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► **To cite this version:**

Pascal Rannou, Maélie Coutelier, Emmanuel D. Rivière, Sébastien Lebonnois, Michael M. Rey, et al..  
Convection behind the humidification of Titan's stratosphere. *The Astrophysical Journal*, 2021, 922  
(2), 10.3847/1538-4357/ac2904 . hal-03439334

**HAL Id: hal-03439334**

**<https://hal.science/hal-03439334>**

Submitted on 22 Nov 2021

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# Convection behind the humidification of Titan's stratosphere

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1 Abstract : On Titan, methane is responsible for the complex prebiotic chemistry, the  
2 global haze, most of the cloud cover and for the rainfalls which model the landscape.  
3 Its sources are located in liquid reservoirs at and below the surface and its sink is  
4 the photodissociation at high altitude. Titan's present and past climates strongly  
5 depend on the connection between the surface sources and the atmosphere upper  
6 layers. Despite its importance, very **little** information is available on this topic. In  
7 this work, we re-analyze two solar occultations made by Cassini before the northern  
8 spring equinox. We find a layer rich in methane at 165 km and at 70°S (mixing ratio  
9  $1.62 \pm 0.1\%$ ) and a dryer background stratosphere (1.1 – 1.2%). In absence of local  
10 production, this reveals an intrusion of methane transported into the stratosphere by  
11 convective circulation. On the other hand, methane transport through the tropopause  
12 at global scale appears quite inhibited. Leaking through the tropopause is an important  
13 bottleneck of Titan's methane cycle at all timescales. As such, it affects the long  
14 term evolution of Titan atmosphere and the exchange fluxes with the surface and  
15 subsurface reservoirs in a complex way. Global climate models (GCMs) accounting for  
16 cloud physics, thermodynamical feedbacks and convection are needed to understand  
17 the methane cycle, and specifically the humidification of the stratosphere, at the present  
18 time and its evolution under changing conditions at geological timescale.

19 **1. Introduction :** **There are details about Titan's methane cycle which remain unknown.** Methane  
20 main reservoirs are in lakes, seas and possibly in the subsurface (Mastrogiuseppe et al. (2014)).  
21 Efforts **have been** made to understand **the main characteristics** of the methane tropospheric cycle  
22 (Lora et al. (2015); Faulk et al. (2020)), **that is the global fluxes in the troposphere and the surface**  
23 **and subsurface sources and sinks.** Radio-occultation measurements were used along with arguments  
24 involving circulation, observed cloud cover and wind measured by Huygens to state that, before the  
25 Northern Spring Equinox in 2009, the troposphere was more humid in the southern hemisphere than  
26 in the northern hemisphere (Tokano (2014)). This is a valuable, although quite loose, constraint.  
27 Using photometric observations collected in 2014, Lora and *Ádámkóvics* (2017) found with certainty  
28 a very humid north polar region and, **consistent** with Tokano (2014) but not in a significant way,  
29 a methane humidity increasing from the north (30-60°N) to the southern hemisphere (30°S to  
30 equator). However clear conclusions at global scale could not be drawn due to uncertainties in

31 methane retrievals. Retrieving methane abundance is extremely difficult because optical properties  
32 of the haze and the tropospheric mist are not well defined. Observations of scattered light at  
33 the limb or in nadir viewing do not allow **the disentanglement** of methane abundance and particles  
34 without ambiguity. Thermal infrared emission observed with the Composite InfraRed Spectrometer  
35 (CIRS/Cassini) allows **the retrieval of** the methane mixing ratio, but with a low spatial resolution  
36 and only in the stratosphere. This is enough to describe latitudinal variations but not vertical  
37 profiles (Lellouch et al. (2014)). In the low and middle stratosphere, they **generally found methane**  
38 **abundances** lower than expected from Huygens (Niemann et al. (2010)) with strong variations and  
39 high values above the tropics and the polar regions, possibly connected to the circulation. With the  
40 Ultraviolet Spectrometer UVIS/Cassini, Yelle et al. (2021) also **found** a low methane mixing ratio,  
41 around 1% at 950 km, around the homopause.

42 We re-analyzed two occultations made by the Visual Infrared Mapping Spectrometer (VIMS/Cassini)  
43 during the flybys *T10* in January 2006 and *T53* in April 2009, previously published and analyzed  
44 (Maltagliati et al. (2015)). These observations are performed before the equinoctial turnover in  
45 2009, while the large scale circulation has remained steady for a Titan’s season and a half ( $\simeq 11$   
46 terrestrial years) (Rannou et al. (2004); Lebonnois et al. (2012)). We used the methane linelist  
47 published by Rey et al. (2018) which significantly improves the quality of the retrievals. Details  
48 about the model, the results and comparisons with the previous analysis are discussed in the Sup-  
49plementary Information. Occultations probe extinction along tangential lines of sight at the limb  
50 and they easily allow to separate methane and haze extinction. In the following, we discuss our  
51 retrieval for the methane mixing ratio.

52 **2. Description of the method :** To proceed to the analysis, we calculate the haze and the gas  
53 properties in Titan’s atmosphere following Rannou et al. (2018) and Coutelier et al. (2021). Since  
54 we deal with occultation data, only extinction properties are needed. Then, we built a simple model  
55 for computing tangential transmissions through an atmosphere, as observed with the occultation  
56 mode. The vertical grid of the model is adjusted to the number of data  $N_D$ . We then define as  
57 much as layers than data points. We set the haze properties and the methane abundance at the  
58 lower boundary (or impact factor)  $z_i$  of each layers, with  $i = 1$  to  $N_D$ , as free parameters. The  
59 forward model is able to produce transmissions  $\mathbf{T}_m(z_i)$  at the impact factors  $z_i$  as a function the haze  
60 properties (extinction  $\mathbf{k}_H(z_i)$  and spectral slope  $\alpha_H(z_i)$ ) and the gas methane mixing ratio  $\mathbf{X}_{CH_4}(z_i)$ ,

61 directly linked to the gas extinction ( $k_{CH_4}(z_i)$ ). The model transmissions  $T_m(z_i)$  are compared with  
62 the observed transmissions  $T_o(z_i)$ . To produce the retrieval, we use a bayesian method along with a  
63 diffusive process to stabilize the algorithm. This is very suitable for this kind of triangular problems,  
64 that is when the transmission at a given level only depends on the atmosphere properties above  
65 this level. It is also convenient because only jacobians of the model are needed and the retrieval is  
66 performed with the iterative resolution of a matricial equation (See Appendix A and B for details).  
67 This method is very close to the one used by Maltagliati et al. (2015) and only the better methane  
68 linelist provided by Rey et al. (2018) brings a decisive improvement in our work.

