EPSC Abstracts
Vol. 6, EPSC-DPS2011-885, 2011
EPSC-DPS Joint Meeting 2011

How to build a reduced chemical scheme for 2D/3D photochemical models?

M. Dobrijevic (1,2), T. Cavalié (1,2) and F. Billebaud (1,2)
(1) Université de Bordeaux, Observatoire Aquitain des Sciences de l'Univers, 2 rue de l'Observatoire, BP 89, F-33271 Floirac Cedex, France) (2) CNRS, UMR 5804, Laboratoire d'Astrophysique de Bordeaux, 2 rue de l'Observatoire, BP 89, F-33271 Floirac Cedex, France

Abstract

One major step in the development of 2D/3D photochemical models and GCMs is to determine a chemical scheme that is both simple (i.e. it contains a small number of compounds and reactions) and valid (i.e. the results of the model are in agreement with a reference model). In the present paper, we present a methodology to build a reduced chemical scheme adapted to the study of hydrocarbons in the atmospheres of giant planets and Titan [1]. As an example, we have built a reduced chemical scheme containing only 25 compounds and 46 reactions (including photolysis), which is well adapted to compute the abundance of the main hydrocarbons observed so far in the atmosphere of Saturn. This scheme gives similar results, within the error bars of the model, as a 1D photochemical model using an initial chemical scheme containing 90 compounds and more than 600 reactions.

1. Introduction

One limitation in the development of 2D/3D photochemical models to study the stratospheres of giant planets and Titan is the complexity of the chemical scheme required to study the evolution of the large number of compounds (mainly hydrocarbons) through hundreds of reactions. In 2D/3D modeling, it is not possible for the time being to include large chemical schemes due to computational time limitations. As a consequence, it is important to determine a reduced chemical scheme, which is known to be representative of the main atmospheric chemical processes. The problem is then to create a reduced chemical scheme which would be simple enough and whose results would remain valid. In the following, we present a methodology that fulfills these objectives. We applied it to Saturn.

2. Uncertainty propagation study

The criterion we use to state that a reduced scheme is valid is to ensure that the results of the model are in agreement with a reference model. The reference model is a 1D photochemical model of Saturn that includes uncertainties of rate coefficients (details are given in [1]). The overall precision of photochemical models is highly sensitive to the uncertainties in the rate coefficients used in the chemical scheme. Since the continuity equations are non-linear and strongly coupled, it is necessary to use global sensitivity methods to study how these uncertainties propagate in the photochemical model [2]. The present model is similar to the model used in [3] and [4].

Fig. 1 shows that our model is in good agreement with CIRS observations at the subsolar point. Observational data are within the 1st and 19th 20-quantiles of our model. This shows that chemical processes included in the initial chemical scheme and physical processes implemented in the model are representative of the main processes that govern the composition of Saturn's stratosphere.

3. Reduction methodology

The reduced chemical scheme is built by removing a set of reactions from the initial chemical scheme. We used the methodology presented by [7]. It is based on the computation of Rank Correlation Coefficients (RCCs) which give the strength of the relationship between outputs (mole fractions) and inputs (rate constants) of the photochemical model. Rate constants with low RCCs (in absolute value) have weak influence on the uncertainty of the mole fraction of a given compound. So, below a given threshold (a given value of RCC), all the reactions can be removed.

Our criterion to validate a reduced chemical scheme is the reproduction of a reference model within a certain confidence interval. The profiles of each target

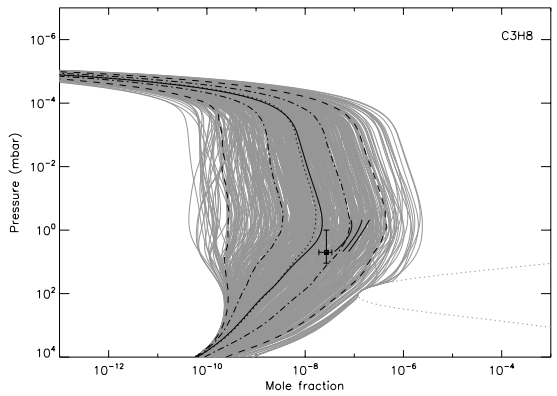

Figure 1: Model. Gray solid lines: Abundance profiles of C_3H_8 obtained after 500 runs. Black solid line: initial profile obtained with the initial chemical scheme. Black dotted line: median profile. Black dashed-dotted lines: 5th and 15th 20-quantiles of the distribution. Black long-dashed lines: 1st and 19th 20-quantiles of the distribution. Gray dotted line: 100% saturation profile. C_3H_8 observations. Bold solid line: CIRS Cassini observations and 1-σ uncertainties at 20° S [6]. Square: IRTF data at -20° planetocentric latitude [5]

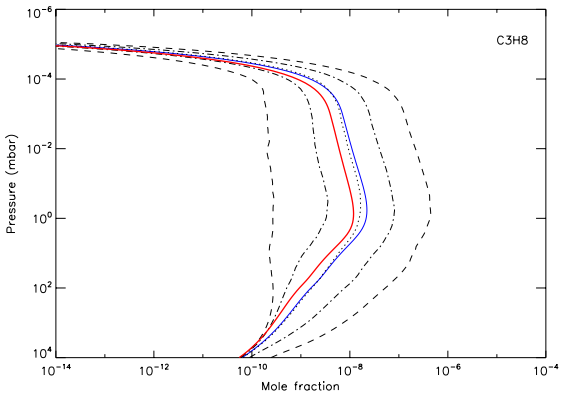

Figure 2: Red line: Abundance profiles of C_3H_8 with the reduced chemical scheme. Blue line: initial profile obtained with the initial chemical scheme. Black dotted line: median profile. Black dashed-dotted lines: 5th and 15th 20-quantiles of the distribution. Black long-dashed lines: 1st and 19th 20-quantiles of the distribution.

molecule should stay within the confidence interval to consider the current reduced scheme as suitable. In the following, the confidence interval is given by the 1st and 19th 20-quantiles in the part of the atmosphere where the compounds are observed.

Following this methodology, we have built a reduced chemical scheme containing only 25 compounds and 46 reactions (including photolysis). This scheme is well adapted to compute the abundance of the main hydrocarbons observed so far in the atmosphere of Saturn. Fig. 2 shows an example for C_3H_8.

Acknowledgements

T. Cavalié wishes to acknowledge for funding from the Centre National d'Études Spatiales (CNES).

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