

Interactive comment on “Repeatability and randomness in heterogeneous freezing nucleation” by G. Vali

Anonymous Referee #1

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The manuscript reports on experimental results of heterogeneous freezing of individual drops of soil suspensions on a cryo stage in temperature ramp experiments. It is elucidating the observed variability in freezing temperatures commonly found in such investigations by a careful statistical analysis of the freezing patterns observed in repeated cooling cycles. The author is able to demonstrate that freezing is primarily determined by the nature of the most effective heterogeneous nucleus present in the germ and that this nucleus often retains its properties over many cycles. This finding is clearly in favour of the singular hypothesis and its statistical analysis allows distinguishing quantitatively between random fluctuations and non random alterations of the freezing temperature. The study is clearly written and represents an important contribution to our understanding of heterogeneous freezing. Three important points of the

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experiment and the evaluation of the data are not clear to me however and should be addressed in a revised version:

Chapter 4, Experimental

In order to judge the results presented it would be very helpful to have at least a crude estimate of the number- and size distribution of the nuclei in the individual droplets.

Chapter 4 Experimental

In order to enhance the significance of the results, an attempt is made to select a single nucleus subset from the data. This concept and its quantification do not become clear to me from the description on P4067, l 13 - P4068, l 13. (what is δ on P4067, l 23; how is eq. (3) derived, is there a typo in eq. (3)?). Looking at the data presented later, I feel that all relevant conclusions are supported by the complete set of data, so that the concept and justification of the SN subset seems doubtful. If it is indeed needed, it should be explained in more detail.

Chapter 6.04 Nucleation rate

I would argue that the nucleation rates, which are derived in this manuscript and bear a unit of $\text{cm}^{-3}\text{s}^{-1}$ can not be compared in magnitude directly to what is observed in homogeneous nucleation studies, as in the former case the unit (per volume) is rather artificial and not related to volume of the droplet but rather to the number of effective nucleating sites. (In a Gedankenexperiment with droplets of larger volume but containing the very same nuclei, heterogeneous freezing would be observed at the same temperatures, but nucleation rates, calculated as described on line 25ff on Page 4075 would be lower.

Minor remarks:

The numbering of the paragraphs is irregular.

P 4069 and table 1: Specify more clearly what is given in the various lines of the table

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P 4096, L. 25 Probably a minus sign is missing in the exponent.

P 4073, L 17 should read: mean freezing temperatures

Fig. 1: Fig. 1b and 1c seem somewhat redundant. I would suggest to display Fig. 1a and 1b as separate figures.

P. 4073 L. 26: and figure 8. The description of the right hand panels is not precise, the unit of the y- Axis should be dimensionless, the standard deviation of δT_{ij} has a dimension of temperature. Probably the ordinate should be labelled according to normal probability paper.

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

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