69 **3. Constrasted stratospheric methane profiles :** The Figure 1 shows the retrieved vertical  
70 profiles for the two observations and the corresponding fits of the data. The methane profile at  
71 70°S (*T10*) displays an enriched layer, with a mixing ratio up to  $1.62 \pm 0.10\%$  at 165 km, isolated  
72 in a relatively dry background atmosphere. Above this layer, the methane mixing ratio is quite  
73 constant with a value  $\simeq 1.05 \pm 0.10\%$ . Below, it drops to  $\simeq 0.70 \pm 0.20\%$  around 125 km. With the  
74 *T53* observation, at 1°N, we find stratospheric values oscillating around 1.15% with error bars and  
75 amplitude both around 0.05% above 200 km and with a wavelength of  $35_{-3.0}^{+4.5}$  km. Huygens found,  
76 near the equator, that methane decreases from 5% to 1.5% (Niemann et al. (2005); Niemann et al.  
77 (2010)) in the troposphere due to condensation. We then find that it further decreases to reach  
78 values around 1.0 to 1.15% in the mid-stratosphere at low latitudes instead of being constant as  
79 generally assumed. Our results are consistent with the effective values retrieved with CIRS in broad  
80 altitude ranges at  $125 \pm 50$  km and  $225 \pm 50$  km. However, our vertical resolutions **allow us** to be  
81 much more assertive and lift ambiguities about the mechanisms behind this methane distribution.

82 The Figure 2 shows our results for the methane mixing ratios along with other values retrieved in  
83 the stratosphere and values evaluated at the cold trap (where the mixing ratio of air is capped due  
84 to the limitation by the saturation vapor pressure) from temperature measurements. Temperatures  
85 are stable with time in the low stratosphere (1 to 10 mbar) between 2005 and 2009 (Achterberg et al.  
86 (2011)) and are expected to be even more stable at higher pressure (Lebonnois et al. (2012)). We  
87 then consider temperatures gathered from data collected between 2005 and 2009, as a consistent set  
88 which depicts a steady state (Lellouch et al. (2014)). On the other hand, post-equinoctial changes  
89 are extremely rapid, even at the scale of a terrestrial year (e.g., Lebonnois et al. (2012); West et al.  
90 (2018); Teanby et al. (2019); Seignovert et al. (2021)). Thus, we ignore in our discussion data taken

91 after equinox.

92 At first glance, the methane mixing ratio at  $125 \pm 50$  km found by Lellouch et al. (2014) are  
93 highly variable with latitude. They appears not related to the latitudinal temperatures at the  
94 cold trap and the corresponding saturation mixing ratios, indicating a disconnection between the  
95 two fields (Figure 2). Near equator (*T53*), the mixing ratio  $1.25 \pm 0.25\%$  around 150 km is also  
96 lower, yet consistent, **compared to** the value found by the GC-MS (Gas Chromatograph and Mass  
97 Spectrometer) at the cold trap. The value  $1.15 \pm 0.05\%$  around 250 km is much lower and not  
98 consistent with the value found at cold trap or in the low stratosphere with the GC-MS. At  $70^\circ\text{S}$   
99 (*T10*), the methane mixing ratio of  $1.62 \pm 0.10\%$  in the enriched layer at 165 km is much larger than  
100 the maximum value allowed by the cold trap temperature at  $70^\circ\text{S}$ , that is  $\simeq 1.05 \pm 0.19\%$ . On the  
101 other hand, this value is comparable to values allowed by the cold trap between tropics. There is  
102 no local source of methane in the atmosphere and we do not expect that an eventual wave system  
103 could concentrate methane up to mixing ratios exceeding the background value by 50%. Such a  
104 layer is then a clear signature of an intrusion coming from a moist environment at another latitude.  
105 These results **undermine** the simple idea of a gaseous vertical flux through the tropopause at global  
106 scale, locally limited by the saturation vapor pressure.

107 **4. Methane profiles decyphered with Titan's circulation :** To understand our results, we  
108 first discuss Titan's circulation with the results of a GCM (Rannou et al. (2006)). Although this is  
109 a 2 dimensional (latitude-pressure) model, it accounts for the haze seasonal transport at global scale  
110 up to the mesosphere, the cloud microphysics and the radiative feedbacks. It predicts well the thick  
111 winter haze polarhood and the haze scavenging by clouds in the troposphere. The corresponding  
112 opacity fields are important drivers of the circulation through the control of the solar flux at the  
113 surface and the cooling to space during the polar night (e.g., Rannou et al. (2004)). To date,  
114 no existing 3-D model accounts for all these couplings although they have major effects on the  
115 troposphere and low stratosphere circulation.

116 GCMs long predicted two opposite circulation cells below the stratosphere temperature inversion  
117 (Rannou et al. (2006)) and a large thermally direct cell in the stratosphere with rapid post-  
118 equinoctial turnover (e.g., Rannou et al. (2004); Lebonnois et al. (2012); Larson et al. (2014);  
119 Lora et al. (2015)) (Figure 3). Before the Northern Spring Equinox in 2009, at the time of *T10*

120 and *T53*, both troposphere cells have a lower branch near surface which converge around 30-40°S  
121 to give an ascending branch emerging from the troposphere (e.g., Rannou et al. (2006)). This is  
122 equivalent to the tropospheric Hadley cells and the Inter-Tropical Convergence Zone (ITCZ) on  
123 Earth, except that cells extend up to the poles because of Titan’s slow rotation speed. Observation  
124 of the tropical clouds on Titan reveals the actual location of the Tropical Convergence Zone (here  
125 after TCZ) around  $\simeq 45 \pm 15^\circ\text{S}$  (Rodriguez et al. (2011); Turtle et al. (2018)). The prediction of  
126 clouds related to the Hadley-like cells in the coupled GCM (Rannou et al. (2006)) at the location  
127 where they are actually observed validates this model for our discussion. However, it should be  
128 reminded that its results are also zonally and time averaged. The sporadic nature of the cloud  
129 events, the short term fluctuations and longitude variabilities of the circulation are then erased.

130 At the TCZ, the ascending winds splits in two branches in the low stratosphere to close the Hadley-  
131 like cells. One cell drives air from the southern tropical tropopause to the south polar region up  
132 to around 150 – 200 km and sink near the south pole. The other cell is forced to return equator-  
133 ward almost horizontally at around 70 km altitude (upper branch) because the strong stratosphere  
134 temperature inversion, with high hydrostatic stability, prevents air from moving upward between  
135 tropics. This circulation leaves a strong dynamical signature with the zonal wind dropping due  
136 to the conservation of angular momentum. This dynamical signature is also observed at 70 km in  
137 reality, with zonal winds of 4 m/s in a thin layer of about 10 km thick while layers above and below  
138 have zonal winds larger than 40 – 50 m/s (Bird et al. (2005)). This specific layer makes a sharp  
139 transition between the Hadley-like cell and the stratosphere cell (Rannou et al. (2006); Lebonnois  
140 et al. (2012); Larson et al. (2014)).

