from V15, a significant increase in the H_2O abundance to 90 ± 15 ppm was detected in the equatorial afternoon region. This difference suggests significant temporal variability in the middle atmosphere of Venus.

The abundance of water vapor at 65–95 km is derived from ground-based microwave observations of the H_2O and HDO lines (see Encrenaz et al., 1995 and references therein). The mean abundance is ~2 ppm. Encrenaz et al. (1995) calculated vertical profiles of H_2O assuming the

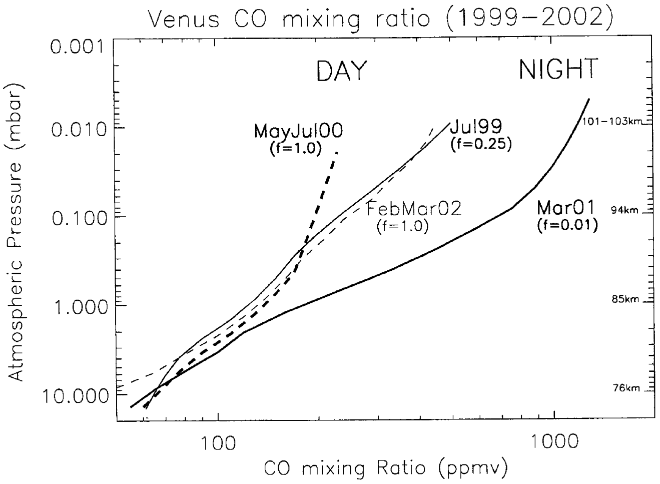

Fig. 5. Variations of the CO mixing ratio from the microwave observations by Clancy et al. (2003).

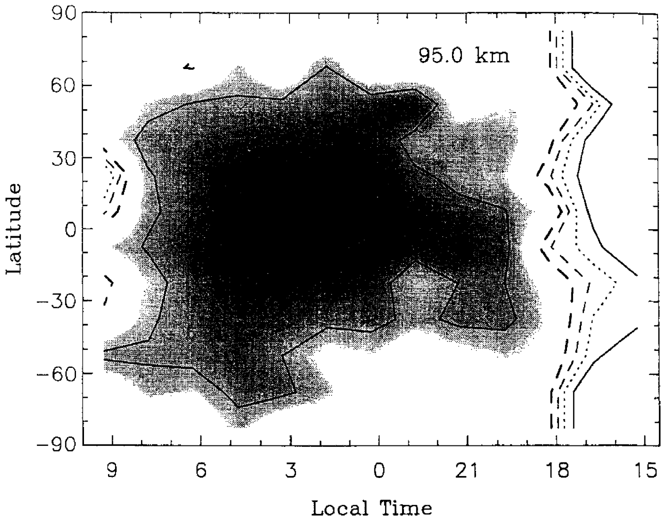

Fig. 6. The CO bulge at 95 km observed with the OVRO millimeter interferometer (Gurwell et al., 1995).

saturation conditions for pure water and obtained $T = 140$ K at the mesopause (95 km) from the observed H_2O line shapes. This temperature is much smaller than $T \approx 170$ K at 95 km in the Venus International Reference Atmosphere (Seiff et al., 1985). Mills (1999b) pointed out that aerosol at 65–95 km consists of sulfuric acid particles which form at 60–65 km and are driven to the mesopause by eddy diffusion. Then the corrected data of Encrenaz et al. (1995) agree with the VIRA temperatures and result in H_2O ~0.1 ppm near 95 km. Sandor and Clancy (2005) studied variations of H_2O at 65–100 km using microwave observations of the HDO lines at 226 and 335 GHz in the period from March 1998 to June 2004. Though the instrument field of view was large (28″ while the diameter of Venus varies from 10″ to 58″), observations at various phases could reveal diurnal variations of H_2O. However, neither long term nor diurnal variability is evident while strong global variations with a time scale of 1–2 months have been unambiguously observed. The H_2O mixing ratios at 65–82 and 82–100 km are different in most of the observations while their mean values are similar at 2 ppm.

The observed variations of water vapor are poorly understood and need further study in both observation and theory.

8. Vertical and spatial variations of CO

The CO mixing ratio near the cloud tops is equal to 50 ppm (Young, 1972) from the spectra of Connes et al. (1968). CO below the cloud layer was measured by the V12 gas chromatograph (Gelman et al., 1979) decreasing from 30 ppm at 42–36 km to 17 ppm at 12 km. Two analyses by the PV gas chromatograph (Oyama et al., 1980) gave 30 and 20 ppm at 42 and 22 km, respectively. A structure of the nightside emission at 2.3 μm resulted in the CO mixing ratio decreasing from 40 to 12 ppm at 46 and 23 km, respectively (Pollack et al., 1993). A recent study of this emission (Marcq et al., 2005) showed a weak increase of CO by 15% between the equatorial region and midlatitudes (±40°).

Microwave observations of the CO and ^{13}CO lines are used to retrieve vertical profiles of CO and/or temperature at 75–100 km (see Clancy et al., 2003 and references therein). Measurements at various phase of Venus with a beam size ~10 arcsec reveal diurnal variations of temperature and the CO abundance (Fig. 5). Using the interferometric technique, it is possible to improve significantly spatial resolution. Gurwell et al. (1995) made mapping of CO at various heights between 70 and 100 km using the OVRO interferometer with six 10.4 m telescopes. The interferometer had a bean size of 2.8 arcsec. The maps show a complicated behavior of CO with a morningside bulge above 90 km (Fig. 6). It would be interesting to model CO at these heights using a three-dimensional global circulation model. Even better results could be achieved using the Very large Array (27 telescopes of 25 m each, spatial resolution 0.04 arcsec) and the Atacama Large Millimeter Array (64 telescopes of 12 m each, spatial resolution 0.01 arcsec, under construction). However, Venus has not been observed with these instruments.

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