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Correlations between cloud thickness and sub-cloud water abundance on Venus

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[1] Past spacecraft observations of Venus have found considerable spatial and temporal variations of water vapour abundance above the clouds. Previous searches for variability below the clouds at 30-45 km altitude found no large scale latitudinal gradients, but lacked the spatial resolution to detect smaller scale variations. Here we interpret results from the VIRTIS imaging spectrometer on Venus Express, remotely sounding at near-infrared 'spectral window' wavelengths, as indicating that the water vapour abundance at 30–40 km altitude varies from 22 to 35 ppmv (±4 ppmv). Furthermore, this variability is correlated with cloud opacity, supporting the hypothesis that its genesis is linked to cloud convection. It is also possible to fit the observations without requiring spatial variation of water abundance, but this places a strong constraint on the spectral dependence of the refractive index data assumed for the lower cloud particles, for which there is as yet no supporting evidence. Citation: Tsang, C. C. C., C. F. Wilson, J. K. Barstow, P. G. J. Irwin, F. W. Taylor, K. McGouldrick, G. Piccioni, P. Drossart, and H. Svedhem (2010), Correlations between cloud thickness and sub-cloud water abundance on Venus, Geophys. Res. Lett., 37, L02202, doi:10.1029/2009GL041770.

1. Introduction

[2] The atmosphere of Venus is optically thick at almost all wavelengths, due to a combination of Rayleigh scattering (in the UV and visible), absorption by cloud particles (in the mid- and far-infrared), and strong absorption bands of CO_2 and other gases. However, there are a few wavelength regions in the near- infrared at which thermal emissions emanating from the deep troposphere escape between strong CO_2 absorption bands [*Taylor et al.*, 1997]. These spectral 'windows' allow remote sounding of the Venusian atmosphere below the clouds to obtain estimates of the abundance of minor species, resulting in a much better understanding of the deep atmospheric composition and chemical cycles present in the Venus troposphere [*Bézard and de Bergh*, 2007].

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[3] Images of Venus obtained at these wavelengths reveal large spatial and temporal variations in peak intensity, due to changes in the total opacity of the cloud layer. In the example shown in Figure 1a, obtained in the emission window at 2.20–2.60 μ m, the cloud layer is 'backlit' by nearly uniform thermal emission from below, and the dark regions represent absorption by thick cloud [Taylor et al., 1997]. Spectra in this window have been used for retrieval of abundances of CO, OCS, H₂O and SO₂ [de Bergh et al., 1995; Marcg et al., 2006, 2008]; in particular, the region around 2.40 μ m is sensitive to the ν_1 and ν_3 , absorption bands of gaseous H₂O. These bands are shifted from the longer wavelengths of their condensed phase within the cloud layers. Apart from one early study which found large variation in sub-cloud water vapour abundance, anti-correlated to cloud opacity [Bell et al., 1991], Earth-based observers [de Bergh et al., 1995; Marcq et al., 2006; Crisp et al., 1991] have not detected any spatial variation in sub-cloud water vapour. However, the instruments used in these observations had spatial resolutions of \sim 500–2500 km at Venus and so would be unable to detect variations on smaller spatial scales. Searches for sub-cloud water exploiting the 1.18 μ m window, which is sensitive to water at altitudes of 0-20 km, also failed to find any spatial variations in water vapour [Crisp et al., 1991; Drossart et al., 1993; de Bergh et al., 1995; Meadows and Crisp, 1996; Bézard et al., 2009], which suggests a lack of sinks/ sources at the surface. This does not rule out water vapour variations higher up in the atmosphere near the cloud base though, given the large vertical wind shear and convectively stable regions found in the 10-40 km altitude range.

