

Interactive comment on “Heterogeneous ice nucleation activity of bacteria: new laboratory experiments at simulated cloud conditions” by O. Möhler et al.

O. Möhler et al.

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We acknowledge the valuable comments from the three referees of our paper. Based on these comments, a revised and improved version of the manuscript will be prepared and submitted for publication in BG. Here we respond to the referee comment by Paul DeMott.

General points: Following the referee comments we will modify the structure of the paper. The section title ‘Cloud simulation experiments’ will be changed to ‘Cloud chamber experiments’. This section will be structured into further subsections entitled ‘The AIDA cloud chamber facility’, ‘Aerosol formation and characterisation’, ‘Spray experiments’ and ‘Cloud expansion experiments’. For the subsection ‘Spray experiments’

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we will include two more figures with time series from two selected spray experiments. These figures and also the former Figures 4 and 5 will include an additional panel with the optical diameters of all individual particles detected by the Welas2 optical particle counter. These plots will clearly show the distinct groups of droplets and ice particles that formed during the spray and expansion experiments. Care will be taken to separate the method description and results into the respective sections and to avoid repetitions of method descriptions.

The droplet freezing experiments were mainly done because the results were used to determine the optimum starting temperature and aerosol cell concentrations for the cloud chamber experiments. The main focus of the present study was not to compare the droplet freezing method to the cloud chamber method. This will be more clearly stated in the revised manuscript. We would not like to remove this piece of information from the paper. We agree to the referee that more comprehensive and systematic measurements would be required to compare both methods on a quantitative basis.

Answer to specific comments:

Abstract, Point 1: First of all, the spray process was necessary to get the bacterial cell from the suspensions into the aerosol phase in the chamber at high enough concentrations. A dry process might have been useful for dispersing the Snomax™ material but not for the suspensions with the living cells. The idea was to keep the cells in their aqueous environment as long as possible before dispersion. For this study we had no means to investigate how many cells really survived the dispersion process with the nozzle. This could be done in future studies, which also should address the effect of different dispersion techniques on the cells and their ice nucleation activity. The specific procedures of the spray and cloud expansion experiments will be more explicitly described in the revised manuscript. As mentioned above an extra subsection about the spray experiments with time series figures from two selected spray experiments will be added. We see no need to modify the abstract in regard of the spray and expansion experiments.

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Abstract, Point 2: What is meant here is that only a minor fraction of the bacterial cells happened to be ice nucleation active, either in the condensation mode or the immersion mode upon further cooling after the CCN activation of the cells. The experiments were not conclusive in regard of time dependent nucleation processes. Time dependent versus singular hypothesis effects should be addressed in further investigations. We will change the wording in the abstract to ‘During these experiments, the bacterial cells first acted as cloud condensation nuclei to form cloud droplets. Then, only a minor fraction of the cells eventually acted as heterogeneous ice nuclei either in the condensation or the immersion mode’.

Abstract, Point 3: Yes, we will mention in the abstract that our results for Snomax™ agree to literature results.

Introduction, Page 1447, lines 3-4: Have changed ‘by several independent research teams’ to ‘by a few research teams’ and added the reference Jayaweera and Flanagan, 1982, Geophys. Res. Lett. 9, 94-97.

Preparation of bacterial cells, Page 1448, lines 11-12: The cell number concentration in the aerosol chamber was determined from the bimodal lognormal fits to the measured aerosol size distributions. This will be mentioned in the revised manuscript.

Droplet freezing studies, Page 1449, Section 3: The droplet freezing studies were done first because the results were used to determine the optimum starting temperature and aerosol cell concentrations for the cloud chamber experiments. This will be stated in the revised manuscript. The main focus of the present study was not to compare the droplet freezing method to the cloud chamber method. We agree to the referee that more comprehensive and systematic measurements would be required to compare both methods on a quantitative basis.

Cloud simulation experiments Page 1451, line 13: The suspensions were sprayed into the chamber with a two-component jet device (model 970 from Düsen-Schlick GmbH, Germany) which uses a particle free synthetic air flow of about 1 l min^{-1} at an abso-

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lute pressure of 2 bar to disperse a liquid flow of about 5 to 10 ml min⁻¹. The spray cloud will certainly not be uniformly distributed during the spray process. However, the internal mixing time scale in the chamber is about 1 min, whereas the spray process lasted about 5 min in most experiments. Uniform distribution of the spray cloud is not necessary to detect frozen spray droplets. Because the chamber was ice supersaturated during the spray process any ice particles formed somewhere in the spray cloud further grow during distribution in the chamber. We know from e.g. the cloud expansion experiments that the residence of ice crystals in the chamber is at least several minutes. Therefore we can assume that any ice crystals formed during the spray process are well distributed within the chamber and that their number concentrations can well be detected with the optical particle counters. Therefore, these experiments tell us the ice nucleation active particle fraction in the immersion mode at the given temperature of the cloud chamber. Nothing can be said or concluded here about the actual time history of the activation behaviour. As mentioned above an extra section with two more figures will be added to the revised manuscript to explain in more detail the spray experiments.

