

Retrieval of Venus' clouds and hazes properties with polarimetric data from SPICAV/VEx

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Abstract

1. Introduction

The study of Venus' cloud layers is important in order to understand the structure, radiative balance and dynamics of the Venusian atmosphere. Polarization measurements have given important constraints for the determination of the constituents of the clouds and haze. From ground based observations, Hansen and Hovenier[3], using a radiative transfer model including polarization, found that the main cloud layers between 50 and 70 km consist of $r \sim 1 \mu\text{m}$ radius spherical droplets of a $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ solution. In the early 1980s, Kawabata[4] used the polarization data from the OCPP instrument on the spacecraft *Pioneer Venus* to constrain the properties of the overlying haze. They found that the haze layer is composed of smaller particles with $r \sim 0.25 \mu\text{m}$ and similar refractive indices.

Our work reproduces the method used by Hansen and Kawabata[3, 4]. We applied a radiative transfer model with polarization on the polarization data of the SPICAV-IR instrument on-board ESA's *Venus Express*. Our aim is to better constrain haze and cloud particles at the top of Venus's clouds, as well as their spatial and temporal variability.

2. SPICAV-IR observations

The SPICAV-IR spectrometer on *Venus Express* is based on an Acousto-Optic Tunable Filter (AOTF) working in the $0.65 - 1.7 \mu\text{m}$ range, with two output beams linearly polarized in perpendicular directions, allowing us to measure the degree of linear polarization for different phase angles[8, 6].

The data give a good latitudinal and phase angle coverage. Latitudinal variations in polarization are visible in the observation data for orbits up to #2700 with a strong increase of polarization towards the poles

(Fig. 1). At lower latitudes, polarization is quite homogeneous and we observe the glory in polarization at low phase angles, in accordance with VMC observations in photometry[7].

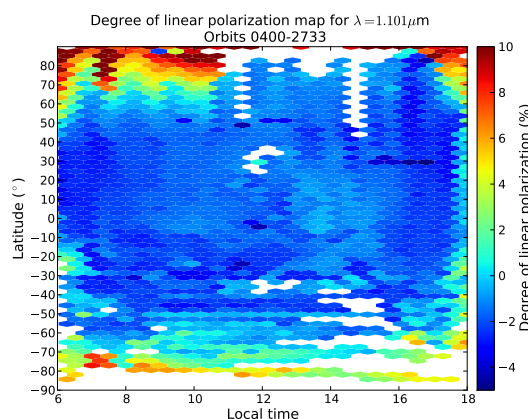


Figure 1: SPICAV data polarization map as a function of local time and latitude for orbits #400 to #2733.

3. Analysis

We use a radiative transfer model taking polarization into account in order to model the clouds[2, 1]. We consider a two layered model: an optically thick cloud layer of micrometric particles made of a concentrated sulfuric acid solution. Above lies the haze layer of $r \sim 0.25 \mu\text{m}$ particles with a varying column density C_h .

3.1. Glory

At low phase angle, the main feature is the glory which gives information about the main cloud particles. We retrieve the effective radius and refractive index of the particles and effective variance of the particle size

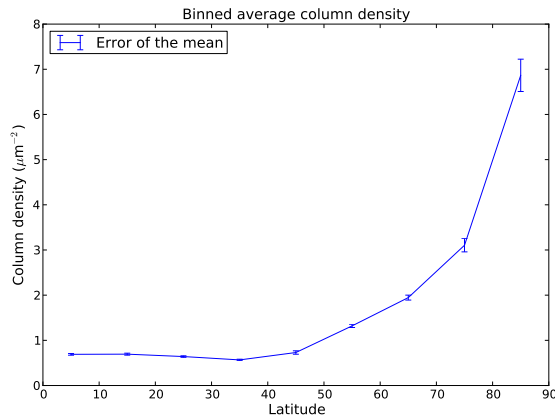


Figure 2: Column density of the haze layer in northern hemisphere of Venus as a function of latitude. The corresponding optical thickness at $1.101 \mu\text{m}$ varies from 0.039 to 0.343

distribution for a dozen glory observations. The retrieved values are in agreement with previous results: the cloud particles are spherical, with radii between 0.8 and $1.3 \mu\text{m}$, $\nu_{\text{eff}} < 0.15$ and refractive indices between 1.39 and 1.44 at $\lambda = 1 \mu\text{m}$.

3.2. High latitudes

At higher latitudes, the main contributor to polarization is the submicrometric haze. The modeling allows us to measure the column density of the haze layer in the northern hemisphere (Fig. 2). We observe a small decrease of C_h with increasing latitude up to 50° followed by a sharp increase of C_h towards the poles. C_h varies from $0.8 \mu\text{m}^{-2}$ at low latitudes up to $7 \mu\text{m}^{-2}$ at higher latitudes.

4. Conclusion

SPICAV-IR provides global measurements of polarization of Venus clouds and allows us to retrieve the parameters of the cloud droplets, in agreement with previous measurements. We confirm that the clouds are made of spherical micrometric droplets of sulfuric acid while the hazes are made of $r \sim 0.25 \mu\text{m}$ particles. The column density of the haze increases towards the pole, in agreement with other studies[5].

5. Perspectives

We aim to generalize the retrievals to both hemispheres and will investigate in more details the latitude and local time dependence of the haze column

density. The long-term variations during the *Venus Express* mission and comparison with OCPP will also be explored. We will also attempt to retrieve the vertical properties of the clouds using the polarization contained in the CO_2 absorption band as illustrated in [9].

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Titan Aerosol Formation as a Sink for Stable Carbon and Nitrogen Isotopes

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Abstract

Stable isotope ratios of major elements can be used to infer much about local- and global-scale processes on a planet. On Titan, aerosol production is a significant sink of carbon and nitrogen in the atmosphere, and isotopic fractionation of these elements may be introduced during the advanced organic chemistry that leads to the condensed phase products. Here we report on the isotopic composition of analogs of Titan's organic aerosol generated in the laboratory via photochemistry of CH₄ and N₂ gas mixtures.

1. Introduction

Several stable isotope pairs, including ¹²C/¹³C and ¹⁴N/¹⁵N, have been measured *in situ* or probed spectroscopically by Cassini-borne instruments, space telescopes, or through ground-based observations. However, the effect of a potentially critical pathway for isotopic fractionation – organic aerosol formation and subsequent deposition onto the surface of Titan – has not been considered due to insufficient data regarding fractionation during aerosol formation. To better understand the nature of this process, we have measured the isotopic fractionation associated with the formation of Titan aerosol analogs via far-UV irradiation of several methane (CH₄) and nitrogen (N₂) mixtures.

2. Experimental Methods

The Titan aerosol analogs used in this study were generated in a photochemical flow reactor (far UV: 115 - 400 nm), described previously [1]. Gas mixtures were prepared by the addition of methane into a mixing chamber with N₂ as a balance gas, with concentrations ranging 0.005 - 1.5% CH₄ in N₂. Previous work indicated chemical composition of the aerosol products should not vary substantially across this concentration range, though production rates do vary by more than 2 orders of magnitude [2].

Aerosols formed in the chamber were collected on a glass fiber filter and analyzed for bulk δ¹³C and δ¹⁵N with a Costech Elemental Analyzer (EA) - Thermo Scientific Delta V isotope ratio mass spectrometer (IRMS). Several filter segments were measured for each sample of collected aerosol. Measurement of the δ¹³C in CH₄ in the starting gas mixture was performed using gas injection on a Thermo Trace Ultra GC Isolink with Carboxen 1006 PLOT column coupled to the IRMS. The δ¹⁵N of the N₂ balance gas was measured by direct injection into a gas/liquid injection port on the EA coupled to the IRMS.

We report the Δ¹³C of the aerosol product, defined as Δ¹³C = δ¹³C_{products} - δ¹³C_{reactants}, to describe the isotopic composition of the products relative to starting gas composition, where

$$\delta(\text{‰}) = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1 \right) \cdot 1000 \quad (1)$$

3. Results and Discussion

An extensive report on the results of this study and discussion of the implications has been reported in a recent publication by Seebree et al [3]. A summary is provided here.

Our results show that the direction of carbon isotope fractionation during aerosol formation is in contrast to the expected result if the source of the fractionation is a kinetic isotope effect (Fig. 1). Explanations for the source of the observed fractionation include: (1) aerosol formation pathways dependent on initial gas phase products with positive δ¹³C; (2) non-kinetic fractionation during photolysis reactions, such as the processes that cause “mass-independent fractionation” in S and O isotopes in Earth's atmosphere; or (3) some combination of these effects.

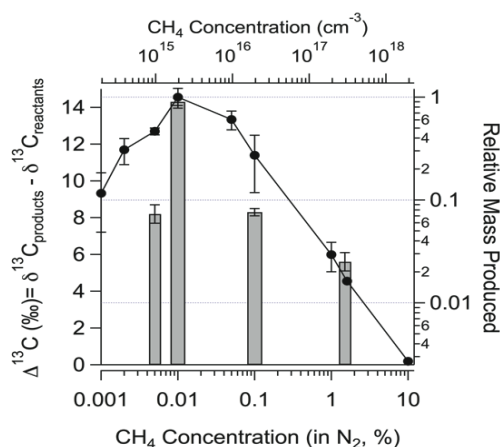


Figure 1: The isotopic composition of carbon in the photochemical aerosol products relative to the reactant gas shows enrichment in ^{13}C (bars, left axis), with a trend similar to the aerosol mass production (circles, right axis) [2].

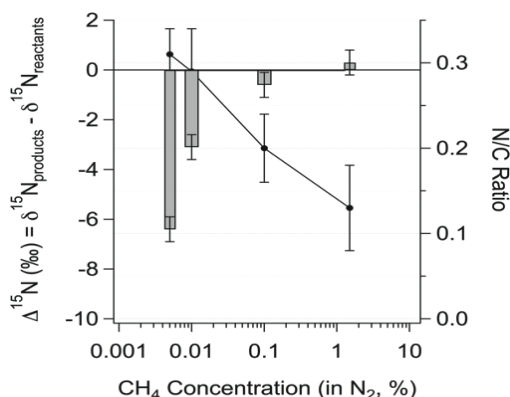


Figure 2: The isotopic composition of nitrogen in the photochemical aerosol products relative to reactant gas shows enrichment in ^{14}N (bars, left axis), increasing with a bulk increase in the N/C ratio (circles, right axis) of the aerosol products.

Our negative $\Delta^{15}\text{N}$ value is the opposite of what is observed for HCN and N_2 in Titan measurements (Fig. 2). Our UV source does not cover the relevant wavelength range for producing the fractionation directly from N_2 , but rather shows that the previously observed incorporation of nitrogen into aerosols via lower energy pathways [4]. The direction of the fractionation is the same as that observed by Kuga et al. [5] from plasma experiments, with a slightly lower magnitude.

4. Summary and Conclusions

In this study, we have analyzed the carbon and nitrogen fractionation of a series of photochemically generated aerosols produced from several mixtures of CH_4 and N_2 [3]. The resulting aerosols were enriched in ^{13}C and ^{14}N . When put in the context of Titan's atmosphere, photochemically-produced aerosols may form a sink for ^{13}C , possibly resulting from secondary reaction pathways that do not depend on the KIE. The enrichment of ^{14}N in the aerosols, if present on Titan, could contribute to an overall enrichment in ^{15}N in Titan's atmosphere as the condensed particles settle out.

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A possible water ice cloud in Jupiter's stratosphere

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Abstract

Jupiter's atmosphere has been sounded in transmission from UV to IR, as if it were a transiting exoplanet by observing one of its satellites, Ganymede, while passing through Jupiter's shadow during a solar eclipse from Ganymede. The spectra show strong extinction due to the presence of aerosols and haze in the atmosphere and strong absorption features from CH₄. In addition, the spectra show two broad features near 1.5 and 2.0 μm that we tentatively attribute to a layer of H₂O ice in Jupiter's stratosphere. While the spectral signatures seem to be unequivocally attributed to crystalline water ice, to explain the strong absorption features requires a large amount of water ice.

1. Introduction

In the search for the characterization of exo-planets atmospheres, there has been recently put emphasis on observing planetary transits in our own solar system [4, 3]. These, in addition of providing insights for future exoplanet characterizations, also serve for exploring our planetary atmospheres themselves.

Here we report on the limb transmission spectra of Jupiter's atmosphere obtained by using ground-based observations of Ganymede, which is in synchronous rotation around Jupiter, when crossing Jupiter's shadow. During the eclipse, the spectral features of the Jovian atmosphere are imprinted in the sunlight that, after passing through Jupiter's planetary limb, is reflected from Ganymede toward the Earth (see Figure 1). The ratio spectrum of Ganymede before and during the eclipse removes the spectral features of the Sun, of the local telluric atmosphere on top of the telescopes, and the spectral albedo of Ganymede. The spectra cover from UV to near-IR and have a high spectral resolution and high signal-

to-noise ratio. The highlights of the observations have been reported by Montañés-Rodríguez et al. [3]. Here we focus on a detailed analysis of the spectral region from VIS to near-IR and particularly on the signatures of water ice.

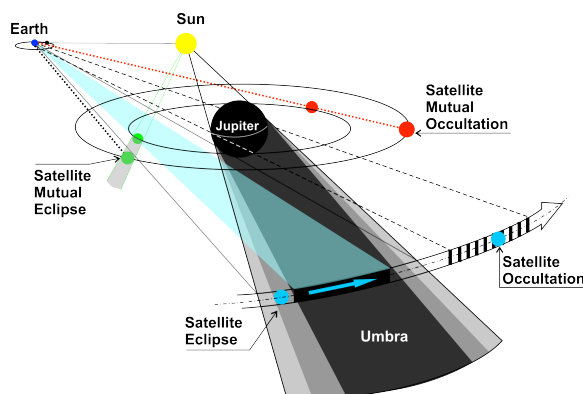


Figure 1: A diagram (not-to-scale) showing the orbital geometry of the Jovian System during the observations [3].

2. Observations

An eclipse of Ganymede was first observed on 06/10/2012 using LIRIS [2] at WHT in La Palma Observatory, Spain. The experiment was repeated later by observing a second eclipse with XSHOOTER [6] at VLT in Paranal Observatory, on 18/11/2012. This work focus on the analysis of VLT data, due to their higher signal to noise ratio, although the WHT observations exhibits essentially the same spectral features.

3. Analysis and Results

Transmission spectra of the occultation of the Sun through Jupiter's atmosphere as observed from

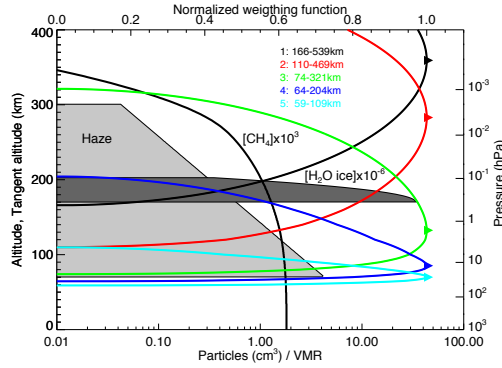


Figure 2: The distribution of haze, CH_4 , and H_2O ice used in the simulations of the transmission spectra (see Fig. 3), together with the tangent altitudes covered in each of our simulations corresponding to the mean times when the (umbra)-penumbral spectra were taken. Altitude is taken as zero at the 1 bar pressure level. Over-plotted are the profiles of the concentrations of aerosols and of the water ice particles cloud [3].

