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LETTERS

Methane drizzle on Titan

Tetsuya Tokano[1], Christopher P. McKay[2], Fritz M. Neubauer[1], Sushil K. Atreya[3], Francesca Ferri[4], Marcello Fulchignoni[5,6] & Hasso B. Niemann[7]

Saturn's moon Titan shows landscapes with fluvial features[1] suggestive of hydrology based on liquid methane. Recent efforts in understanding Titan's methane hydrological cycle have focused on occasional cloud outbursts near the south pole[2–4] or cloud streaks at southern mid-latitudes[5,6] and the mechanisms of their formation. It is not known, however, if the clouds produce rain or if there are also non-convective clouds, as predicted by several models[7–11]. Here we show that the *in situ* data on the methane concentration and temperature profile in Titan's troposphere point to the presence of layered optically thin stratiform clouds. The data indicate an upper methane ice cloud and a lower, barely visible, liquid methane-nitrogen cloud, with a gap in between. The lower, liquid, cloud produces drizzle that reaches the surface. These non-convective methane clouds are quasi-permanent features supported by the global atmospheric circulation, indicating that methane precipitation occurs wherever there is slow upward motion. This drizzle is a persistent component of Titan's methane hydrological cycle and, by wetting the surface on a global scale, plays an active role in the surface geology of Titan.

The descent of the Huygens probe into Titan's troposphere on 14 January 2005 provided a direct means of determining the condensation state of methane in regions where no clouds have been recognized before. The methane mixing ratio measured by the Huygens Gas Chromatograph Mass Spectrometer (GCMS) is characterized by an essentially uniform methane mixing ratio of $(4.92 \pm 0.25) \times 10^{-2}$ between the surface and 6–8 km altitude, and a monotonic decrease with altitude above this level down to 1.62×10^{-2} at the tropopause[12,13]. Using the pressure profile measured by the Huygens Atmospheric Structure Instrument (HASI)[14], this yields a column methane abundance of $2{,}040 \pm 100\,\mathrm{kg\,m^{-2}}$, equivalent to 2.86 km amagat or ~5 m of liquid methane, which is intermediate between previous values mostly ranging from ~2 km amagat to ~4 km amagat (refs 15, 16), ruling out both excessive supersaturation in the mid-troposphere and a largely subsaturated atmosphere.

We inferred the detailed structure of the implied condensation layer by analysing the relative humidity (RH) of methane (Fig. 1), calculated from the measured methane mixing ratio, temperature and pressure. In the Earth's atmosphere the RH of water rarely exceeds 100%, so regions with an RH of 100% are almost always accompanied by the presence of clouds, fog and/or precipitation. Pure CH_4 freezes at 90.6 K (~2.5 km altitude at Titan), but dissolved nitrogen depresses the freezing point by 10–15 K under Titan conditions—that is, liquid CH_4-N_2 can stably exist up to ~15 km altitude (refs 17, 18). When this effect is taken into consideration, the calculated RH increases almost linearly from ~45% at the surface to 90% at 6 km altitude and then approaches 100% (curve 2 in Fig. 1).

Thus the atmosphere is nearly saturated, implying a cloud above ~8 km altitude that consists of liquid CH_4-N_2 extending at least to the freezing level. The behaviour of this binary CH_4-N_2 mixture below the freezing point is not well determined from experimental data—that is, it is not clear a priori whether the binary mixture can exist as a supercooled liquid, as is often the case in terrestrial clouds at

