Parameterization of homogeneous nucleation within the GEM model

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Introduction

Ice nucleation can be separated within four categories: heterogeneous deposition, homogeneous deposition, heterogeneous freezing and homogeneous freezing. Deposition processes are phase changes from vapor to ice while freezing is for changes from liquid to ice. Heterogeneous nucleation processes describe phase changes with the help of an aerosol while homogeneous mechanisms are phase changes with only pure water or a solution. Our interest lies in the homogeneous processes and more precisely in the homogeneous freezing since homogeneous deposition is impossible under normal atmospheric conditions.

The Arctic is very interesting to study for ice nucleation since the temperatures reach below -38 ˚C, which is necessary for homogeneous freezing, and also because pollution will affect the composition of the aerosols in this region which will modify the nucleation processes like homogeneous freezing. During winter, the Arctic is polluted by aerosols coated with sulfuric acid. Meanwhile, when the region is less or not polluted, the aerosols are principally covered by ammonium sulfate.

Objectives

Simulations with GEM have been resulting in highly overestimated crystal concentration values in the pressure levels where the temperature is below -38 ˚C meaning that the results from the model become poor starting from when homogeneous freezing is activated. This might be caused by a problem in the simulation of homogeneous freezing which leads to our goal: testing new parameterizations for homogeneous freezing in the GEM model to improve the simulation of this process and to take into account the chemical composition of non activated solution droplets.

Model[1]

The model that will be used in this study will be the Global Environmental Multiscale (GEM) model. The physics package is composed of a radiation scheme (Li and Barker, 2005) and a land-surface scheme known as Interactions Soil-Biosphere-Atmosphere (Noilhan and Planton, 1989). The part of the model which interests us is the microphysics scheme used which is Milbrandt and Yau (2005) for GEM.

This is a two-moment microphysics scheme which divides the hydrometeors into six classes: cloud droplets, cloud ice crystals, raindrop, snow, graupel and hail. The parameterizations used to simulate the microphysical processes can be found in Milbrandt and Yau (2005). This scheme uses different parameterizations for the four types of heterogeneous freezing (deposition, condensation, contact and immersion) and homogeneous freezing. The latter, which is the process that we want to obtain better results for, currently uses the parameterizations proposed by DeMott et al. (1994).

Method[1]

- Integration domain: Centered over the Arctic and will be a rectangle covering 9100 km by 7700 km. This domain will cover all areas beyond 50˚N including the North Atlantic Ocean, the Pacific Ocean, the Arctic Ocean, Northern Canada, Greenland, Siberia, Northern Asia, Iceland and most of Europe.

- Horizontal resolution: 0.25˚ with 364 latitudinal grid points and 308 meridional grid points which includes a 12-point sponge zone. To eliminate the latter, results will be analyzed on a sub-domain of 8500 km by 7100 km.

- Vertical levels: 53 ranging from 1000 to 10 hPa with a higher resolution in the lower levels.

- Boundary: The initial and lateral boundary conditions will be obtained from analysis data of the European Centre Meteorological Weather Forecast (EC-MWF). The analyzed fields are available on 18 pressure levels on a longitude/latitude grid with a 2º by 2º spatial resolution at every 6h for December 2006 and January 2007.

- Sea: Sea ice and sea suface temperature are from AMIP2 (Atmospheric Model Intercomparison Project). They are available on a grid at a horizontal resolution of 1º by 1º.

Acknowledgements

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Parameterizations

After reviewing many articles, a few ways to modify the parameterization of homogeneous ice nucleation caught our interest. We will test the following ones within the GEM model to see if they improve the results of the simulations when we compare them to in situ observations.

- **Water activity[4,5]**:

The idea behind using water activity, which depends on the temperature, the concentration and the pressure of the solution, as the determinant for homogeneous ice nucleation in solutions is that the freezing temperatures for any type of solution fit on a curve, as we can see in figure 1. Also, the water-activity criterion (a_w-a_w^i) can be used to parametrize the homogeneous nucleation rate coefficient (J) which is needed to calculate the nucleation rate. Water activity under atmospheric conditions can be expressed in terms of relative humidity which means that we can calculate saturation values and compare them with observations. For example, figure 2 shows the ice saturation ratio as a function of temperature for different aerosol radius for a nucleation rate of 1 min^{-1}.

