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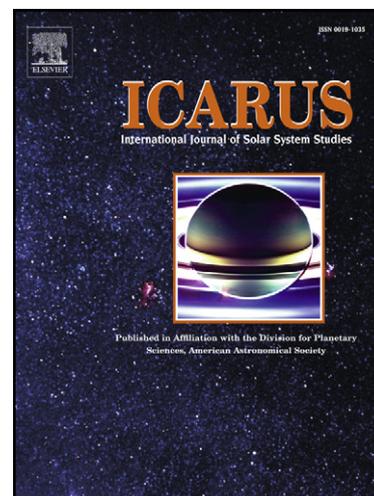
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# Far-infrared opacity sources in Titan's troposphere reconsidered

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**ABSTRACT**

We use Cassini far-infrared limb and nadir spectra, together with recent Huygens results, to shed new light on the controversial far-infrared opacity sources in Titan's troposphere. Although a global cloud of large CH<sub>4</sub> ice particles around an altitude of 30 km, together with an increase in tropospheric haze opacity with respect to the stratosphere, can fit nadir and limb spectra well, this cloud does not seem consistent with shortwave measurements of Titan. Instead, the N<sub>2</sub>-CH<sub>4</sub> collision-induced absorption coefficients are probably underestimated by at least 50% for low temperatures.

*Keywords:* Titan ; Atmospheres, composition ; Infrared observations

## 1 Introduction

Titan's troposphere has an active methane cycle that resembles the Earth's hydrological cycle. Unfortunately, there are only a few small regions of the electromagnetic spectrum that can penetrate Titan's atmosphere as deep as the troposphere, making remote sensing of the methane cycle somewhat troublesome. The wavelengths that probe Titan's troposphere include a few narrow windows in the methane absorption in the visible and near-infrared, and the far-infrared, where there are only very few gas absorption lines (Tomasko et al., 2008a). Interpretation of the latter wavelength region ( $\sim 200\text{-}600\text{ cm}^{-1}$ ) has proven difficult due to uncertainties in tropospheric opacity sources. Early analysis of Voyager Infrared Interferometer Spectrometer and Radiometer (IRIS) spectra (Thompson and Sagan, 1984; Toon et al., 1988; McKay et al., 1989) invoked methane clouds to account for (part of) the additional opacity needed to fit the data. When more reliable collision-induced absorption (CIA) coefficients became available this additional opacity source was concluded to result from an increase of  $\text{N}_2\text{-CH}_4$  CIA, caused by supersaturated methane concentrations (Courtin et al., 1995; Samuelson et al., 1997). Furthermore, it was shown by Samuelson et al. (1997) that large ethane particles near the tropopause could also be used to fit the far-infrared spectra, although methane supersaturation was still needed to improve the fit in this case. Courtin et al. (1995) also found a marginal agreement when clouds near the tropopause are included, but they obtained a better agreement with the data by including supersaturation. However, *in situ* measurements by the Huygens probe in early 2005 at  $10^\circ\text{S}$  showed tropospheric  $\text{CH}_4$  not to be supersaturated at all (Niemann et al., 2005). Since the arrival of Cassini, far-infrared spectra that probe the troposphere have not been analysed in detail again. Hence, the source of the far-infrared tropospheric

opacity source remains unknown. Here, we will re-assess the issue of Titan's far-infrared opacity sources, using data from instruments onboard the Cassini mission and strong constraints placed by the Huygens Gas Chromatograph Mass Spectrometer (GCMS) and Atmospheric Structure Instrument (HASI) measurements. This enables us to place new constraints on possible tropospheric opacity sources.