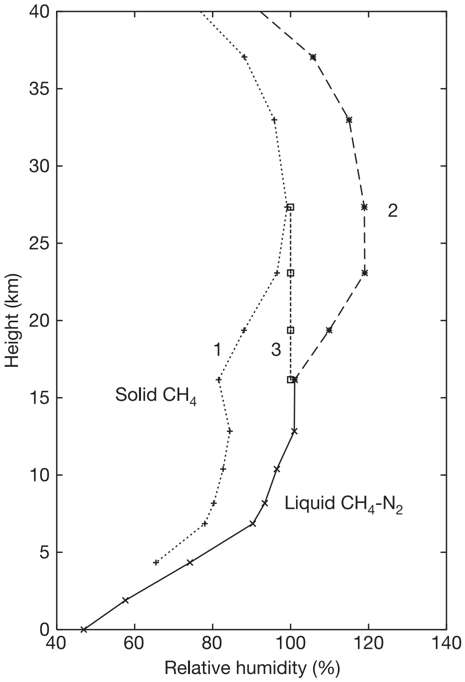

Figure 1 | Vertical profile of the methane relative humidity at the Huygens entry site under different assumptions. The relative humidity (RH) is calculated from the measured methane mixing ratio[12,13], temperature[14] and pressure[14]. Dashed curve 1 shows the RH with the saturation vapour pressure over pure solid CH_4 (that is, without dissolved N_2). This curve is shown only down to the freezing point of CH_4. The solid curve shows the RH with the saturation vapour pressure over a liquid binary CH_4-N_2 mixture[18], and is shown only up to the freezing level. Dashed curve 2 is the RH with the saturation vapour pressure over a supercooled liquid binary CH_4-N_2 mixture. Dashed curve 3 is a hypothetical RH profile under the assumption that a phase change from liquid to solid and a compositional change from CH_4-N_2 to CH_4 take place. A constant RH of ~100% is possible, even if there is a gap in the cloud. Near the cloud edge turbulent mixing can cause entrainment of drier air from outside the clouds[28], so clouds can sometimes exist in subsaturated regions. Therefore, it is not possible to determine the exact altitude of cloud top and bottom solely from the RH profile.

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[1]Institut für Geophysik und Meteorologie, Universität zu Köln, Albertus-Magnus-Platz, 50923 Köln, Germany. [2]NASA Ames Research Center, MS 245-30, Moffett Field, California 94035-1000, USA. [3]Department of Atmospheric, Oceanic, and Space Sciences, University of Michigan, 2455 Hayward Street, Ann Arbor, Michigan 48109-2143, USA. [4]CISAS "G. Colombo", Università di Padova, Via Venezia 15, 35131 Padova, Italy. [5]LESIA, Observatoire de Paris, 5 Place Jules Janssen, 92195 Meudon, France. [6]Université Denis Diderot–Paris 7, UFR de Physique, 2 Place Jussieu, 75006 Paris, France. [7]NASA Goddard Space Flight Center, Code 915, Greenbelt, Maryland 20742, USA.

temperatures below 0 °C. But as upon freezing a substantial fraction of nitrogen should be exsolved from the CH_4 condensate, the saturation vapour pressure would lie somewhere between that over solid pure CH_4 (curve 1 in Fig. 1) and that over liquid CH_4-N_2 (curve 2). Assuming that the CH_4-N_2 condensate freezes near 15 km, we can construct a hypothetical RH profile (curve 3) beginning at 16 km with 100%, and staying constant at this value until it merges with the RH profile for solid CH_4 near 21 km, implying that the N_2 content of the condensate decreases with decreasing temperature. In this case, the RH is virtually uniform from 8 to 30 km, representing one single extensive condensation layer with either an abrupt or successive change in the chemical composition from CH_4-N_2 to CH_4 associated with a phase change above 15 km.

A Titan cloud model that assumes liquid CH_4-N_2 and its freezing[11] predicts nearly uniform RH (~100%) extending over several tens of kilometres across the freezing level in the presence of clouds. The resulting upper ice cloud and lower liquid cloud are separated from each other by a narrow gap, although this gap does not manifest itself in a local minimum of RH or methane mixing ratio. The lower liquid cloud is mainly a result of melting of the upper ice cloud rather than a product of *in situ* condensation into the liquid phase. With our assumption of compositional and phase change, the upper cloud ice consists of nearly pure solid CH_4, while the lower cloud below 16 km consists of a liquid CH_4-N_2 mixture with a nitrogen concentration of ~20%. The sudden increase in the CH_4 count rate measured by the GCMS near 16 km (refs 12, 13) seems to support this liquid–solid transition; the entry of liquid droplets into the heated inlet of the GCMS can better explain the CH_4 count rate jump than the entry of solid particles. This scenario is consistent with the detection of an optically thin methane haze near 21 km by the Huygens DISR (Descent Imager Spectral Radiometer)[1]. The apparent disappearance of the observed methane haze below 20 km indicates a gap in the condensation layer as predicted[11], while the top of the methane haze near 21 km suggests a drop in the cloud mass because of decreasing air density with height. However, a subvisible cloud should extend up to the altitude at which the RH drops below 100%, that is, ~30 km. The presence of a cloud gap below 20 km can be understood as an immediate consequence of the phase change: this transitional region is too cold to sustain liquid cloud, but slightly too dry for pure CH_4 to condense.

