Review of “Using freezing spectra characteristics to identify ice nucleating particle populations during the winter in the alps” by Creamean et al. (2019)

The work by Creamean at el. analyzes the ice nucleating particle (INP) activity within samples of aerosol, rime, and snow collected at the High Altitude Research Station in Jungfraujoch. An interesting and key finding of the work is that samples originating from the southeast of the sampling location contained appreciable INP activity at temperatures above -15 °C. The authors refer to this as a “warm mode” and quantify it with the differential INP function, $k(T)$, as well as the derivative of the frozen fraction with respect to temperature, df/dT. In a previous review of this paper, Gabor Vali made a convincing argument that the df/dT metric misrepresents the existence of a bi-modality in the data. I echo this argument and will try to use a synthetic INP case to illustrate why the authors' use of df/dT is problematic.

For my example I will consider a population of V = 1 μL droplets containing 1 μm desert dust particles having the INP activities measured by Niemand et al. (2012). I will also assume 10% of the particles contain Snomax, the INP activity of which can be quantified using the empirical relation from Wex et al. (2015). The left hand panel of Fig. 1 shows the resultant frozen fraction as a function of temperature with 10% of droplets freezing at high temperatures due to the presence of Snomax and the rest of the droplets freezing at colder temperatures following the INP activity of dust. I note that this example has been synthesized to have a warm mode. Now if we analyze $V * k(T)$ and df/dT (the right hand panel of Fig. 1), we can see that at warm temperatures the two metrics are virtually equivalent but diverge at lower temperatures. We can understand why this is the case by investigating the relationship of f to the cumulative INP function, $K(T)$ and $k(T)$ from the original equation by Vali (1971):

$$f = 1 - \exp(-K(T)V) \quad (1)$$

Differentiating f with respect to T we obtain:

$$\frac{df}{dT} = Vk(T)\exp(-K(T)V) \quad (2)$$

Equation 2 implies that when $f \approx 0$:

$$\frac{df}{dT} \approx Vk(T) \qquad (3)$$

However, when $f \approx 1$:

$$\frac{df}{dT} \approx 0 \qquad (4)$$

(4) must be satisfied for exactly the reason cited by Gabor Vali in his review: "Since fewer and fewer drops are left as the sample drops freeze and $f(T)$ approaches unity at the lowest temperatures in an experiment, the increase in $f(T)$ per temperature interval, $[df(T)/dT]$, can be expected to drop off, just as it seen in these figures."

Based on the argument made previously by Vali and reiterated here, the statement made by the authors on page 8 line 63 is misleading as it is attributing what is analogous to the cold temperature peak of the blue line in the right hand plot of Fig. 1 to a "cold mode". This is simply not the case. However, coincidently, the attribution of the warm peak to the warm mode is valid but only because of the approximation in Eqn. (3).

In their revision, the authors have added the $k(T)$ analysis which is consistent with previous work and is better equipped to analyze the INP activity of the collected samples. $k(T)$ exhibits the warm mode signature the authors are valuably reporting. I therefore urge the authors to remove the df/dT analysis as it would avert any future confusion on how to properly diagnose freezing spectra while preserving the key findings of the paper.

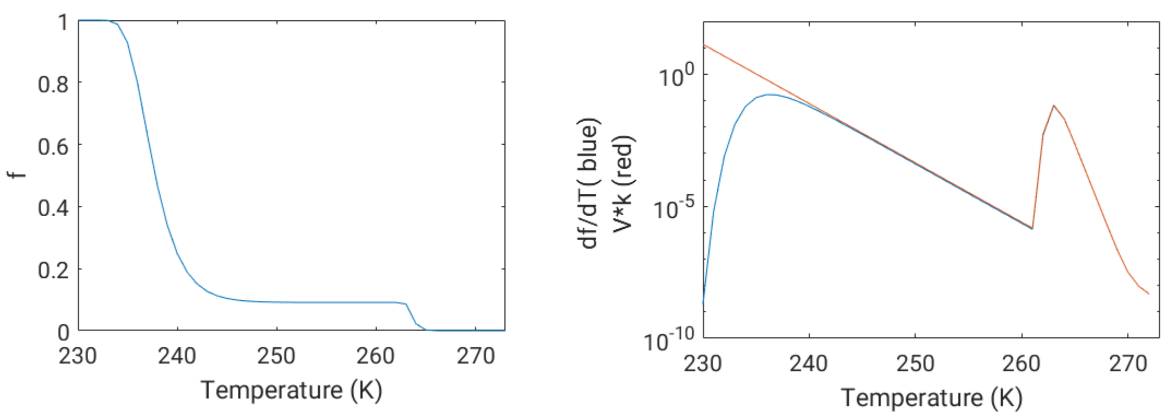

Figure 1. Left: Frozen fraction vs. temperature of a synthetic data set comprised of a population of droplets containing a mixture of dust and biological particles. Right: df/dT (blue) and $k(T)$ of the INP mixture.

References

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., … Leisner, T. (2012). A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles. *Journal of the Atmospheric Sciences*, *69*, 3077–3092. https://doi.org/org/10.1175/JAS-D-11-0249.1

Vali, G. (1971). Quantitative Evaluation of Experimental Results an the Heterogeneous Freezing Nucleation of Supercooled Liquids. *Journal of the Atmospheric Sciences*. https://doi.org/10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2

Wex, H., Augustin-Bauditz, S., Boose, Y., Budke, C., Curtius, J., Diehl, K., … Stratmann, F. (2015). Intercomparing different devices for the investigation of ice nucleating particles using Snomax® as test substance. *Atmospheric Chemistry and Physics*, *15*(3), 1463–1485. https://doi.org/10.5194/acp-15-1463-2015