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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct quantification of total and biological ice nuclei in cloud water" *by* M. Joly et al.

M. Joly et al.

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General comments: This paper adds very valuable new information to the current debate about bio-precipitation. In particular, the authors found that (1) Biogenic ice nuclei were active at <= -6°C and were the dominant active ice nuclei between -6 and -10°C. At -11°C and below the non-biological IN accounted for more than 1/2 of the ice nucleation activity, at -13°C reaching more than 90 % of total activity. (Fig. 2). (2) The number of total biogenic IN (Table 2) that are active at high T (-6 to -8°C) was larger than expected based on earlier ïňĄndings, e.g. approx. 100 x of Christner et al. The authors also speculate about the total biogenic IN being made up largely or even totally by intact bacteria. While that part of the paper is not convincing, it is still an interesting

discussion – nevertheless the paper could stand alone without that part.

Critical to the quality of the paper is in my view the severe absence of statistical analysis.

We do not agree that no statistics was done in the original manuscript. Among many examples, we presented a PCA analysis of the data in Figure S2. In the revised manuscript, we extended this analysis with new parameters that we were able to collect afterwards (precipitation, cloud event duration, . . .), and we performed new stats. Correlation matrices are shown in Table S2, and text has been added accordingly (Lines 232-234, 251-253, 264-281, notably).

In addition, as nice and new as the data are, a principle limitation of this work is that the mountain peak can produce orographic clouds, and thus the clouds can be contaminated by bacteria derived from the side of the mountain. Without addressing this issue, the authors cannot remove my suspicion that not all the clouds investigated were true high altitude clouds. It would be better to discuss this matter (instead of not mentioning the problem): Maybe the authors could have a meteorologist make an educated estimate how large the contribution from the surface of mountain side possibly could be?

It is right that we cannot exclude the possible influence of local sources, so a sentence has been modified in the text to indicate it (line 289-290). However, concerning the fact that clouds could have been orographic, great care was taken for avoiding such situation. Note that new variables have been added in the analysis (see section 2.1 and answer 1) to referee 1).

SpeciïňĄc comments:

Page 3709, line 26 - : It appears that the authors have not considered the papers by Santl-Temkiv on hailstones' bacterial content and bacteria's origin (FEMS Microbiol Ecol 81: 684–695; PLoS ONE 8(1): e53550).

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Those references are not about IN, so we think that citing these would be out of purpose here. The only reference by this group that deals with IN that we found (Šantl-Temkiv et al., 2009) originated from a conference:

Šantl-Temkiv, T., Gosewinkel-Karlson, U., Finster, K., and Munk Hansen, B.: The diversity and proportion of ice nucleation active bacteria in rain and their ability to produce extracellular ice nucleation active particles, 18th International Conference on Nucleation and Atmospheric Aerosols (ICNAA), 1460–1466, Prague, Czech Republic, 10–14 August 2009.

Page 3716, line 23: The statement on the bacteria targeted by lysozyme is not correct. See also the interactive comment by C. Morris on this matter.

See answer 2) to C. Morris's comments.

Page 3719, line 1-2: OVERREPRESENTATION does not seem to be what the authors mean here. Either the use of this term should be explained, or the expression deleted.

We agree that the term "overrepresentation" was not appropriate as this referred to an inferred maximum possible value, rather to an actual value. The corresponding sentence has been removed.

Pages 3718-3719: Otherwise I agree with the authors' conclusions. See my general comments, above.

Page 3722, lines 23-25: I should think that this conference presentation can be replaced by citable publications originating from the same authors.

See answer 3) above. No other publication concerning IN from these authors seems to exist.

Table 1: I strongly suggest that the sample volumes should be added as a separate column. This will enable the readers to fully appreciate the work done, and to use the data for their own considerations and estimates in the future. Another information that

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I feel is missing is the type of cloud(s) encountered in each event, e.g. convective, stratus, orographic.

Samples volumes are now presented in Table 1. Concerning the type of clouds sampled, we now show satellite images which show overviews of the meteorological situation over Europe and France at the moments of sampling.

Table 2, and corresponding discussion in the text: I miss a calculation of the number of IN per cloud droplet. Based on their LWC, the authors should be able to provide such an estimate. For the readers, such numbers would aid in appreciating the signiïňĄcance of the authors' ïňĄndings for cloud physics.

We agree that the number of IN per droplet is an interesting feature so we added a simple estimation based on the droplet diameter to the manuscript (lines 291-295). We considered cloud droplets as spherical objects, with a volume (mL) = 4/3 π rˆ3 , where r is the droplet radius (cm). From this value, we calculated the concentration of IN per droplet ([IN]d) as [IN]d= V $\times$[IN] with [IN] the concentration of ice nuclei per mL of cloud water.

