ICE NUCLEATION IN THE MARTIAN ATMOSPHERE.

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Introduction

In the Martian atmosphere dust and other types of aerosol particles (most importantly carbon dioxide and water ices) scatter and absorb both short- and longwave radiation. This affects the thermal structure of the atmosphere and flow fields therein. Flow patterns affect the aerosol particle distribution by lifting dust from the surface and advecting dust and ice particles in the atmosphere, which again modifies its thermal and flow structure. This feedback between aerosols and flow patterns may, for example, have a great influence in giving rise to global (and also local) dust storms. For understanding this feedback, both the boundary layer processes (*Savijärvi*, 1999; *Savijärvi et al.*, 2004) being involved in lifting and advecting dust particles and creating atmospheric circumstances favorable to the onset of nucleation, and the aerosol processes themselves (*Noppel et al.*, 2002; *Korhonen et al.*, 2004; *Määttänen et al.*, 2005) should be studied in detail.

On one hand, Martian aerosol processes are simpler than the ones in the atmosphere of Earth since there are practically no organic vapours (altough Mars Express has detected signs of methane) or anthropogenic emissions influencing the aerosol processes. On the other hand, the Martian atmosphere is nearly pure CO_2 vapour, which takes part in the processes of nucleation and condensation. The results of these processes are largely controlled by the amount of the participating vapour. In a near-pure situation, nucleation becomes nonisothermal and strong coupling of fluxes arises in multicomponent condensation. These effects need to be taken into account in the theories used.

The ultimate goal of developing an aerosol dynamical model for Mars is to apply it with an atmospheric model to enhance the model's ability to include the effect of aerosols. One development at hand is also the possibility to enhance the accuracy of simple atmospheric condensation schemes, which normally use a certain treshold saturation ratio, after which all the excess vapour is condensated. More accurate nucleation models can calculate the treshold saturation ratio, which then can be used in atmospheric models with a more simple approach.

The ongoing binary nucleation studies will tell about the coupling or the two nucleating species in the Martian atmosphere, and will give hints about the importance of coupling in condensation.

One-component nucleation

We studied homogeneous and heterogeneous nucleation of carbon dioxide based on the work by *Wood* (1999), and water nucleation following the same schemes. We have compared our results on CO_2 nucleation to the laboratory measurements of *Glandorf et al.* (2002). The H_2O nucleation model was compared to the calculations made by *Inada* (2002). Model simulations have been conducted in average atmospheric near-surface conditions for 95.3% CO_2 atmosphere. Water vapour concentration was 300 ppm, and the inert gas consisted of N_2 (2.7%), Ar (1.6%) and O_2 (0.13%) together with either H_2O or CO_2, depending on which one was not nucleating. Temperature range in the simulations was 100-300 K and the surface number concentration of the condensation nuclei (dust particles) was defined as $N_{CN} = \tau/(Q\pi H) \cdot (1/r_{CN}^2)$ where τ is the dust optical depth, Q the Mie extinction coefficient, H scale height and r_{CN} the radius of the dust particles. The dust profile was calculated as in *Conrath* (1975) for a very low dust loading of the atmosphere (τ=0.1). In our basic cases the r_{CN} was 1.0 μm, but simulations have been conducted also with varying r_{CN}. The variation in r_{CN} was calculated as a linear decrease from the surface value with a coefficient given by model simulations with the dust deposition model of *Merikallio* (2003).

For both substances homogeneous nucleation is very unlikely to happen in the Martian atmosphere because of the very high supersaturation required (saturation ratios of the order of 10^8), but instead heterogeneous nucleation on dust particles seems probable. The onset saturation ratio (the saturation ratio for nucleation rate of 1 s^{-1}) of heterogeneous nucleation for CO_2 on dust particles of 1.0 μm radius is 1.32. This is in fair agreement with measured values (*Glandorf et al.*, 2002). For constant concentration of CO_2 it means that heterogeneous nucleation starts when temperature drops below 145 K. For water with average concentration of 300 ppm the onset saturation ratio would be 1.18 and the corresponding temperature 200 K.

Nucleation profile simulations

We modelled nucleation as a function of height at different locations, e.g. Mars Pathfinder landing site, and compared results with observations. We conducted the nucleation experiments using atmospheric profiles aquired from modelling and observations. In some cases (The

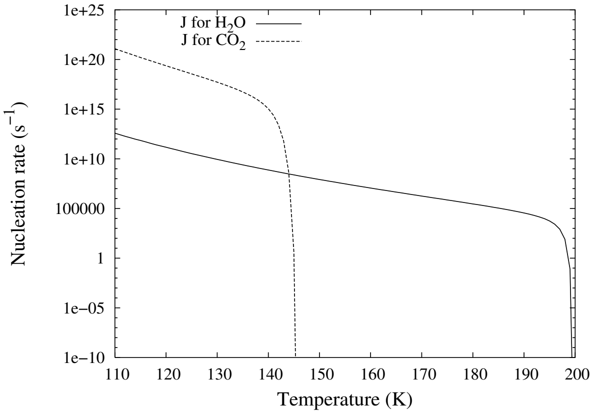

Figure 1: Heterogeneous nucleation rates for H_2O and CO_2 in the Martian atmosphere.

Mars Pathfinder landing site and the Memnonia region) the 1-dimensional (1D) model of the University of Helsinki (*Savijärvi*, 1999; *Savijärvi et al.*, 2004) was initialized with temperature profiles obtained from the Mars Climate Database (MCD, *Lewis et al.*, 1999) to produce the needed moist profiles (since the MCD does not include water). We also used the unmodified landing profile of the Mars Pathfinder (personal communication, Dr. Anthony Colaprete) to compare with previous work by *Colaprete et al.* (1999). Our modelling results are in fair agreement with observations and previous modelling studies.