141 Methane mole fractions superimposed to the circulation pattern (Figure 3) show that the methane-  
142 rich layer observed during *T10* is directly linked to an ascending branch which comes from the  
143 moist troposphere at the TCZ. It can convey methane to the south polar region up to about 160  
144 km, above the cold and dry polar tropopause and explains the intrusion layer. For *T53*, we know that  
145 in the inter-tropical region, the transition in methane abundance from 1.5% to 1.15% (and  $\simeq 1\%$  in  
146 Lellouch et al. (2014)) takes place at the interface between the Hadley-like cell and the thermally  
147 direct stratosphere cell. The sharp drop in the zonal wind at the same altitude (Bird et al. (2005))  
148 associated to horizontal winds and to a strong temperature inversion indicates inhibited vertical  
149 exchanges. It marks the transition between a moist troposphere and low stratosphere, on one side,

150 and the dryer background stratospheric air **fed** in methane through intrusions at some latitudes and  
151 not globally, on the other side. Clearly, the equatorial region is not one of these intrusion zones.

152 **5. Humidification by convection on Titan and on Earth :** The dynamical origin of the  
153 stratospheric methane distribution is obvious in observations, however GCMs accounting for non  
154 convective cloud microphysics and transport of species are not able to produce such distribution  
155 patterns (e.g., Rannou et al. (2006)). Instead, they predict a constant methane mixing ratio in  
156 the stratosphere and the mesosphere that directly depends on the mixing ratio at the tropopause.  
157 This discrepancy underlines the role of cloud convection in transferring methane upward above the  
158 cold trap and into the stratosphere. Between 2004 and 2010, clouds are frequently observed at the  
159 TCZ (Rodriguez et al. (2011); Turtle et al. (2018)) and briefly near equator during the equinoctial  
160 transition (Turtle et al. (2011)). A photometric analysis shows that the core of these cloud systems  
161 can reach the high troposphere and the tropopause (Griffith et al. (2005)). Mesoscale cloud models  
162 also show that with a surface humidity larger than 50% or 65%, convective clouds triggered by  
163 a perturbation in a moving air mass (Barth (2010)) or in a static atmosphere easily reach high  
164 altitudes (Barth and Rafkin (2007, 2010)). Large scale ascending motions in the Hadley-like cells  
165 could trigger and facilitate convection. Convective clouds also inject methane ice particles, rather  
166 than vapor, without limitation due to the saturation vapor pressure at the cold trap. This would  
167 further enhance contrasts in the stratospheric methane distribution.

168 On Earth, by comparison, the entry of water vapour in the stratosphere between tropics and at mid-  
169 latitudes is due to a slow gaseous advection, capped by cloud condensation, and also to overshooting  
170 of convective ice clouds in the lower stratosphere (e.g., Liu et al. (2010)). Convective overshoots  
171 marginally account for several percents of the total flux of water, less than 18% according to the  
172 largest estimates (e.g., Ueyama et al. (2018); Dauhut et al. (2015)). As a direct consequence,  
173 average water abundance in the tropical stratosphere is relatively homogenous and well related  
174 to the tropopause temperature. Only oxidation of methane further produces inhomogeneities in  
175 the water stratospheric abundance. The Earth's case enlighten Titan's case ; on Earth, transport  
176 dominated by slow advection yields a quite uniform water mixing ratio in the stratosphere. On the  
177 contrary, on Titan, a substantial transfer by convection and **an inhibited slow convection** explains  
178 the sharp distribution in the stratosphere and a low background mixing ratio. **In both atmospheric**  
179 **systems, slow advection and strong convective transports at the tropopause exist, but they have not**



180 the same relative strengths. This difference in transports through the tropopause causes different  
181 relationship between the tropospheric and stratospheric mixing ratio.

182 **6. Consequence on short and long term climate :** After Huygens observations, the strato-  
183 spheric methane abundance 1.4% is often used as a fiducial value to study other components of  
184 Titan (haze, clouds, temperature, other species, surface reflectivity, thermal equilibrium). This is  
185 incorrect in two ways. First, the methane stratospheric abundance is around 1 to 1.1% in average  
186 rather than 1.4%. Secondly, its value is highly variable with space and probably with time. A  
187 simple test using the model of Coutelier et al. (2021) shows that a low methane abundance affects  
188 the retrieved haze opacity in stratosphere by  $\simeq -4\%$  and the tropospheric mist opacity by  $\simeq +7\%$ .  
189 Although apparently moderate, these changes are about twice the accumulated errors produced by  
190 the instrumental uncertainties and the model bias on the retrieved values. Such a low methane  
191 abundance also sets new conditions for the amount and the distribution of methane in the up-  
192 per atmosphere (Yelle et al. (2021)), with probable consequences on the escape rates and on the  
193 composition of aerosols and minor species.

194 More generally, humidification by convective entry and inhibition of slow vertical transfer also sets  
195 new conditions for the evaluation of the long term climates of Titan (Lunine and Atreya (2008);  
196 Hayes et al. (2018)). This way to regulate the stratosphere humidification strongly depends on  
197 the thermodynamical conditions behind cloud formation and convection, known to have onset and  
198 cut-off thresholds (e.g., Barth and Rafkin (2007, 2010), Barth (2010)). Other climatic conditions,  
199 in the past or in the future, may radically change the way methane is transported upward due to  
200 convective motions. This would dim or enhance the flux of methane available for photochemistry  
201 much more and in a more complex way than anticipated with simple models based on slow advective  
202 transport (e.g., Lorenz et al. (1999)). Of course, it would also change our perception of Titan history  
203 in shaking the exchange timescales and fluxes between the different methane reservoirs in Titan.  
204 Future progress in understanding the methane cycle on Titan will strongly rely on efficient and  
205 complete global climate models, associated to mesoscale models, able to treat the cloud microphysics  
206 and the moist convection process in detailed aspects.

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352 **Appendix A The retrieval model:** Occultations probe extinction along lines of sight at the  
 353 limb of the planet and are not sensitive to multiple scattering due to lower layers or to surface  
 354 reflectivity. This kind of data easily allows to separate methane and haze extinction since they  
 355 extinct light in very different ways. To match data, we divide atmosphere according to the vertical  
 356 sampling of observations and we compute transmitted flux accounting haze and gas extinction in  
 357 each layer. We use data between  $0.8 \mu\text{m}$  and  $2.0 \mu\text{m}$ , where the signal is largely dominated by  
 358 haze and methane extinction. Other methane bands, at  $2.3 \mu\text{m}$  and  $3.4 \mu\text{m}$ , are mixed with other  
 359 undefined absorptions. This has been long remarked for the  $3.4 \mu\text{m}$  fundamental band (e.g., Malt-  
 360 agliati et al. (2015)) but, in this work, we also find a prominent unknown absorption that we discuss  
 361 later. Methane absorption is given by Rey et al. (2018) linelist and is treated with the correlated-k  
 362 method (Goody et al. (1989)) with 16 terms. Other gas properties are set with linelists published  
 363 in the Hitran database (Gordon et al. (2017)). The mixing ratio  $X_{CH_4}$  is the only free parameter  
 364 relative to methane given for each layer. Haze extinction is computed with a model of scattering  
 365 and absorption by fractal aggregates (Rannou et al. (1997)) and the main characteristics of haze  
 366 could be summarized as a vertical profile of extinction coefficient  $k_H$  at a reference wavelength,  $\lambda_0$ ,  
 367 and the spectral slope  $\alpha_H = \partial k_H(\lambda)/\partial \lambda$ .