Ganymede have been simulated for the observations of the eclipse in the penumbra and within the first stages of the umbra by using the Karlsruhe Optimized and Precise Radiative Transfer Algorithm (KOPRA; [5]). Besides the CH_4 absorption, the simulations also include: Rayleigh scattering by molecular hydrogen and helium, collisions induced absorption (CIA) for H_2 - H_2 and H_2 -He, and Mie scattering by water ice and aerosols. The distribution of CH_4 , haze, and H_2O ice used in the simulations are shown in Fig. 2, and the simulated spectra together with one of the penumbral observed spectra are shown in Fig. 3.

The spectra show the most prominent CH_4 bands, the extinction of the aerosol particles (haze), and two distinct absorption features of water ice at 1.5 and 2.0 μm . All major features of the measured spectra are simulated. The absorption spectral features at 1.5 and 2.0 μm can be very well reproduced in our model with crystalline water ice at 150 K, but needs a total vertical column of about 10^{13} particles/ cm^2 with a size of $\sim 0.01 \mu\text{m}$ located near the 0.5 mbar level (see Fig. 2). If sublimated, this leads to a much larger water amount (about a factor of 500) than that measured by HERSCHEL [1] at and above that pressure level. While the spectral features fit perfectly to those of water ice, the required large amount of water ice to reproduce the

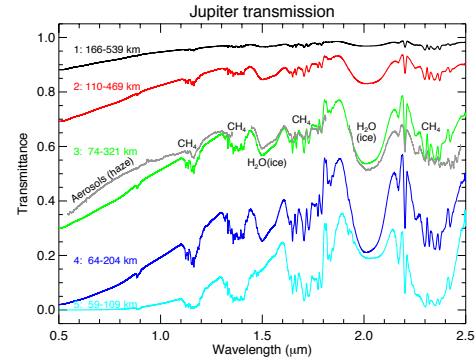


Figure 3: Transmission spectra of Jupiter calculated for the early phase of the penumbra and during the umbra over the 0.5–2.5 μm spectral region. The grey line is one of the observed penumbral spectra [3].

spectra is still not understood.

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Photochemical aerosols on Titan and the giant planets

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Abstract

Our ideas about the nature of photochemical aerosols on Titan and the giant planets is evolving thanks to new data coming in from the Cassini spacecraft, ground-based and space-based telescopes, and theory and modeling. Aerosol formation begins at altitudes around 1000 km on Titan and around 800 km above the 1-bar pressure level in the polar thermospheres of Jupiter and Saturn where auroral energy is available to form ions and radicals. We have evidence that hydrocarbon chemistry is important in aerosol formation for all of these bodies and we believe that hydrazine on Jupiter and phosphine on Saturn may lead to aerosol production. Aerosols have a fractal aggregate structure on Titan and in the polar regions of Jupiter and Saturn. Their vertical and horizontal distributions reflect a balance between local production and horizontal and vertical transport governed by eddies and jets. They are important for radiative energy balance in ways that have only recently come to light.

1. Introduction

Photochemical aerosols play an important role in the energy budgets of the stratospheres of Titan, Jupiter and Saturn. As data accumulate from ground-based and space-based telescopes and instruments we are learning about the optical, physical, and chemical properties of these particles and their spatial distributions. With these data and with recent developments from theory and modeling we are also learning about the processes responsible for their formation and life cycles. The following paragraphs briefly summarize the background and recent developments.

2. Jupiter and Saturn

Photochemical models of Jupiter's stratosphere [6], predict aerosol formation based on nitrogen and hydrocarbon chemistry initiated by photolysis of NH_3 and CH_4 . For Saturn replace NH_3 with PH_3 . Models

specific to the auroral regions [3],[15] predict hydrocarbon chemistry leading to the formation of polycyclic aromatic hydrocarbons (PAHs). There is some observational evidence for benzene [2]. So far there is no evidence for hydrazine or (for Saturn) diphosphene.

Two aerosol regimes are apparent from observations [1]. Jupiter appears to have polar caps in images in strong methane absorption bands. At near-UV wavelengths the polar regions are dark. Both of these can be explained by a polar stratospheric aerosol that is optically thick at the slant viewing angles observed from earth. The distribution of aerosols is asymmetric, with the north polar aerosol extending to lower latitudes ($\sim 45^\circ$) in the north versus $\sim 67^\circ$ in the south. These differences most probably reflect the asymmetry in the auroral footprint, although confinement by zonal jets also plays a role. The boundaries of these regions for Jupiter are marked by Rossby waves [9].

Polar aerosols in Jupiter's and Saturn's stratospheres are both highly polarizing and forward scattering at visible and near-IR wavelengths. These combined properties can only be understood if the aerosols are composed of aggregates of small (~ 40 nm radius) monomers [12]. The altitude and latitude distributions of these aerosols and their optical properties play a pivotal role in the radiative energy balance of the stratosphere.

An unusual feature has been seen only twice and only in near-UV images of Jupiter. It was first seen as a dark oval with the same size and shape as Jupiter's Great Red Spot at high northern latitude in a images from the Hubble Space Telescope obtained in 1997 [13]. It was seen again in 2000 in near-UV images obtained by the Cassini Imaging Science Subsystem where it was observed to evolve over several weeks [7].

Saturn has a feature known as the polar hexagon near latitude 75° N., close to the latitude where auroral energy is deposited. Polarization images and UV

images show that the hexagon is a boundary that dynamically confines the aurorally-generated fractal-aggregate haze, much as the terrestrial winter polar vortex confines ozone [10].

3. Titan

Photochemistry converts methane and nitrogen in Titan's atmosphere to photochemical products whose composition is some form of $H_xC_yN_z$. This still leaves considerable uncertainty in the proportionality of H, C, and N, and it is likely that their proportionality changes with altitude as the reaction rates and mixing ratios of the gaseous photochemical products depend on altitude.

Titan haze particles are composed of fractal aggregates [12]. The DISR experiment on the Huygens probe provided data inside the atmosphere to refine our understanding of these particles (average particles have ~4000 monomers with average radius 40 nm) [14]. At altitudes higher than where the DISR sampled the aerosol is thought to be in the form of small monomers. The Cassini INMS and CAPS instruments showed that aerosol formation begins at very high altitudes (~1000 km).

An unusual feature of the haze is a gap in the vertical profile that makes it appear as a 'detached' haze layer. This layer moved in altitude from 500 km when Cassini first arrived in 2004 to about 300 km in 2012, with most rapid change near equinox. Several ideas have been put forth to explain this [8], [5], [4]. See [11],[14] for more detail on these and other observational, theoretical/modeling and laboratory studies of the Titan haze.

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Mass spectrometry investigation of Titan aerosols analogs formed with traces of aromatic compounds

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Abstract

We present a laboratory analysis of Titan aerosol analogs formed with traces of aromatic and heteroaromatic compounds as precursors. We observe an influence of the precursor aromatic compound on the composition and growth process of the aerosol.

1. Introduction

The detection of benzene at ppm levels in Titan's atmosphere [1] by Cassini's Ion and Neutral Mass Spectrometer (INMS) supports the idea that aromatic and heteroaromatic reaction pathways may play an important role in Titan's aerosols formation. In laboratory studies it has been shown that these aromatic molecules are easily dissociated by ultraviolet radiation and can therefore contribute significantly to aerosol formation [2] and be used to dope the production of aerosol analogs [3].

In this work we investigate the effect of the chemical nature of the aromatic reactant on the aerosol composition and growth pattern using Laser Desorption-Time of Flight mass spectrometry (LD-TOF) and Fourier Transform Infrared Spectroscopy (FTIR)

2. Experimental

Samples were prepared using a photochemical reactor developed at GSFC presented in Figure 1. The gas mixture used consisted of nitrogen – methane with traces (100 ppm) of benzene, pyridine, naphthalene or quinoline.

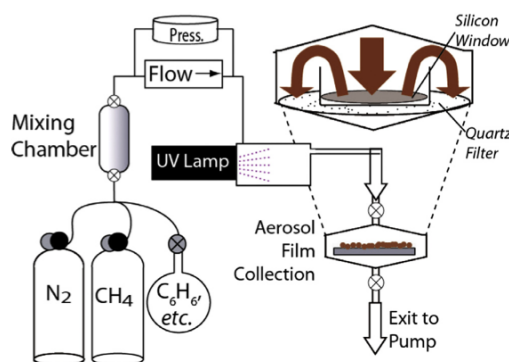


Figure 1: The photochemical reactor is used for the generation and collection of the Titan aerosol analogs.

Samples are collected under inert atmosphere and analyzed both with FTIR and LD-TOF.

3. Preliminary results

Infrared analysis of our samples shows that inclusion of aromatic compounds as trace precursors allows to better fit laboratory data to Titan aerosol spectra observed by Cassini [3,4]. The improvement is especially visible on the far infrared (~ 200 cm⁻¹) bands observed by CIRS [5] and on the 3.28 μ m band reported at an altitude of 950 km by VIMS [6]

Figure 2 presents preliminary LDMS results of some aerosol samples produced with trace amount of aromatic compounds.

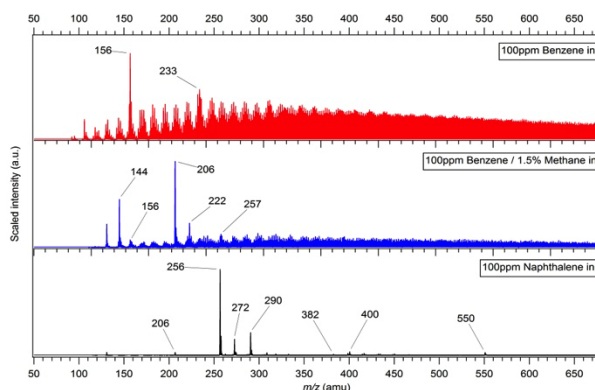


Figure 2: LD-TOF mass spectra of Titan aerosol analogs produced with 100ppm of benzene in N_2 (top); 100 ppm of benzene in $N_2:CH_4$ 98.5:1.5 (middle) and 100 ppm of naphthalene in N_2 (bottom)

LDMS results show that the aerosol growth patterns depend both on the number of rings and on the nitrogen content of the trace precursor used.

We also perform MS/MS analysis on some prominent peaks of aerosol mass spectra. This MS/MS approach allows us to identify some of the key compounds in the aerosol growth processes.

Acknowledgements

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Stratospheric Ices in Titan's Atmosphere

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Abstract

Observations from Cassini, Voyager, and ground-based data point to the condensation of trace species in Titan's atmosphere, including HCN, C_2H_5CN , HC_3N , C_2H_2 , C_2H_6 , and C_4N_2 . These and a dozen other species have now been added to the Titan CARMA microphysics model, which shows condensation occurring between about 60 and 100 km in Titan's atmosphere. Results on condensation altitudes as well as particle size will be presented, and implications for the optical properties of Titan's stratospheric aerosol particles will be discussed.

1. Introduction

Above the optically thick haze layers in Titan's atmosphere, methane and nitrogen molecules are broken apart by uv radiation, cosmic rays, and energetic electrons. A number of photochemical reactions occur, resulting in the creation of many trace hydrocarbon and nitrile species. When these trace gases reach Titan's stratosphere, many become supersaturated and condense out as ices surrounding a haze particle core. The condensation curves for 14 of these species are shown in Figure 1.

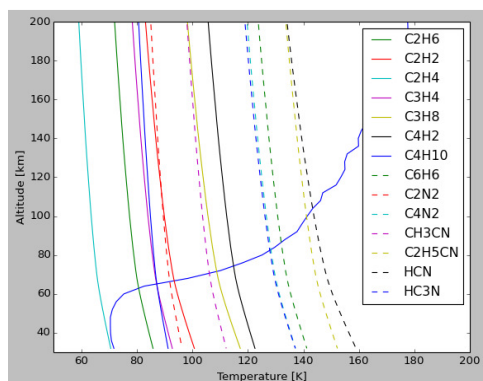


Figure 1: Condensation curves for trace species found in Titan's atmosphere. Saturation occurs where

each (nearly vertical) mixing ratio line intersects Titan's temperature profile (shown in blue).

2. Modeling & Results

The ice condensation is explored using a 1-D microphysics model, which is an extension of the model described in [1] and [2]. Both haze and cloud particles are transported vertically through sedimentation and eddy diffusion. All particles are subject to coagulation. Angstrom-sized haze particles are introduced through a production function related to the photochemical destruction of methane. Cloud particles are created through nucleation following the classical theory. Cloud particles then interact with the volatiles through condensational growth and evaporation.

Both ice and droplet particles can be treated, as well as melting and freezing. Most of the species shown in Figure 1 condense below their freezing point, however propane (C_3H_8) initially forms droplet cloud particles around 70 km and then freezes at about 60 km. Both propane and ethane (C_2H_6) ice particles will melt near the surface. Condensation timescales for many of the ices are long, resulting in particles $\sim 1\text{-}5\text{ }\mu\text{m}$ in radius. HCN, which begins to condense around 105 km, initially grows to $\sim 20\text{ }\mu\text{m}$, and the C_3H_8 droplets can reach $\sim 40\text{ }\mu\text{m}$.

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This work was supported by the NASA Outer Planets Research program.

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Observations of Titan's haze and clouds by Cassini VIMS

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Abstract

The study describes recent observations of Titan's atmosphere by the Visual and Infrared Mapping Spectrometer (VIMS) onboard the Cassini spacecraft. These observations include solar occultation observations, observations of North polar clouds and specular reflections on Titan's seas. The goal is to determine the optical depth of Titan's haze as a function of wavelength, latitude and season, and to better constrain the organic cycle.

1. Introduction

Titan is the only satellite in the solar system with a dense atmosphere with methane constituting the second largest component. Methane is irreversibly transformed into ethane by photolysis. The carbon cycle includes the replenishment of the atmosphere with methane [1,2], the formation of clouds, the precipitations of methane and ethane, the formation of organic molecules in the upper atmosphere, their coalescence to form the haze particles [3], the sedimentation of those heavy organic molecules that are eventually swept by surface winds to form the dunes [4], the formation of lakes at polar latitudes [5] and the interaction of liquid hydrocarbons with the icy porous regolith [2,5]. Since Titan entered spring, the VIMS instrument has observed the formation of clouds forming on the North Pole (Fig. 1). Specular reflections on the large seas provide information on the waves and winds [6]. Finally, solar occultation observations provide constraints on the haze opacity on the North Pole and can be compared with previous solar occultation observations at other latitudes.

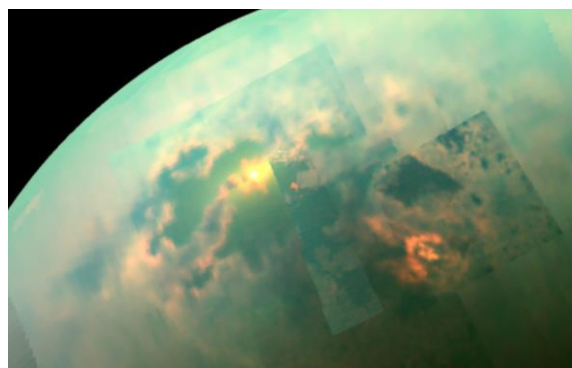


Figure 1: Titan flyby T104 showing a cloud (V shape) over Ligeia Mare, specular reflection on Kraken Mare (large yellow spot), and 5- μ m bright shorelines along Kraken Mare.

