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A non-monotonic eddy diffusivity profile of Titan's atmosphere revealed by Cassini observations

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ABSTRACT

Recent measurements from the limb-view soundings of Cassini/CIRS and the stellar occultations from Cassini/UVIS revealed the complete vertical profiles of minor species (e.g., C_2H_2 and C_2H_4) from 100 to 1000 km in the atmosphere of Titan. In this study, we developed an inversion technique to retrieve the eddy diffusion profile using C_2H_2 as a tracer species. The retrieved eddy profile features a low eddy diffusion zone near the altitude of the detached haze layer (~ 550 km), which could be a consequence of stabilization through aerosol heating. Photochemical modeling results using the retrieved eddy profile are in better agreement with the Cassini measurements than previous models. The underestimation of C_2H_4 in the stratosphere has been a long-standing problem in planetary photochemical modeling, and the new eddy diffusion profile does not solve this problem. In order to match the observations, we suggest a new expression for the rate coefficient of the key reaction, $H+C_2H_4+M \longrightarrow C_2H_5+M$. The new reaction rate coefficient is estimated to be ~ 10 times lower than that used by Moses et al. (2005)'s model, and should be validated in the laboratory and tested against the hydrocarbon chemistry of giant planets.

1. Introduction

In one-dimensional photochemical models, the vertical transport of a species is often parameterized as a diffusion process. The diffusion coefficient, referred to as K_{zz} or eddy diffusivity, incorporates various scales of turbulent processes arising from nonlinear wave breaking and instabilities. Many empirical studies estimated this parameter from the amplitude of gravity waves (e.g. Strobel, 1974). Lindzen (1981) first theoretically concluded that the eddy diffusivity was proportional to the inverse square root of background atmospheric density and his finding served as a basis for the estimation of eddy diffusion in early photochemical models (e.g. Yung et al., 1984). The macroscopic eddy mixing resulting from dynamical instabilities is much more difficult to characterize. Two primary sources of these instabilities are convective instability and shear instability. The stability parameter is the Richardson number:

$$R_i = \frac{N^2}{\left(\frac{\partial U}{\partial z}\right)^2 + \left(\frac{\partial V}{\partial z}\right)^2} \tag{1}$$

where N^2 is the square of the Brunt–Väisälä frequency; U, V are the mean zonal and meridional winds, respectively. The atmosphere is subject to convective instability if R_i is negative and is subject to mechanically driven turbulent flow if R_i is positive but less than 0.25 (Taylor, 1931). Detailed calculation of eddy diffusivities relies on sophisticated dynamical models, such as the large-eddy simulation (LES) model, and high-order closure formulations of turbulent kinetic energy (TKE), which have been elaborated in boundary layer meteorology (Stull, 1988; Wyngaard, 1992). More practical ways to estimate the eddy diffusivity are based on observations. For example, oceanographers usually inject non-reactive tracers into the ocean and observe their evolution. Fitting the spreading of the tracer to a diffusive equation measures the eddy diffusivity.

In photochemical modeling of planetary atmospheres, such experiments are difficult to implement and high-resolution dynamical modeling coupled with chemical sources and sinks has not yet matured. Instead, modelers adopt the eddy diffusivity required to fit the measured abundance of a species whose vertical structure is controlled primarily by transport. This empirical approach not only facilitates the modeling of vertical transport but sheds light on unknown dynamical processes as well. For instance, Allen et al. (1981) found that a sudden decrease of eddy diffusivity at 92 km in Earth's mesosphere was required to produce the atomic oxygen peak. This hypothesis was later confirmed and

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explained by the theory of breakdown of gravity waves (Lindzen, 1981).

A significant number of photochemical models has been developed to investigate the distribution of hydrocarbons in Titan's atmosphere (Yung et al., 1984; Lara et al., 1996; Wilson and Atreya, 2004; Lavvas et al., 2008a, 2008b; Krasnopolsky, 2009). It has been accepted wisdom that the eddy diffusivity increases monotonically from the stratosphere to the thermosphere. Though this argument is rooted in the monotonic growth of the amplitude of gravity waves, it ignores the eddy mixing resulting from convective and shear instabilities, which depend on local properties whose strength is not guaranteed to behave monotonically with respect to altitude. In fact, Titan's atmosphere exhibits strong thermal variations and wind shear. At several altitudes, the temperature lapse rate exceeds the adiabatic lapse rate (Fulchignoni et al., 2005). Therefore, it is entirely possible that the eddy diffusivity could reach a maximum (minimum) in the interior when the lapse rate is largest (smallest) or when the wind shear is strongest (weakest).

In this work, we explore the possibility of a non-monotonic eddy diffusion profile. We develop an inversion technique that uses C_2H_2 as the tracer species for inverting the required eddy diffusion profile that agrees with the latest Cassini/CIRS, UVIS and INMS observations. We also discuss the revision of the rate coefficients for the chemistry of hydrocarbons given the retrieved eddy diffusion profile.

