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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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General Comments:

This study presents detailed results on freezing experiments carried out with drops of dilute soil suspensions and distilled water. The importance of random variations vs. systematic alterations of nucleation temperatures is analysed. The results are discussed in terms of the singular and stochastic hypotheses and compared with findings from other studies. This study contributes to the understanding of the factors that influence heterogeneous nucleation temperatures even though it goes without any characterization of the heterogeneous nuclei present in the sample and is in this respect very empirical. The paper is clearly written, although at some instances more detailed explanations would facilitate the reading and understanding of the evaluation procedure, the tables and the figures. I therefore recommend some changes to improve the

readability of the manuscript. Moreover I have some specific questions and remarks.

Specific comments:

Page 4061, line 21: a nucleation rate of 1/(cm3s) is considerably lower than the range found in homogeneous ice freezing experiments. See e.g. Pruppacher and Klett but also more recent publications that show nucleation rates from 10ˆ4 to 10ˆ13 1/(cm3s) in the temperature range from -35 to -40 C.

Page 4065 - 4066, experimental and analysis procedure: a scheme of the cold stage would be helpful.

Page 4067, line 23: the equation at the beginning of the line should read: n(T)=K(T)*V

Page 4067: Equation (3) should be derived or explained better.

Page 4068, line 20: a reference would be appropriate here.

Page 4069, lines 21 - 25: a more detailed explanation of the Spearman rank correlation would be helpful.

Page 4070, line 10 and Figure 3b: does the panel really show how many drops retained freezing temperatures above the SN threshold through all previous runs or just how many drops had a freezing temperature above the threshold during a specific run?

Page 4070, lines 11-16 and Table 1: the different lines of the table should be explained better. The numbers cited in the text can not be found in the table. Maybe there has been a mix-up with different versions of the paper?

Page 4071, line18 and page 4072, line11: are run-to-run changes meant?

Section 6 and Figure 9, left and middle panel: It is not clear, why Gaussian pdfs with sigmas of 0.2 and 0.42 are shown in Figure 9 although the experimentally determined sigmas for the soil samples and the distilled water are 0.28 C and 0.6 C, respectively.

Section 6 and Figure 9, right panel: The nucleation rate for heterogeneous nucleation

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is given in terms of 1/(cm3s1) instead of 1/(cm2s). A heterogeneous nucleation rate per droplet volume is only valid for a fixed suspension concentration. While it is clear that the soil dust characterization at hand, does not allow the determination of the nucleation rate per heterogeneous surface, this drawback of the applied experimental procedure should at least be discussed. The nucleation rates given in the right-hand panel of Figure 9 seem to be too low to match the freezing frequency of the drops at the characteristic temperature: from the left-hand panel it can be estimated that the probability of a drop to freeze at Tc +/-0.1 K is around 30 % for the sigma = 0.2 case. Based on the nucleation rate given in the right-hand panel, the probability of a 0.01 cm3 drop to freeze at Tc +/-0.1 K (i.e. within 12 s for a cooling rate of 1 K/min) is ca 0.05 1/(cm3s)*12 s * 0.01 cm3 = 0.006. This value is 50 times smaller!

Page 4081, lines 25 and 26: the rate curves of the Zobrist et al. freezing experiments are given in their figures 2 and 3 in terms of 1/(cm2s). A comparison should therefore be possible.

Page 4082, lines 10-13: it does not make any difference under the stochastic hypothesis whether the nucleation rate is determined based on experiments made with steady cooling rate or at fixed temperatures. Whether freezing experiments comply with the stochastic hypothesis may depend on the heterogeneous nucleus under investigation. There is therefore no obvious contradiction between Zobrist et al. (2007) and Vali (1966, 1994) when the studies were performed with different types of ice nuclei.

Page 4082, lines 14-21: Marcolli et al. (2007) claim that the occurrence probability of contact angles follows a log-normal distribution, not an exponential increase. The use of contact angle does not exactly parallel the role of the characteristic temperature since the contact angle distribution is used within classical nucleation theory, thus combining the stochastic and the singular hypotheses.

Technical comments:

Page 4061, line 10: perception would be more appropriate than percept.

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Page 4066, line 1: tests instead of test.

Pages 4068 - 4080: there is an unnecessary zero in the numbering of the subsections: e.g. 5.0.1 has to be changed to 5.1. In section 6, the numbering starts with 6.0.4 instead of 6.1.

Page 4067, line 2 and references: Vali (1971) is missing in the reference list.

Page 4074, line 13: the quotation mark before fluctuations is missing.

Page 4075, line 21: the at the end of the line has to be deleted.

Page 4078, line 16: by any means instead of my any means.

Page 4081, line 21: 40 micrometer instead of 40 mm.

Tables 1 and 2: temperature instead of temparature in the figure heading.

Figures 1 and 2: (a), (b) .. should be added to the figures.

Figures 2(a) and 7(d): to what temperature interval does the contour plot scale refer to?

Figures 5 and 6: horizontal lines should be added to guide the eye (as e.g. in figure 3). Figure caption of figure 5: average freezing temperature instead of average temperature; figure caption of figure 6: freezing instead of frezing

Figure 7: horizontal lines should be added to panels (a)-(c) to guide the eye.

Figures 8 and 9: (a)-(c) should be added to the panels and the caption.

Figure 10: horizontal lines should be added to guide the eye. It does not become clear from the figure captions whether the three panels refer to three different drops. To illustrate better the proceeding of the experiment, spheres and diamonds should be interconnected. Is it correct that at the end two runs with 4.0 K/min are followed by two runs with 0.7 K/min?

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 4059, 2008.

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