368

369 **Appendix B Method related to the retrieval procedure:** To produce our analysis, we devel-  
 370 oped a retrieval model following the technique of the bayesian inference (Rodgers (2000)). Using  
 371 the Bayes theorem, this technique allows for retrieving a parameter set  $\mathbf{x}$  along with an estimated  
 372 error  $\sigma_{\mathbf{x}}$  from a set of observation  $\mathbf{y}$  with observation errors  $\sigma_{\mathbf{y}}$ . Each set is represented by a vector.  
 373 The strength of this approach essentially relies on the fact that we only need a knowledge about  
 374 the direct relationship between  $\mathbf{y}$  and  $\mathbf{x}$ . This relation is formally given by  $\mathbf{y} = F(\mathbf{x})$ , where  $F$   
 375 stands, in our case, for the radiative transfer model that the links observed transmissions ( $\mathbf{y}$ ) and  
 376 the atmosphere parameters ( $\mathbf{x}$ ) such as the haze extinction or gas mixing ratios.

377

378 To proceed, the function  $F$  has to be linearized around a given solution  $\mathbf{y}(\mathbf{x}_0) = \mathbf{y}_0 + \partial F(\mathbf{x})/\partial \mathbf{x} \times$   
 379  $(\mathbf{x} - \mathbf{x}_0)$ , and where  $\mathbf{y}_0 = F(\mathbf{x}_0)$ . In this way, the set of partial derivatives  $\partial F(\mathbf{x}_0)/\partial \mathbf{x}$  yields a matrix  
 380  $\mathbf{K}$  and the estimation for the best value of  $\bar{\mathbf{x}}$  can be written as (Rodgers (2000)) :

381



$$\bar{\mathbf{x}} = \mathbf{x}_0 + (\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{K})^{-1} \mathbf{K}^T \mathbf{S}_\epsilon^{-1} (\mathbf{y} - \mathbf{y}(\mathbf{x}_0)) \quad (1)$$

382 where  $\mathbf{S}_\epsilon$  is the diagonal error matrix ( $\mathbf{S}_{\epsilon,(ij)} = \delta_{i,j}/\sigma_{\mathbf{y}_i}^2$ ),  $\mathbf{S} = (\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{K})^{-1}$  is the covariance matrix  
383 and  $\mathbf{y}$  is the observation vector. If  $\mathbf{x}_0$  is far from the best solution, the first estimated value  $\bar{\mathbf{x}}$  will  
384 not be the best possible solution but will be better than  $\mathbf{x}_0$ . So we can start an iterative process to  
385 converge toward a solution which will be the most likely solution. In general, 20 to 40 iterations are  
386 needed to obtain an acceptable solution. It depends on the size of the vector  $\mathbf{x}$ . Notably, although  
387 we start with an initial guess for  $\mathbf{x}_0$ , we do not include any *a priori* value in the retrieval matrix.  
388 The error on the retrieved parameter  $\mathbf{x}$  are given by the square root of the diagonal terms in the  
389 covariance matrix  $\sigma_{x_i} = \sqrt{\mathbf{S}_{ii}}$

390

391 In order to avoid instabilities in the converged solution, we used a Tikhonov regularization. This  
392 consists in adding a process analogous to a diffusion in the  $\mathbf{x}$ -space to attenuate spurious oscilla-  
393 tions. It takes the form of a matrix,  $\mathbf{H}$ , inserted in the covariance matrix as:

394

$$\mathbf{S}^\dagger = (\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{K} + \beta \mathbf{H})^{-1} = (\mathbf{S}^{-1} + \beta \mathbf{H})^{-1} \quad (2)$$

395 where  $\mathbf{H}$  is defined from the second order derivative matrix  $\mathbf{L}$  as  $\mathbf{H} = \mathbf{L}^2$  (e.g, Quémerais et al.  
396 (2006); Koskinen et al. (2011)) and  $\beta$  is analogous to a diffusion coefficient. The order of mag-  
397 nitude of the matrix terms in  $\mathbf{L}$  is around  $(\Delta Z)^{-2}$ , with  $\Delta Z$  the vertical distance between two  
398 consecutive observations. Setting  $\beta$  is a matter of tuning since there is no related physical process  
399 behind. We scale  $\beta$  against the larger term of  $\mathbf{S}^{-1}$  and we defined a free factor  $\gamma$  so that we have  
400  $\beta = \gamma \Delta Z^4 \times \max(\mathbf{S}_{i,j}^{-1})$ . We tested several values of  $\gamma$  around 1 in order to remove small scale oscil-  
401 lations but keeping the variations comparable to the atmosphere scale height ( $\simeq 40$  km). We found  
402 that  $\gamma \simeq 0.316$  gives the requested effect for retrieval performed with VIMS data at wavelengths  
403 shorter than  $3 \mu\text{m}$ .

404

405 To retrieve haze and methane, we proceed in two steps. We first use the methane windows where

406 haze extinction alone acts on transmission. This allows to fix the haze vertical and spectral extinc-  
407 tion. With this information about the haze layer, we then retrieve the vertical profile of methane  
408 mixing ratio. we also need to account for the spectral shift of the VIMS instrument (e.g. Maltagliati  
409 et al. (2015)) that we evaluated as part of the method. We use a subset of the data between 200  
410 and 300 km, in the middle atmosphere, and we seek for the best fit with different values of the shift.  
411 With a cost function ( $\chi^2$ ) computed in the methane bands only, we are able to draw the value of  
412 a  $\chi^2$  as a function of the wavelength shift. We then evaluate the best value of the shift and the  
413 error bars. Wavelength shifts for VIMS channels giving the best matches of data are  $2.64^{+0.16}_{-0.21}$  nm  
414 for *T10* and  $9.40^{+0.10}_{-0.09}$  nm for *T53*. This significantly differs from RC19<sup>1</sup> prescriptions.