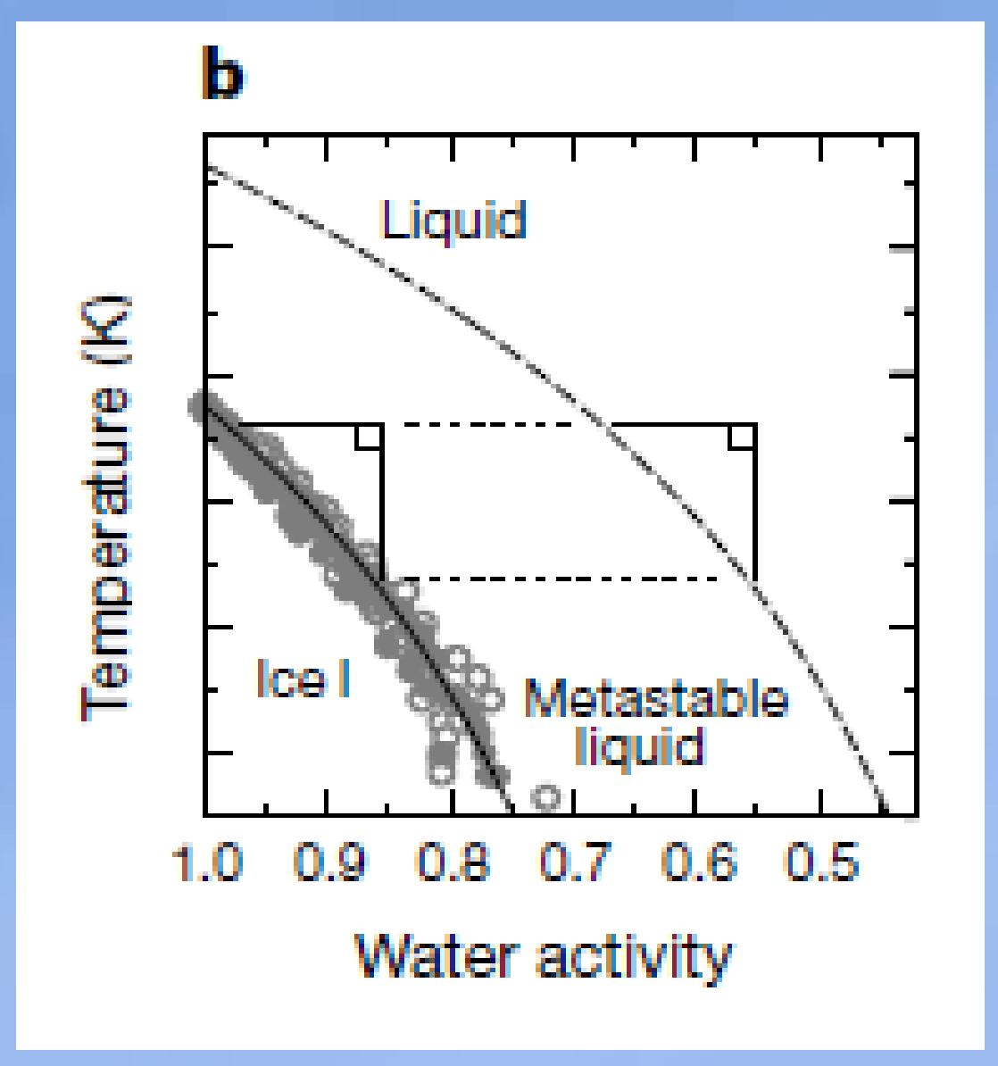

Figure 1[4]