In Section 2, we describe the strategy for choosing an appropriate tracer species. In Section 3, we provide the inversion techniques and compare the modeling result using the retrieved eddy profile with the observations. In Section 4, we analyze the chemical pathways and propose updates for hydrocarbon chemistry. In the final section, we discuss the role of CH_4 escape and heterogeneous reactions that might affect our retrieval. We also discuss a possible mechanism that gives rise to the retrieved eddy profile.

2. Choice of tracer species

In the chemical modeling literature of Titan's atmosphere, the choice of tracer species progresses with the available observations. When Voyager (Coustenis et al., 1989) and ground-based millimeter observations (Tanguy et al., 1990) first detected HCN, its abundance was used to constrain the eddy diffusivity in the lower atmosphere (Toublanc et al., 1995; Lara et al., 1996) for HCN was thought to possess low reactivity with other species. After the arrival of Cassini spacecraft in 2005, subsequent measurements provided new constraints on the eddy diffusion profile. Lavvas et al. (2008a, 2008b) constructed the first comprehensive photochemical model based on Cassini measurements. In their model, the eddy diffusion profile was adjusted to fit the abundance of C_2H_6 (Vinatier et al., 2007) and ^{40}Ar (Waite et al., 2005). In addition, Yelle et al. (2008) suggested an asymptotic expression for the eddy diffusion profile based on the thermospheric profile of ^{40}Ar, CH_4 (above 1000 km) and the stratospheric abundance of C_2H_6 ($100 \sim 300$ km). This eddy diffusion profile was then widely used in recent chemical models (Vuitton et al., 2008; Hörst et al., 2008; Krasnopolsky, 2009). However, the asymptotic expression relies on a free parameter γ, which is 0.9 in Yelle et al. (2008) but 2.0 in the model of Krasnopolsky (2009, Appendix), and the modeling results produced by different choices of γ were inconsistent with the observations for some important species (e.g., C_2H_2 in the model of Krasnopolsky, 2009; C_4H_2 in Vuitton et al., 2008). The discrepancies are likely caused by the unconstrained eddy diffusion profile in the mesosphere of Titan ($500-1000$ km). Changes in the altitude of the fall-off region in the asymptotic

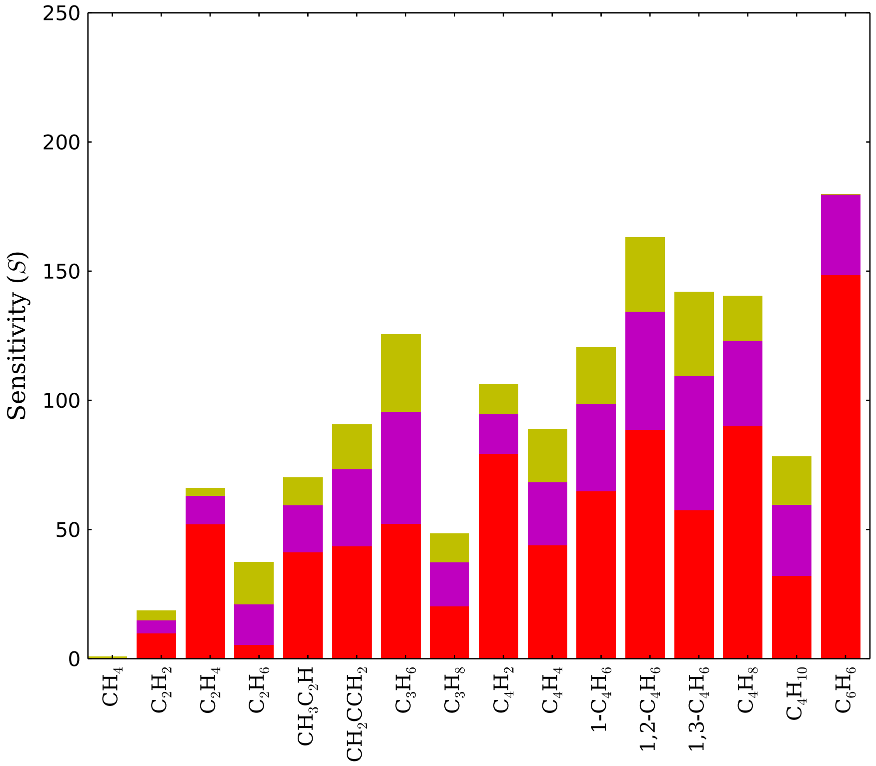

Fig. 1. Summation of the square of fractional changes over 82 levels and 297 reactions for 18 species when the rate coefficient for each reaction is doubled. See Eq. (2) for the definition of sensitivity. For clarity, radicals are not shown in the figure because their abundances are not affected by transport due to their short chemical lifetimes. The total sensitivity is divided into three parts: red part is the contribution from 50 km to 500 km; magenta, from 500 km to 1000 km; and yellow, above 1000 km. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this article.)