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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Activation of Intact Bacteria and Bacterial Fragments Mixed with Agar as Cloud Droplets and Ice Crystals in Cloud Chamber Experiments" *by* Kaitlyn J. Suski et al.

Anonymous Referee #1

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Suski et al present single-particle mass spectrometry (SPMS) measurements of bacteria and fragments that served as CCN and INPs in the AIDA cloud chamber during immersion cloud freezing experiments. This work tackles the important question of the role of primary biological particles in cloud formation and properties through laboratory experiments. I have questions below regarding interpretation of the data that may impact the results. Otherwise, revisions are recommended below primarily to increase clarity of the manuscript.

The main finding of this work is that bacteria fragments (mixed with agar) serve as cloud droplet nuclei, whereas intact bacteria rarely nucleate cloud droplets. The miniSPLAT

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Discussion paper

size distributions of particles prior to cloud formation (expansion) within the chamber compared to the cloud droplet residuals support this conclusion. However, I have several questions that may impact the interpretation of the results. 1) Could intact bacteria burst upon droplet activation or freezing? (Is there any support for this from previous studies?) Or, could intact bacteria burst during drying within the CVI? 2) What are the size cuts of the PCVI and IS-PCVI and transmission efficiencies? This information needs to be provided in the experimental section. For each expansion, what fraction of the droplets and ice crystals were and were not transmitted through the CVIs? (Provide in results & discussion.) 3) It is stated that the pressure changes during expansions impacted the miniSPLAT aerosol velocities. What was the dependence of the size-dependent miniSPLAT inlet transmission efficiency on these pressure changes during expansions? If particles >0.5 um were not as efficiently transmitted through the aerodynamic lens at these lower pressures, this would explain why intact bacteria were not observed in the cloud particle residues.

Additional Major Comments: - Experimental (Page 5): Please provide additional information about the miniSPLAT operation. What was the LDI power, and did it vary during the study (since ion fragmentation is dependent on this)? What was the size-dependent inlet transmission efficiency as a function of inlet pressure? How were size calibrations conducted (PSLs?) and over what particle size range? What is the size range of efficient inlet transmission, and does it depend on pressure? (Figure 1 only gives a lower detection limit – no upper limit is provided, although it is clear in this figure that transmission appears to drop off above 1 um.) How were number concentrations obtained from the miniSPLAT data (what calibration/data processing was done)?

- Page 5, Line 31: In addition to different relative peak intensities, there appear to be differences in the individual ions present at > m/z 50. This should be discussed.

- To aid interpretation of the mass spectra, please label all ions in Figures 2, 5, 7, 10, and 14. If the chemical identity is not known, please list possibilities and/or at least the numerical m/z. Also, please label in the captions whether these mass spectra

correspond to averages or representative examples.

- Figure S1 is a very informative figure, and the authors may consider moving this to the main text. In particular, the labeling of the ions is useful, and the magnification of the small peaks is helpful. Regarding the comparison here, what differences are observed between the 'agar + bacterial fragments' and 'agar'? This should be discussed in the main text to support the inclusion of agar in the aerosols (Page 6, Line 8). It appears that the main difference may be the presence of m/z 131 in the bacterial fragments. Is this observed in the aerosols and cloud residues? The mass spectra shown in the main text are only shown up to m/z 120, so the reader cannot evaluate this. If m/z 131 is indeed a primary difference between the mass spectra, then I recommend that the authors show the mass spectra in the main text up to at least m/z 135.

- Page 6, Line 3: It is stated here that "intact cells show relatively lower intensities for the metal ions (23Na+, 39/41K+, 40Ca+)", but I do not see this in Figure 2.

- Page 6, Line 4: Please discuss the organic ions present, as they are not stated here in terms of m/z or ion formula, making interpretation of this statement by the reader not possible. There are many previous SPMS papers on primary biological particles that could aid in this mass spectral interpretation. A greater interpretation of the ions above m/z 41 may aid in the interpretation of the results. Example manuscripts to consult for SPMS biological particle mass spectra include (but are not limited to): Fergenson et al 2004 (Analytical Chem), Czerwienec et al 2005 (Analytical Chem), Srivastava et al 2005 (Analytical Chem). Peaks unique to the bacteria (fragments and intact) and not agar should be highlighted in this discussion.