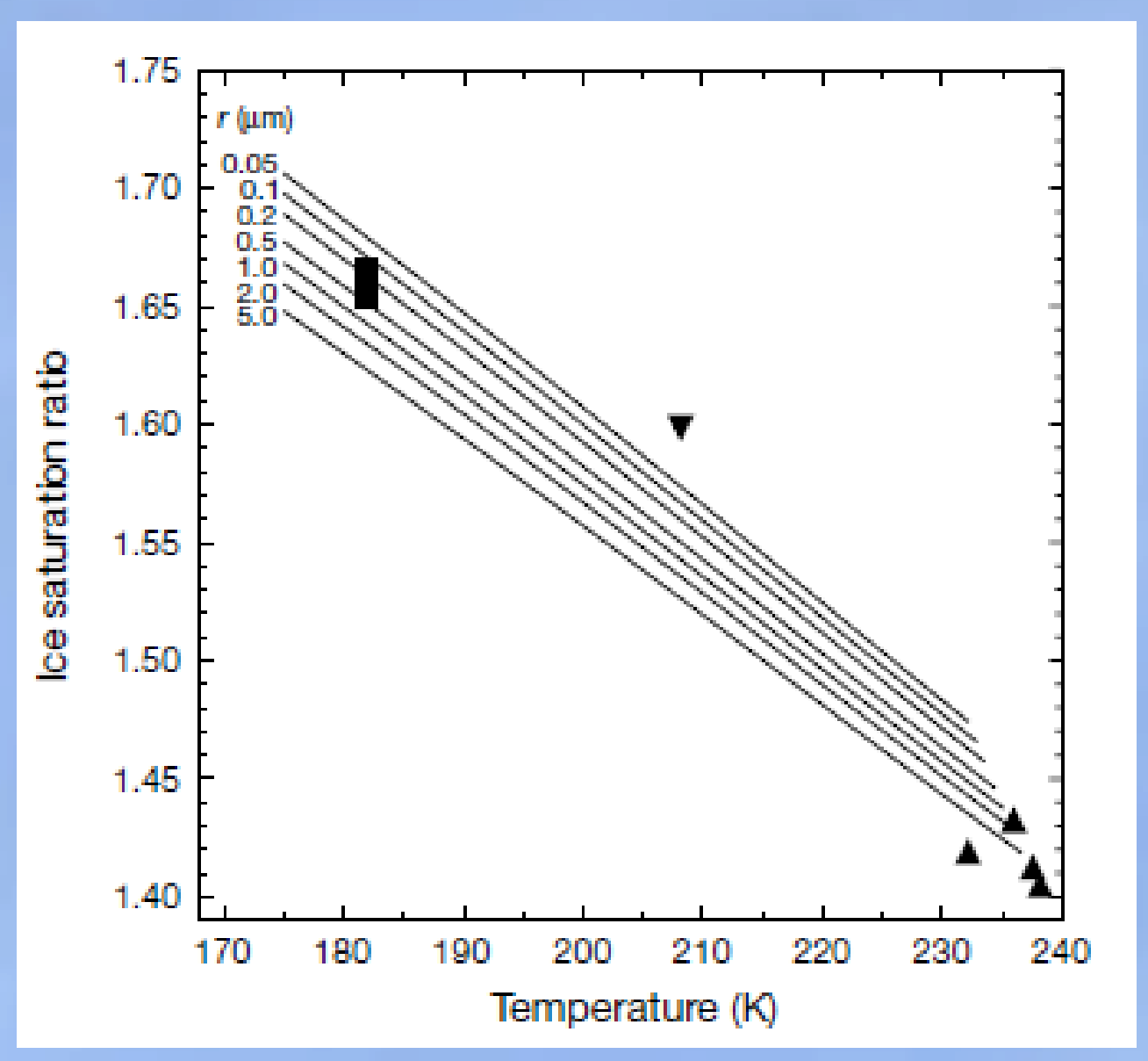

Figure 2[4]

However, this theory is not in agreement with other studies. For example, the results of Swanson (2008) show a large difference for the H_2SO_4, as seen in figure 3, compared to the curve shown by Koop et al. (2000) which means that a single $T_f(a_w)$ curve shouldn't be sufficient to describe every aqueous solution. This doubt is further strengthened by the fact that Swanson is able to obtain ice saturation ratios similar to observations with his results while Koops are too low in the case of sulfuric acid, as we can see in figure 4.