2. Solar occultation observations

During flyby T103 in July 2014, the VIMS instrument recorded solar light curves through Titan's atmosphere for an impact parameter located at 66S and 55N during ingress and egress, respectively. These observations complement previous observations obtained at different latitudes since the T10 observations obtained in January 2006 [7]. These new observations show that the opacity at the polar caps is less than the opacity at the equator (Fig. 2).

At shorter wavelengths (0.9- to 1.6- μ m), the VIMS values and the DISR values [3] are in very good agreement. However, the extrapolation of the DISR trend to longer wavelengths predicts larger opacities than the values inferred from the VIMS observations.

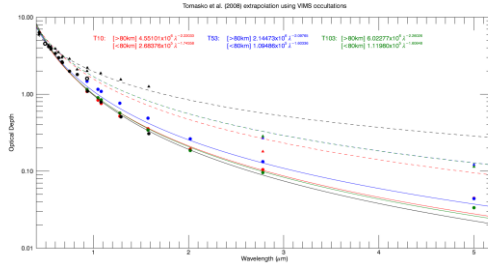


Figure 2: Comparison of the opacities obtained by the Huygens probe [3] and the VIMS instrument.

3. Clouds

Few clouds have been observed on the North Pole although their formation due to the evaporation of methane from the lakes was predicted by Global Circulation Models [8]. However, during the T104 flyby (Fig. 1), the VIMS instrument recorded a large V-shape cloud over Ligeia Mare. The characteristics of this cloud are being investigated to determine its origin.

4. Specular reflections

The VIMS instrument has acquired several observations of specular reflections on the large seas present on Titan's North Pole such as the one detected during T104 (Fig. 1). Besides providing information on the roughness of Titan's surface (waves), these observations also provide spectra that can be used to infer the opacity of Titan's atmosphere and to compare with the values obtained by the solar occultation observations [9].

5. Spectral information

The spectra of the clouds, the specular reflections, the solar occultation observations can be compared with spectra of the surface. Of particular interest are the absorption features in the two broad atmospheric windows at 2.7- and 5- μm . Three absorption features can be identified in the 2.7- μm window at 2.57-, 2.74-, and 2.97- μm . The 2570 nm feature is present in the specular reflection spectra, the surface spectrum and the solar occultation observations and is a methane band. The 2740 nm feature is present in the surface spectrum, slightly visible in the T110 specular reflection observation and almost invisible in the T84 specular reflection. It is also not visible in

the solar occultation observation. It is therefore interpreted as a surface feature. There is debate onto whether this absorption could be related to the presence of water ice at the surface. However, water ice remains dark at higher wavelengths, which is clearly not the case of the surface spectrum. Finally, the 2970 nm feature is visible in the surface spectrum and the specular reflection observations but not in the solar occultation observations

5. Summary

The diversity of the VIMS observations provide important information that help understand Titan's organic cycle. The good agreement between the DISR observations and the VIMS observations gives confidence in the retrieved opacity of the atmosphere. This is important for radiative transfer models that are used to retrieve the surface albedo.

Acknowledgements

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Heterogeneous CO₂ nucleation in the Martian mesosphere – laboratory experiments

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Abstract

We present measurements on heterogeneous CO₂ nucleation on sub 4 nm radius iron oxide and silicon oxide particles as analogues to naturally occurring Meteoric Smoke Particles (MSP). From the particle growth, we infer the particle surface desorption energy of CO₂. Additionally, evaluation of nucleation rates at controlled particle temperature and CO₂ concentration gives us the contact parameter of CO₂ on these particles, resulting in a full nucleation parameterization for MSPs in the Martian mesosphere. We find strong indications that the same parameterization can be used for upward propagated Martian Dust Particles (MDPs). We apply this nucleation parameterization to conditions of the Martian mesosphere which results in characteristic nucleation onset temperatures. Furthermore, the Sticking Coefficient of CO₂ on solid CO₂ is evaluated.

1. Introduction

CO₂ ice clouds have been detected in the Martian mesosphere [e.g. 1, 9, 10, 11]. These clouds mainly appear during pre- and post- aphelion season and are believed to be caused by heterogeneous nucleation on nanoparticles during supersaturated conditions induced by thermal tides and gravity waves [4, 12]. Although great progress has been made in the last century in monitoring and modeling mesospheric CO₂ clouds on Mars, large uncertainties especially in parameterizing the microphysical formation process of the ice particles remain. Main ice nucleation candidates are Meteoric Smoke Particles (MSPs) or upward propagated Martian Dust Particles (MDPs). Currently a single parameterization [5] is used to describe nucleation on either of these particle types in the mesosphere. It leads to CO₂ nucleation at temperatures only slightly below the saturation

temperature. This result is in disagreement with night time observations of temperatures well below saturation temperature in absence of clouds [3, 8]. One possible explanation for the discrepancy between observation and theory is that the nucleation ability of MSPs and MDPs is actually lower than assumed.

We recently designed a novel experiment which allows us to investigate CO₂ nucleation and growth processes on MSP analogues under controlled conditions being reasonably close to conditions in the Martian mesosphere. This allows us to determine important surface properties of the MSP analogues such as CO₂ desorption energy, sticking coefficient, contact parameter and critical temperature for nucleation as a function of the particle size.

2. Experimental Method

We produce free sub 4 nm radius iron oxide and silicon oxide particles representing MSP analogues with a microwave plasma particle source. These particles are transferred into the vacuum setup TRAPS [7] in which we trap singly charged particles of selected mass in a supersaturation chamber called MICE [2]. In MICE, we can control the particle temperature and ambient CO₂ concentration. Heterogeneous nucleation and subsequent growth of CO₂ ice is observed by extracting a small sample from the trapped particle population at defined time steps and analyzing its mass distribution with a time of flight spectrometer.

3. Results and Discussion

Figure 1 shows an exemplary series of measurements of CO₂ deposition on silicon oxide particles having an initial radius of about 2.5 nm. The measured

particle mass is shown as function of the residence time in MICE. The CO_2 concentration is set to a constant value, in this case to $5\text{E}15 \text{ m}^{-3}$.

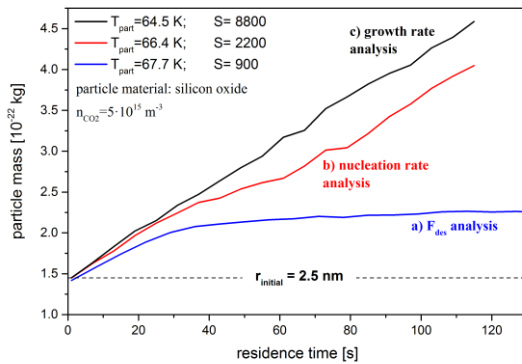


Figure 1: exemplary dataset with 2.5 nm silicon oxide particles used for desorption energy (a), nucleation rate (b), and growth rate (c) analysis.

The particle temperature and therefore saturation is varied and allows us to adjust conditions with and without nucleation taking place. The blue line (a) represents a condition at which no nucleation took place. The temporary increase of particle mass represents an adsorption process of CO_2 molecules on the particles surface allowing us to determine the desorption energy of CO_2 molecules on the investigated particle material. The red line (b) represents conditions which allow us to evaluate nucleation rates and determine the contact parameter m . Finally, at high saturations as represented by the black line (c) the growth rate can be evaluated to retrieve the sticking coefficient α of CO_2 on solid CO_2 . The evaluation of such measurements allow us to determine a full parameterization of nucleation and growth of CO_2 on MSPs at conditions being reasonably close to conditions in the Martian mesosphere. We will present this new parameterization and show that it probably can be used for MDPs as well. We will discuss the results and highlight the consequences when applying this parameterization to the Martian mesosphere resulting in defined temperatures at which nucleation gets induced.

Acknowledgements

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A novel experimental setup to study the nucleation of atmospheric vapours on small nanoparticles

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Abstract

We present a novel supersaturation chamber which allows us to expose nanoscale particles to supersaturated vapors in the wide range of temperatures occurring in planetary atmospheres. This chamber, the molecular flow ice cell (MICE), is integrated in the vacuum setup TRAPS and enables us to study adsorption, nucleation and growth processes of condensable vapours as for instance water vapour and carbon dioxide. We will present the experimental setup with focus on MICE. The general function principal of MICE and its limitations will be highlighted and we will elaborate that this new device is able to study adsorption, ice nucleation and growth processes exemplified with CO₂ nucleation experiments in the mesosphere of Mars.

1. Introduction

Heterogeneous nucleation on nanometer sized aerosol particles is able to initiate the formation of clouds in the atmosphere of planets. An example are ice clouds in the higher atmosphere of Earth, so called Noctilucent Clouds (NLCs). These clouds have been detected in the polar summer mesopause region of Earth at heights of 80-90 km [e.g. 3, 7]. They are believed to be caused by heterogeneous nucleation of H₂O on sub 2 nm meteoric smoke particles (MSPs). Surprisingly, similar clouds have been detected in the mesosphere of Mars as well [e.g. 1, 5, 6, 8]. In contrast to NLCs on Earth, they consist of CO₂ ice and occur at low latitudes mostly during pre- and post- aphelion season. Here, the main candidates acting as ice nuclei are MSPs and Martian Dust particles (MDPs). Scientists dealing with the formation of NLCs struggle with large uncertainties in describing the nucleation processes taking place due to a lack of experimental data at the extreme

conditions of the mesosphere which states the need of laboratory measurements.

We recently designed a new supersaturation chamber which allows us to expose charged nanoscale particles to supersaturated vapors. Within the chamber we can control the particle temperature and vapor concentration of basically any condensable vapor. The chamber consists of a linear ion trap and is an integral part of the TRAPS Apparatus [4]. Among other things it allows to study nucleation and growth processes on nanoparticles at the very low temperatures in planetary atmospheres. Until now, we have studied nucleation and growth of H₂O and CO₂ on sub 4 nm radius iron oxide and silicon oxide particles as an analogue of MSPs. These measurements allow us to parameterize the growth and nucleation process of mesospheric CO₂ and H₂O clouds on Mars and on Earth. We will use this contribution to show our experimental setup and highlight the functioning of the molecular flow ice cell.

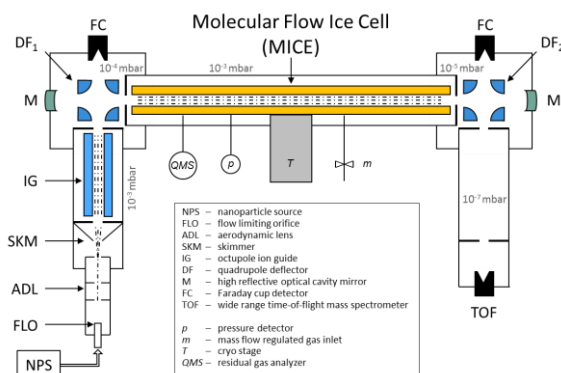


Figure 1: Schematic setup of the TRAPS apparatus including MICE [2].

2. Experimental Setup

Singly charged nanometer sized (2-4 nm in radius) MSP analogues are produced in a microwave plasma reactor and transferred via an aero-dynamic lens into the TRAPS chamber which is shown in Figure 1. Within the chamber the particles are further guided with an octupole ion guide. Particles of one polarity and a defined mass are selected into MICE using a quadrupole deflector DF₁. MICE is a combination of a linear ion trap with a supersaturation chamber working in the molecular regime: Temperature controlled surfaces are placed between the quadrupole electrodes. These surfaces are covered with a thin layer of the condensable vapour prior to an experiment. The vapour concentration is adjusted by controlling the temperature of the ice covered surfaces whereas the additional control of the electrode temperature allows us to control the particle temperature by collisions with the buffer gas helium. In MICE up to 10⁸ particles are held under controlled particle temperature and vapor concentration. Heterogeneous ice nucleation and growth processes then can be examined by analyzing the mass distribution of the particles with a time-of-flight mass spectrometer (TOF) as function of the residence time under supersaturated conditions.

3. Results

We will show that the MICE is able to store sub 4 nm radius particles without significant loss in time. Homogeneous conditions are applied within the trap and we are able to produce saturations higher as 10⁵. We will proof the functioning in the molecular regime and expound the operation regimes of MICE in terms of adjustable particle temperature and vapor concentration of CO₂ and H₂O. We will show exemplary that this new device is able to study adsorption, ice nucleation and growth processes exemplified with CO₂ nucleation experiments in the mesosphere of Mars.

Acknowledgements

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Aerosol charging processes in planetary and terrestrial atmospheres

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Abstract

Interactions between the ions and aerosols cause charge exchange, which can lead to substantial aerosol charge and ion removal in the atmosphere. Aerosol charging plays an important role in various processes such as aerosol scavenging by droplets and aerosol growth by affecting aerosol-aerosol coagulation rates. Ions are removed in regions with abundant aerosol, which may modify charge flow in an atmosphere, such as that associated with an atmospheric electrical circuit. A review will be made of the charging processes and the consequences occurring in atmospheres of Mars, Venus and Titan and compared with terrestrial atmosphere [1], [2], [3], [4], [5]. Some recent results on charging of aerosols in the lower and upper atmosphere of Titan will be presented and consequences will be discussed.

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Titan's aerosol optical properties with VIMS observations at the limb of Titan.

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Abstract

The study of Titan properties with remote sensing relies on a good knowledge of the atmosphere properties. The in-situ observations made by Huygens combined with recent advances in the definition of methane properties enable to model and interpret observations with a very good accuracy. Thanks to these progresses, we can analyze in this work the observations made at the limb of Titan in order to retrieve information on the haze properties as its vertical profiles but also the spectral behaviour between 0.88 and 5.2 μm .

1. Introduction

To study the haze layer and more generally the source of opacities in the stratosphere, we use some observation made at the limb of Titan by the VIMS instrument onboard Cassini. We used a model in spherical geometry and in single scattering, and we accounted for the multiple scattering with a parallel plane model that evaluates the multiple scattering source function at the plane of the limb.

Our scope is to retrieve informations about the vertical distribution of the haze, its spectral properties, but also to obtain details about the shape of the methane windows to disentangle the role of the methane and of the aerosols.

2. Results

We started our study at the latitude of 55°N, with an image taken in 2006 with a relatively high spatial resolution (for VIMS) (**Figure 1 - left**). Our preliminary results show the spectral properties of the aerosols are the same whatever the altitude (**Figure 2**). This is a consequence of the large scale mixing. From limb profile between 0.9 and 5.2 μm , we can probe the haze layer from about 500 km (at 0.9 μm) to the ground (at 5.2 μm) (**Figure 1 - right**).

We find that the vertical profile of the haze layer shows three distinct scale heights with transitions around 250 km and 350 km. We also clearly see a transition around 70-90 km that may be due to the top of a condensation layer.

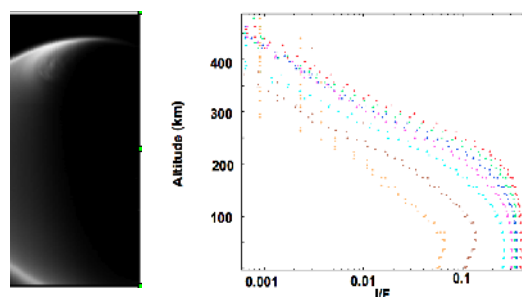


Figure 1: (Left) Image of Titan northern polar region with an extended polar cloud. The intensity coming out from the atmosphere can be used to study the atmosphere properties in the stratosphere. (Right) Vertical profiles of I/F in the 7 windows between 0.9 μm and 5.2 μm .

