Atmos. Meas. Tech. Discuss.,
doi:10.5194/amt-2018-134-AC4, 2018

Atmospheric Measurement Techniques
Discussions

Interactive comment on "Cleaning up our water: reducing interferences from non-homogeneous freezing of "pure" water in droplet freezing assays of ice nucleating particles" *by* Michael Polen et al.

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Received and published: 27 August 2018

We thank Dr. Vali for his extensive and highly insightful comments. The k(T) derivative method of analyzing droplet freezing spectra is certainly a valuable approach that is not typically used by the ice nucleation community. Making the community aware of this alternative analysis approach first introduced by Vali (1971) is a worthwhile endeavor and thus we have included a discussion of this in the revised manuscript. We note there are two small typos in the original comment provided by Gabor Vali that we would like to correct here for clarity: Eq. 1 that is referred to from Vali (1971) is actually Eq. 11, and ΔT was accidentally used instead of ΔN in the natural logarithm term. Here is the

original Eq. 11 from Vali (1971), using T instead of theta for temperature:

k(T) = -1/(Vd*ΔT)*ln[1-ΔN/N(T)]

where T is temperature, V_d is the droplet volume, ΔN is the number of drops that froze during the temperature interval ΔT, and N(T) is the number of drops still unfrozen at temperature T.

We have engaged in a series of discussions with Gabor Vali regarding the use of k(T) to analyze our droplet freezing spectra. Through this discussion Gabor realized there are some important but not obvious details involved in properly using k(T), such as the width of the temperature step ΔT (or Δtheta) used. ΔT should be large enough such that more than one droplet freezing event occurs during a ΔT interval, but not so large such that important features of the k(T) spectrum are not observable. In our initial k(T) analysis we found a ΔT of 0.05 or 0.1 C to be an appropriate choice. Gabor plans to write a tutorial fully describing this analysis, using some of our droplet freezing data to illustrate the correct application of the k(T) method. We will also demonstrate the use of k(T) in a forthcoming paper that describes the design and evaluates the performance of our new microfluidic droplet freezing approach. To add k(T) analysis to this current manuscript would require rather significant additions to fully explain and illustrate this rather nuanced analysis. Instead we have added a detailed discussion of the k(T) analysis method with a link to Gabor's original comment, as follows:

"Alternatively, retrieval of the differential nucleus concentration, referred to as k(θ) in Vali (1971), is also recommended to assess the INP concentration in the sample versus that caused by background freezing. This approach can be used as a means of quantitatively attributing the INP signal to the sample versus the background for each droplet over the entire freezing spectrum. The differential nucleus concentration can be calculated using:

k(T) = -1/(V_d*ΔT)*ln[1-ΔN/N(T)] (2) where k(T) is the differential ice nucleus concentration, V_d is the droplet volume, ΔT is a temperature step that must be prescribed in the analysis, ΔN is the number of droplets that froze in that ΔT temperature step, and N(T) is the total number of unfrozen droplets at T. An important aspect is that ΔT is not the temperature step of the actual measurements, such as from the frequency at which images are acquired. To produce meaningful k(T) spectra the ΔT should be large enough such than more than one droplet typically freezes in a given temperature step. In our initial k(T) analysis we found a ΔT interval of 0.05 or 0.1 °C to work well for our experimental conditions. ΔT should be varied until a reasonable representation of the droplet freezing spectrum is produced that displays the important features of the spectrum and allows the sample to be distinguished from the background freezing of a control. Realizing that this is an important and nuanced detail, Gabor Vali is planning to produce a tutorial explaining the use of k(T) and selection of ΔT, using some of our data to illustrate this method. Referring back to Eq. (2), as an example, given an array of 100 droplets and a specified ΔT of 0.1 °C intervals, if the first 2 droplets freeze within one measurement interval, ΔT = 0.1 °C, ΔN = 2, and N(T) = 98. Using this metric, each freezing event in the interval ΔT is the result of at least one active INP, but given a small ΔT and a large N the interval can be approximately attributed to a single active INP.

Inherent to all droplet freezing methods is the assumption that the freezing of any droplet at a given temperature interval is caused by the combination of INPs present from the sample plus any background freezing due to impurities and substrate artifacts. The differential ice nucleus method, k(T), provides a quantitative assessment of the sample versus the background INP concentration at each temperature interval. k(T) is an alternative approach to the more commonly used method of just subtracting the cumulative K(T) or cINP background spectrum from the cumulative sample spectrum. This k(T) analysis method is discussed in detail by Gabor Vali in the comment (doi: 10.5194/amt-2018-134-SC1) he provided on the discussion version of this manuscript (https://www.atmos-meas-tech-discuss.net/amt-2018-134/amt-2018-134-SC1- supplement.pdf), based on the framework originally laid out in Vali (1971)."

Reference:

Vali, G.: Quantitative Evaluation of Experimental Results an the Heterogeneous Freezing Nucleation of Supercooled Liquids, J. Atmos. Sci., 28(3), 402–409, doi:doi.org/10.1175/1520-0469(1971)028<0402:QEOERA>2.0.CO;2, 1971.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2018-134, 2018.