415 **Appendix C Retrieval with wavelengths shorter than 2  $\mu\text{m}$ :** We show here comparisons  
416 between our results and results from the previous analysis (Maltagliati et al. (2015)) obtained with  
417 the same dataset. The Figure 4 shows the retrieved vertical profiles of methane mixing ratio for  
418 the two observations *T10* and *T53* as retrieved in this work and as retrieved by Maltagliati et al.  
419 (2015) in the bands at 1.4  $\mu\text{m}$  and 1.7  $\mu\text{m}$ . We also display the retrieval that we can perform with  
420 the three bands at 1.2  $\mu\text{m}$ , 1.4  $\mu\text{m}$  and 1.7  $\mu\text{m}$  separately. This shows that results concerning the  
421 methane mixing ratio differ from the one made by Maltagliati et al. (2015), especially concerning  
422 the enriched layer at 165 km for *T10* which was not retrieved in the previous analysis. This feature  
423 is real since it appears separately in the three methane bands, in our analysis, with about the same  
424 contrast relative to the background. The background itself is also consistently found with the three  
425 different bands separately. The main difference between our work and the work by Maltagliati et al.  
426 (2015) is due to the high quality of the methane linelist that we use (e.g. Rey et al. (2018)). The  
427 corresponding fits of the data are shown in the Figure 5.

428 **Appendix D Retrieval in the 2.3  $\mu\text{m}$  methane band:** The Figure 6 shows the retrieved ver-  
429 tical profiles for the two observations *T10* and *T53*, in the two wavelength intervals : between 2.0  
430 and 2.8  $\mu\text{m}$  (band at 2.3  $\mu\text{m}$ ) and 0.88 to 2  $\mu\text{m}$ . The modeled and retrieved spectra for the 2.3  $\mu\text{m}$   
431 band are also shown. In our results, we immediately remark that the 2.3  $\mu\text{m}$  band can not allow for  
432 a safe retrieval of the methane abundance because another species strongly interacts with methane  
433 absorption. This clearly appears in the spectra that can not be match with the same quality of fit

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<sup>1</sup>[https://pds-imaging.jpl.nasa.gov/data/cassini/cassini\\_orbiter/vims-calibration-files/clark-et-al\\_vims-radiometric-calibration-pds-2018-v2.0.pdf](https://pds-imaging.jpl.nasa.gov/data/cassini/cassini_orbiter/vims-calibration-files/clark-et-al_vims-radiometric-calibration-pds-2018-v2.0.pdf)

434 than the spectra in between 0.88 to 2  $\mu\text{m}$  (Figure 1 of the article and Figure 5). Moreover, these  
435 poor fits are obtained with an excessive amount of methane. It was already well known that extra  
436 absorptions exist in the C-H fundamental band at 3.4  $\mu\text{m}$  where methane does not absorb enough  
437 to explain observations (Bellucci et al. (2009); Maltagliati et al. (2015)). Here, we clearly see that  
438 there is also an undefined absorption in the 2.3  $\mu\text{m}$  band. As suggested by Maltagliati et al. (2015),  
439 ethane could be the absorbing gas, although it would appear surprising that a trace gas could com-  
440 pete with methane in one of its bands. If ethane is not responsible for the extra absorption in the  
441 2.3  $\mu\text{m}$  band, it would mean that something is not yet understood in Titan's atmosphere opacity.  
442 For now, we consider that retrieval of methane with the 2.3  $\mu\text{m}$  band is not reliable and is only  
443 shown here for demonstration.

444

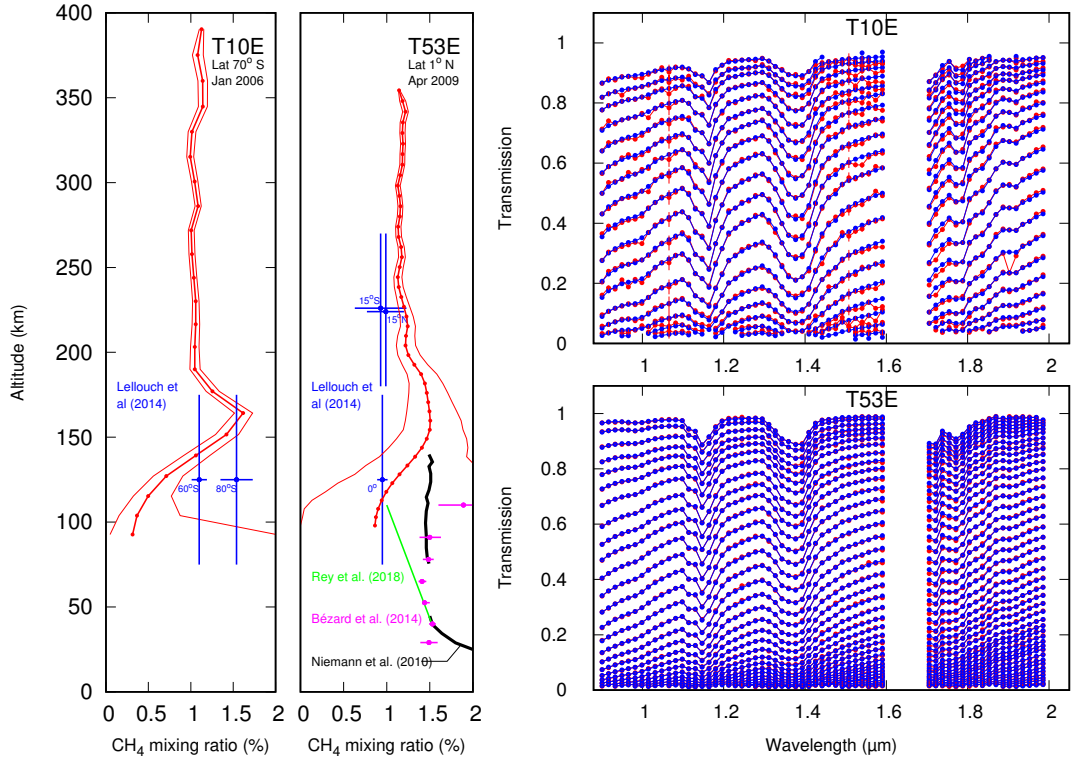


Figure 1: *Two plots at left : Retrieved methane mixing ratio (red dots) and the related error envelope (red curves) with the two observation sets, between 0.88 and 2  $\mu\text{m}$ . For the observation T10, we also plot the retrieval made with the CIRS observations at  $125 \pm 50$  km (Lellouch et al. (2014)) at  $60^\circ\text{S}$  in Jul. 2009 and at  $80^\circ\text{S}$  in Nov 2007. For T53, we plot the methane mixing ratio retrieved with CIRS at  $125 \pm 50$  km at equator in Dec. 2007, and at  $225 \pm 50$  km at  $15^\circ\text{N}$  in March 2007 and at  $15^\circ\text{S}$  in Sept 2006. We also show the methane mixing ratio retrieved by the Gas Chromatograph and Mass Spectrometer (GC-MS) (Niemann et al. (2010)) and with the Descent Imager Spectral Radiometer (DISR) (Bézard (2014); Rey et al. (2018)), both onboard Huygens which landed in January 2005 at  $\simeq 10^\circ\text{S}$ . The spatial resolutions of our vertical profiles are  $\pm 6.25$  km and  $\pm 2.5$  km for T10 and T53 respectively. At right, comparison between observed transmissions (red dots with error bars) and the model retrieval (blue dots) in the spectral range selected to retrieve methane mixing ratio. The gap in spectra between 1.59 and 1.68  $\mu\text{m}$  is due to uncorrect spectels in VIMS instrument.*

