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Interactive Comment

Interactive comment on "Repeatability and randomness in heterogeneous freezing nucleation" *by* G. Vali

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Additional observations with other materials

Data included in the paper under discussion were obtained with two slightly different soil suspensions and with a distilled water sample. Key results are consistent for all three samples even though they contained materials of various chemical composition and physical state. This fact raises the possibility, but with a question mark, that the concepts arrived at in the paper are perhaps applicable to most ice nuclei, independent of composition. The purpose of this short note is to reinforce that expectation by extending the list of materials for which experiments have yielded similar results. The experiments with these samples were conducted essentially the same way as that described in the paper but had different primary goals and therefore only statistical data are available not the individual drop histories. Consequently, these data could not be used as primary evidence for the repeatability of freezing temperatures, but they do support that finding well.

The table below summarizes the observations. Samples of silver iodide (AgI), a soil sample (MP-81), *Pseudomonas syringae* bacteria (strain T2304), and a cell-free bacterial preparation were tested. The silver iodide sample was a commercial powder preparation. The soil sample was similar to the one used for the experiments reported in the paper. The cell-free sample was produced as described in Phelps et al. (*J. Bacteriology*, **167,** 496-502, 1986) and provided for these tests by Dr. Ray Fall;this sample retained fragments that included the ice nucleating protein from the outer membrane of *Ps. syr.* bacteria, but had most of the original cell material removed. The samples were suspended in distilled water at concentrations that led to freezing temperatures for all drops, $T_f > -10$°C, as indicated in the second column of the table. The third column shows the temperature, T_w, to which the drops were warmed between freezing cycles until all drops were melted. The number of drops tested simulteneously, N, and the number of freezing cycles executed with the sample, n, are shown in the two right-hand columns of the table.

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Table 1: Standard deviation of observed run-to-run temperature changes, σ_{obs}, and values that would be expected for random variations, σ_{rand}.

SAMPLE	T_f(˚C)	T_w (˚C)	σ_{obs}	σ_{rand}	N	n
Silver iodide, AgI	−3...−5	+5	0.39	0.99	80	11
Soil MP-81 (filtered)	−6...−10	+5	0.43	0.72	50	4
		+5 (40 min)	0.38	0.79	50	4
Ps. syringea	−4...−6	+5	0.36	0.68	50	7
		+20	0.85	1.48	40	7
Cell-free	−4...−8	+5	0.83	1.20	50	7

Results are expressed here in terms of the standard deviation of the run-to-run changes of the freezing temperatures of individual drops, designated as δT_{ij} in the paper, for all the drops and all the runs. The observed value, σ_{obs}, is compared to the value σ_{rand} that this parameter would be expected to have if the sample drops froze in a random sequence but with the frequency distributions of freezing temperatures as observed. A smaller observed value than the value for random variations, $\sigma_{obs}<\sigma_{rand}$ can be interpreted as an indication that the freezing temperatures of the drops remained close to the characteristic temperature T_c of the most active nucleus in each drop, as discussed in the manuscript. As the data in Table 1 indicate, this is the case for all but the last sample.

For a comparison of the magnitudes of variability in freezing temperatures between these data and those presented in the paper, σ_{obs} has been calculated for drops of Sample A with $T_f>-10$˚C. This value is σ_{obs}=0.67, similar to the values shown in Table 1.

Two further aspects of these data deserve comment. For the soil sample MP-81, the length of time at +5˚C between runs was extended to 40 minutes. For the *Ps. syr.* sample, re-freeze cycles were made with the temperature between runs raised to +20°C instead of the normal +5°C. From the data in Table 1 it appears that neither the additional time in the liquid state, nor the exposure to warmer temperatures inflluenced the results greatly for these samples. These observations represent further evidence for the robustness of the nucleating sites.

In light of these results, and with a caveat attached to the limited extent of the tests here described, it seems warranted to extend the conclusions of the paper to the samples here reported. The fact that these additional observations included a simple inorganic (AgI) of well-known properties, a bacterial ice nucleant, and a cell-free derivative of that, is quite reassuring for the general validity of the singular character of heterogeneous freezing nucleation.

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