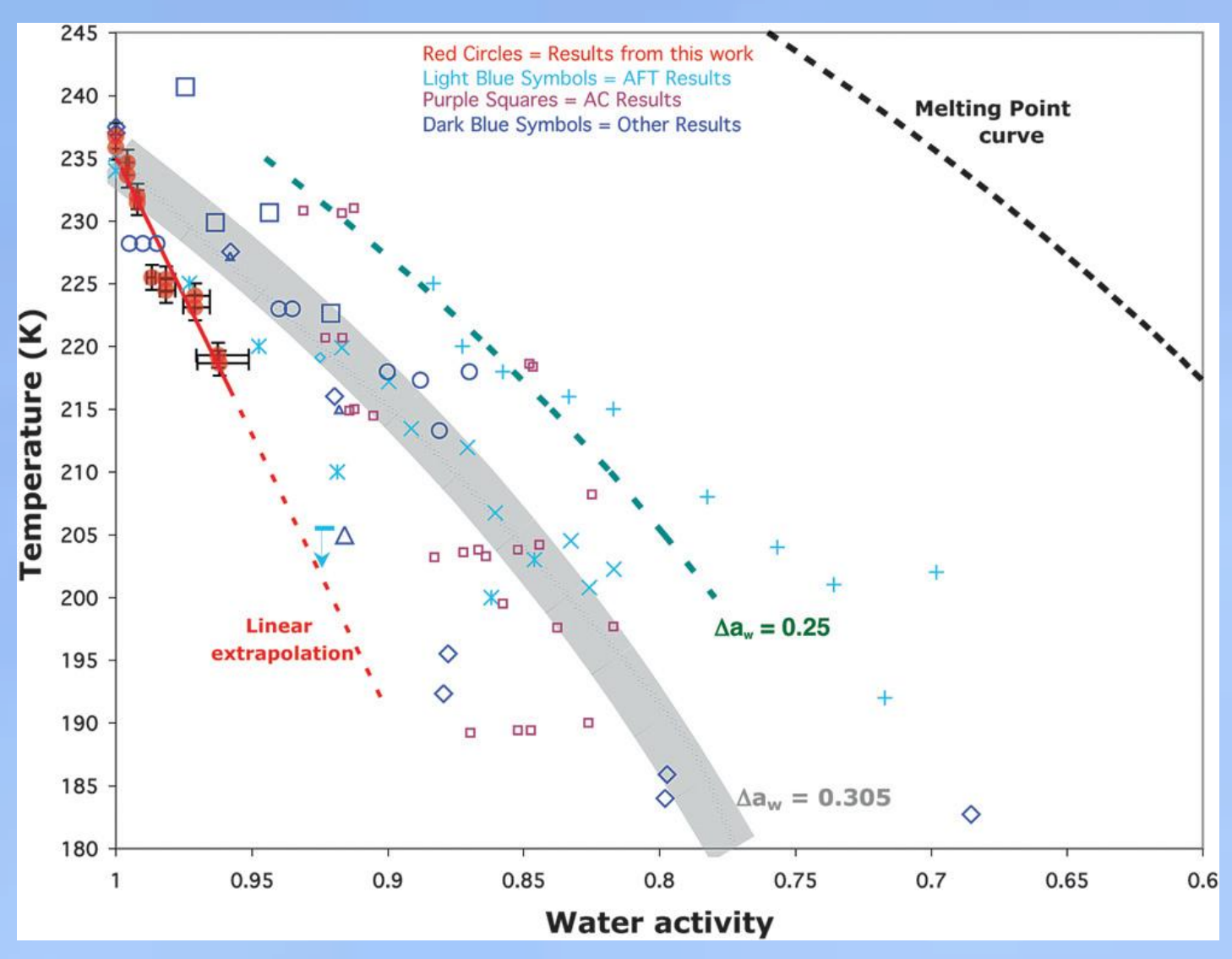

Figure 3[5]