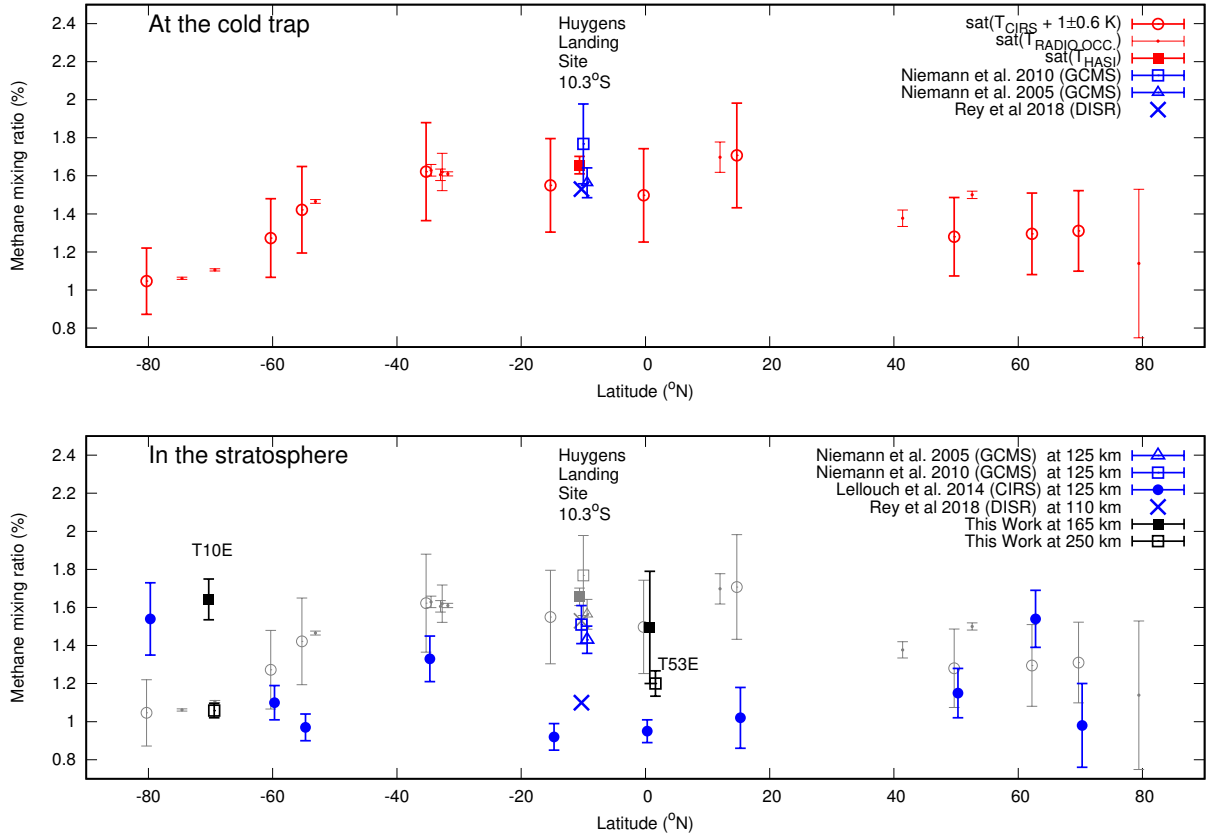


Figure 2: *At top* : Methane mixing ratio as a function of the latitude at the cold trap (altitude  $\simeq 30$  km). We show values measured with the GC-MS (Niemann et al. (2005); Niemann et al. (2010)) and retrieved with the DISR (updated values by Rey et al. (2018)), both onboard Huygens. We also show saturation methane mixing ratio inferred from temperature measurements around the cold trap made by the Huygens Atmospheric Structure Instrument (HASI) (Fulchignoni et al. (2005)), from temperatures at  $30 \pm 1$  km retrieved with the Radio Occultation Experiment (Schinder et al. (2011, 2012, 2020)) and from temperatures retrieved by CIRS (Lellouch et al. (2014)) around the cold trap. We only selected dates before the North Spring Equinox. We add  $+1 \pm 0.6$  K to CIRS temperatures in order to account for a systematic bias in retrieved temperatures at the cold trap, compared to HASI and to the radio-occultation temperature retrievals (Lellouch et al. (2014)). This makes the *saturation* methane mixing ratios calculated from CIRS at the cold trap to be consistent with the 'in situ' measurements made by Huygens. At the cold trap, methane condensate is expected to be pure ice (Tokano et al. (2006)), then we use vapor pressures as published by Fray and Schmitt (2009). *At bottom*: Methane mixing ratios retrieved in the stratosphere for this study along with the retrieval made by CIRS at 125 km and DISR at 110 km. The light grey symbols are those displayed in the top panel and reported here to facilitate comparisons.