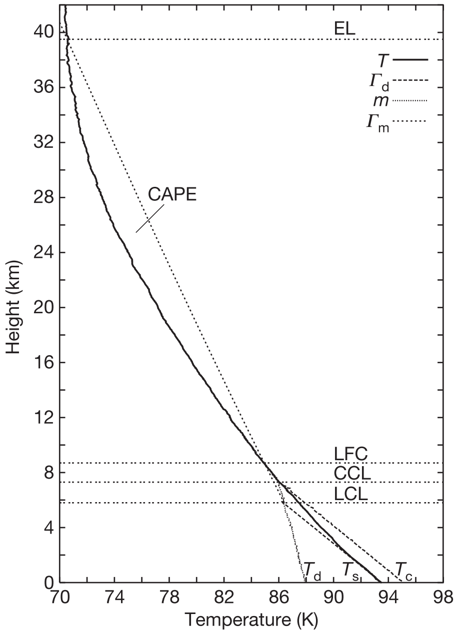

Figure 2 | Thermodynamic diagram at the Huygens entry site. T_s ($= 93.6$ K) is the surface temperature, T_d (~88 K) is the surface dew point, T_c (~95 K) is the convective temperature (the threshold temperature the surface temperature would have to exceed to enable unforced moist convection for the given methane abundance), LCL (~6 km) is the lifting condensation level, CCL (~9 km) is the convective condensation level, LFC (~7 km) is the level of free convection, EL (~40 km) is the equilibrium level, Γ_d is the dry adiabat, Γ_m is the moist adiabat, T is the measured temperature profile and along the line m the saturation mixing ratio is constant. CAPE (convective available potential energy) is the buoyant energy that is available for rising air parcels that can be converted to kinetic energy of moist convection in the case of successful triggering, and corresponds to the area bounded by the temperature curve and the moist adiabat between LFC and EL. In calculating the dry adiabatic lapse rate, the real gas equation (virial equation)[29] is used, considering the atmospheric composition measured by the GCMS[12] and the vertical variation of it; the moist adiabatic lapse rate is calculated with the Redlich-Kwong equation of state[30], which includes the effect of the binary CH_4-N_2 mixture. CAPE amounts to ~960 J kg^{-1}, corresponding to weakly unstable convective potential by terrestrial standards, so the chance of severe storms is rather low at the time and place of Huygens landing. The equilibrium level (EL), where ascending air experiences negative buoyancy, is located near 40 km (close to the mean cloud top height of mid-southern-latitude clouds recently observed[5]), indicating that convective clouds, if they develop at all, could indeed extend up to ~40 km. Both clouds are located in a stably stratified environment, so they do not represent inversion clouds of stratocumulus type, but may be classified as stratus-type clouds.

If we assume that liquid CH_4-N_2 does not freeze but exists as supercooled liquid, the corresponding RH increases above 16 km and attains a maximum of 120% between 23 km and 27 km; that is, there would be substantial supersaturation, as was suggested after the Voyager mission[19,20]. However, the presence of a vertical gradient in the methane mixing ratio in this altitude region[13] points to coexistence with liquid condensates. There are two difficulties in supporting this RH profile. First, the detection of a thin methane haze near 21 km (ref. 1) conflicts with the assumption of supercooled liquids in this altitude region. Second, the increase of RH in two separate altitude regions is not predicted by any methane condensation model[8–11] under any assumption. Furthermore, the presence of supercooled

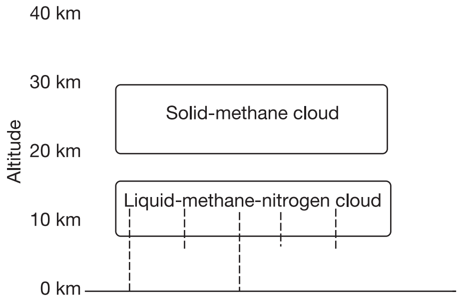

Figure 3 | Diagram of the vertical cloud structure at the Huygens entry site. The upper cloud consists of solid particles, mainly CH_4, and most resembles a terrestrial cirrostratus. The lower cloud consists of a liquid binary CH_4-N_2 mixture and resembles a terrestrial stratus. Either cloud may or may not contain small amounts of C_2H_6 ice as condensation nuclei. However, considering the low downward flux of C_2H_6 ice[13] and the capability of bare tholins to serve as condensation nuclei for methane[11], the role of C_2H_6 may not be as significant as thought before. The upper and lower cloud are separated from each other by a gap because no supercooled droplets exist. CH_4 ice particles falling from the upper cloud passing this gap supply the lower cloud upon melting. Dashed lines indicate rainfall that partly arrives at the surface. Falling drizzle gradually becomes poorer in nitrogen by preferential evaporation of dissolved nitrogen. Except for latitudinal variation in cloud heights, this basic structure may be representative of approximately half of Titan that experiences slow large-scale upwelling. In a colder environment (for example, the winter pole), the lower liquid cloud may extend to higher altitudes, as the freezing level increases[17]. If the global circulation pattern changes with season[9,10], it will induce a monsoon with a wet season during upwelling and a dry season during downwelling.

liquid CH_4-N_2 means that neither a phase change nor a compositional change of the cloud would take place, but the kink at the bottom of curve 2 (Fig. 1) near 16 km is suggestive of an abrupt change, contradicting our assumption of no transition.