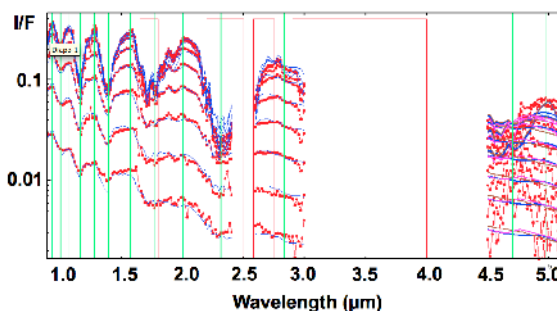


Figure 2: I/F spectra taken on the image at 7 altitudes (red spots) and a model fit (blue lines). The red boxes show the wavelengths excluded from the fit (VIMS default, and C2H6 absorption feature). The green lines show the wavelengths where the limb profiles are studied. **Figure 1** only displays the profiles inside the windows.

Retrieving the aerosol particle distribution in Titan's detached layer from ISS limb observations

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Abstract

The study of the detached haze layer above Titan's thick atmosphere is one of the key elements to understand the growth of the aerosols in the upper atmosphere of Titan. In this work we will present the results of a radiative transfer inversion of the vertical profile distribution of aerosols in the detached haze layer (from 300 to 600 km) by using the I/F ratio observed by Cassini ISS camera. The analyses will focus on the derivation of the particle size distribution.

1. Introduction

Titan's atmospheric detached haze layer was first observed in 1983 by Rages and Pollack [1] during the Voyager 2 exploration with the first full coverage of Titan's atmosphere at intermediate and high phase angles. The analyses of these images showed the presence of an aerosol opaque thick layer between 300 and 350 km over the main haze of Titan. Since 2005, the Imaging Science Subsystem (ISS) instrument on board the Cassini mission performs a continuous survey of the Titan's atmosphere almost every month. In 2011, West *et al.* [2] confirm the persistence in time of the detached haze layer but with an important variability in its height (over 500 km in 2007, under 380 km in 2010). Detail analyses showed, that this layer corresponds to the transition area between small spherical aerosols and large fractal aggregates [3, 4]. Then, the characterization of the aerosols' size distribution along vertical profiles in the detached haze layer is one of the key elements to understand their growth.

2. Observations

In this study, we focus our analysis on the I/F ratio observed by ISS at the limb of Titan for different phase angles. We have restricted the observations on the detached haze layer most probable location between 600 km to 300 km, in the UV filter ($\lambda = 338$ nm, where

the multiple scattering is low) and in a short period of time (2006).

Table 1: List of ISS images used in CL1-UV3 filter

| ISS ID | Date | Phase | Sun lat. |
|-------------|------------|---------|----------|
| N1525327324 | 2006/04/03 | 146.9 ° | -32.9 °N |
| N1540314950 | 2006/10/23 | 120.2 ° | -1.9 °N |
| N1521213736 | 2006/03/16 | 68.0 ° | -19.3 °N |
| N1546223487 | 2006/12/31 | 66.5 ° | -5.9 °N |

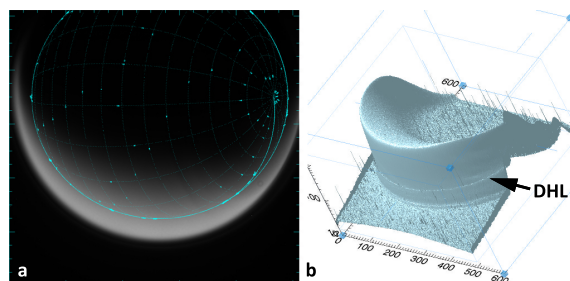


Figure 1: (a) ISS image N1540314950 with its coordinates grid. (b) 3D plot of I/F ratio to see the detached haze layer.

3. Model

Considering the atmosphere of Titan homogeneous longitudinally, we use the vertical profile according from HASI' measurements [5] for the temperature, the pressure and the density. Then, we modeled the effect of the Rayleigh scattering, the gas absorption [6] and the spherical/fractal aggregate aerosols [3] opacity. We calculate I/F at the limb, assuming a spherical geometry [1]:

$$\frac{I_n}{F} = \sum_{i=1}^{2n} \int_{z_{i-1}}^{z_i} \frac{\langle \omega_0 P(\theta) \rangle_j}{4} \exp(-\tau_{0i}(z) - \tau_i(z)) \beta_j dz \quad (1)$$

With $\langle \omega_0 P(\theta) \rangle_j$ the product of the single scattering albedo and the phase function for scattering angle of $\theta = 180 - \phi$, and β_j the extinction coefficient in the layer j .

Therefore, the aerosol size distributions are constrained layer by layer from the top to the bottom of the detached haze layer. The inversion is performed by a Levenberg-Marquardt algorithm (provided by More *et al.* [7] in the *minpack.f90* package)

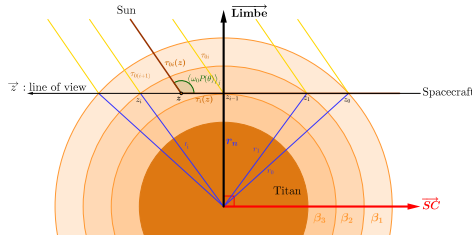


Figure 2: Integration of the light along the line of sight in a spherical atmosphere with the onion peeling method.

4. Perspectives

Analyses of Cassini ISS observations with the use of the model will provide a temporal survey of the evolution of the detached haze layer in term of aerosol distributions in height and in radius. We will also be able to constrain the latitudinal variations between the equator and the poles. The next development of this model will take into account the multiple scattering to perform the inversion on the whole profile at different wavelengths to apply it to the other filters of ISS and VIMS.

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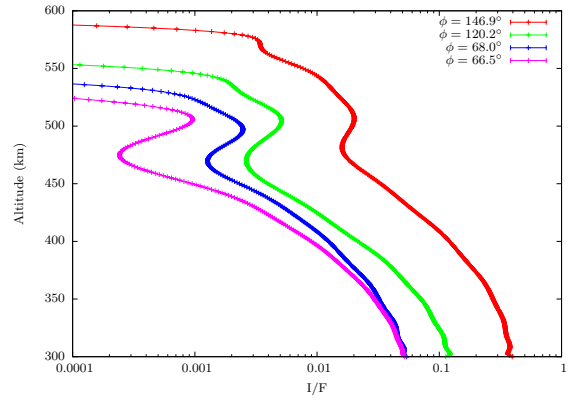


Figure 3: Observations of the I/F ratio profile for different phase angles at the geographic equator (0 to 5°N).

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Saturn's cloud structure and particle phase function based on Cassini ISS observations

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Abstract

Cassini ISS observations of Saturn provide images with high spatial resolution that allow dynamical analysis of many Saturnian meteorological phenomena, such as the giant storms [1] or the Hexagon wave [2]. Observations in selected narrow filters in and out the methane absorption bands in the near infrared also provide information on the vertical distribution of clouds and hazes in the lower stratosphere and upper troposphere, in particular regarding micron-sized or smaller populations. In this work, we use Cassini ISS observations to constrain the vertical distribution of particles in the Northern Hemisphere in the 2011-2013 period with particular emphasis in the Equatorial Zone and the Polar Region, where the Hexagon is located. We also focus on the phase function describing the scattering of light by tropospheric particles, which is a substantial piece of information only accessible with observations at high phase angles.

1. Observations

The observations used in this work are all images taken in the methane bands at 727nm and 890nm (MT2 and MT3 filters) and an adjacent continuum at 752 nm (CB2). These filters are sensitive mostly to tropospheric levels [3], moving deeper as the methane absorption decreases from MT3 to CB2. At the same time, these filters have wavelengths similar enough to assume that other characteristics (such as the phase function parameters) are the same for all of them.

The observations were selected based on the following criteria: (1) to cover partially or completely the Northern Hemisphere, (2) to span as much phase angles as possible, (3) to be taken in as short as possible amount of time at a given latitude to avoid temporal variations in the atmosphere. In this way, more than 70 images were selected, navigated and

calibrated to retrieve maps of absolute reflectivity as a function of latitude and scattering angles. Phase angles covered from as low as 10° to values as high as 160° for some filters and latitudes.

2. Modelling approach

In order to reproduce the observed maps of absolute reflectivity, the NEMESIS retrieval code [4] and its underlying radiative transfer correlated-k algorithm was used. The multiple-scattering radiative transfer code is based in the doubling/adding scheme and includes gas absorption.

NEMESIS is designed to use fixed spectral parameters and iterate over the possible vertical atmospheric profiles. Since we aimed to retrieve phase function parameters, an initial scanning of the free parameter space using a Nelder-Mead simplex algorithm was performed. Using the best-fitting result for the spectral parameters, NEMESIS provided the optimal solution for the vertical distribution of particles.

In our model, we included three layers of particles: stratospheric and tropospheric hazes (with different properties) and the tropospheric upper cloud, putatively formed by ammonia ice, following common assumptions in similar previous works [3,5].

The particle phase function for the troposphere was assumed to be a double Henyey-Greenstein phase function, in order to compare later with previous works based on Pioneer flyby observations on the late 70s that determined the Saturnian phase function for first and only time [6].

Retrieved free parameters include the particle density as a function of height for the hazes, average particle density for the cloud, particle phase function parameters for the tropospheric haze and particle size distribution for the stratosphere.

3. Particle phase function

The double Henyey-Greenstein phase function (2HG) is described by a forward scattering asymmetry (g_1), a backward scattering asymmetry (g_2) and the relative contribution of each (f). We show some preliminary results for the latitudinal depend of these parameters in Figure 1 below.

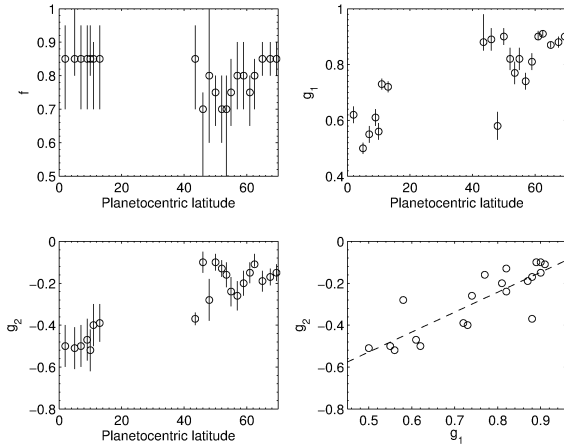


Figure 1: Sample retrievals of the 2HG phase function parameters as a function of latitude for the 2011 observations. Bottom right panel shows the correlation between forward and backward parameters. The gap at mid-latitudes is due to the 2010 GWS, whose rapid evolution makes impossible to work with several phase angles at the same time.

4. Conclusions

In general terms, we find the particles to be more forward scattering at equatorial latitudes than at the mid-latitudes, suggestive of larger sizes. The phase function for the particles at mid-latitudes is more similar to the one retrieved using previous missions [6].

We also find a distinct vertical cloud structure in the equatorial region, with higher and more vertically extended hazes in the low latitudes. This kind of vertical distribution of particles agrees well with the observed differences in the dynamics as seen in methane absorption bands or the adjacent continuum, which could be sounding dynamically decoupled levels in the atmosphere.

We are currently undergoing a detailed analysis of the vertical cloud structure of Saturn's North Polar

Region and we will also show the phase function results for the Hexagon and adjacent latitudes.

Acknowledgements

We gratefully acknowledge the work of the Cassini ISS team that made the data available. This work was supported by the Spanish MICIIN project and AYA2012-36666 with FEDER support, Grupos Gobierno Vasco IT765-13, and UPV/EHU UFI11/55. S.P.-H. acknowledges support from the Jose Castillejo Program funded by Ministerio de Educación, Cultura y Deporte, Programa Nacional de Movilidad de Recursos Humanos del Plan Nacional de I-D+i 2008-2011.

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Evolution of Titan's atmospheric aerosol under high-altitude ultraviolet irradiation

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Abstract

The Cassini-Huygens space mission revealed that the chemistry initiating Titan's aerosols formation starts in the upper layers of the atmosphere (~1000 km). The grains then sediment until deposition at the surface. Despite all the data collected, the photochemical evolution of the aerosols between 1000 and 600 km is still unknown, especially regarding their optical properties. The aim of this work is to investigate the photochemical aging processes of those aerosols and how they interact with the VUV solar radiations. We put our results in perspective with the Cassini data for different atmospheric altitudes.

1. Introduction

Titan is the biggest satellite of Saturn whose atmosphere is mainly composed of molecular nitrogen (N_2) and methane (CH_4) with an average ratio of 98/2 %. [1]

The Cassini/Huygens mission revealed that the interaction between those neutral molecules and the UV solar light leads to a complex photochemistry that produces heavy organic molecules. When those molecules condense, they will then become the solid aerosols, which are responsible for the brownish haze surrounding Titan. [2, 3]

Between 1000 and 600km, the VUV solar radiations are still significant and will continue to modify the physical, chemical and optical properties of those grains. A change in these parameters can impact the radiative budget of Titan's atmosphere.

Then, the aim is to identify and understand the photochemical evolution of the aerosols. In order to compare our data to the ones of Cassini's instruments such as CIRS or VIMS, we will focus on their infrared signatures and how they can be modified after being exposed to VUV radiations.

2. Method

To do so, we irradiate thin films of Titan's atmospheric aerosols analogues with VUV synchrotron radiations provided by the DESIRS beamline at the SOLEIL synchrotron facility. The analogues are produced by submitting a 95-5% N_2 - CH_4 molecular mixture to a radio-frequency electron discharge.[4]

In Titan's ionosphere, the aerosols are exposed to the full VUV-solar spectrum so the experiment needs to reproduce those conditions in terms of wavelength and flux. The effect on the aerosols may vary according to the wavelength. This is why we chose three typical wavelengths representative of irradiation effects according to the altitude:

1. 95 nm to test possible ionizing effects
2. 121.6 nm ($Ly-\alpha$), as an important solar contribution
3. 190 nm, to probe the soft VUV irradiations

The solar VUV-UV photon flux reaching the top of Titan's atmosphere is about 10^{14} photons/s/cm² [5] while the DESIRS line provides a monochromatic flux of 10^{16} photons/s/cm².

Nevertheless, our irradiation time is of few hours and it has been calculated that the residence time of the

aerosols in the thermosphere (between 1000 and 600 km) is about the duration of one Titanian day (10^6 s). [6] So we counterbalance our higher photon flux by a shorter irradiation time.

The diagnostic of the irradiated films is then performed using infrared absorption spectroscopy (ATR technique).

2. Results

Figure 1 describes the evolution of the infrared absorption signature of the tholins films as a function of the irradiation wavelength. It seems that the main difference happens at 95 nm. The N-H amine signature (3250 cm^{-1}) decreases while the saturated-hydrocarbon ones increases (2920 and 2850 cm^{-1}), which is consistent with a transfer of H from N to C. This leads us to think that VUV irradiation, firstly, dishydrogenates the amines functions and, secondly, increases hydrogenation of olefinic structures in the aerosols.