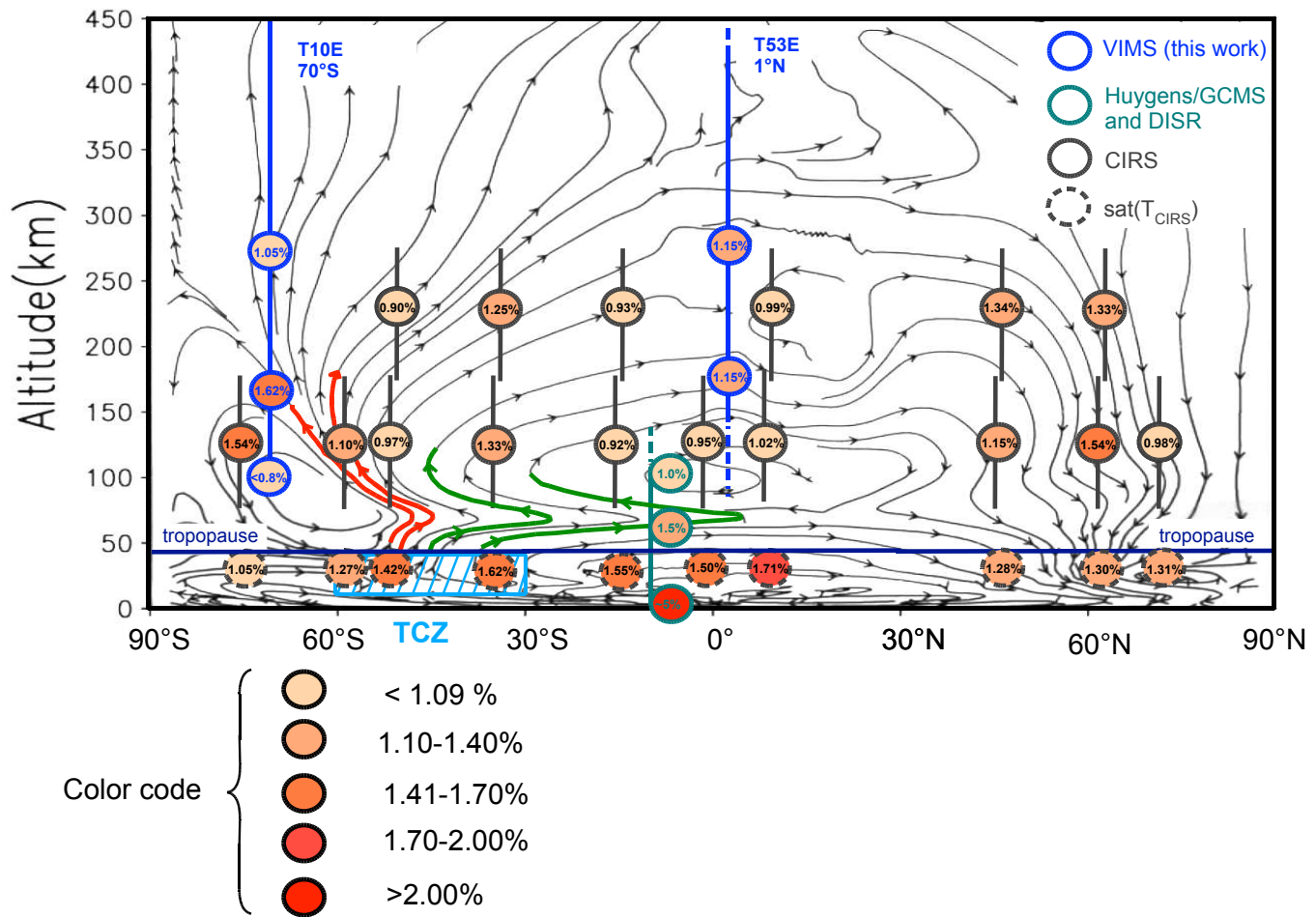


Figure 3: Circulation pattern averaged over one terrestrial year before the Northern Spring Equinox as predicted by the Titan IPSL-GCM (Rannou et al. (2006)) shown along with values of methane mixing ratios (values and color code) measured with Huygens (Niemann et al. (2010); Bézard (2014); Rey et al. (2018)), with CIRS (Lellouch et al. (2014)) and found in this work. Note, when comparing values, that CIRS values are averaged on broad vertical intervals of  $\pm 50$  km while other values are local (less than  $\pm 6.3$  km). We also show the saturation mixing ratios for the cold trap temperature evaluated with CIRS (dashed circles). The mixing ratios shown here are reported, without their error bars, from Figure 2 except for the CIRS values at 225 km. In this case, error bars are between 0.15% and 0.47%. Two couples of wind streams are highlighted in red and green. They show how the methane-rich south tropical ascending branch at the TCZ, where convective clouds are frequently observed before equinox at latitude  $45 \pm 15^\circ S$  (Cyan hatched box, Rodriguez et al. (2011); Turtle et al. (2018)), is connected to the methane rich layer at  $70^\circ S$  and to the transition layer around 70 km in the equatorial region. High density of the stream lines is related to high mass flux. Therefore, the flux from the TCZ is quite constant and remains strong up to the edge of south polar region while the branch toward the equator rapidly diverges and weakens. It should be kept in mind that the circulation fluctuates with time around average patterns, as those shown here.