## 2 Observations

We use far-infrared spectra between  $250\text{-}550\text{ cm}^{-1}$  from the Cassini Composite Infrared Spectrometer (CIRS) (Flasar et al., 2004), with a spectral resolution of  $13\text{ cm}^{-1}$ . Below  $250\text{ cm}^{-1}$  the spectra probe the tropopause region and more variables come into play (e.g. various haze and cloud signatures (de Kok et al., 2007), trace gas emission lines, and potential errors in  $\text{N}_2\text{-N}_2$  CIA). Hence, the wavenumber region below  $250\text{ cm}^{-1}$  does not add significant constraints on this issue is not considered. The latitude of these observations ( $15^\circ\text{S}$ ) is close to the latitude of the Huygens landing site, since most variables are constrained by Huygens observations there. We use both a nadir spectrum and six limb spectra with central tangent heights lower than 50 km. The six limb spectra are obtained by fitting splines through radiance versus altitude points at each measured wavenumber of the 119 individual spectra between 0-170 km taken during this limb observation (see Teanby, 2007; de Kok et al., 2007). The field-of-view (FOV) of the limb measurements is 53 km, which is taken into account by calculating spectra at 5 tangent altitudes within the FOV and weighing them according to the spatial response of the CIRS detector (Teanby and Irwin, 2007). The nadir spectrum is an average of 36 spectra taken within several minutes spanning  $5^\circ$  of latitude. Data were taken on 22 July 2006 (nadir) and 7 September 2006 (limb), which are the T16 and T17 Titan flybys. No

significant temporal or spatial variations at this latitude are expected in this period and hence all these observations can be fitted simultaneously.

### 3 Model

We use the NEMESIS radiative transfer code for spectral calculations and retrievals (Irwin et al., 2008). This code uses the correlated- $k$  method (Lacis and Oinas, 1991) to calculate spectra and it includes an optimal estimation retrieval algorithm (Rodgers, 2000). When CH<sub>4</sub> clouds are considered, multiple scattering is used, since large CH<sub>4</sub> particles are very effective scatterers in this wavelength region. For the nadir spectrum, a doubling-adding routine is used in a plane-parallel atmosphere (e.g. Plass et al., 1973). For the limb spectra, this doubling-adding routine is used to calculate the internal radiation field, which describes the upward and downward going radiances between each layer at a number of emission angles. The equation of radiative transfer that includes both thermal emission and scattering (Hanel et al., 2003) is then numerically solved in a spherical geometry, using the following scattering source function, which is the integral over solid angle  $\Omega$  of the phase function  $P(\theta)$ , with phase angle  $\theta$ , multiplied by the internal radiation field  $I'$ , scaled by the single-scattering albedo of the atmospheric layer  $\tilde{\omega}$ :

$$J_s = \tilde{\omega} \frac{1}{4\pi} \int_{4\pi} I' P(\theta) d\Omega \quad (1)$$

Hence, our limb scattering model is a pseudo-spherical approximation, since the internal radiation field is approximated in a plane-parallel atmosphere. The continuum opacity sources are CIA of pairs of N<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>, calculated according to Borysow and Frommhold (1986a,b,c, 1987), Borysow (1991) and Borysow and Tang (1993). Pressures, temperatures and CH<sub>4</sub> volume mixing ratio (VMR)

are taken from Huygens GCMS and HASI measurements (Niemann et al., 2005; Fulchignoni et al., 2005). A  $\text{H}_2$  VMR of 0.1% is assumed (Courtin et al., 2008). Also, photochemical haze and potentially clouds contribute to the far-infrared opacity. Haze is assumed to be non-scattering, using the extinction cross-sections of de Kok et al. (2007). Haze is expected to be highly absorbing in this wavelength region, due to high imaginary refractive indices and small particle sizes (Khare et al., 1984), and hence the non-scattering nature of the haze particles seems a reasonable assumption. The vertical profile is consistent with the southern profiles of de Kok et al. (2007) and with Tomasko et al. (2008b). Above an altitude of 80 km this profile has a scale height of 65 km, whereas below 80 km the haze scale height is identical to the atmospheric pressure scale height. Solid  $\text{CH}_4$  clouds are modelled using Mie theory and the optical constants of Martonchik and Orton (1994). We use a log-normal distribution of particle sizes, with an effective radius of  $100 \mu\text{m}$  and a  $\log(\sigma)$  value of 0.3 (after Barth and Toon, 2006). This distribution includes particles sizes between  $\sim 40\text{-}250 \mu\text{m}$  and results in a flat extinction cross-section between  $250\text{-}550 \text{ cm}^{-1}$  and single scattering albedos close to unity. This behaviour is also seen for similarly broad distributions around mean particle radius larger than  $50 \mu\text{m}$ , so the exact particle size is not well constrained when cloud particles become this large. Similarly, our exact choice of particle size is also of little consequence. For easy computations of the phase function, double Henyey-Greenstein functions are fitted to the Mie results.