2. Data

[4] With the arrival of Venus Express around Venus in 2006, we have for the first time a Venus orbiter equipped to observe the near-IR window regions, offering far better spatial and temporal coverage than Earth-based observations as well as freedom from telluric absorption features. The VIRTIS instrument includes an infrared mapping spectrometer ('VIRTIS-M') which observes Venus from 1.0 to 5.0 μ m, creating a 256 × 256 pixel image with a spectral sampling of $\Delta \lambda = 0.01 \ \mu$ m, as well as a non-imaging 'high resolution' spectrometer ('VIRTIS-H') whose spectral sampling is $\Delta \lambda \sim 0.001 \ \mu$ m in the 2.3 μ m region [*Drossart et al.*, 2007; *Piccioni et al.*, 2008]. Recently, VIRTIS-H was used to retrieve gas abundances from the 2.3 μ m window region, and found some small variations in H₂O abundance, with reported values ranging from (29 ± 0.6) ppm to (32.5 ±

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1.0) ppm [*Marcq et al.*, 2008]. However, the VIRTIS-H spatial resolution was limited by the need to co-add multiple spectra in order to achieve a sufficient signal-to-noise ratio. This process also introduces a bias towards brighter regions of the planet where the 2.3 μ m radiance is stronger. In the present work we use VIRTIS-M data, which is less subject to these limitations, in order to further explore the spatial



variation water vapour in the sub-cloud region at 35-40 km altitude.

3. Method

[5] The VIRTIS-M spectra were analysed using the Nemesis radiative transfer model [*Irwin et al.*, 2008; *Tsang et al.*, 2008a], assuming four different size distributions (or 'modes') [*Esposito et al.*, 1997] for the H₂SO₄/H₂O cloud particles making up the Venus cloud layers. We fit variations in brightness by varying the amount of cloud particles in the lower-middle cloud layer [*Tsang et al.*, 2008b], using refractive index data for 75%H₂SO₄/25%H₂O [*Palmer and Williams*, 1975] for all cloud particles; the sensitivity to refractive index data is discussed more below.

[6] Spectra of the 2.20–2.50 μ m emission feature have the form shown in Figure 1b. The region from 2.20–2.30 μ m is sensitive only to cloud opacity, while the longer-wavelength section $(2.30-2.50 \ \mu m)$ is also sensitive to the abundance of minority gas species CO, OCS, H₂O and SO₂ at an altitude of 35–45 km. We therefore follow a two-stage process to fit the spectra: first we retrieve cloud opacity by matching the 2.20–2.30 μ m region of the spectrum, and secondly we retrieve the minor gas abundances using the 2.30–2.50 μ m spectrum. It is important to first fix the cloud structure before retrieving the gas abundances, because the cloud scattering properties are wavelength dependent and therefore affect the retrieved abundance of H₂O [Bell et al., 1991; Marcq et al., 2006]. The CO, H₂O and OCS abundances are retrieved simultaneously by minimizing the difference between the modelled synthetic spectra and the observed spectra [Irwin et al., 2008; Tsang et al., 2008a, 2008b]. The main mode of variability is cloud optical depth, with regions of thicker cloud resulting in a much lower radiance emerging from the cloud tops. It can be seen in Figure 1b that allowing the water vapour to spatially vary allows a good spectral fit to be achieved. Applying this procedure to a VIRTIS-M data qube allows maps of water vapour distribution to be produced, as shown in Figures 1c and 2.

4. Results

[7] Water vapour retrievals were carried out for a total of seven observations. These avoid observations at latitudes poleward of 60° , because the cloud structure at these high latitudes is, as yet, still poorly constrained, but otherwise include all observations made with the longest possible integration time for the VIRTIS-M instrument (18 s). Data

Figure 1. Imaging spectroscopy in the near-infrared from VIRTIS. (a) Night side thermal emissions at 2.3 μ m show variations in total cloud optical depth. (b) Two measured spectra (black) in regions of low and high cloud opacity are shown with their associated errors (shaded area). Synthetic spectra generated using 34 ppmv of water vapour (blue) provides the best fit for the first (brighter) but not for the second (darker) spectrum. A synthetic spectrum with 23 ppmv (orange) provides a better fit for the second spectrum. The spectral regions of CO, H₂O, and OCS absorptions are indicated. Fitting across the entire observation results in **c**, a map of the H₂O abundance.