At the landing site of Mars Pathfinder the nucleation model predicted H_2O nucleation in a slightly thinner (40 m) layer than the 1D model (80 m). This is related to the higher treshold saturation ratio calulated by the nucleation model (1.18 vs. 1.0), and thus nucleation only happens closer to the surface where temperatures are cold enough for the critical saturation ratio to be reached. The model results are anyhow in good agreement with eachother and the observations by Mars Pathfinder (*Schofield et al.*, 1997).

In the Memnonia region, where surface fog was observed by the Viking Orbiter 1 in late 70's (*Briggs et al.*, 1977), the model shows clear H_2O nucleation throughout the night reaching up to the height of 100 m. The result is agreeing with the model of *Inada* (2002), who predicted an 800 m thick saturated layer. Since the critical saturation ratio for nucleation in our model is 1.18, we see nucleation only in the 100 m layer close to the surface.

The nucleation model predicted nucleation also higher in the atmosphere, when using the unmodified landing profile of Mars Pathfinder (which does not reach below 8 km). Our nucleation results agree well with the cloud model results of *Colaprete et al.* (1999) at 55 km and below.

Current status: two-component nucleation

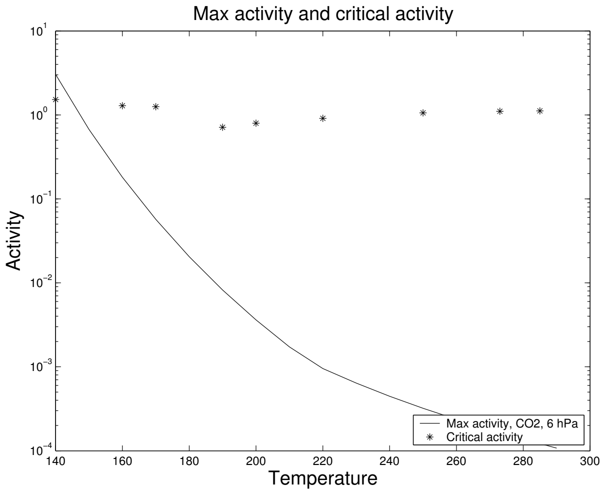

Figure 2: The maximum achievable activity of CO_2 and the onset activity required for binary nucleation at 6 hPa.

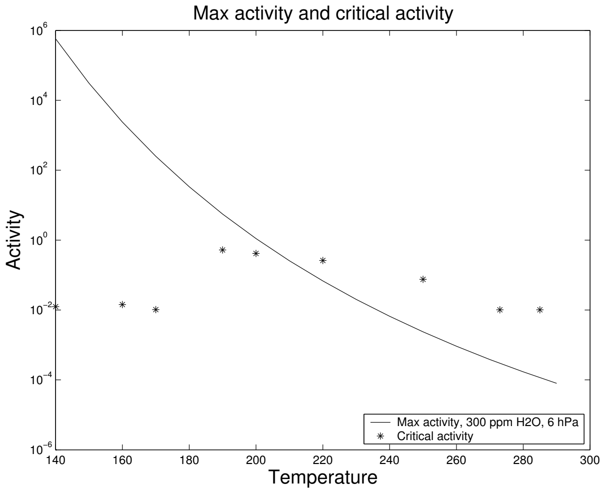

Figure 3: The maximum achievable activity of CO_2 and the onset activity required for binary nucleation at 6 hPa.

We have studied the two-component (binary) nucleation of CO_2 and H_2O. We assumed an ideal mixture of CO_2 and H_2O as a first approximation but tested also two non-ideal parametrisations. The original model (*Noppel et al.*, 2002) did not include heterogeneous nucleation which was thus added. The theoretical possibilities of nucleation of the binary system in the Martian conditions have been studied.

For the binary system, we have to consider the thermodynamic variable *gas phase activity*. In a binary system, the saturation ratio measures how saturated a vapour (one of the components) is with respect to the *mixture* surface. The *gas phase activity* tells how saturated a vapour in the system is with respect to the *pure* substance. This activity is the one used in the Figures 2 and 3.

Figure 2 and Fig. 3 illustrate the possibility of the process in the Martian atmosphere: the solid line is the maximum achievable activity and the stars the required onset activities for binary nucleation. For onset to happen, the star for one temperature needs to be below the line in both plots (so for both substances). The plots show, that theoretically at 140 K in about the average surface pressure of Mars binary nucleation of CO_2 and H_2O is possible, but as we know, reaching that temperature is not. This temperature can not be achieved, because the components start nucleating one by one already in warmer temperatures, and thermodynamically the temperature can not go below the CO_2 condensation temperature. Higher in the atmosphere even 140 K is not enough, since the needed activities can not be achieved because of the thinness of the gas (basically the solid line would fall towards lower values, but the stars would stay at about the same values).

According to our models, binary homogeneous nucleation does not happen in the Martian atmosphere, and is not extensively dealt with here.

With this theoretical testing and analysis, we can conclude that binary nucleation, homogeneous or heterogenous, does not happen in the Martian atmosphere. The results will be published soon.

Future work

In the future also a condensation model taking into account the coupling of species (*Vesala et al.*, 1997) will be transformed into Martian conditions. Deposition and coagulation will be treated when a complete aerosol model will be modified into Martian conditions using the new nucleation and condensation schemes. Eventually the aerosol model or a lighter parametrisation will be implemented into the 1D and 2D Mars atmospheric models of the University of Helsinki (*Savijärvi and Siili*, 1993; *Savijärvi et al.*, 2004), and possibly into the 3D mesoscale model MLAM. Thus the aerosol model will be improving the atmospheric models, for example, by describing more precisely cloud formation and aerosol distribution which are closely connected to the radiative transfer of the model.

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