Similarly, the presence of only pure solid CH_4 is internally inconsistent. The calculated RH is roughly 20% smaller and reaches a plateau (7–16 km) of only ~80%, but increases again and eventually reaches saturation (100%) between 25 and 30 km. We might envision a recent occurrence of a large, non-steady condensation event extending from 8 km to 16 km that temporarily suppressed the RH to 80%, but such an event requires or induces a temporal anomaly of several kelvin in the temperature profile that is not observed (Fig. 2). Moreover, the Huygens DISR[1] did not find an optically thick methane cloud around 10 km that would point to such an event. Therefore, we cannot find a physically meaningful mechanism to keep the RH at ~80% for some kilometres below 16 km and to allow an increase of RH to 100%. The only internally consistent scenario is the separate presence of an upper CH_4 ice cloud between about 20 km and 30 km and a lower liquid CH_4-N_2 cloud between ~8 km and 16 km (Fig. 3). Optical detection of the lower liquid cloud by Huygens has not been reported so far, indicating that this cloud may be subvisible. Modelling[11] suggests that subvisible clouds are established by the cloud mass balance if the upward methane flux is slow ($10^{-7}\,\mathrm{g\,cm^{-2}\,s^{-1}}$, corresponding to an eddy diffusivity of $5{,}000\,\mathrm{cm^2\,s^{-1}}$) and the droplets are large (0.01–2 mm, "rain without clouds"[7]), and is an indication that this cloud is a steady-state feature, in contrast to many clouds observed elsewhere[2–6]. The size of cloud droplets varies between less than 0.1 mm (ref. 10) and between 0.1 mm and 1 mm with more particles on the lower side[11], so they may be best characterized as drizzle.

The thermodynamic diagram (Fig. 2) reveals that the temperature at the surface was 1–2 K too low to enable free moist convection at the convective condensation level. Forced moist convection at the lifting condensation level near 6 km cannot be definitely ruled out, but we are unable to find a suitable lifting mechanism: orographic forcing is unlikely near the Huygens site, given the flat topography[1] and the weak wind near the surface[1,21]. Lifting of air along a cold front could not occur owing to the lack of baroclinic instability on Titan[9]; also, the vertical profile of temperature and dew point does not show any anomaly that would point to substantial horizontal advection of heat or moisture. Cryovolcanic activity[22] could trigger mid-latitude clouds[6], but no such features are known in the vicinity of the Huygens site. There is also no evidence of strong updrafts typical of thunderstorms[23].

The driver of this type of condensation is the slow upward transport of methane by large-scale atmospheric circulation[9,10]. The presence of upward wind in the troposphere at the Huygens site was directly confirmed by HASI[23], and is also predicted by general circulation models (GCMs)[9,10] for this season.

We note that the vertical temperature profiles at two separate near-equatorial regions retrieved by Voyager 1 were almost identical[24], the equator-to-pole temperature contrast was merely 3 K (ref. 20) and the methane abundance determined by remote sensing[20] and predicted by GCMs[9,10] is almost uniform within at least ±30° latitude; together, these observations imply that the single measurement by Huygens may be representative of at least 60% of Titan's globe. GCMs[9,10] predict the widespread presence of such thin stratiform clouds anywhere that slow mean upward motion is found—that is, over about half of Titan's globe at any instant, even if the methane abundance decreases with latitude. Therefore, the clouds encountered by Huygens possibly cover half of Titan.

Previous model predictions[8–11] can be used to estimate the precipitation rate at the landing site from the measured methane humidity profile. The most important constraint on the precipitation is the uniform methane mixing ratio below 6 km (refs 12, 13), which we interpret as evidence of slow evaporation of falling rain, as evaporation would produce a vertical gradient in the methane mixing ratio[8,9,11]. Hence the rain from the liquid cloud must eventually pass through the lower troposphere and reach the surface to achieve mass balance, because otherwise the cloud would grow sufficiently to be detected optically. We estimate the surface precipitation rate at the Huygens landing site as $\sim 50\,\mathrm{mm\,yr^{-1}}$ (see Supplementary Information).

On Earth, regions with this annual precipitation rate are considered deserts, but the rainfall on Titan is inferred to be more persistent than in terrestrial deserts, given the stratiform character of the clouds. With the measured column methane abundance and this precipitation rate, the atmospheric residence time is estimated as 80 years, which is 5–6 orders of magnitude shorter than the photochemical lifetime of Titan's atmospheric methane. Non-zero surface precipitation means that the hydrological cycle of atmospheric methane regularly involves the surface, contrary to some previous expectations. Although the erosive potential may be quite limited with such a low precipitation rate[25], it would be at least sufficient to wet the surface material, and may explain the generally wet character of the surface material[26]. Atmospheric methane can probably be supplied by (cryovolcanic) outgassing[13,22,27], but the atmospheric circulation is required to globally distribute methane and precipitation[9,10] and thus produce global wetting of the surface. Large-scale stratiform precipitation—drizzle—may constitute a more persistent component of Titan's whole methane cycle[13] than the optically thick but sporadic clouds.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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