Figure 2. Correlation of cloud thickness and water vapour concentrations. (a, c) Observations of 2.3 μ m thermal emissions taken on the 7 and 21 March 2007, during orbits 320 and 344, respectively. (b, d) The corresponding sub-cloud H₂O abundance. To first order, the regions of high cloud thickness are co-located with regions of low water vapour abundances.

were spatially co-added in 20×20 pixel bins in order to further increase the signal-to-noise ratio, which degrades the spatial resolution of the water maps to ~100 km. The H₂O abundances retrieved from these data range from 22 to 35 ppm, with an error of $\pm 3-4$ ppm on each individual retrieval. This variation is for the most part anti-correlated with cloud opacity, as is shown in Figures 2 and 3, although there are regions where water vapour abundance anomalies do not correspond directly to cloud features (Figures 1 and 2). Even where water and cloud features do coincide, the highest H₂O abundances do not necessarily correlate to the region of highest cloud optical depth, suggesting a complex mechanism is at work.

[8] We found that the observed variations in the spectra could not be reproduced without varying the abundance of water vapour (Figure 1c), but we investigated several possible alternatives, specifically (1) temperature fluctuations, (2) cloud particle size changes and (3) variations in cloud base altitude. Fluctuations in the temperature profile in the deep atmosphere have been measured over the course of many months using radio occultation from Venus Express [*Tellmann et al.*, 2009] and found to be ~5 K at the 1 bar level (~50 km) and less than this at the cloud base and in the H₂O line-forming region (40–45 km). Radiative transfer

modelling shows that a ~ 20 K difference would be needed at this height level in order to have any significant effect on the spectrum [*Tsang et al.*, 2008a]. Cloud particle size, as



Figure 3. Correlation of 2.3 μ m thermal emission with H₂O abundance at 40 km from seven high exposure spectral images, spread across three months of observation.

diagnosed by examining the $1.74/2.30 \ \mu m$ ratio, was not found to vary appreciably within the observations used in this work, which is as expected for observations away from the polar regions [*Wilson et al.*, 2008]. Variations in the cloud base altitude were also tested, from 43 to 48 km, in 1 km increments, since a lowering in cloud base altitude would put aerosol particles in the line-forming region. The current cloud model has a nominal cloud base altitude of 45 km, taken from descent probe aerosol volume scattering measurements and photochemical/condensation models [*Esposito et al.*, 1997]. No model scenario produced a satisfactory fit without the need to also alter the water vapour mixing ratio.

5. Refractive Indices

[9] We do find that the retrieved water vapour abundance is sensitively dependent on the imaginary component of the refractive index assumed for the lower cloud particles. There are only a few attempted measurements of this quantity, and some of those do not report imaginary refractive indices for H₂SO₄ at this wavelength range because they are so low ($k \sim 2 \times 10^{-3}$) and thus difficult to measure; reported values for k at 2.4 μ m range from 2 to 6×10^{-3} [Palmer and Williams, 1975; Niedziela et al., 1999; Lund Myhre et al., 2003]. The most reliable refractive index data in the 2.2–2.6 μ m range [Palmer and Williams, 1975] has been used in all analyses of Venus nightside emissions to date [de Bergh et al., 1995; Marcq et al., 2006, 2008; Crisp et al., 1991; Bézard et al., 2009; Meadows and Crisp, 1996] including this work. We have assumed 75%H₂SO₄:25%H₂O composition by weight for all cloud particles, but we still find spatial variation of water vapour if the 85% or 96% H₂SO₄ refractive index data [Palmer and Williams, 1975] are used. If 96% H₂SO₄ is assumed, as suggested by chemical models including that by Krasnopolsky [2007], the mean contrast change in water vapour drops to 10 ± 4 ppm. Altitude variations of these higher concentration cloud particles also has little effect, since most of the radiation is coming from well below the cloud layer.

