Atmospheric Photochemistry from a Perspective of Ab Initio Simulations

PETR SLAVÍČEK[1,2]

[1]*Department of Physical Chemistry, Institute of Chemical Technology, Prague, Technická 5, Prague 6, Czech Republic*

[2]*J. Heyrovský Institute of Physical Chemistry, Academy of Sience of the Czech Republic, v.v.i., Prague 8, Czech Republic*

petr.slavicek@vscht.cz

In my talk, I will summarize the work of our laboratory on theoretical modeling of atmospheric photochemical processes. The atmosphere of the Earth can be viewed as a chemical reactor exhibiting a rich chemistry. Many of the chemical processes in the atmosphere are initiated by light. The atmospheric models therefore need data describing the interaction between light and matter (absorption cross sections, quantum yields). While laboratory measurements are able to provide high-quality data, the assistance of theoretical chemistry is often needed. This is the case of unstable radicals or processes taking place on the interfaces as well as in the case of finite size particles

In the first part, I will focus on modeling of UV absorption spectra for floppy molecules using reflection principle method combined with path-integral molecular dynamics or quantum thermostat sampling [1]. The approach will be demonstrated on the atmospherically important nitrate anion on the ice surfaces.

Non-adiabatic photodynamical simulations using techniques of Full Multiple Spawning or Surface Hopping will be demonstrated in a second part of the talk. Special emphasis will be put on the photochemical processes following the excitation of halogen halides on the surfaces of ice particles [2].

In the final part of the talk, I will briefly show how photochemical methods can be used for a characterization of water particles with tens to hundreds of water units, allowing for a direct identification of water nanocrystallization [3].

Acknowledgement: P.S. acknowledges project number 208/10/1724 by the Grant Agency of The Czech Republic and project Barrande 7AMB12FR016 by the Ministry of Education of the Czech Republic.

References

[1] M. Ončák, L. Šištík, P. Slavíček J. Chem. Phys. **133**, 174303 (2010)

[2] M. Ončák, P. Slavíček, M. Fárník, U. Buck J. Phys. Chem. A **115**, 6238 (2011)

[3] C. C. Pradzynski, R. M. Forck, T. Zeuch, P. Slavíček, U. Buck Science **337**, 1529 (2012).