## 4 Results

Using only the known gas and haze opacity, the calculated spectrum is shown as the thin solid line in Fig. 1. This synthetic spectrum clearly overestimates the nadir

radiances, meaning that an additional opacity source is needed in the troposphere. We also found that tropospheric opacity was needed when fitting other low latitude nadir data from other flybys. Also the Voyager IRIS data is consistent with the CIRS data, meaning that previous conclusions based on Voyager data were not the result of instrumental artefacts. As the past has shown (e.g. Courtin et al., 1995; Samuelson et al., 1997), far-infrared nadir spectra can be fitted using a range of assumptions. For instance, instead of an increased  $\text{CH}_4$  abundance, the  $\text{N}_2\text{-CH}_4$  CIA coefficient might be in error. Multiplying the coefficients by a factor of 1.5 gives excellent agreement with nadir observations at low latitudes (Tomasko et al., 2008a), which is also shown in Fig. 1 (dotted lines). This additional opacity causes one to probe higher in the troposphere, where temperatures are lower, giving rise to lower radiances. However, Fig. 1 also shows that the limb measurements are poorly reproduced with this assumption. We find that we cannot fit the limb spectra by only scaling the  $\text{N}_2\text{-CH}_4$  CIA coefficients, even if we scale different altitudes differently. This is because the higher limb spectra probe high in the troposphere, where atmospheric density (and thus CIA opacity) is low, reducing the sensitivity to CIA. The atmosphere is optically thin there and hence adding opacity will increase radiances. At present, we cannot rule out large temperature-dependent and wavelength-dependent errors on the  $\text{N}_2\text{-CH}_4$  CIA coefficients of Borysow and Tang (1993).

Alternatively, we can fit the limb measurements reasonably by increasing tropospheric haze opacity in the upper troposphere by a factor of 5 or more compared to our nominal model (see Fig. 2). However, such an increase in haze opacity does little to reduce the nadir radiances. A combination of increased CIA and increased tropospheric haze opacity can fit both limb and nadir data reasonably well (see Fig. 1, long dashed lines).

A third plausible variable is a potential global  $\text{CH}_4$  cloud. We retrieved a cloud profile by allowing the cloud density to vary between altitudes of 20 and 40 km. If only this cloud is retrieved, nadir radiances can be brought to the correct magnitudes using a thin cloud around 30 km (see Fig. 2), although the limb spectra are not fitted very well. Adjusting the haze opacity as well yields good fits to all spectra (see Fig. 1, dot-dashed lines). Again, haze opacity in the high troposphere has to be increased in this case (see Fig. 2).

## 5 Discussion

We have shown that large  $\text{CH}_4$  cloud particles at 30 km can be used to fit far-infrared nadir spectra that probe Titan's troposphere. An increase in tropospheric haze opacity with respect to our nominal model is needed to fit the limb spectra. Based on Huygens temperature (HASI) and  $\text{CH}_4$  (GCMS) measurements, Tokano et al. (2006) and Barth and Toon (2006) show that a global  $\text{CH}_4$  cloud should exist between 20-30 km, but there have been no conclusive observations of this cloud. Only Ádámkovics et al. (2007) mention they observe a global cloud, yet Penteado et al. (2010) fit Cassini Visual and Infrared Mapping Spectrometer (VIMS) data at the same wavelengths well without requiring such a cloud. There is further indirect evidence for this cloud from Huygens Surface Science Package data, which indicate atmospheric turbulence, possibly caused by clouds, between 20-30 km (Lorenz et al., 2007). On the other hand, flux and polarisation measurements from Huygens Descent Imager-Spectral Radiometer (DISR) show no indication of any cloud particles between 20-30 km (Tomasko et al., 2008b, 2009). The number density of cloud particles needed to fit the far-infrared spectra (Fig. 2) is several orders of magnitude lower than the haze number density found by Tomasko et al. (2008b). This number