Implications for Titan's aerosols

Because of absorption, the atmospheric penetration depth varies from one wavelength to the other. For Titan, the 95 nm radiations are more likely to occur above 1000 km of altitude, while the 121.6 and 190 nm ones will be more absorbed in lower parts. Our results imply then a chemical evolution of the aerosols right after their appearance in the thermosphere. The growth process in the ionosphere will enrich the nanoparticles with N-bearing groups, which will be simultaneously affected by the short VUV radiations, as in our experimental conditions, in order to favor the aliphatic groups. Those results are in agreement with the Cassini-VIMS[7] and CIRS[8] measurements in the stratosphere, showing a main saturated-hydrocarbon signature of Titan's aerosols in the mid-IR range.

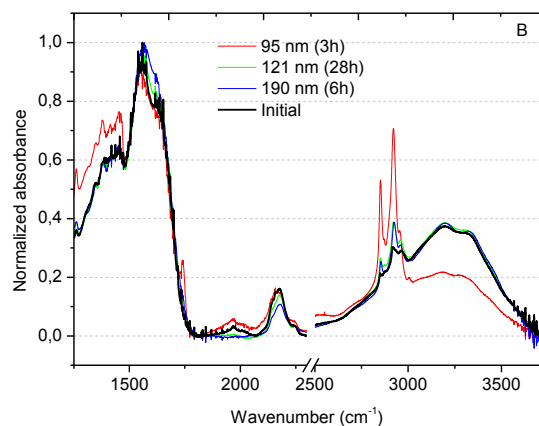


Figure 1: Infrared absorption spectra recorded from samples irradiated by photons for 3h (95 nm), 28h (121.6 nm), and 6h (190 nm). The thick black spectrum is the mean of the non-irradiated spectra.

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Implications of hazes in observations of exoplanet atmospheres

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Abstract

The current inventory of exoplanet observations provide information for the composition and temperature conditions of these environments. Moreover they indicate that hazes can be an important component of their atmospheres. From studies on solar system atmospheres (e.g. Titan) it is well established that the presence of hazes can have significant implications on the thermal structure and composition of a planetary atmosphere. In this work we will present a study for the properties of photochemical hazes in exoplanet atmospheres in terms of their potential size and density distributions. Furthermore we will discuss their implications on the atmospheric heating and photochemistry in regard to the available observations.

Properties of particles in the upper clouds of Venus in the UV-dark and -bright regions as retrieved from the UV and near-IR VMC/VEx images

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Abstract

The nature of ultraviolet contrasts observed on the upper cloud deck of Venus is still not known. To constrain better the properties of particles that may cause the UV contrasts, the phase dependences of brightness of the Venus clouds measured by the UV (0.365 μm) and near-IR (0.965 μm) channels of the Venus Monitoring Camera (VMC of the *Venus Express* mission) in the UV-dark and -bright regions are jointly analyzed. It was found that:

(1) Variations in the composition of submicron particles in the clouds play a key role in the UV contrasts at low latitudes near the local noon. (2) In the pairs of UV-dark and -bright regions, the sizes of the 1- μm mode of cloud particles are the same. (3) The radius of particles in the upper clouds at mid latitudes decreases with latitude: from 1.05-1.2 μm at $\sim 36^\circ\text{S}$ to 0.8-0.9 μm at $\sim 62^\circ\text{S}$. (4) An additional amount of nonabsorbing 0.9- μm particles at the cloud top produces the UV-bright bands at $\sim 50^\circ\text{S}$.

1. Introduction

The range of small phase angles, where the glory phenomenon is observed, is of key importance for the phase-function analysis of the clouds composed of liquid aerosols, because the angular position and the shape of this feature allows of reliable estimates of the properties of spherical particles. Our previous papers were focused on the near-IR (NIR) profiles [1-3], since the interpretation of the data for shorter wavelengths meets the problem of a large number of unknowns (a noticeable contribution of submicron particles and the presence of absorbing material in different modes of cloud particles). At the same time, it was shown that the joint analysis of UV and NIR phase profiles may yield useful results. Unfortunately, only in ten orbits of the mission the images were taken simultaneously in UV and NIR channels at small phase angles. The measured phase profiles were fitted with the radiative-transfer models that took into account (i) the submicron particles of

different composition homogeneously mixed with the 1- μm mode in the clouds and (ii) the overcloud haze.

2. UV contrasts near the equator

We selected the pairs of small UV-dark and -bright regions ($0.2^\circ \times 0.2^\circ$) observed at approximately the same phase and zenith angles, i.e., their different brightness cannot be caused by different observational conditions. Then, the brightness of these regions in two channels was traced along the whole set of images of the orbit, which yielded the phase profiles (Fig. 1, upper thick profiles).

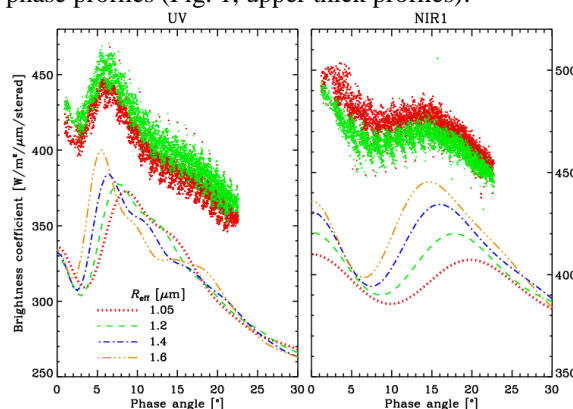


Figure 1: The phase profiles measured in the UV and NIR channels for the groups of UV-dark (red) and -bright (green) regions between 5°S and 10°S (orbit #2146). The UV-dark regions are brighter in NIR. The single-scattering models (thin curves below) are presented for several particle radii (the refractive index is $1.51+0.00005i$ and $1.46+0.0i$ for 0.365 and 0.965 μm , respectively).

Since the position of a glory strongly depends on the particle sizes, a comparison of the measured phase profiles with the single-scattering phase functions of spherical particles immediately yields the estimate of the radius of cloud droplets. The humps in the phase profiles measured for UV-bright and -dark regions are at the same angular position (Fig. 1), which indicates the same sizes of cloud particles mostly contributing to the scattering at the probed level in

the UV-dark and -bright clouds. In the equatorial region near the local noon, the derived radii of cloud particles turned out to be rather high, 1.3-1.6 μm . No unambiguous connection between the UV contrasts and the NIR brightness was found. In some cases, the regions that appear contrasting in UV show no difference in NIR. This means that the properties of 1- μm mode particles are the same there and only the contribution of submicron particles differs. The difference in the composition of 10% of the number of submicron particles (sulphur versus sulphuric acid) is enough to produce the observed UV contrasts. In the other cases, the UV contrasts are accompanied by the NIR brightness differences, which suggests that the cloud particles of the 1- μm mode also contribute to these contrasts. However, the modeling showed that exactly the variations in the composition of submicron particles produce a key effect on the UV contrasts observed. Moreover, a portion of submicron particles with a high refractive index, when incorporated into the sulphuric acid aerosols during the condensation process, may provide the higher (relative to that of sulphuric acid) refractive index for the 1- μm mode particles derived from modeling.

3. UV-bright strip at 50°S

The images with glories acquired in the so-called transition region, where a wide UV-bright strip divides mottled clouds at low latitudes and quasi-laminar flow at higher latitudes, are of special interest. To analyze the properties of particles in this strip, the phase dependences for the regions located along several meridians were built (Fig. 2, upper sets of symbols).

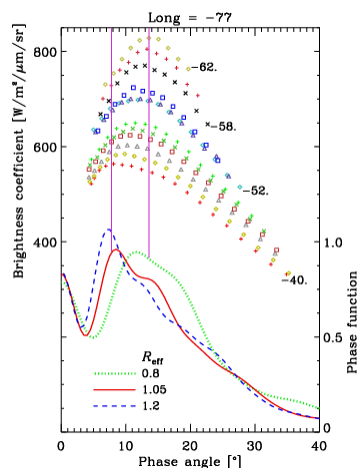


Figure 2: Angular positions of the glory maxima observed in the UV phase profiles for the regions along the -77° meridian with a 2° latitude step (orbit #1918) are compared

to those in the single scattering phase functions (lower curves, right axis) calculated for sulfuric acid droplets of different radii.

Practically all of the phase profiles of the meridian sets, where the glory maximum can be found, show that it moves to larger phase angles for the regions at higher latitudes. This means that the size of particles in the upper clouds decreases toward the pole in the considered latitude range (Fig. 2). Moreover, a sharp increase of brightness in the bright band often occurs with no change of the particle sizes (the maximum remains practically at the same angular position). The modeling with varying properties of particles (of both submicron and 1- μm modes) in the clouds and in the above attached haze confirmed that the profiles for a the UV-bright belt can be well described only by the models containing the 0.9- μm particles in the main upper clouds or in the overcloud haze.

4. Summary

1. Variations in the composition of submicron particles, that inhabit the clouds together with the 1- μm mode, play a key role in the UV contrasts observed at low latitudes near the local noon.
2. In the pairs of UV-dark and -bright regions, the sizes of the 1- μm mode of cloud particles are the same.
3. The effective radius of particles in the upper clouds at mid latitudes decreases with increasing latitude: from 1.05-1.2 μm at 35-40°S to 0.8-0.9 μm at 60-62°S.
4. An additional amount of nonabsorbing particles about 0.9 μm in radius at the cloud top produces the UV-bright bands often observed at $\approx 50^\circ\text{S}$.

Acknowledgements

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Analysis of Titan's haze from Cassini/ISS observations

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Abstract

In-situ observations made by Huygens combined with recent analysis of the optical properties of the haze enable to model the radiative transfer in the Titan atmosphere and interpret observations with a very good accuracy. However, the haze layer presents spatial and temporal inhomogeneities in its optical properties as a result of the particle horizontal transport. These variations in the haze distribution are evident from the observed variations of its geometric albedo along the last decades (see e.g. Lorenz et al. 2004). In this work we will describe how we analyze variations in the optical properties of the haze through the analysis of images taken by the Cassini Imaging Science Subsystem (ISS). The analysis of the images is undertaken by radiative transfer simulations.

1. Description of the model

Since the analysis of images is undertaken for any viewing geometry, the simulations require the use of a full three-dimensional radiative transfer model in spherical geometry. In this work we use a Monte-Carlo radiative transfer model in spherical geometry (Tran, 2005). The phase function, single scattering albedo and density of the particles present in the main haze layer are taken from Tomasko et al. 2008.

2. Analysis

In order to analyze the optical properties of the haze, we simulate light scattered from Titan's atmosphere and then we compare the model results with images taken by the ISS cameras. I/F measurements at the limb can provide data to derive vertical profiles of haze optical properties. As example, **Figure 1** shows the ratio between I/F simulated with the Monte-Carlo model for different values of the scale height (H) that describes the vertical haze opacity above 80 km, and an image taken by ISS cameras in the violet filter. In this example, best results are obtained for H=50 km.

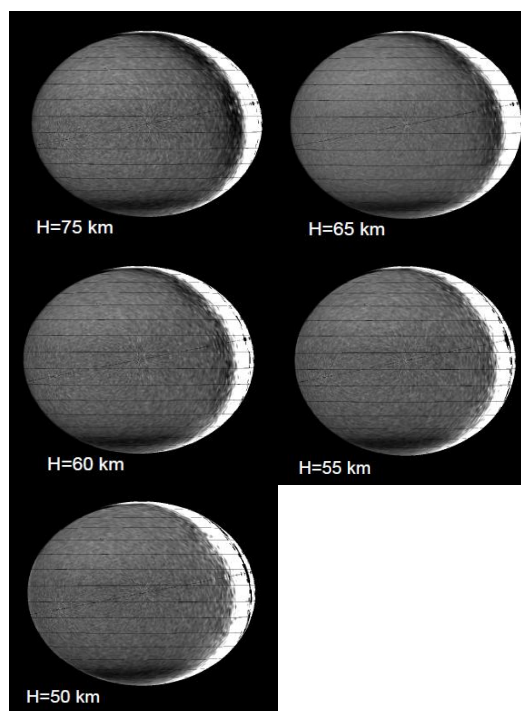


Figure 1: Ratio between I/F simulated with the Monte-Carlo model and measured by ISS in the violet filter.

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Modeling Venus' clouds with the moment method: paving the way for 3D GCM simulations

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Abstract

We describe a project to model the microphysics of Venusian clouds. The goal of the project is to complete the IPSL Venus 3D GCM with a cloud microphysics module.

1. Introduction

Venus is a terrestrial planet, which is enshrouded by clouds. The thickness of this cloud layer is more or less 20 km. The clouds are thin, like cirrus on Earth but they are stratified and create a large opacity.

The cloud layers are surrounded by haze above and below. Moreover, this cloud system is divided by properties of particle size distribution into three layers: the upper cloud deck (56.5 to 70 km), the middle cloud deck (50.5 to 56.5 km) and the lower cloud deck (47.5 to 50.5 km) [2]. The aerosols that constitute the clouds are composed of a $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ solution. The solid state of aerosols is still debated [2,8]. There is only one complete in-situ profile on cloud droplets measured by Pioneer Venus during its descent [2]. The upper cloud deck and the upper haze were studied by several missions like Venus Express [11]. The droplet radii distributions can be divided in several size modes. The mode 1 (mean radius $\bar{r} \simeq 0.2\mu\text{m}$) is the smallest but has the largest number concentrations. Modes 2 ($\bar{r} \simeq 1.0\mu\text{m}$) and 3 ($\bar{r} \simeq 3.5\mu\text{m}$) hold most of the condensed mass [2]. The division in modes 2 and 3 of the largest particles and the existence of mode 0 and 2' are still debated [2,12,10].

The cloud top and base and the thickness change with latitude and the particle size has a latitudinal dependence [12,5]. In addition, an unknown UV absorber is present in the cloud layers and may be related to clouds.

At last, the clouds affect the radiative balance, the sulfur chemical cycle, the dynamics and the atmospheric structure of Venus.

2. Modelling

2.1. The IPSL Venus GCM

The Venus Global Climate Model has been developed at the Laboratoire de Météorologie Dynamique (LMD, France) [4]. The characteristics of this model include radiative transfer, dynamics, atmospheric chemistry, diurnal cycle and a full topography defined by Magellan mission's data. With this full GCM, the Venusian atmosphere is simulated between 0 and 150 km.

However, there are still some problems with vertical temperature description and with the representation of the cold collar. They may be due to the simple description of the cloud layers in the model [4]. Thus, to achieve better simulations of the Venus climate, the GCM needs a microphysical model.

2.2. VenLA

The Venus' cloud model VenLA is developed at LATMOS [6]. It is a 1D sectional microphysics model based on [3]. VenLA is computationally too demanding to be integrated in the IPSL Venus GCM, which is why we need to develop another method.

2.3. The moment method

The moment method is a statistical method to describe a distribution function with few parameters called moments. On the I interval ($I = \mathbb{R}_+$) with the n^{th} moment, the distribution $f(x)$ is defined with the moment scheme by:

$$M_n(f) = \int_I x^n f(x) dx \quad (1)$$

When applied to a particle size distribution, each moment M_n is associated with a meaningful parameter of the distribution. With the equation (1), the moment of order 0 is the total number of particles N and the moment of order 3 is the total volume of the particle

population.