Page 1451, lines 24-25: In fact sampling losses can be neglected for the full aerosol size range, i.e. the smaller residual particles and the larger bacterial cells. Therefore we changed this sentence to 'Particle losses in the sampling tubes can be neglected for all aerosols used in this study'.

Page 1452, lines 6-8: The SIMONE experiment was sensitive enough to detect both the depolarisation of the non-spherical bacterial cells and of the ice particles. There was also a difference in depolarisation of Snomax™ and of living cells, as can be seen in the aerosol depolarisation of Figures 4 Snomax™) and 5 (bacteria type PS2). This is already mentioned on page 1454, lines 20 to 24.

Page 1452, line 16-17: Please note that the equivalent sphere diameter is given here. Both the size distribution analysis and the large backscatter depolarisation ratio measured for strain 31R1 bacteria (type PS2, see above) indicate the marked aspherical

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shape of the cells. This means they still can be as long as $1\text{ }\mu\text{m}$ or more which is typical for such cells. Unfortunately we do not have independent size or shape analysis, e.g. from electron microscopy.

Page 1452, line 26-27: Yes, the number concentration of bacterial cells is indeed determined from the size distribution fits. This will now also be mentioned in section 2 (see comment above).

Page 1453, line 17: Yes, of course there are diabatic effects because of the warmer chamber walls. Thanks for the hint. We change 'adiabatic' to 'quasi-adiabatic'.

Results and Discussion Page 1455, line 16: The Welas instruments measured all the time but did not detect larger ice crystals. The detection limit is of the order of 0.1 cm^{-3} . If no ice particles are detected during an experiment, the detection limit defines an upper limit of the ice number concentration that may have formed during this experiment, and from that we obtain an upper limit of the ice nucleation active particle fraction.

Page 1455, lines 19-20: We refer here to the onset of ice nucleation in the cloud chamber experiments by Ward and DeMott (1989). The major ice nucleation peaks around $-6\text{ }^{\circ}\text{C}$ and colder are mentioned later in the discussion.

Page 1456, line 15: Yes, we argue here about deposition nucleation because the relative humidity was below 100 % and no liquid water droplets were present in the chamber.

Page 1457, general comments: As mentioned above, we just made use of the aerosol spray formation to obtain additional information about the ice nucleation efficiency of the bacterial cells. We do not see why the processes in the spray should be that much different at the different temperatures. We very carefully discussed the results from the spray experiments and will, as already mentioned above, add another chapter to explain these experiments in some more detail. We agree that inferences made to deactivation of cells from the present study is speculative. Such processes deserve

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further investigations.

Conclusions, Page 1458, line 19-20: 'No significant ice activity' means, in our view, the same as 'No ice formation within the detection limit'. Any experiments have their limits. The detection limits for ice formation are described in the paper. We see no reason to repeat the detection limits again here. Anyway, we suggest to change this sentence to 'Within the detection limits of our experiments, no ice activity of the bacteria species was observed above -7°C '.

Technical corrections:

1) *Page 1447, lines 12-15:* 'growth' was replaced with 'grows' in the first sentence. Next sentence replaced with 'The warmer the freezing temperature the more time the ice particles have to take part in this sequence and the more likely they are to grow to precipitation size.'

2) *Page 1447, line 19:* 'number' added before 'concentrations'.

3) *Page 1448, line 7:* 'industrial secret' replaced with 'proprietary information'.

4) *Page 1448, line 22:* Yes, we mean 'The samples were tested for their INA...'

5) *Page 1449, line 25:* Remove the word 'from'.

6) *Page 1452, line 6:* 'Also' is now spelled correctly.

7) *Page 1454, line 27-29:* This is possibly the third mention of the Welas instruments, again suggesting more careful attention needed toward organization and details. TO BE DONE

8) *Page 1456, line 12:* 'approved' replaced with 'confirmed'.

9) *Page 1456, line 16:* Thanks for the hint. Of course the ice number concentration decreased with time due to settling losses of the larger ice crystals.

10) *Page 1456, line 26:* 'at least' is now omitted.

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11) *Page 1457, line 6*: bacterial cells have 'been' found.

12) *Figures 4 and 5*: The figures 4 and 5 have been replotted with larger label font sizes. The plotted parameters will be described in the first figure caption. We have also included another panel to the time series figures that shows the optical diameters of all individual particles detected by the Welas2 optical particle counter. These plots clearly show the distinct groups of droplets and ice particles that formed during the spray and expansion experiments.

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