Available online at www.sciencedirect.com
ScienceDirect
Planetary and Space Science 56 (2008) 27–66
Planetary and Space Science
www.elsevier.com/locate/pss

Coupling photochemistry with haze formation in Titan's atmosphere, Part I: Model description

P.P. Lavvas[a,c], A. Coustenis[b], I.M. Vardavas[a,c,*]

[a]Department of Physics, University of Crete, Vassilika Voutes, P.O. Box 2208, 71003 Heraklion, Crete, Greece
[b]Observatory of Paris at Meudon, 5 Place J. Janssen, 92195 Meudon, Cedex, France
[c]Foundation for Research and Technology Hellas (FORTH), Heraklion, Crete, Greece

Received 13 March 2007; received in revised form 24 April 2007; accepted 30 May 2007
Available online 12 September 2007

Abstract

We introduce a new 1D coupled Radiative/Convective-Photochemical-Microphysical model for a planetary atmosphere and apply it to Titan. The model incorporates detailed radiation transfer calculations for the description of the shortwave and longwave fluxes which provide the vertical structure of the radiation field and temperature profile. These are used for the generation of the photochemistry inside the atmosphere from the photolysis of Titan's main constituents, nitrogen (N_2) and methane (CH_4). The resulting hydrocarbons and nitriles are used for the production of the haze precursors, whose evolution is described by the microphysical part of the model. The calculated aerosol and gas opacities are iteratively included in the radiation transfer calculations in order to investigate their effect on the resulting temperature profile and geometric albedo. The main purpose of this model is to help in the understanding of the missing link between the gas production and particle transformation in Titan's atmosphere. In this part, the basic physical mechanisms included in the model are described. The final results regarding the eddy mixing profile, the chemical composition and the role of the different haze precursors suggested in the literature are presented in Part II along with the sensitivity of the results to the molecular nitrogen photoionization scheme and the impact of galactic cosmic rays in the atmospheric chemistry.

Keywords: Titan; Planetary Atmospheres; Photochemistry; Microphysics; Haze

1. Introduction

From the initial observation by Huygens (1655) and the methane detection by Kuiper (1944) to the recent Cassini/Huygens space mission, Titan has been the subject of many studies that have aimed towards understanding its climate and the important role of the haze that is formed in its atmosphere and obscures its surface from direct observations in the visible. The processes that control haze formation and its radiative properties have been the least understood to date. The recent success of the Cassini/Huygens mission has provided valuable validation data that supplement the earlier Voyager mission data and many years of ground-based observations.

The most prominent characteristic of Titan's atmosphere is the well-defined haze structure, observed since the Voyager era (Rages and Pollack, 1980). The haze is directly observed since it provides the orange color of the Titan's atmosphere in the visible images and its origin is linked to the photochemistry taking place in its atmosphere. Nitrogen and methane, the most abundant constituents in Titan's atmosphere, are photodissociated by solar ultraviolet radiation, energetic particles from Saturn's magnetosphere and galactic cosmic rays (GCR), leading to the initiation of a complex organic photochemistry, which finally produces the haze. This coupling between the photochemistry and haze formation is the subject of the present work.

*Corresponding author. Department of Physics, University of Crete, Vassilika Voutes, P.O. Box 2208, 71003 Heraklion, Crete, Greece.
E-mail address: vardavas@iesl.forth.gr (I.M. Vardavas).

0032-0633/$ - see front matter
doi:10.1016/j.pss.2007.05.026

The neutral photochemistry in Titan's atmosphere was investigated even before the Voyager era (Strobel, 1974; Allen et al., 1980). The Voyager mission provided data that led to the first detailed photochemical model developed by Yung et al. (1984) which described the basic photochemical schemes that control the abundance of the observed hydrocarbons and nitriles in Titan's atmosphere. Based on this early work and further analysis of Voyager data and ground-based observations, more advanced photochemical models were developed. Toublanc et al. (1995) used an elaborate Monte Carlo description for solar radiation transfer within the atmosphere to investigate the possible production of oxygen-containing species arising from an influx of water vapor at the top of the atmosphere. Lara et al. (1996) used an ablation profile for the water vapor influx, included the effects of GCR and presented a physical description of the condensation processes taking place in Titan's lower stratosphere. Lebonnois et al. (2001) investigated the seasonal variation of the composition in Titan's stratosphere using a 2-D (latitude–altitude) model. Beyond neutral species chemistry, models have included ionospheric chemistry, as in the recent work of Wilson and Atreya (2004) where the contributions of energetic electrons and photoelectrons were included.