- Page 7, Lines 23-25: It is stated here that "The small differences between the MS of residuals and the small particle mode suggest a slightly higher content of organics in the residuals", but I do not see this in Figure 5. Please provide additional support/description (e.g. label and discuss specific peaks).

- Page 9, Line 3: This sentence implies that peaks > m/z 44 are all organics, which

contradicts Page 6, Lines 1-2 and previous SPMS biological particle studies that show higher mass inorganic peaks as well. This discussion should consider specific peaks and fix this statement.

- Page 9, Lines 28-30: Typically CVIs concentrate cloud residuals during sampling and this transmission is size-dependent. This information needs to be stated in the experimental section, and this needs to be considered in the interpretation of the number concentrations measured after the CVI, as discussed on this page. The reader also does not have knowledge of the transmission differences between the IS-PCVI and PCVI to evaluate the transmission of cloud droplets vs ice crystals as stated here.

- Page 11, Line 31 – Page 12, Lines 1-2: This result was not shown or discussed in the results & discussion, and if included here in the conclusions, it should be shown and discussed.

- To aid in comparison of the results between the different expansions, the authors are encouraged to combine (into a multi-panel plot or by stacking the mass spectra) the following mass spectral figures: 5, 7, 10, and 14. Similarly, interpretation would be improved with combining the following size distribution figures: 4, 9, and 13. Figures 1 and 6 could be similarly combined.

- Figure 11 and associated discussion: Since ion intensities in laser desorption ionization are dependent on ionization energies (e.g. Gross et al 2000, Analytical Chem.; Reinard & Johnston 2008, J. ASMS), the ion ratios used here need to be discussed in greater detail as they do not represent quantitative mole or mass ratios. In particular, inorganic ions typically have much lower ionization efficiencies compared to organic ions, which also undergo significant fragmentation at 193 nm. In addition, the organic ions included here in the ratio need to be stated here, in the experimental, or in the results & discussion. While it is clear that the organic contribution increases with size (and this is a useful result), clarification needs to be provided for the reader not familiar with LDI. Additionally, the text on page 9 refers to the dva distribution here as a

'schematic representation'. Please clarify in the caption and show actual data instead.

Minor Comments: - Introduction: Many sentences do not include references, which should be added to support the statements. In particular, references are needed on page 2, lines 1, 12, 14, 20, and 21.

- Page 3, Line 23: Move this sentence to the end of the previous paragraph, as one sentence does not constitute a full paragraph.

- Page 4: RH is defined twice here as well as on page 2.

- Page 4, line 15: Why are the SMPS and APS data converted to volume-equivalent diameters? It would seem more appropriate, for comparison to the miniSPLAT data (in vacuum aerodynamic diameter), to simply convert the SMPS mobility diameters to aerodynamic diameter and leave the APS data in aerodynamic diameter. Also, please provide a reference for the chosen particle density.

- Results & Discussion: It would be useful for the reader for sub-headers to be added, for example, 1) Bacteria mass spectral signatures, 2) Expansion 1, 3) Expansion 2. . ..

- Page 5, Line 18: Space needed after (dve).

- Page 6, Lines 11-13: This sentence is not clear as written. Laser power should be provided in the experimental section.

- Page 7, Lines 26-27: Move this sentence to later in the results & discussion where Expansion 2 is discussed.

- Page 7, Last paragraph: This paragraph is all repetition (summary) and could be removed as it doesn't seem necessary.

- Page 9, Lines 7-10: Please clarify this sentence.

- Page 9, Line 19: Should Figure 13 be referred to here instead of Figure 6a (typo?)?

- Page 9, Line 24: Add mention of the number concentration prior to expansion for

context.

- Page 10, Lines 3-4: Move “(light blue)” to after “expansion” in this sentence.

- Page 10, Lines 10-12: Move sentence to the end of the previous paragraph.

- Page 11, Line 23: Please provide a reference for the hygroscopicity of intact bacteria.

- Figure 9 caption: Mention the type of bacteria used. Make sure this is included in each figure caption.

- Figure 12: Please clarify the legend of the bottom plot.

- Figure 13: Please provide the sample timing for the ‘cloud residuals’ vs ‘cloud residuals late’, so that the reader can refer back to Figure 12 for context.

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