In our case, we consider a log-normal size distribution function (2) [9]:

$$f(x) = \frac{N}{\sqrt{2\pi}\sigma_x} \exp\left(-\frac{(x - \bar{x})^2}{2\sigma_x^2}\right) \quad (2)$$

Where x is a radius, \bar{x} is the mean radius of the aerosol distribution and σ_x is the variance.

The moments will be the tracers in the 3D GCM. A tracer is a quantity that we follow in the modeling calculation. The calculation time of a simulation is proportional to the number of tracers in the model: with few tracers is faster than lots of tracers. With a sectional model like VenLA, each bin is a tracer, which means tens or hundreds of tracers would be added to the GCM. This is why the moment method with two or three moments may be a good method to develop a microphysical module for a global model like the Venus GCM.

2.4. Modeling approach

We are developing a 1D cloud model with the equations of microphysical processes solved with the moment scheme.

Then we will make comparison between our model and the high and low resolution VenLA simulations. With these tests, we will study the ideal number of moments that we need in the model and the moments that we will use: the mean radius, the variance or/and the total number of particles in the distribution.

We will present the first results of our 1D model with the moment scheme and a comparison with the results of high and low resolution VenLA 1D.

3. Summary and Conclusions

The moment method is already used in the IPSL Mars GCM [7] and the IPSL Titan GCM [1] to describe the cloud microphysics. Therefore, it is interesting to use it also in the Venus GCM.

Here we present a status report on the development of the moment method cloud module. The development of this model will allow us to have a better understanding of Venusian climate with a complete GCM.

4. Perspectives

The goal of this model development is to simulate in three dimensions the formation and the evolution of clouds on Venus. It will be integrated in the IPSL

Venus 3D GCM to obtain the more complete Venusian climate model.

Acknowledgements

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VenLA: The LATMOS Venus cloud model

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Abstract

The LATMOS Venus cloud model VenLA (Venus Liquid Aerosols) is based on a terrestrial Polar Stratospheric Cloud (PSC) model [7]. VenLA models the formation, growth and decay of sulfuric acid - water droplets. The model has undergone several updates. We will present results of reference runs and sensitivity tests based on input profiles from VIRA [5] for temperature and pressure and from occultation data concerning the vapors, as in [8]. We compare the results to the available observations on cloud particle number densities and sizes, and to other modeling studies. The VenLA model will define the baseline for a parallel project on development of a moment method scheme to be used in a global climate model (see abstract Guilbon et al., this conference [2]).

1. Introduction

The clouds of Venus and the aerosols in the Earth's stratosphere are close relatives: they are mainly composed of sulphuric acid droplets. The Venus clouds are an important climatic element. The clouds are optically thick and absorb most of the insolation arriving at Venus, letting only few percent of sunlight reach the surface. The clouds on Venus are divided in three layers of differing properties (particle size distributions, number density). Even though the upper clouds can be directly studied with satellite instruments, only a handful of observations have acquired information on the conditions and properties inside the clouds (in particular [6] on cloud properties) and thus the study of the lower and middle cloud layers relies largely on modeling.

2. Methods

The PSC model [7] is a sectional microphysical model that can describe a multimodal particle size distribution discretized with tens or hundreds of radius bins. In its original configuration it is able to model all of the

microphysical processes relevant to the PSCs including the multiple phase transitions related to the particle types including liquid and solid phases of water, sulfuric acid, nitric acid and their mixtures. The PSC model required several modifications when becoming VenLA. The major modifications include: removal of nitrous species and related cloud particle types from the model, addition of homogeneous and heterogeneous nucleation parameterizations, inclusion of the condensation nucleus particle type, addition of coagulation accounting for both single-type and multi-type particle coagulation, and parametrization of vertical mixing via eddy diffusion. Because of the extreme dryness of the Venus atmosphere, we also needed to add iteration in the calculation of the weight fraction of sulfuric acid in the droplet in order to correctly account for the change in total water content.

VenLA receives as input atmospheric profiles of temperature, pressure, vapor concentrations and condensation nucleus properties (a lognormal size distribution defined in a given altitude range [3]). The vapors are consumed during nucleation and condensation and replenished when the droplets evaporate or when mixing brings in vapor-rich air. In these simulations the temperature and pressure profiles are not changed and thus the simulated clouds are considered as formed in average conditions and do not reflect effects of large-scale dynamics. In principle the model can also be used with varying input profiles, but here we focus on using time-independent VIRA profiles [5] only.

Homogeneous nucleation is described with the parameterization of [14] and heterogeneous nucleation with a simple parameterization as in [3]. Condensation/evaporation is treated in two steps: simple (fast) equilibration whenever a droplet experiences a change in environmental conditions (temperature, partial pressure of water vapor) causing a change in the equilibrium composition, and (slow) condensation/evaporation during which the droplet grows/shrinks conserving the equilibrium composition [9]. We account for Brownian and gravitational coagu-

lation (coalescence) and we use the numerical method of [4]. The coagulation kernels are calculated as in [1, 12] and the sum of the Brownian and gravitational kernels is corrected as in [10, 11]. Vertical transport (sedimentation and eddy diffusion) is treated following the method of [13] and settling velocity is calculated using [9, 1] and corrected to account for mixing.

3. Results

We will focus on reproducing the [6] in-situ observations of the cloud properties. We will also probe the variations of the clouds by using the VIRA profiles from different latitudes. One of the main sensitivity tests will be the effect of CN on the cloud properties. The nucleation pathway may prove significant in the simulations since it defines the number of formed particles. This regulates the particle size for a constant condensable mass. In our preliminary test runs, when using only the homogeneous nucleation parameterization, we reach the observed condensed mass load, but the droplet number concentrations are too low and consequently the droplets are too large. Using heterogeneous CN activation it is much easier to attain observed number concentrations and sizes, however, the used initial CN concentration profile plays a role in the development of the cloud. We will initialize our CN profiles following previous studies to enable a direct comparison.

4. Summary and Conclusions

The VenLA cloud model and reference and sensitivity run results will be presented. The results will be compared with published modeling studies and observations. The role of CN will be put in particular focus.

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A radiative model for Titan's atmosphere in the IR

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Abstract

The aim of this work is the development of a model of Titan atmosphere between 1 and 5 micron, using data from Cassini-Huygens mission. The simulations will be useful to remove the atmospheric features from the measured spectrum, to study the surface. The radiative transfer model is performed with ARS (Atmosphere Radiation Spectrum), a a group of Fortran 77 routines, able to calculate absorption coefficients, radiance and other parameters about gas and aerosols at LTE (Local Thermal Equilibrium) [5] and considering multiple scattering in nadir geometry. Our study covers the IR spectral range but it would be extended also to the visible spectrum.

1. Introduction

1.1 Physical and chemical characteristics

With a mean density of 1.8 g/cm^3 , Titan is likely formed by rocks and ice, like Dione and Enceladus but with an higher density for the gravitational compression probably due to a salty ocean present under the surface [7]. The pressure and the temperature at the surface are 1500 hPa (1.5 times Earth pressure) and 95 K, respectively.

The atmosphere is mainly composed by nitrogen but also by methane that is 5% at the surface and decreases to 2% in the upper layers. [9]

Methane plays an important role in atmosphere dynamics and structure, since photoionization forms different organic compounds. It was demonstrated by the Huygens probe that provided precious information about atmosphere dynamics and the interactions between surface and atmosphere.

Also complex hydrocarbons (ethane, acetylene), PAHs (benzene) and nitriles are present and form haze of reddish-brown organic compounds (tholin).

Hydrocarbons and compounds are formed by the photodissociation and ionization of molecular nitrogen and methane. They are carried by precipitation to the surface, so methane and ethane condense. [12]

1.2 VIMS and HASI instruments

In this work we compare the spectrum obtained by VIMS (Visible and Near-Infrared Mapping Spectrometer) with the simulated spectrum. VIMS is a spectrometer aboard the Cassini spacecraft, aimed at the study of Saturn system. It acquires images in 352 channels with a angular resolution of 0.25×0.25 mrad between 0.85 and $5.2 \mu\text{m}$ for the IR channel. Its spectral resolution is 16 nm [1].

HASI (Huygens Atmospheric Structure Instrument) was part of the probe Huygens a multi-sensor package designed to measure the physical properties of Titan's atmosphere. [3]

2. Proceedings and data

The ARS code uses the main equation of the radiative transfer to produce the synthetic spectrum. It takes into account also Mie scattering, valid when a particle is bigger than an incident wave, as for the haze layers in our case. We extracted the molecular information from the HITRAN 2012 and Sromovsky et al. 2012. [10,11] databases. [2]

Despite methane is considerably less than nitrogen, it characterizes the main features in the IR spectrum. All other molecules (N_2 , HCN, C_2H_2 , C_2H_4 , C_2H_6 , CO_2 , CO) can be neglected for the aim of this work.

We use the temperature-pressure profiles taken from HASI (data on PDS nodes) and observation geometries of a specific observed spectrum as input for the simulations. The surface albedo is difficult to interpret for the optically thick and complex atmosphere. Therefore, we used an approximated constant albedo and then we tried with Negrao's profile [8]. The atmosphere has been split in 101 layers, with a constant layering step of 10 Km. Moreover, we assumed constant all the calculated physical parameters in each single layer. Measured spectra used for our comparison are average over 7 point, taken by the different flybys of Cassini. [2]

2.1 Aerosols

Titan's aerosol spreads in a main haze under 300 km and in other separated layers at 520 km and at 1000

km. These particles have a fractal-aggregate and also spherical nature. The spectrum at different wavelength is strongly dependent by them. For the model we used the particle size distribution by Lavvas [6], that is unimodal in the upper atmosphere, then it become bimodal. We calculated the MIE scattering properties of assumed spherical particles using the refractive index data by Khare. [4] [2]

3. Results

We are producing the radiative transfer model of Titan atmosphere in the infrared range. At first we tested the compatibility of ARS with Titan atmosphere changing different parameters. During the testing we added other methane bands by Sromovsky improving the comparison with the measured spectrum. We introduced collision-induced absorption of the N_2 and CH_4 molecules. Lavvas distribution improved a lot the quality of our model. The synthetic spectrum is in good agreement with the measured one, but the fit must be still improved at certain wavelength. Our work will continue changing values of albedo in each wavelength and also using different measured spectra. [2]

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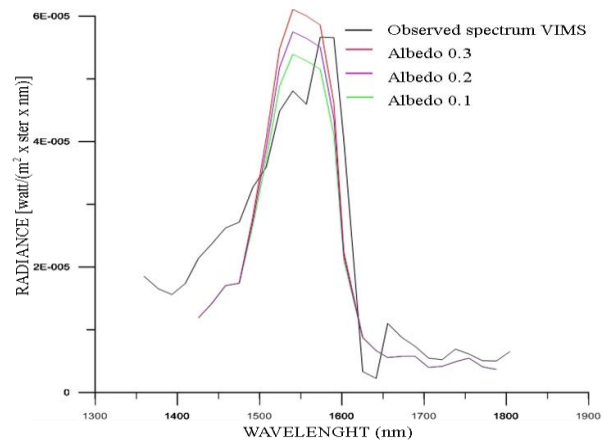


Figure 1: Simulated (only CH_4) and observed radiance spectrum of Titan at different albedo, range 1350-1800 nm.

Six-year operation of the Venus Monitoring Camera (Venus Express): spatial and temporal variations of the properties of particles in upper clouds of Venus from the phase dependence of the near-IR brightness

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1. The VMC data

Since May, 2006, the Venus Monitoring Camera (VMC) [1] has been imaging Venus in four narrow spectral channels centered at the wavelengths of 0.365 μm (UV), 0.513 μm (VIS), 0.965 μm (NIR1), and 1.010 μm (NIR2). It took around 300 000 images in four channels covering almost all the latitudes, including night and day sides.

We analyze the whole set of the VMC data processed to October, 2012, *i.e.* the data from orbits 60–2352 obtained in the phase angle range $\alpha = 0^\circ - 140^\circ$. The data for the fixed incidence angles Z_0 and latitudes $((30^\circ\text{N} \dots 70^\circ\text{S}) \pm 2^\circ$ with step 10°) were selected and distributed among the 1° -intervals of the emission angle Z and the local solar time (LST) interval 6–18 h. For every Z , the 1° -binning by a phase angle was done, and the mean value and the standard deviation in the bins were calculated. The latter turned out to be very small, usually less than 0.01%. Each value of brightness obtained in such a way corresponds to a specific combination of photometric angles Z_0, Z, α , and the modeling was performed for each of them.

2. Modeling the phase dependence of brightness

To estimate the properties of cloud particles, we modeled the phase dependence of brightness retrieved from the daytime images obtained in NIR1 VMC channel. The choice of the NIR wavelength decreases the number of free parameters, since there is no NIR absorption in the Venus clouds. The radiative transfer calculations were performed for the plane-parallel atmospheric layers, and the

single scattering phase functions were found with the Mie theory. The standard gamma size distribution of cloud particles was assumed; the effective radius R_{eff} was varied from 0.9 to 1.4 μm (the 1- μm mode), the effective variance ν_{eff} was mainly fixed at 0.07. The real refractive index m_r of the 1- μm mode was varied from 1.44 (typical of H_2SO_4) to 1.49.

Since submicron particles are known to be ubiquitous in the Venus clouds and hazes, their presence was also taken into account. Their properties were assumed as $R_{\text{eff}} = 0.23 \mu\text{m}$, $\nu_{\text{eff}} = 0.18$, and m_r corresponding to H_2SO_4 . For submicron particles in the clouds the sulfur composition was considered as well. For the haze submicron particles, the larger radii, up to 0.9 μm , were also tested. The percentage of submicron particles N_{sub} in the main cloud layer, and the optical depth of the overcloud haze τ_{haze} were varied [2, 3].

The total number of models was 1024 (see the on-line supplementary material of [3]). Most of them were calculated only for one value of the optical depth of the clouds $\tau = 30$, since the sensitivity of the phase profiles to this parameter was found to be relatively small. The best fits were chosen automatically by the least square method.

The examples of the best fits are shown in fig. 1. They illustrate the tendency in the behavior of submicron particles: the submicron particles are detected in the haze and/or clouds mostly in the morning. Of course, this fact does not mean that there is no submicron mode in the afternoon and in the evening in the Venus clouds; this only means that its amount diminishes, and the NIR1 channel cannot distinguish it, because its sensitivity to submicron particles is low (as compared to UV).

