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Heterogeneous ice nucleation: Bridging stochastic and singular freezing behavior

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Much of the dispersed water in atmospheric clouds is in a metastable, supercooled state, and often freezing is stimulated by relatively rare aerosol particles known as heterogeneous ice nuclei. Heterogeneous ice nucleation directly influences cloud physical processes, precipitation formation, global radiation balances, and therefore Earth's climate (Cantrell and Heymsfield, 2005; Pruppacher and Klett, 1997). It is important to understand the heterogeneous freezing process at a fundamental level in order to describe this process in a physically-based way that will behave robustly in weather and climate models.

For several decades there has been conflicting evidence as to whether heterogeneous ice nucleation is a stochastic process (i.e. ice nucleation is time-dependent) or whether nucleation takes place on specific particle surface sites, so-called active sites, at characteristic (i.e. deterministic) freezing temperatures, known as the singular hypothesis (i.e. freezing is assumed to be time-independent). The apparent conflict between these descriptions of nucleation is drawn into focus by considering results from two ice nucleation experiments conducted within our research groups. The experiments are distinguished by the two approaches to forming a statistical ensemble of freezing temperatures: by repeating a measurement of one system many times (freezing of a droplet containing a single volcanic ash particle) versus measuring many similar systems independently (freezing of a large number of water droplets each containing a size-selected Arizona Test Dust particle). The single-particle ensemble exhibits clear stochastic behavior, while the multi-particle ensemble apparently exhibits singular behavior.

To reconcile the seeming contradiction and more generally to better understand the competing ideas and the somewhat bewildering range of interpretations and applications of stochastic and singular ice nucleation we have developed a model that treats statistically similar particles as being covered with surface sites characterized by different nucleation barriers, but with each surface site following the stochastic nature of ice embryo growth. To do so we consider a large number of spherical 'ice nucleus' particles of identical size, each particle immersed in a water droplet, and where the properties (e.g., interfacial free energy) of individual particles are not necessarily identical, but are selected from a common probability distribution.

With this so-called 'soccer-ball' model we can phenomenologically explain the seemingly contradictory experimental results. For the multi-particle ensemble the variability of the surface properties across the population of Arizona Test Dust particles is most plausibly responsible for the broad temperature range over which droplets freeze and for the apparent missing time dependence for freezing. Since in the single-particle ensemble a single particle was used repeatedly, the variability of the surface properties is eliminated so that the results reflect only the purely stochastic freezing nature.

The central conclusion of this work is that the emergence of singular or nearly singular behavior can be explained without appeal to active sites possessing characteristic freezing temperatures. Rather, an idealized population of statistically similar but individually different particles, characterized by a relatively wide distribution of interfacial free energies, and subject to purely stochastic freezing behavior, can manifest seemingly singular behavior: weak time dependence of freezing probability, and wide freezing temperature distributions. If this general approach survives subsequent experimental scrutiny, it may provide a useful means for representing ice nucleation in computational models.

References:

Cantrell, W. and Heymsfield, A.: Production of ice in tropospheric clouds – A review, B. Am. Meteorol. Soc., 6, 795–807, 2005.

Pruppacher, H. R. and Klett, J. D.: Microphysics of Clouds and Precipitation, Kluwer Academic Publishers, Dordrecht, The Netherlands, 1997.