can even decrease by an order of magnitude if particles with effective radii of 200  $\mu\text{m}$  are considered instead of 100  $\mu\text{m}$ . However, the extinction at near-infrared and visible wavelengths of the cloud, regardless of the particle size, is far larger than the haze extinction (0.23 versus 0.025  $\text{km}^{-1}$  at 1  $\mu\text{m}$ ) and hence they should have been visible in the DISR measurements if such a cloud were present. Also if the cloud is vertically more extended than the retrieved profile, the extinction is at least as large as the haze extinction at 1  $\mu\text{m}$ . A cloud with an optical depth of  $\sim 0.1$  should also be visible in VIMS data. Contamination of the clouds by haze particles can remedy this problem somewhat, since the haze particles are expected to be strongly absorbing in the far-infrared and less so at shorter wavelengths (Khare et al., 1984). Because solid  $\text{CH}_4$  is non-absorbing in the far-infrared, a small contamination of haze can increase the absorbance of the cloud particles by several orders of magnitude. Although less particles of the same size are needed to fit the far-infrared data in this case, a haze contaminant will only reduce the cloud extinction by several tens of percent at most. The potential non-sphericity of the ice particles is also not expected to make a large difference for the required cloud extinction (e.g. Mishchenko et al., 1996). Thus, despite obtaining good fits to the CIRS data with a physically plausible cloud, a global cloud does not seem to be the main additional opacity source in Titan's troposphere needed to fit the far-infrared nadir spectra, since it does not agree with shorter wavelength observations by DISR and VIMS. However, the data discussed here cannot rule out a more optically thin methane cloud.

This implies that the low temperature  $\text{N}_2\text{-CH}_4$  CIA coefficients of Borysow and Tang (1993) probably underestimate the absorption by at least 50%, as indicated by our model fits. In addition, tropospheric haze opacity should show an increase with respect to that at higher altitudes. This latter result does not seem unreasonable,

since DISR shows a decrease in the steepness of the opacity versus wavelength below 80 km, and again below 30 km, indicative of larger haze particles (Tomasko et al., 2008b). These larger particles will then also produce higher opacities at far-infrared wavelengths, and relatively more so than at short wavelengths.

In order to give more insight into Titan's tropospheric opacity sources, new low-temperature laboratory measurements of  $N_2$ - $CH_4$  are needed down to temperatures of 70 K. This issue can also be further constrained by repeating this study at other latitudes, especially where temperatures or  $CH_4$  concentrations are different from the Huygens landing site. Radio occultation measurements will give some of the required constraints at these latitudes, but they do not simultaneously constrain both  $CH_4$  and temperature. The best constraints on the missing far-infrared opacity source would be obtained by dedicated *in situ* measurements.

### Acknowledgements

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**Figure Captions**

Figure 1: Fits (thin lines) to three of the measured spectra (thick solid lines) for different model cases (thin solid line – nominal case based on Huygens results; dotted line – scaled N<sub>2</sub>-CH<sub>4</sub> CIA; short dashed line – retrieved CH<sub>4</sub> cloud; dot-dashed line – retrieved CH<sub>4</sub> cloud and haze profiles; long dashed line – retrieved haze profile and scaled N<sub>2</sub>-CH<sub>4</sub> CIA). For the limb observations the tangent height at the centre of the detector is shown. Error bars are shown as shaded areas.

Figure 2: Particle density (left) and extinction at 1 μm (right) of the CH<sub>4</sub> ice cloud needed to fit the far-infrared spectra. Also plotted are the initial (dotted line) and retrieved haze extinction profiles at 500 cm<sup>-1</sup> for the cases of retrieved CH<sub>4</sub> cloud and haze profiles (dashed line) and retrieved haze profile and scaled N<sub>2</sub>-CH<sub>4</sub> CIA (dot-dashed line).

