Possible Impact of Homogeneous Freezing Nucleation on in Situ Measurements

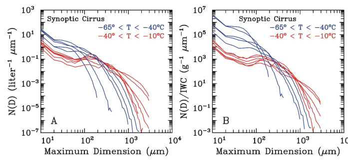
Research Highlight

For a given aerosol loading, the aerosol indirect effect for cirrus clouds may have the greatest radiative impact at the coldest cirrus temperatures, where their greenhouse effect is greatest. Thus it is critical to understand the relative contributions of homogeneous and heterogeneous nucleation processes at temperatures below -40°C. That is, the higher nucleation rates from homogeneous freezing could decrease ice particle size and fall speed, and for a given ice water content (IWC), increase ice crystal concentration N and cloud optical depth, relative to heterogeneous nucleation processes. The lower fall speeds increase the cloud IWC and lifetime, with cloud optical depth increasing due to smaller particle size and greater IWC. For semi-transparent to moderately thick cirrus, cloud emissivity increases more rapidly with optical depth than cloud albedo such that the above changes produce a net warming. Since only a few global climate models (GCMs) (e.g., CESM1 and ECHAM5) carry supersaturation with respect to ice, homogeneous nucleation is generally not treated in GCM simulations. Thus the global indirect effect for cirrus clouds is essentially unknown at this time, although preliminary CESM1 simulations indicate it is substantial.

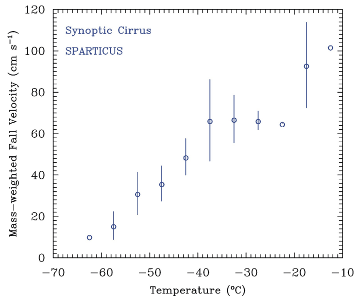
Ice particle shattering at sampling probe inlets has often produced ice artifact concentrations ~ 102 times greater relative to N measured by recently developed probes designed to minimize shattering. Thus historical measurements of the ice particle size distribution (PSD) and N in particular were often masked by shattering artifacts, making it impossible to measure nucleation effects on the ice PSD. The TC4 and SPARTICUS field campaigns in 2007 and 2010, respectively, used the 2D-Sterio probe and ice particle interarrival time data processing software that were both designed to minimize shattering. TC4 (sponsored by NASA) sampled mostly maritime anvil cirrus clouds off the coast of Costa Rica while SPARTICUS (funded by the Atmospheric Radiation Measurement Climate Research Facility) sampled midlatitude cirrus clouds. No evidence of liquid water was found during the periods sampled.

The PSD analysis from these two field campaigns produced the following findings for fresh anvil and synoptic cirrus clouds: a relatively abrupt change near -40°C in PSD shape, N/IWC ratio, and mean size. For the synoptic cirrus, an abrupt change in ice particle shape and fall speed near -40°C was also found. These changes were attributed to the onset of homogeneous nucleation which occurs near -38°C, which predicts higher nucleation rates, lower mean sizes and fall speeds, and possibly more isometric ice crystal growth at colder temperatures for non-anvil cirrus (as was observed here). The nucleation rate coefficient J, having units of number of ice germs formed per unit volume (i.e. mass) of condensate per unit time, is related to the N/IWC ratio, which can be viewed as the time integration of J in a cloud parcel from nucleation onset to sampling time. However, this is oversimplified as it ignores aggregation and ice fallout. Ice PSDs were approximately unimodal for T $<$ -40°C and bimodal for T $>$ -40°C. This might be explained through two approximate equilibrium conditions regarding ice production and removal rates through nucleation and aggregation, respectively, separated at -40°C. Some of these results are shown in Figures 1 and 2.

The results for synoptic cirrus suggest a cirrus formation mechanism whereby once the relative humidity (RHi) threshold for homogeneous nucleation is attained, vigorous ice crystal production ensues and the RHi levels rapidly drop to near ice saturation due to intense competition among ice crystals for water vapor. Such growth conditions produce near isometric ice crystal shapes (i.e., short columns, thick plates, and compact, high density crystals), consistent with our observations.



A: Temperature dependence of the synoptic PSD during SPARTICUS, with PSD averaged over five-degree C temperature intervals. PSD are color-coded by temperature regime. B: The same PSD as in panel A, but divided by their corresponding IWC to express the impact of ice nucleation rates on the PSDs.



Temperature dependence of Vm corresponding to the mean PSDs. Vertical bars indicate standard deviations. Data points without bars are based on fewer than three PSD samples.

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In conclusion, we feel these results could play a pivotal role in developing and testing new parameterizations of ice nucleation in climate models, noting that 174 PSDs were analyzed for SPARTICUS between -10°C and -65°C. The abrupt change in ice particle shape for synoptic cirrus will have a strong impact on cloud optical properties and fall speeds, with both strongly affecting cloud radiative forcing. However, size changes appear to trump ice particle shape changes, as is evident for the ice fall speeds. An unresolved issue is whether ice nucleation for T $<$ -40°C in fresh maritime anvils and synoptic cirrus in the northern hemisphere is primarily due to homogeneous nucleation or whether heterogeneous nucleation processes, although likely secondary, are still contributing significantly to ice production. The answer to this question will impact the water vapor concentrations in the upper troposphere, and important climate feedback.

Reference(s)

Mitchell DL, S Mishra, and RP Lawson. 2011. Cirrus Clouds and Climate Engineering: New Findings on Ice Nucleation and Theoretical Basis. In Planet Earth 2011 - Global Warming Challenges and Opportunities for Policy and Practice, Ed. by Elias G. Carayannis, InTech.

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Working Group(s)

Cloud Life Cycle, Cloud-Aerosol-Precipitation Interactions