Table 3, legend: (1) DETECTION limit is the wrong term, as the authors are referring to an upper limit of quantiïňĄcation (accidentally) caused by how much the droplets were diluted.

We replaced "our detection limit" by "experimental quantification limit"

(2) Please change ïňĄrst sentence to "Inferred maximum possible fraction of INA bacteria among total bacteria . . ."

This has been changed.

Fig. 2: (1) Should the y-axis label not read ". . .frozen droplets", instead of ". . . samples"?

The Y-axis was referring to the proportion of cloud samples for which at least one

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droplet froze during droplet freezing assays.

2) Please change ïňĄrst sentence to “Cumulative proportion of frozen samples (droplets?) at speciïňĄc freezing temperatures . . .”.

It is now changed by “Cumulative proportion of cloud samples for which at least one freezing event was observed during IN assays, in the absence of treatment (shaded bars) or after heating at 95°C for 10 minutes (black bars).”

Please include error bars.

It is not possible to include error bars here as this figure shows absolute frequencies.

Fig. 3: What is the reason for showing every single data point, i.e. individual samples? The whole paper lacks statistics. Here is one obvious place to offer to the reader a regression, with corresponding statistics, over all samples.

We showed, already in the original manuscript, both every single data point, in Figure 3, to allow the reader to figure out the variability of profiles, and consensus, i.e. averages, in Figure 4.

Fig. 4: Also here the use of statistics would improve the paper. Here the reader would appreciate seeing error bars, conïňĄdence intervals, or similar measures of statistical signiïňĄcance.

Error bars have been inserted in Figure 4.

Fig. 5: (1) Again, what is the reason for showing every single data point, i.e. individual samples? Also here is one obvious place to offer to the reader a regression, with corresponding statistics, over all samples.(2) The data points are the same as in Table 3. There is no need to show the data twice – unless . . . see remark no. 1). (3) The y-axis scale is correct, but confusing. Please eliminate the “%” and change the numbers on the scale accordingly (e.g. 10% becomes 10e1). (4) Please change the sentence to “Inferred maximum possible fraction of INA bacteria among total bacteria .

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. ."

Figure 5 is not presented anymore in the manuscript.

Technical corrections:

Please re-work the whole manuscript for using grammatically and syntactically correct English. Here is a non-exhaustive list of spelling errors and wrong use of words: CloudY air, several places in the paper, including abstract and legends Back trajectory PLOTS (these are called back trajectories, not plots) Cloud'S microphysics At –10°C there WERE . . ., seen in the abstract (correct to say "were observed, were measured, but not just "there were") guarantY, page 3710 wrong use of "nor", page 3710 MaterialS, Page 3712, line 1 CIN, page 3713, line 12, (please put IN in subscript) HITTING the puy de . . ., page 3714 (better: "reaching" or "arriving at") otherS, page 3716, line 23

All these have been corrected.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C2577/2014/acpd-14-C2577-2014-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 3707, 2014.

Table 1. Main characteristics of the cloud events sampled. Samples recovered as ice formed upon impaction in the sampler are indicated in italic. See detailed ion composition in Table SM1.