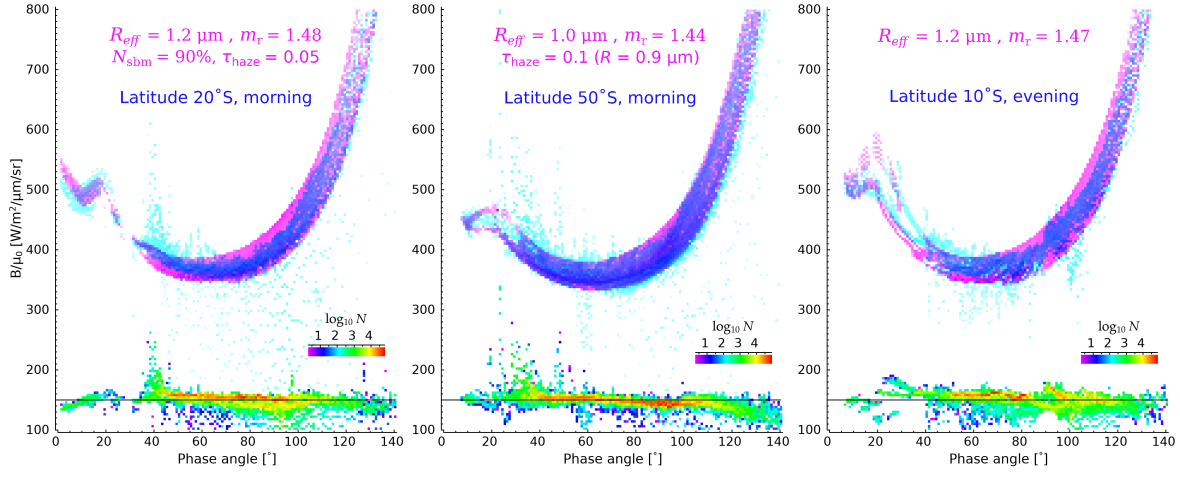


Figure 1: Examples of the best model fitting for the data obtained in the morning (LST = 6–10 h; two left panels) and in the evening (LST = 16–18 h; right panel) at $Z_0 = 70^\circ$ and different latitudes. Superposition of cyan (observation) and magenta (model) colors produces dark blue color. The parameters of the models are specified in the plots. Every point in the observational profile (cyan color) corresponds to the specific combination photometric angles Z_0 , Z , and α . The model (magenta color) was calculated for each of these points according to its photometric conditions. Deviations of the model from the observations ($\frac{1}{2}(Data - Model) + 150 \text{ W m}^{-2} \text{ sr}^{-1} \mu\text{m}^{-1}$) are shown at the bottom of each plot, where N is the number of observations at the given photometric conditions (see color scale). Orbits 60–2352.

3. Summary

The following temporal and spatial variations of the sizes and refractive index of cloud particles were found from the phase dependencies of brightness measured by the NIR1 VMC channel:

- The presence of particles with $R_{\text{eff}} \approx 0.9 \mu\text{m}$ in the clouds and/or the overcloud haze is pretty confident at latitudes $40^\circ\text{S} - 60^\circ\text{S}$.
- In general, the particles at low latitudes are somewhat larger than in the regions closer to the southern pole ($R_{\text{eff}} = 1.2 - 1.4 \mu\text{m}$ versus $0.9 - 1.05 \mu\text{m}$).
- The refractive index of cloud particles at latitudes $40^\circ\text{S} - 60^\circ\text{S}$ is usually smaller than that closer to the equator ($m_r = 1.44 - 1.45$ versus $1.45 - 1.47$ with sporadic spikes up to 1.49).
- No clear tendency in the temporal behavior of the refractive index and sizes of cloud particles during the local day is observed. The exception is the presence of small submicron particles ($R_{\text{eff}} = 0.23 \mu\text{m}$) detected mostly in the morning within the clouds and/or in the haze above the clouds.

The procedure and results of this analysis are described in detail in [3].

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The atmospheric vertical structure in the Saturn polar hexagon: Insights from Cassini and ground-based data in the visible and near-infrared

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Abstract

Saturn's polar hexagon is a very stable atmospheric feature discovered by the Voyager spacecraft at the boundary of the northern polar cap. Many studies have been devoted to the measurement of wind speed across this structure, but detailed quantitative retrievals of atmospheric parameters and their vertical distribution is actually missing. We will report a work in progress based on the analysis of data from Cassini (VIMS, ISS) and ground-based observations (IRTF-SpeX). We will mainly focus on the variations of clouds and hazes vertical profiles and microphysics, and those of the abundances of trace gases to which the near-infrared wavelengths are sensitive (phosphine, ammonia, CH₃D, water, etc.). These quantities can also be used as tracers of atmospheric motions and can help in constraining a comprehensive dynamical framework.

1. Introduction

The northern latitudes of Saturn host a remarkable atmospheric structure which survived exceptionally stable for at least the 30 years passed since the Voyager encounter: the polar hexagon, which marks the boundary of the northern polar cap at a latitude of about 75°N. When the Cassini spacecraft reached the Saturn system, allowing unprecedented investigations of the atmosphere in terms of spatial resolution and coverage, the regions poleward of about 80°N were in permanent night, and new information about the polar cap only came from thermal and non-LTE emissions. These studies allowed comprehensive descriptions of the atmospheric thermal field in the troposphere and stratosphere by means of mid- and far-infrared wavelengths (Cassini-CIRS [1]) as well as the measurement of tropospheric wind velocities by tracking the motion of the deeper small discrete clouds revealed by Cassini-VIMS [2] in the 5 micron

window [3]. Dynamical implications were deduced from these investigations (e.g. the confirmation of a stable jet stream embedded in the hexagon track [4]) and moderate seasonal trends were inferred from temperatures retrieved passing the 2009 northern spring equinox [5].

The better and better illumination condition taking place by approaching the next 2017 northern solstice makes very effective the investigation of atmospheric properties by means of reflected sunlight spectroscopy, in the whole UV to near-infrared spectral range. This kind of observations are mostly devoted to the measurement of radiative absorption by atmospheric gases and anisotropic scattering by gases and particulates. They are made very sensitive to the vertical distribution of atmospheric components in the tropo-stratosphere by the very large spectral variation of the methane absorption coefficient in this spectral range.

However, recent studies of the polar hexagon in reflected light (using VIMS [3] and ISS [6] data) were mainly devoted to describe the horizontal motions and the meridional wind profile, while a comprehensive inclusion of the changes in vertical structure and vertical motion tracers (minor gases and particulates) is still lacking.

2. Data and methods

The primary dataset useful for our investigation is that of Cassini-VIMS. We have searched the VIMS data relative to the polar hexagon in order to select a set of observations covering a wide range of observing conditions and times. Particular attention has been paid to observations at high solar phase angle, since spatial and temporal changes in vertical distribution are more evident in forward scattering configurations.

VIMS data usually offer good spatial resolution at Saturn due to the close observing range, but on the other hand suffer of a moderate spectral resolution (about 17 nm in the IR). Ground-based observations of Saturn at NASA-IRTF Telescope at Hawaii have been acquired in the last months, using the SpeX spectrograph, in order to achieve a better understanding of the VIMS spectra in the 3-4 micron range. Thanks to the higher spectral resolution of this instrument (about 11 times the VIMS one) we are able to investigate subtle changes in the methane bands shape and discriminate the effects of variations of temperature and vertical cloud structure.

Plane-parallel multiple-scattering 1D radiative transfer models have been used to explore the dependencies of spectral variations through sensitivity studies, and to retrieve the most significant atmospheric parameters. Retrievals are based on both chi-square best fit and Bayesian inversion approaches [7].

3. Summary and Conclusions

Here we will report about in progress investigations of spectral data (both from spacecraft and ground-based) covering the Saturn's polar hexagon in different times and observing conditions, aimed to describe the distribution of particulates at the upper troposphere-lower stratosphere altitudes. The retrieved quantities can put further constraints to the dynamical framework of the hexagon. The synergistic use of data from several instruments (e.g. Cassini-VIMS, Cassini-ISS, and ground-based) is expected to strengthen the outcomes of retrieval models at all latitudes and hence also improve our understanding of Saturn's atmospheric dynamics and seasonal changes.

Acknowledgements

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Wind Shear May Produce Long-Lived Storms and Squall Lines on Titan

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Abstract

The impact of CAPE and wind shear on storms in a Titan-like environment are explored through numerical simulation. Model results indicate that Titan storms should respond to changes in the Richardson Number. Very long-lived storms (>24 hours) propagating for 1000 km or more might be possible. Varying amounts of shear in the Titan environment might explain the variety of convective cloud expressions identified in Cassini orbiter and ground-based observations. The resulting distribution and magnitude of precipitation as well as surface winds associated with storms have implications on the formation of fluvial and aeolian features and on the exchange of methane with the surface and lakes.

1. Introduction

The presence of convective clouds on Titan is now well established, e.g. [1][2][3][4]. Cloud resolving simulations are able to reproduce the observed cloud top heights, the rate of cloud top rise, and the overall timescale of typical convective cells [5]. An increasing number of observations of Titan's convective clouds suggest, however, that not all the deep convective clouds behave the same. There are indications that convective clouds may be able to organize into mesoscale systems [6] or long-lived linear systems [7][8].

Besides convective available potential energy (CAPE), which is a measure of the total energy available to convection, vertical shear of the horizontal wind is known to strongly control the morphology and dynamics of convection on Earth. Given a fixed amount of CAPE, terrestrial storms transition from short-lived single cellular convection to long-lived multi-cellular convection as shear increases. Under some conditions, individual convective cells can organize into long-lived squall lines. This study focuses on the combined impact of

CAPE and shear on Titan's storms, as determined from numerical modeling.

2. Numerical Experiment Design

A series of idealized two-dimensional numerical simulations were conducted that closely mirror the experiments done by [9] for terrestrial convection. The numerical model is the Titan Regional Atmospheric Modeling System (TRAMS), as described in [10]. The thermal environment for all simulations is identical and is taken from the retrieved Huygens HASI temperature profile [11]. Nonzero CAPE scenarios were generated by increasing the methane mixing ratio in the lowest 4 km of atmosphere of the profile from [12]. When these CAPE scenarios are combined with four different low level shear profiles a total of eight storm scenarios are generated (Table 1).

3. Results

Numerical modeling indicates that both large-scale shear and CAPE environment control the dynamics of the clouds. This response to the large-scale environment is analogous to the behavior of deep convective clouds on Earth. The balance between shear and CAPE, as expressed through the bulk Richardson Number (N_R) is a good indicator of the response of a storm to its environment. Large N_R results in short-lived single cell storms (Figure 1). As shear increases for a given CAPE, and N_R decreases, the storms transition to a multicellular regime (Figure 2). Multicellular storms are longer-lived and are characterized by a downdraft generated cold pool that interacts with the background shear vorticity to initiate cells along the leading edge of the storm gust front. The most intense multicellular systems simulated in this study behave similar to terrestrial squall lines. Cloud outbursts and linear cloud features observed from ground and Cassini may be the result of these organized storm systems.

Surface winds beneath storms can exceed 10 m/s and total precipitation may be measured in meters; the erosion potential by wind and precipitation is extremely large. Multicell storms can translate over 1000s of kilometers during their lifetime of 12 or more hours. Winds from storms should be more than sufficient to produce waves on any nearby lakes, and the wind stress should also be sufficient to initiate aeolian activity, including over the widespread sand and dune seas. Wind and precipitation from intense but episodic storms may be the primary mechanism shaping the surface of Titan.

3. Figures

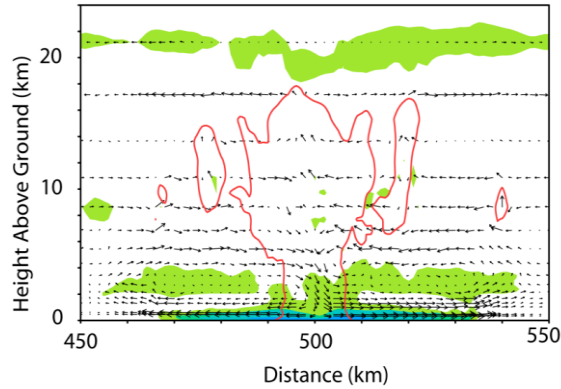


Figure 1: The mature phase of a single cell storm. Red lines indicate cloud boundaries. Vectors show storm-relative motion. Shading indicates sub-cloud cold air mass. The shallow cold air spreads away from the storm and cuts off the supply of convectively unstable air.

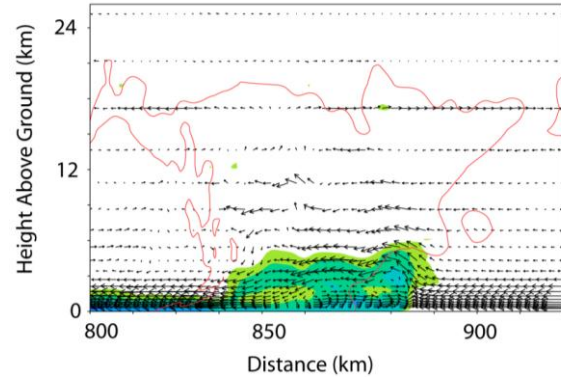


Figure 2: The mature phase of a multicell storm. The cold air lifts convectively unstable air into the storm and produces a continuous train of new cells that maintain the storm for hours.

4. Tables

Table 1: CAPE and SHEAR Scenarios

| | | CAPE (J/Kg), Mixing Ratio Perturbation (g/kg) | |
|---------------------------------------|----|---|----------------|
| | | 250, 5.0 | 500, 10.0 |
| Shear m(s ⁻¹) per 5 km | 0 | U0R5; Ri=∞ | U0R10; Ri=∞ |
| | 1 | U1R5; Ri=1250 | U1R10; Ri=2500 |
| | 5 | U5R5; Ri=250 | U5R10; Ri=500 |
| | 10 | U10R5; Ri=125 | U10R10; Ri=250 |

Acknowledgements

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The Clouds of Venus – an overview of Venus Express results

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Abstract

Venus is completely enveloped by clouds. The main cloud layers stretch from altitudes of 48 – 75 km, with additional tenuous hazes found at altitudes 30 – 100 km. Clouds play a crucial role in governing atmospheric circulation, chemistry and climate on all planets, but particularly so on Venus due to the optical thickness of the atmosphere. The European Space Agency's Venus Express (VEx) satellite has carried out a wealth of observations of Venus clouds since its arrival at Venus in April 2006. Many VEx observations are relevant to cloud science – from imagers and spectrometers to solar, stellar and radio occultation – each covering different altitude ranges, spectral ranges and atmospheric constituents.

We have formed an International Team at the International Space Science Institute to bring together scientists from each of the relevant Venus Express investigation teams as well as from previous missions, as well as those developing computational and analytical models of clouds and hazes. The aims of the project are (1) to perform intercomparisons of cloud parameters measured using different

techniques, (1) to create self-consistent reference cloud/haze models which capture not only a mean cloud structure but also its main modes of variability; and (2) to bring together modelers and observers, to reach an understanding of clouds and hazes on Venus which matches all observables and is physically consistent.

This talk will present an overview of Venus Express cloud observations of all different types, and discuss progress towards a new reference cloud model to be submitted to an update of the Venus International Reference Atmosphere.

Acknowledgements

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Radiation-induced near-surface atmospheres of Europa and Titan

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Abstract

We will discuss and compare photochemical processes on Europa and Titan, focusing on the near-surface and lower-atmospheric organic composition on both of these moons. Europa's surface receives high doses of radiation that can easily oxidize organics in the presence of water-ice. Titan's atmosphere is depleted of oxygen, but enriched with organic molecules, including condensed aerosols.

In this contribution we will focus on photochemistry caused by longer wavelength UV-VIS photons (>250 nm) photons that can pass through Titan's atmosphere to the haze region (~ 100 km) and onto the surface of Titan [1, 2] and electron-induced processes of organics on Europa's surface, leading to the formation of Europa's tenuous atmosphere [3-6]. We then compare the role of organics on both of these astrobiologically-important icy bodies in our solar system.