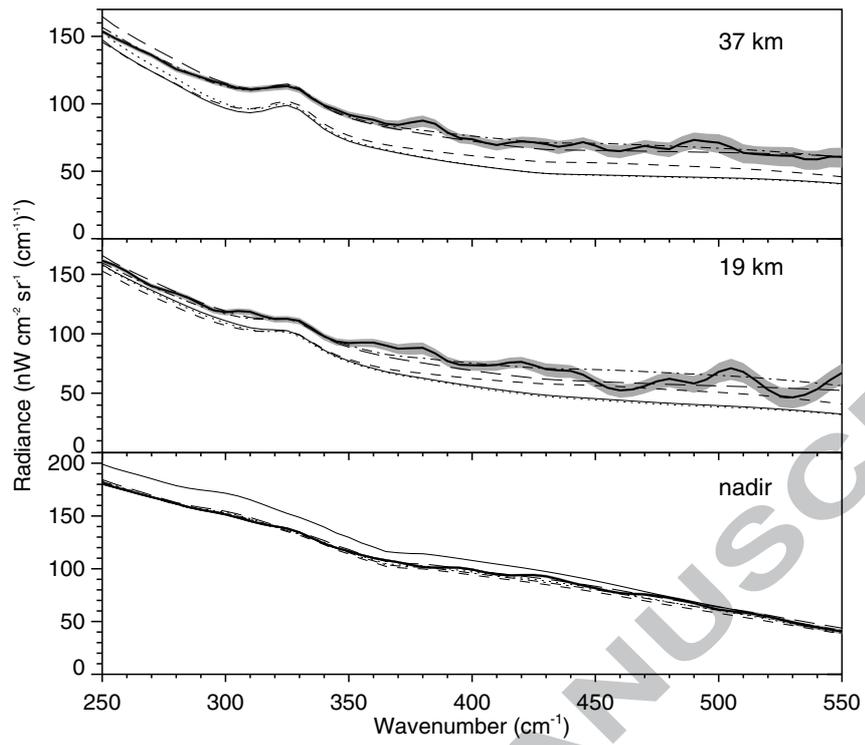


Fig. 1. de Kok et al.; Titan's tropospheric opacity

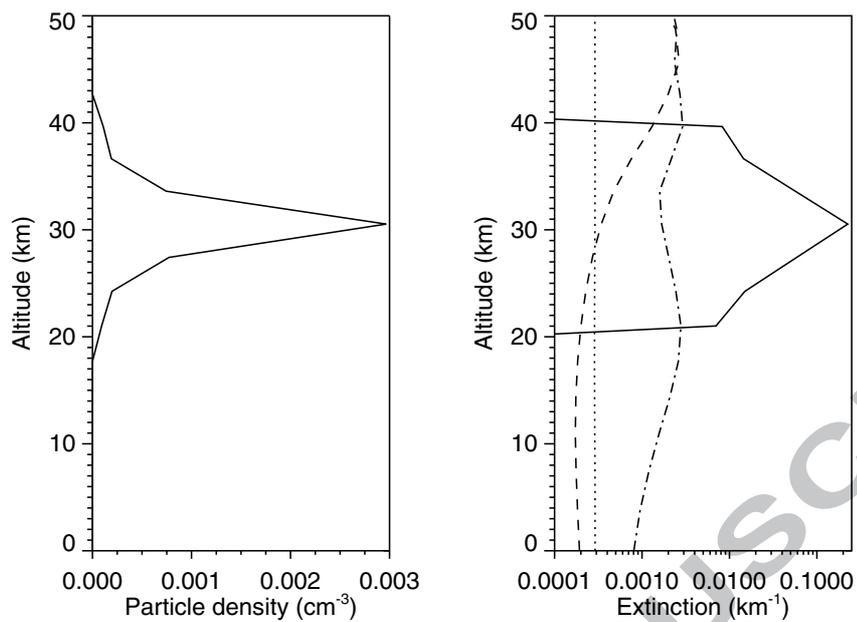


Fig. 2. de Kok et al.; Titan's tropospheric opacity