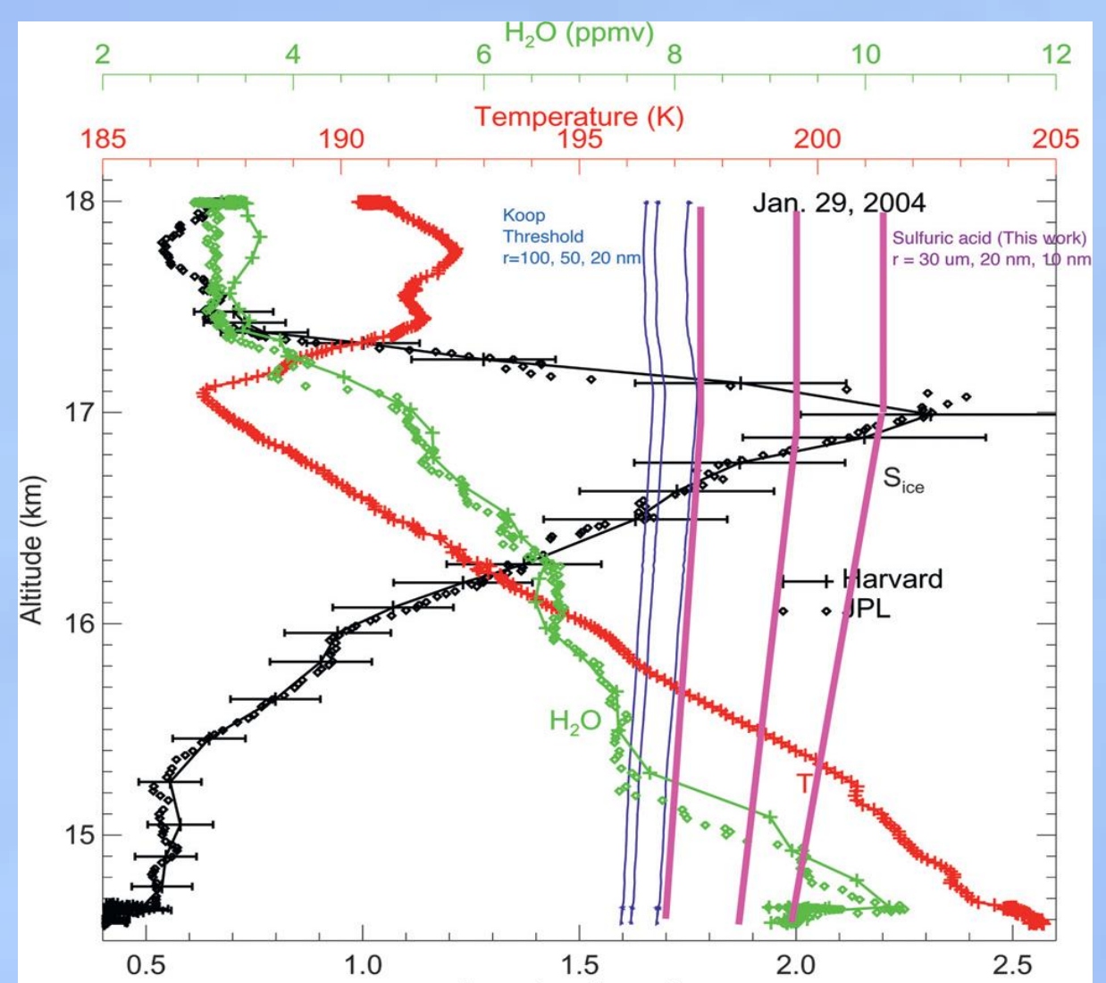

Figure 4[5]

- **Coating of an ice crystal[3]**:

Bogdan and Molina (2009) also proposed an interesting theory that could result in a slowing of 10^3 on the growth of ice crystals compared to uncoated ice. This would be due to aqueous droplets becoming mixed-phase particles made of a pure ice core with a residual solution coating. This could explain the fact that relative humidity with respect to ice has been observed to be much higher then 100% for extended periods, even within cirrus ice clouds. The slower ice growth could result in greenhouse warming due to the longer lifetime of water vapor in the upper troposphere and could also change the radiative properties of cold cirrus.