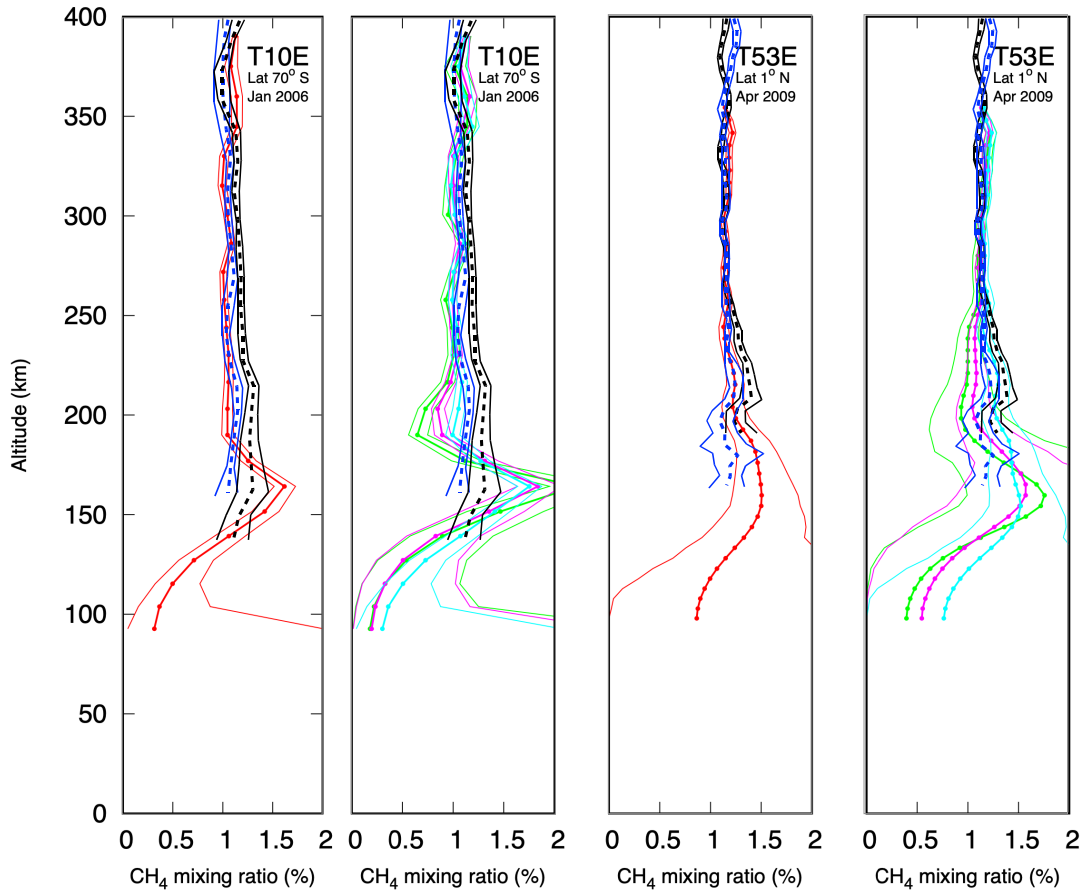


Figure 4: (First and third graphs from left) Methane mixing ratio (red dots) and the related error envelope (red curves) retrieved with the observation sets *T10* and *T53*, and with data at wavelengths between 0.88 and 2.03  $\mu\text{m}$  shown along with the profiles retrieved by Maltagliati et al. (2015) with the 1.4  $\mu\text{m}$  (dash black) and the 1.7  $\mu\text{m}$  (dashed blue) methane bands. (Second and fourth graph from left) Methane mixing ratios (color dots) and the related error envelopes (color curves) obtained with the observation sets *T10* and *T53*, and using the methane bands at 1.2  $\mu\text{m}$  (light green), 1.4  $\mu\text{m}$  (pink) and 1.7  $\mu\text{m}$  (cyan), shown along with the profiles retrieved by Maltagliati et al. (2015) with the 1.4  $\mu\text{m}$  (dash black) and the 1.7  $\mu\text{m}$  (dashed blue) methane bands.

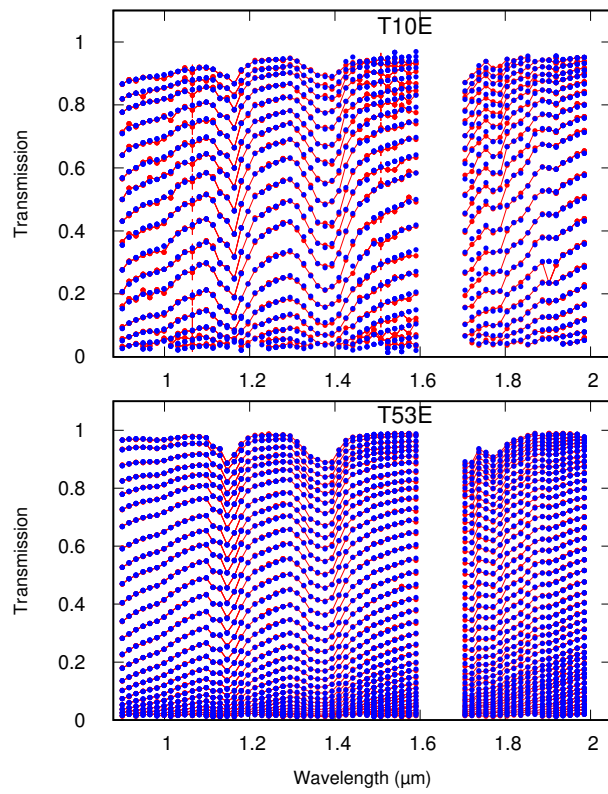


Figure 5: Comparison between observed transmissions (red dots with error bars) and the concatenation of the model retrieval (blue dots) made with the three methane bands separately. The gap in spectra between 1.59 and 1.68  $\mu\text{m}$  is due to uncorrect spectels in VIMS instrument.



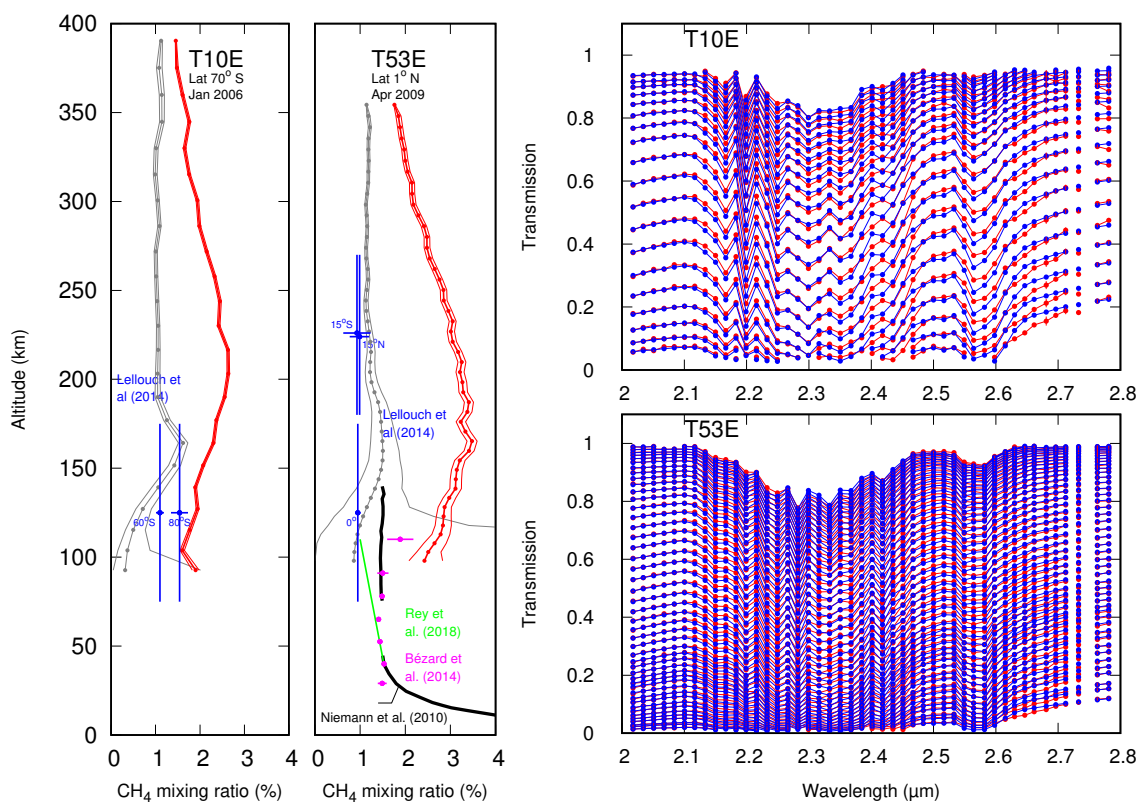


Figure 6: Two plots at left : Retrieved methane mixing ratio (red dots) and the related error envelope (red curves) with the observation sets  $T10$  and  $T53$ , with wavelength between  $2.03$  and  $2.8 \mu\text{m}$ . The grey profile are those plotted in the Figure 1 of the article, with retrievals performed with parts of the spectra between  $0.88$  and  $2 \mu\text{m}$ . We also plot methane mixing ratios from other works (Lellouch et al. (2014), Niemann et al. (2010), Bézard (2014); Rey et al. (2018)). At right, comparison between observed transmissions (red dots with error bars) and the model retrieval (blue dots) in the spectral range selected to retrieve methane mixing ratio. The gap in spectra corresponds to wavelengths where ethane is known to absorb, but can not be modelled.