A common approach to modelling the photochemistry has been to generate the vertical temperature distribution, from the surface to the thermosphere, using temperature vertical profiles that were synthesized by combining measurements from Voyager I (Lindal et al., 1983) and model results for the temperature structure at different altitudes (Lellouch et al., 1989; Yelle, 1991; Yelle et al., 1997). Further, in order to include the effects of the aerosols in the radiation transfer calculations, vertical profiles of haze opacity were either specified by a simple exponential decrease with altitude or in more recent work generated by microphysical models using a specified vertical haze production rate (Yung et al., 1984; Lebonnois et al., 2001; Wilson and Atreya, 2004). Using this approach, photochemical models have managed to fit most of the atmospheric species concentrations available from observations before the Cassini/Huygens mission (Coustenis et al., 1995). In the present work we develop a modelling approach that generates the thermal structure, the atmospheric composition and the haze structure, in a self-consistent manner. The haze is produced from polymer production governed by the photochemistry, which is determined by and determines both the radiation field and atmospheric temperature structure. However, we do not address the complex problem of non-LTE effects on the temperature structure in the upper atmosphere.

The microphysical models used to derive the haze vertical structure and its optical properties are usually validated against Titan's spectral geometric albedo from the ultraviolet to the near-infrared based on ground-based and space observations. As was first shown by McKay et al. (1989), the fit to the spectral geometric albedo depends mainly on three parameters; the haze particles' optical properties (refractive index, size, shape and amount), the methane profile and Titan's surface reflectivity; using different spectral domains of the above region, constraints can be set on the values of parameters controlling the haze structure.

The haze particles' refractive index is based on laboratory measurements, while the size and amount is generated by the microphysical models, assuming their shape. The shape of the haze particles in Titan's atmosphere has been the subject of debate for a long time. Photopolarimetry measurements of scattered light from the Pioneer 11 (Tomasko and Smith, 1982) and Voyager (West et al., 1983) space missions have given high polarization at ~90° phase angle, which if the particles are spherical, constrains their size to 0.1 μm. On the other hand, high phase angle brightness measurements from Voyager (Rages et al., 1983), required particles between 0.2 and 0.5 μm with the upper limit more plausible. In order to overcome this problem, two possible solutions were suggested; one of a bimodal distribution (Courtin et al., 1991; Toon et al., 1992a,b) and the other of fractal aggregates constructed from spherical units (West and Smith, 1991; Rannou et al., 1995, 1997). Since then many microphysical models using the fractal aggregates have been published (Rannou et al., 2003 and references therein). The advantage of the fractal approach is that it provides, in general, a good fit to the geometric albedo both in the UV, visible and near-IR regions while at the same time matches the polarization data. However, fractal models have been unable to provide a good fit to the methane absorption feature at 0.62 μm in comparison with the success of the spherical particle models. The fit to the data was improved by applying a haze cut-off below 100 km, (Tomasko et al., 1997; Rannou et al., 2003) as suggested by HST measurements at that time (Young et al., 2002). A haze clearing was also included in the spherical particle models but at lower altitudes (below 30 km in McKay et al., 1989). The recent results from the DISR instrument on board the Huygens probe (Tomasco et al., 2005) show that the haze opacity extends down to the surface. Here we assume the particles to attain a spherical shape, starting from the monomer's size which corresponds to the smallest aerosol particle generated by the photochemical description. No fractal structures are considered.

In addition to the shape/size, the haze particles' refractive index is an important parameter in the model calculations. Until recently, most models used the first laboratory measurements for the refractive index of Titan haze-type analogs (tholins) made by Khare et al. (1984a), scaled by a factor which depends on the wavelength and the type of particles used; for spherical particles $\frac{4}{3}$ in the shortwave region of the geometric albedo (McKay et al., 1989), while for fractal particles 3 in the UV and 1.5 in the visible (Rannou et al., 1995). More recent measurements have shown that the optical properties of the laboratory haze analogs depend significantly on the experimental