1. Introduction

The Cassini-Huygens mission revealed that the nitrogen- and methane-dominated Titan atmosphere of Titan is complex and more active than previously thought [7, 8]. Until recently it has been assumed that photochemistry is only confined to the upper atmosphere, where high-energy UV photons, electrons, Saturn's magnetospheric particles, and solar wind can penetrate. At lower altitudes, due to the lack of high-energy radiation sources (other than cosmic rays), it is expected that no further photochemistry could occur. Recent studies in our

laboratory clearly demonstrated that ices and aerosols in Titan's atmosphere could further undergo photochemical transformations that result in the formation of larger organics [9-11] in Titan's haze region in its lower atmosphere (<100 km). These covalently-bonded organic aerosols containing predominantly C, H, and N elements finally rain down onto Titan's surface [12, 13].

Unlike Titan (with close to a 1.5 bar atmosphere), Europa does not have an atmosphere. Sputtering through radiation, ion, and micrometeoroid bombardment results in the formation of only a tenuous atmosphere on Europa. The survival of organic matter on Europa's surface under these conditions is not well understood. Our work on radiation-processed organics in ice sheds more light on the fate of organics on Europa and what kinds of ions and neutrals are generated near the surface that could form the tenuous atmosphere of Europa.

2. Laboratory Experimental Studies

Most of this work has been conducted at the *Ice Spectroscopy Lab (ISL)* and at the *Titan's Organic Aerosol Spectroscopy and chemisTry (TOAST)* lab of Gudipati at JPL.

This talk focuses on photochemical processes of volatile condensates on pre-formed Titan organic aerosols, simulating the lower-atmospheric and surface processes on Titan. We carried out laser irradiation studies using 532 nm, 355 nm, and 266 nm (Nd-YAG laser 2nd, 3rd, and 4th harmonics). IR and UV spectra were monitored simultaneously to follow the reaction kinetics and reaction products.

In Europa simulations, we irradiated organics such as polycyclic aromatic hydrocarbons (PAHs) and subjected them to electron and photon bombardment. We conducted UV, IR, and mass-spectrometric analysis on these ices.

Acknowledgements

This research was carried out partly at the Jet Propulsion Laboratory (JPL, California Institute of Technology) and partly in France at various laboratories. The JPL portion was carried out under a contract with the National Aeronautics and Space Administration. Funding through the NASA Planetary Atmospheres Program, an earlier NASA Astrobiology Program "Titan as a model prebiotic system", and JPL's funding for the infrastructure is gratefully acknowledged.

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Photochemistry of condensed species on Titan's aerosols analogues

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Abstract

The formation of the aerosols of Titan starts in the upper layers of the atmosphere at about 1000 km. Then, they aggregate and sediment to the surface. Despite the intense work made on the comprehension of the aerosols formation, their chemical evolution during their sedimentation through the stratosphere and the troposphere remains unknown. In this work we investigate how the aerosols can interact with condensed species present in these atmospheric layers and chemically evolve when irradiated by UV solar photons.

1. Introduction

Titan, the largest satellite of Saturn, has a dense atmosphere made of molecular nitrogen (98 %) and methane (2 %) [1]. Dissociation and ionization of N_2 and CH_4 in the ionosphere of Titan at about 1000 km, by VUV solar photons and energetic particles from the Saturn's magnetosphere produce heavy organic molecules [2]. This chemistry initiates the formation of the brownish haze surrounding Titan. Then, the particles aggregate and sediment to the ground.

The measurement of the temperature profile in Titan's atmosphere by the HASI instrument onboard the Huygens probe shows a drop of temperature in the lower atmosphere with a minimum about 70 K at 44 km of altitude [3]. These low temperatures allow a possible condensation of photochemical products on the aerosols surface during their sedimentation through the stratosphere and the troposphere [4]. Different emission bands in the far infrared have been observed by the IRIS instrument of the Voyager spacecraft and by the CIRS instrument of the Cassini spacecraft and attributed to condensed species [5] [6].

If high-energy solar photons initiating the organic chemistry in the ionosphere of Titan's have been significantly absorbed at the altitude where species can condense, photons with longer wavelengths are still present at altitude as low as 100 km. A chemical evolution of aerosols can then be initiated by the reactivity of aerosols coated with condensed species and irradiated by solar photons in the UV-visible range.

Here we present a first experimental study of the aging processes of Titan's aerosols coated with condensed species and irradiated by UV-visible solar flux in the lower atmospheric layers of Titan.

2. Material and methods

To study this phenomenon, we irradiate condensed species coated on a blank sapphire window or coated on thin films of Titan's aerosols analogues, named tholins, with a UV laser.

The analogues are produced by subjecting a N_2 - CH_4 mixture to a radio-frequency discharge [7].

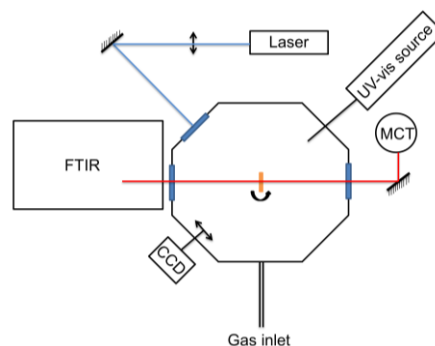


Figure 1: Scheme of the Acquabella experimental setup at the JPL's Titan Organic Aerosol Spectroscopy and chemisTry (TOAST) laboratory.

A blank sapphire window or a tholins sample is mounted in the Acquabella experiment [8]. C_2H_2 is deposited at 50 K to form an ice film on the top of the tholins sample. C_2H_2 is one of the most abundant species in the atmosphere of Titan's with a mixing ratio about 2 ppm_v as measured by the CIRS instrument at 120 km [9] and solid C_2H_2 has been observed in Titan's stratosphere [10].

The acetylene consumption during the irradiation is monitored using absorption infrared spectroscopy. The evolution of the tholins sample, after the sublimation of the remaining ice, is monitored by absorption infrared spectroscopy.

3. Results

We conducted a series of experiments by depositing a thin layer of acetylene ice over the tholins film and subjected these samples to various solar spectra wavelengths using a tunable laser system. We found that while at longer wavelengths no significant chemistry was noticeable, but at shorter wavelengths, particularly 355 nm a significant amount of acetylene was depleted, indicating chemical bonding of acetylene to the tholins polymer.

Controlled experiments showed that a higher consumption of C_2H_2 coated on a tholins sample occurred compared to the irradiation of C_2H_2 ice film alone. Further experimental results and analysis of the data will be presented.

Acknowledgements

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On the reactivity of anions in Titan's atmosphere: Synthesis of their precursors and low-temperature experiments

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Abstract

The presence of numerous negative ions in the upper atmosphere of Saturn's biggest satellite Titan has been revealed by the Electron Spectrometer, one of the sensors making up the Cassini Plasma Spectrometer. The presence of anions was not expected, and their formation mechanisms remain mostly unknown. Anions may play some important role in the production of the aerosols which in turn contribute to the formation of a dense haze which blackens Titan's surface.^[1] The investigation of their reactivity appears indispensable.

The first objective of our work is to synthesize precursors of anions of astrophysical interest, mainly C_xH^- (where $x = 2, 4, 6$) and C_xN^- ($x = 1, 3, 5$) selected for their probable presence.^[2] The second objective is to investigate the reactivity of these molecular anions down to low temperatures in gaseous phase with the help of the CRESU apparatus (French acronym for Kinetics of Reactions in Supersonic Uniform Flows). Reactions between anions and abundant heavy molecular species are likely to contribute to the growth of molecular anions in Titan's upper atmosphere.

For choosing the appropriate precursors of the anions, there are certain criteria that should be satisfied. First of all, the synthesized and purified precursor should be stable. That is, it shouldn't polymerize, decompose, or become self-reactive due to pressure or temperature. Also, the vapor pressure of the precursor should be rather high. Another important criterion is related to the dissociative electron attachment of the precursor ($AB + e^- \rightarrow A^- + B$). The electron attachment on the precursor should be efficient at low electron energy (close to 0 eV) and

upon dissociation, the dominating exit channel should be the one that leads to the desired anion.

The reactivity of the molecular anions is then investigated with the CRESU which is designed to operate in the gas phase and down to low temperatures (from 300 down to 50 K). It consists of a Laval nozzle in which the gas expands to generate a supersonic beam. Its temperature is set according to the design of the nozzle and the pressure in the chamber and reservoir. An electron beam which crosses the flow generates a plasma which initiates ion chemistry. The anions of interest are produced by dissociative electron attachment onto the synthesized precursors. A moveable quadrupole mass spectrometer coupled to a Langmuir probe measure the ion population and the electron density respectively. The molecular co-reactant is introduced in large excess in the flow through the reservoir to perform pseudo first order kinetic experiments.^[3]

This study aims to determine the rate coefficients and branching ratio of selected anion-molecule reactions down to low temperatures. Our work should contribute to the understanding of complex chemistry taking place in Titan's atmosphere. Farther from us, this is also of interest for the cold chemistry of molecular clouds and circumstellar envelopes in which anions have been recently detected.

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Towards further understanding of the negative ion and nanograin dataset acquired by Cassini at the moons Titan and Enceladus

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Abstract

The detections of negative ions up to $\sim 13,800$ amu/q in Titan's ionosphere by the Cassini Plasma Spectrometer (CAPS) Electron Spectrometer (ELS) was remarkable in that the observations were made by an instrument designed and calibrated for measuring electrons [1, 2, 3]. These species have been detected whenever the CAPS-ELS is oriented in or close to the spacecraft 'ram' velocity vector at altitudes below 1450 km, where they revealed a complex and chemically dynamic atmosphere rich in organic molecules. In addition to this, at Enceladus, negative water clusters [4] and fluxes of nanometer-sized water-ice grains were detected at the top of the instrument's E/q range [5] within the Enceladus plumes. These were similarly detected in the spacecraft ram direction whenever Cassini traversed the moon's 'plume ionosphere' at high southern latitudes. In this study we present further examination into these negative ion and nanograin data-sets through an increased understanding of the instrument response to these unexpected species.

The intrinsic energy resolution, $\Delta E/E=17\%$, of the instrument [6, 7, 8], as well as spacecraft potential effects, limit the positive identification of specific species, as the instrument was designed for electrons. This corresponds to $\Delta m/m$ for mass spectra when observed.

In earlier analyses at Titan, the negative ion observations were presented in broad 'mass groups' based on the observed spectra [1], [3]. We explore the idea that as the CAPS-ELS scans through the spacecraft ram direction, oversampling of Titan's atmosphere occurs due to the relatively high rate at which the scans in energy are performed compared to the rate at which the sensor is actuated, potentially adding an extra dimension to the data-set.

Convolving ELS calibration data with the Titan data, we examine trends in the spectra which may be interpreted as being due to the presence of specific masses. We discuss these in relation to corresponding negative ion species identified as likely to be present in Titan's upper atmosphere [1, 9].

The ELS Microchannel Plate (MCP) efficiency to negative ions and nanograins has previously been estimated as a uniform 5% over the ELS energy range, using a study conducted by Fraser [1, 10, 11]. In this analysis we present updated MCP efficiency curves relevant to the Titan and Enceladus negative ion and nanograin detections.

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Kick-starting aerosol formation on Titan with ion chemistry

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Abstract

The Cassini Huygens probe has revealed the presence of abundant and heavy ions in the upper atmosphere of Titan [1, 2]. These so-called macromolecules are now considered to be key intermediates in the production of aerosols observed at lower altitudes [3]. Under this scenario, macromolecules are formed and grow by gas phase ion-neutral reactions in the ionosphere. There they attach efficiently free electrons and recombine with positive ions leading to a rapid gain in mass. As they settle down in the cold atmosphere, they continue to grow until they reach a size where coagulation takes over. However, the composition of these macromolecules and their precise formation mechanisms are still largely unknown. It includes the first steps leading to the production of the macromolecules, which govern the growth rate.

In particular, negative ion cold chemistry has not been explored systematically. Although there have been a number of experimental studies conducted to determine the kinetics of anion-neutral reactions, a fraction only has simultaneously led to the determination of the nature of products and even less to the branching ratio into the different exit channels.

In the laboratory, we recently engaged in kinetics studies of anion-molecule reactions starting with the reaction of CN^- and C_3N^- with cyanoacetylene HC_3N over the 50-300 K temperature range using the CRESU technique (French acronym standing for Reaction Kinetics in Uniform Supersonic Flow). The results show that the $\text{CN}^- + \text{HC}_3\text{N}$ reaction contributes directly to the growth of larger anions [4] whereas $\text{C}_3\text{N}^- + \text{HC}_3\text{N}$ does not [5].

The investigation is now extended in the laboratory to other anions (such as C_4H^-) through the synthesis

of adapted molecular precursors. The development of a versatile selected anion source, which will be combined with the CRESU apparatus, will be also presented.

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First steps of neutral hydrocarbon cluster formation in Titan's atmosphere: a laboratory kinetics approach

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Abstract

Titan's dense atmosphere is the siege of a complex photo-chemistry initiated by the dissociation of its two most abundant components, nitrogen N_2 and methane CH_4 . This cold chemistry generates a plethora of hydrocarbons and nitriles and takes part in the production of a thick orange haze. The detection of heavy neutrals and positive ions in Titan's upper atmosphere by the Ion and Neutral Mass Spectrometer (INMS) embarked onboard Cassini [1] and measurements of significant amounts of negative ions with masses up to 13 000 amu with the Cassini Plasma and Electron Spectrometer (CAPS/ELS) [2] suggest that low temperature chemical reactions and physical processes occurring at high altitudes near 1000 km could be the haze source. This haze material could act as a nucleus for the condensation of organic vapors in Titan's stratosphere and troposphere.

In any event, evidences to support this hypothesis are still partial. The pathways leading to the formation and growth of haze aerosols remain far to be well understood. From a chemical viewpoint, for instance, the low temperature kinetics and the nature of the products of negative ion-molecule reactions are poorly known. From a physical viewpoint, the precise steps of hydrocarbon condensation remain elusive too.

Hydrocarbons, which are formed in Titan's cold atmosphere, starting with ethane C_2H_6 , ethylene C_2H_4 , acetylene C_2H_2 , propane C_3H_8 ... up to benzene C_6H_6 , play not only some role in aerosol production, but also in cloud processes, rain generation and Titan's lakes formation.

Our goal is to study in the laboratory the kinetics of the first steps of condensation of these hydrocarbons. Several studies have investigated the phase of e.g. ethane and propane and their spectral signatures. At

the exception of our studies on the dimerization of benzene (C_6H_6) [3], pyrene ($C_{16}H_{10}$) [4] and anthracene ($C_{14}H_{10}$) [5] performed over the 15-300 K temperature range, there is no other work on the first elementary steps of the kinetics of nucleation of hydrocarbons. Rate coefficients however, are very sensitive to the description of the potential interaction surfaces of the molecules involved. Combined theoretical and experimental studies at the molecular level of the homogenous nucleation of various small molecules should improve greatly our fundamental understanding. This knowledge will serve as a model for studying more complex nucleation processes actually taking places in planetary atmospheres.

Here we present the first experimental kinetic study of the dimerization of two small hydrocarbons: ethane (C_2H_6) and propane (C_3H_8). We have performed experiments to identify the temperature and partial densities ranges over which small hydrocarbon clusters form in saturated uniform supersonic flows. Using our unique reactor based on Laval nozzle expansions [6], the kinetics of their formation has also been investigated down to 23 K. The chemical species present in the reactor are probed by a time of flight mass spectrometer equipped with an electron gun for soft ionization of the neutral reagents and products.

This work aims at putting some constraints on the role of small hydrocarbon condensation in the formation of haze particles in the dense atmosphere of Titan.

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