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H_2 abundance and ortho-to-para ratio in Titan's troposphere

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Abstract

We have analyzed spectra recorded between 50 and 650 cm^{-1} by the Composite Infrared Spectrometer (CIRS) aboard the Cassini spacecraft at low and high emission angles to determine simultaneously the H_2 mole fraction and ortho-to-para (o-p) ratio in Titan's troposphere. We confirm that the N_2-CH_4 collision-induced absorption (CIA) coefficients used up to now need to be strongly increased at temperatures of 70-85 K. We find that the N_2-N_2 CIA coefficients are also too low in the N_2 band far wing, beyond 110 cm^{-1}. We derived a tropospheric H_2 mole fraction equal to $(0.88 \pm 0.13) \times 10^{-3}$, which agrees with a previous determination based only on the H_2-N_2 dimer transition in the $S_0(0)$ line. We find that the H_2 para fraction is close to equilibrium in the 20-km region. We have investigated different mechanisms that may operate in Titan's atmosphere to equilibrate the H_2 o-p ratio and we have developed a one-dimensional model that solves the continuity equation in presence of such conversion mechanisms. We conclude that exchange with H atoms in the gas phase or magnetic interaction of H_2 in a physisorbed state on the surface of aerosols are too slow compared with atmospheric mixing to play a significant role. On the other hand, magnetic interaction of H_2 with CH_4, and to a lesser extent N_2, can operate on a timescale similar to the vertical mixing time in the troposphere. This process is thus likely responsible for the o-p equilibration of H_2 in the mid-troposphere implied by CIRS measurements.

1. Introduction

Molecular hydrogen (H_2) is formed in the upper atmosphere of Titan from the photodissociation of methane (CH_4) and other hydrocarbons. It exists in two forms: a singlet state with an anti-parallel configuration of the two proton spins (para-hydrogen) and a triplet state with the proton spins aligned parallel (ortho-hydrogen). A previous analysis of measurements with the Cassini/CIRS yielded a globally averaged H_2 mole fraction around 1×10^{-3} in Titan's upper troposphere [1], in agreement with the Huygens/GCMS determination. This abundance is hard to reconcile with the 3-4 times larger value inferred from Cassini/INMS measurements around 1000 km, assuming no surface sink of H_2 and standard eddy mixing profile [2]. The previous CIRS determination was based on N_2-H_2 dimer transitions occurring in the region of the $S_0(0)$ rotational transition of para H_2 and had to assume ortho-para (o-p) thermodynamical equilibrium [1]. The goal of this study is i) to infer simultaneously the H_2 mole fraction *and* o-p ratio using both the $S_0(0)$ (para) and $S_0(1)$ (ortho) H_2 lines, and ii) investigate the o-p equilibration in Titan's atmosphere.

2. Analysis of Cassini/CIRS observations

We used low-latitude selections of CIRS spectra between 50 and 650 cm^{-1} at 15-cm^{-1} resolution recorded at low and high emission angles (Fig. 1). We analyzed the whole spectral range with a radiative transfer model that incorporates results from the Huygens probe measurements to better constrain the different opacity sources (gases and aerosols). We found that we need to increase the N_2-CH_4 collision-induced absorption (CIA) coefficients used up to now by about 52%, in agreement with previous analyses of CIRS spectra, and also increase the N_2-N_2 CIA coefficients beyond 110 cm^{-1}, in agreement with recent quantum mechanical calculations. We derived a H_2 mole fraction equal to $(0.88 \pm 0.13) \times 10^{-3}$, which pertains to the ~1-34 km altitude range probed by the $S_0(0)$ and $S_0(1)$ lines. We found that CIRS spectra can be fitted assuming either ortho-to-para H_2 thermodynamical equilibrium at all levels or a constant para fraction in the range 0.49-0.53.

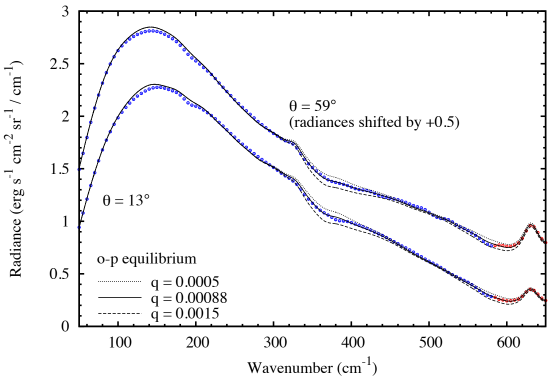

Figure 1: Low (13°) and high (59°) emission angle spectra are compared with synthetic spectra assuming o-p H_2 equilibrium and uniform H_2 mole fractions of 0.5, 0.88 and 1.5 × 10^{-3}. The high-emission spectra are shifted by 0.5 erg s^{-1} cm^{-2} sr^{-1} / cm^{-1} for clarity.

3. H_2 ortho-para conversion in Titan's atmosphere

From a careful survey of the mechanisms that can result in ortho-para (o-p) equilibration, we conclude that magnetic interaction with CH_4, and to a lesser extent N_2, is the only one that can operate in Titan's troposphere on a timescale comparable with that of dynamical mixing. H exchange and conversion through adsorption and magnetic interaction on aerosol surface are not efficient enough in regard to atmospheric mixing (Fig. 2).

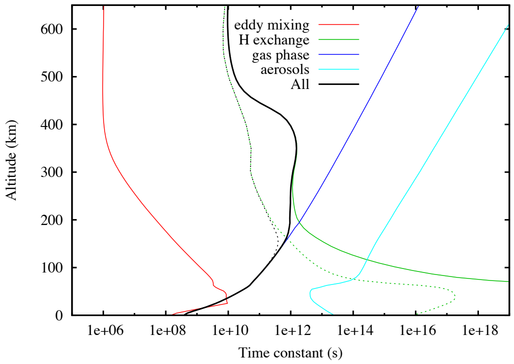

Figure 2: Ortho-para H_2 conversion times due to H exchange in the gas phase (green lines), magnetic interaction with N_2 and CH_4 in the atmosphere (blue line), magnetic interaction on aerosol surface (cyan line), and all these processes together (black lines). The dashed and solid lines correspond to different H density profiles. Also shown is the dynamical mixing time calculated from the eddy mixing profile used in our model (red line).

We have developed a 1-D model for the H_2 para fraction that solves the continuity equation and incorporates the conversion mechanisms in terms of time constants. Our nominal model produces a para fraction profile that is about 10% smaller than required to fit the observations. To reproduce the inferred o-p ratio in the 20-km region, low atmospheric mixing in the troposphere down to 15-20 km and conversion rates with CH_4 or N_2 slightly larger than extrapolated from o-p conversion rate measured in natural H_2 are required.

4. Conclusions

Our study confirms that the H_2 mole fraction in the troposphere is 3 to 4 times smaller than inferred from in situ measurements by Cassini/INMS. If those are correct, this discrepancy points to some unidentified physical or chemical mechanism at work to deplete H_2 in the lower atmosphere, or a peculiar choice of the eddy mixing profile [3]. The H_2 o-p ratio is close to equilibrium in the 20-km region. Equilibration results from magnetic interaction of H_2 with CH_4 and N_2 in the gas phase.

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