Sample	Date	Sampling period (UTC)		Sampling duration (h)	Volume sampled (mL)	Cloud period (UTC)[a]		Cloud event duration (h)[a]	Time in cloud before sampling (h)[a]	Time in cloud after sampling (h)[a]	Precipitation accumulated in the vicinity (mL)[b]	Mean sampling temperature (°C)	Mean LWC during sampling (g m^{-3})	Bacteria concentration (mL^{-1})
		From	To			From	To							
# 76	29-Jun-11	6:30 AM	11:45 AM	5.25	> 200	6/28/11 10:00 PM	6/30/11 0:00 AM	26	8.5	12.3	1.6	11.5	0.6	n.d.*
# 77	7-Jul-11	1:50 PM	3:00 PM	1.17	15	7/7/11 9:00 AM	7/8/11 6:00 AM	21	4.8	15	7	12.0	0.1	n.d.*
# 78	20-Jul-11	7:30 AM	9:10 AM	1.67	47	7/19/11 3:00 PM	7/23/11 4:00 PM	97	16.5	78.8	0.2	8.3	0.3[c]	12355
# 79	7-Nov-11	1:00 PM	2:30 PM	1.50	193	11/6/11 8:00 AM	11/8/11 11:00 AM	51	29	20.5	0.4	7.0	0.6[c]	10825
# 80	*20-Jan-12*	*12:45 PM*	*3:00 PM*	*2.25*	*55*	*01/18/12 11:00 PM*	*01/26/12 0:00 AM*	*169*	*37.7*	*129*	*0*	*-0.4*	*0.3[c]*	*9980*
# 81	*23-Jan-12*	*1:00 PM*	*4:00 PM*	*3.00*	*53*	*01/18/12 11:00 PM*	*01/26/12 0:00 AM*	*169*	*110*	*56*	*0*	*-1.2*	*0.1[c]*	*33724*
# 82	*19-Mar-12*	*12:10 PM*	*4:10 PM*	*4.00*	*45*	*3/17/12 11:00 PM*	*3/21/12 11:00 AM*	*84*	*37.2*	*42.8*	*0.2*	*-1.5*	*0.1[c]*	*1648*
# 83	*4-Apr-12*	*6:10 AM*	*9:20 AM*	*3.17*	*29*	*4/3/12 11:00 PM*	*4/6/12 12:00 PM*	*61*	*7.2*	*50.7*	*0.25*	*-0.4*	*0.1[c]*	*14914*
# 84	*18-Apr-12*	*8:10 AM*	*12:15 PM*	*4.08*	*31*	*4/17/12 6:00 PM*	*4/25/12 6:00 AM*	*180*	*14.2*	*161.8*	*0*	*0.2*	*0.1[c]*	*3902*
# 85	25-Jun-12	1:35 PM	5:00 PM	3.42	66	6/25/12 01:00 AM	6/26/12 12:00 AM	35	12.6	19	0	13.3	0.3[c]	4474
# 86	13-Sep-12	7:50 AM	9:50 AM	2.00	75	9/12/12 7:00 PM	9/13/12 3:00 PM	20	12.8	5.2	0.8	6.0	0.6[c]	5199
# 87	10-Oct-12	8:40 AM	9:50 AM	1.17	70	10/08/12 9:00 PM	10/11/12 0:00 AM	63	35.7	26.2	0	9.4	0.6[c]	19658

a: Defined as RH > 95% based on hourly average (see Fig. S1).
b: Sum of precipitation accumulated at 5 rain gauge stations in the vicinity of puy de Dôme (Royat, Farnette, Sayat, Trois Ponts and Blanzat) (see Fig. S1).
c: Estimation from sample collection rate and puy-de-Dôme data archive.

* n.d.: not determined.

Fig. 1.

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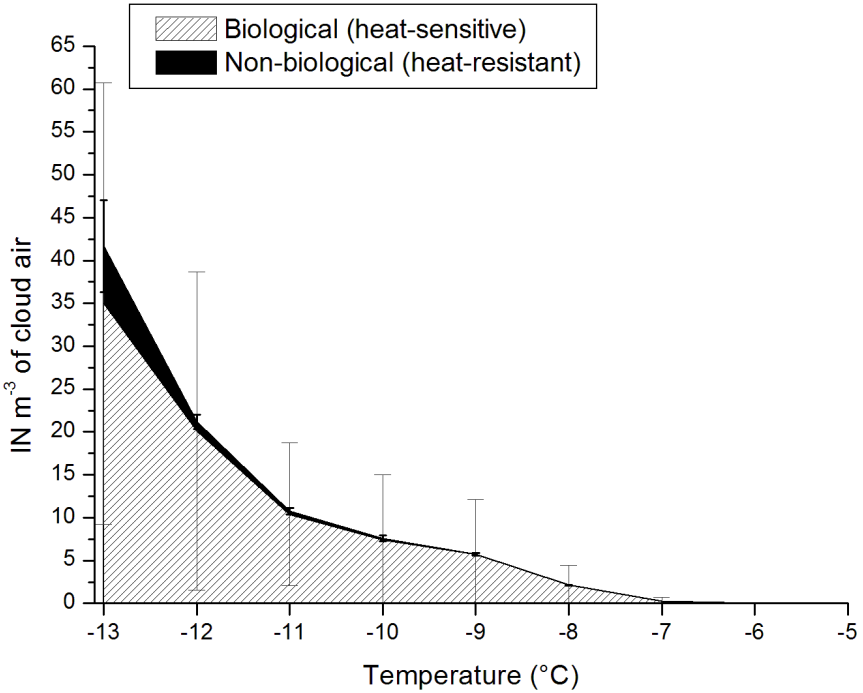

Fig. 2. Fig 4. Mean cumulative concentrations of biological (heat-sensitive. shaded area) and non-biological (heat-resistant, black area) IN in clouds (n=12) per volume of air.

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