

Heterogeneous CO₂ nucleation in the Martian mesosphere – laboratory experiments

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Abstract

We present measurements on heterogeneous CO₂ nucleation on sub 4 nm radius iron oxide and silicon oxide particles as analogues to naturally occurring Meteoric Smoke Particles (MSP). From the particle growth, we infer the particle surface desorption energy of CO₂. Additionally, evaluation of nucleation rates at controlled particle temperature and CO₂ concentration gives us the contact parameter of CO₂ on these particles, resulting in a full nucleation parameterization for MSPs in the Martian mesosphere. We find strong indications that the same parameterization can be used for upward propagated Martian Dust Particles (MDPs). We apply this nucleation parameterization to conditions of the Martian mesosphere which results in characteristic nucleation onset temperatures. Furthermore, the Sticking Coefficient of CO₂ on solid CO₂ is evaluated.

1. Introduction

CO₂ ice clouds have been detected in the Martian mesosphere [e.g. 1, 9, 10, 11]. These clouds mainly appear during pre- and post- aphelion season and are believed to be caused by heterogeneous nucleation on nanoparticles during supersaturated conditions induced by thermal tides and gravity waves [4, 12]. Although great progress has been made in the last century in monitoring and modeling mesospheric CO₂ clouds on Mars, large uncertainties especially in parameterizing the microphysical formation process of the ice particles remain. Main ice nucleation candidates are Meteoric Smoke Particles (MSPs) or upward propagated Martian Dust Particles (MDPs). Currently a single parameterization [5] is used to describe nucleation on either of these particle types in the mesosphere. It leads to CO₂ nucleation at temperatures only slightly below the saturation

temperature. This result is in disagreement with night time observations of temperatures well below saturation temperature in absence of clouds [3, 8]. One possible explanation for the discrepancy between observation and theory is that the nucleation ability of MSPs and MDPs is actually lower than assumed.

We recently designed a novel experiment which allows us to investigate CO₂ nucleation and growth processes on MSP analogues under controlled conditions being reasonably close to conditions in the Martian mesosphere. This allows us to determine important surface properties of the MSP analogues such as CO₂ desorption energy, sticking coefficient, contact parameter and critical temperature for nucleation as a function of the particle size.

2. Experimental Method

We produce free sub 4 nm radius iron oxide and silicon oxide particles representing MSP analogues with a microwave plasma particle source. These particles are transferred into the vacuum setup TRAPS [7] in which we trap singly charged particles of selected mass in a supersaturation chamber called MICE [2]. In MICE, we can control the particle temperature and ambient CO₂ concentration. Heterogeneous nucleation and subsequent growth of CO₂ ice is observed by extracting a small sample from the trapped particle population at defined time steps and analyzing its mass distribution with a time of flight spectrometer.

3. Results and Discussion

Figure 1 shows an exemplary series of measurements of CO₂ deposition on silicon oxide particles having an initial radius of about 2.5 nm. The measured

particle mass is shown as function of the residence time in MICE. The CO₂ concentration is set to a constant value, in this case to 5E15 m⁻³.

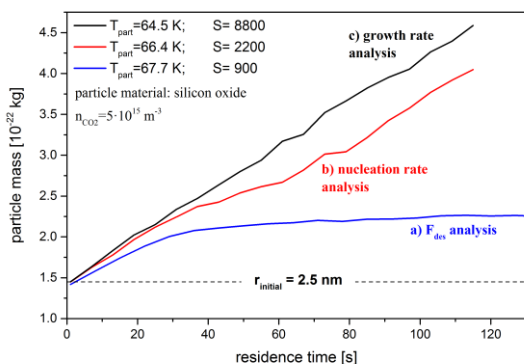


Figure 1: exemplary dataset with 2.5 nm silicon oxide particles used for desorption energy (a), nucleation rate (b), and growth rate (c) analysis.

The particle temperature and therefore saturation is varied and allows us to adjust conditions with and without nucleation taking place. The blue line (a) represents a condition at which no nucleation took place. The temporary increase of particle mass represents an adsorption process of CO₂ molecules on the particles surface allowing us to determine the desorption energy of CO₂ molecules on the investigated particle material. The red line (b) represents conditions which allow us to evaluate nucleation rates and determine the contact parameter m . Finally, at high saturations as represented by the black line (c) the growth rate can be evaluated to retrieve the sticking coefficient α of CO₂ on solid CO₂. The evaluation of such measurements allow us to determine a full parameterization of nucleation and growth of CO₂ on MSPs at conditions being reasonably close to conditions in the Martian mesosphere. We will present this new parameterization and show that it probably can be used for MDPs as well. We will discuss the results and highlight the consequences when applying this parameterization to the Martian mesosphere resulting in defined temperatures at which nucleation gets induced.

Acknowledgements

We gratefully acknowledge financial support by the German Federal Ministry of Education and Research (BMBF) under grant number 05K13VH3 and would like to thank the EU QualityNano Research

Infrastructure Program for funding a transnational access research campaign.

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