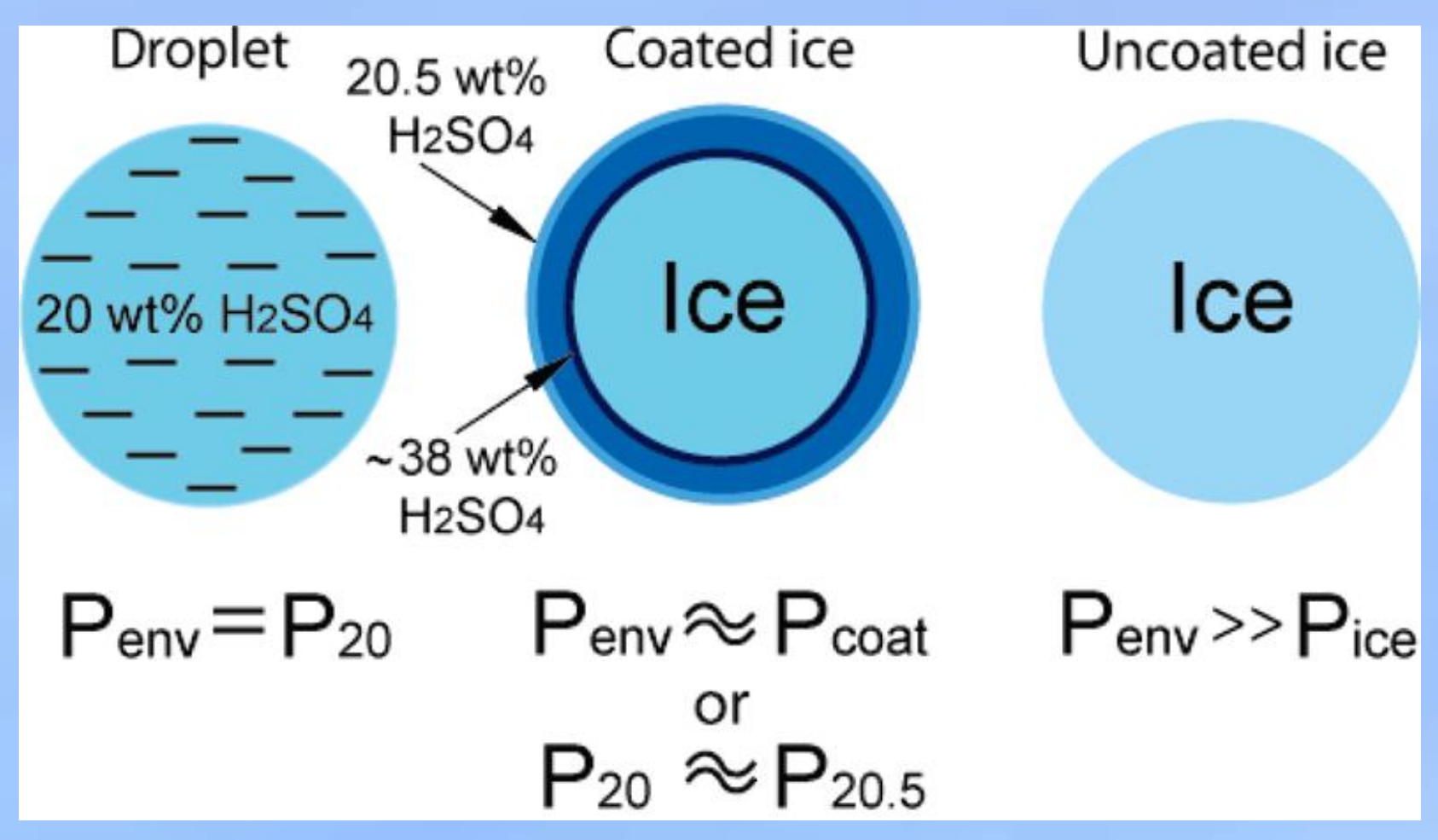

Figure 5[3]

Conclusion

This study will help us understand the impact of sulfuric acid and ammonium sulfate coated aerosols for homogeneous freezing which is essential to improve cloud microphysics simulations. It will also allow us to perform better simulations for the aerosols-cloud interaction. For example, further knowledge regarding this type of nucleation could confirm hypotheses such as the one presented in Girard et al. (2014) which means that we could associate the formation of TIC-2B clouds from the ISDAC mission to the acid coating of the aerosols.

References

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