[10] The only way in which we can satisfactorily match the observations without invoking spatial variation of water vapour is to propose a specific form of $k(\lambda)$ for lower cloud particles where $k(\lambda)$ exhibits a gradient $dk/d\lambda$ of $(1 \pm 2) \times$ 10^{-3} per unit μm from 2.2 μm to 2.4 μm , followed by a steeper gradient of $(1.3 \pm 0.6) \times 10^{-2}$ per unit μm at 2.4– 2.5 μ m. We find it impossible to match both the H₂Osensitive and OCS-sensitive portions of the spectrum for constant H₂O vapour and OCS concentrations unless this change of gradient in $k(\lambda)$ is invoked. It is possible that H₂SO₄/H₂O mixtures exhibit this form of $k(\lambda)$. The accuracy and spectral resolution of the H₂SO₄/H₂O refractive index data is low enough that this ad hoc form of $k(\lambda)$ lies within the range of reported experimental values. Alternatively, one could postulate that this form of $k(\lambda)$ is due to an unknown component of the lower cloud particles, occurring either in the entire lower cloud deck or only in regions of thicker cloud, which might include exotic candidates such as FeCl₃ and P₂O₅. Further laboratory measurement of $k(\lambda)$, both of H₂SO₄/H₂O mixtures and other candidate materials are needed to resolve this ambiguity. Thus we note that it is possible to match spectra from the Venus observations

without requiring horizontal variations of water vapour abundance. If this is indeed the case, then it represents a stringent spectral constraint on the optical properties of the lower cloud particles.

6. Discussion and Conclusions

[11] If on the other hand the spectral variations seen in the VIRTIS data are indeed indicative of H₂O abundance variation at 30-45 km altitude, what could be causing them? The few existing measurements of gaseous H_2SO_4 below the clouds show great spatial and temporal variability [Butler et al., 2001]. There is a net transport of H₂SO₄ from the cloud-tops, where it is formed by the action of solar UV photons on gaseous SO₂ CO₂ and H₂O, to the sub-cloud atmosphere where it dissociates at altitudes of 35-45 km due to the high temperatures to form H₂O and SO₃ [Krasnopolsky and Pollack, 1994]. This downwards H₂SO₄ flux is expected to be spatially inhomogeneous in the lower cloud, because of vertical motions associated with convection, and this may be what leads to the observed variability in gaseous H₂SO₄ abundances observed at 35-45 km (1–14 ppm at 45 km altitude [Kolodner and Steffes, 1998]). It seems likely that this variability in sulphuric acid abundance could give rise to variability in its decomposition product, H_2O , at the altitude of its decomposition. Cloud measurements from the Pioneer Venus Large Probe show 20 mg/m³ of cloud mass in the lower cloud [Knollenberg and Hunten, 1980], which if evaporated and dissociated would create approximately 6 ppmv of H₂O vapour at that altitude, similar in magnitude to the 12 ppmv variation in water vapour abundance observed at 40 km altitude (noting though, that the density is almost three times higher at the lower altitude). This explanation for the H₂O variability is not completely satisfactory, though, for several reasons: For example, it is not clear why other tracers such as CO at 35-40 km, which are understood to indicate vertical transport [Marcq et al., 2008; Tsang et al., 2008b], do not also show spatial variation associated with cloud variability. A further possibility is that the H₂O and H₂SO₄ variability is associated with photochemical processes below the clouds [Yung et al., 2009]. Another possible explanation could arise from the assumption that sulphuric acid is known to be highly hygroscopic. In the clouds, liquid water could be condensed, trapped and hidden in the sulphuric acid droplets. In the presence of downwelling, the sulphuric acid descends and the trapped water may become free in the form of vapour due to the higher temperature conditions.

[12] Turning from infrared to radio frequencies, radio occultation can be used to measure vertical profiles of gaseous H_2SO_4 at altitudes down to ~35 km; a recent reanalysis of six individual occultations from the Magellan and Mariner 10 orbiters found H_2SO_4 abundances at 41 km s ranging from 0 ppm to 8 ppm [*Kolodner and Steffes*, 1998], and it certainly seems reasonable to suggest that a larger range of variation might be found if more occultations were analysed. The decomposition of each H_2SO_4 molecule would yield a molecule of H_2O , so this range of >8 ppm of H_2SO_4 at 41 km would be expected to yield a variation of >8 ppm in water vapour abundance at this altitude, and thus is entirely consistent with the present results from VIRTIS.

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