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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice nucleation by water-soluble macromolecules" *by* B. G. Pummer et al.

Anonymous Referee #1

Received and published: 23 October 2014

The paper entitled "Ice nucleation by water-soluble macromolecules" by Pummer et al. gives some insights into the potential role of macromolecules including proteins, polysaccharides. . . in atmospheric microphysics as ice nuclei.

This paper presents a real interest because it is the first one to give an overview of this kind of biological IN which are water-soluble and which present IN properties per se, independently of the biological organisms (fungi, pollen,..) excreting or releasing them. The discovery of these INMs (Ice Nucleating Macromolecules) is rather recent, previous reviews were related to mineral IN, or biological IN such as bacteria, spores, fungi...

However I have major concerns about this paper:

1) The structure of the paper is not coherent. It is presented as a research article with an experimental part, results and discussion sections but it looks more like a review on the domain. Also the supplement part is unusual, closer to a patchwork of book chapters, the idea being to help some readers to understand the rest of the paper.

2) The content, although interesting and informative, is not strong enough to be published in ACP. The experimental results and new data are limited in the paper; they only complement data published elsewhere, often by the same authors. One can compare this paper with the data published previously in ACP by the same authors, for instance Pummer et al. (2012, 12, 2541- 2550) or Augustin et al. (2013, 13, 10989-1103) on the same topic and which are much more consistent. Also the long supplementary information presents notably very basic definitions or discussions which are too general for being presented in such research paper (for instance the description of macromolecules could be part of any chapter of a biochemistry book for undergraduate students...).

I would suggest to completely rewriting this paper as a review. The new results and protocols could be shifted in the Supplement section, and the long discussions and information present in the actual supplementary part S1 should be deleted, incorporated in the review in a shorter form when possible (for instance the discussion about the motivation for the expression of biological INMs) or supported by references to general book chapters, reviews or other papers (for instance the descriptions of macromolecular chemistry, of basic physics of INA ...).

The authors may have two alternatives:

1) They could write a "mini-review" that takes into account the information presented in this paper, but in this case it should be published elsewhere, for instance in "Atmospheric research" or "Atmospheric environment". In my opinion this paper would be too short for a review in ACP when compared to other reviews on similar topics (see for instance of Hoose and Möhler, 2012)

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C. Hoose, O. Möhler. Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments. Atmospheric Chemistry and Physics 2012; 12(5):12531-12621. DOI: 10.5194/acpd-12-12531-2012

2) Alternatively they could write a deeper review that could include more aspects, for instance which would present in more details the atmospheric context, other macro-molecules of interest for atmospheric sciences (HULIS, biosurfactants, EPS, SOAs etc...) including CCN aspects. This new review could be then resubmitted to ACP.

Finally they could "re-use" the information present in the Supplement section for writing a book chapter.

Other comments:

P 24282 Methods of characterization of INMs

Although basic methods such as heating or the use of guanidinium chloride, boric acid and enzymes are valid to determine to presence of proteins or saccharides in INMS they are limited and should be completed by more powerful analytical methods. Why did the authors not use NMR and/or mass spectroscopy, which are the common tools to assess the structure of macromolecules? These techniques can be applied to purified compounds and also on complex mixtures through to 2D NMR (1H-1H or 1H-13C) or LC-MS and Maldi-TOF MS. The idea is not necessarily to determine the exact structure but to determine the chemical functions present in the molecules. NMR for instance easily detect aromatic functions, sugar characteristic signals (anomeric 1H), amino acids signals, carboxylic or aldehyde functions etc.... These techniques are much more informative and reliable than those used in this manuscript and can give indications on structural motifs which are not polysaccharides or proteins, and with no a priori.

P 242887 Critical cluster size

Although Fig 4 presents interesting results, the discussion about PVA (p 24288) is

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long and rather useless; it is quite evident that this simple oligomer has nothing to do with a complex protein structure. The necessary molecular arrangement to make an ice crystal for different proteins has already been well described from models (see Garnham et al., 2011). Note that this paper, which is cited P 24277 line 5, should rather be cited when describing Fig 1c or within this paragraph. From this, it was expectable that PVA would remain a rather inefficient IN, whatever its size.

P 24291 Atmospheric impacts

P 24292 line 6: the sentence should be changed to " At last, microorganisms were found in cloud waters....." .The reference Amato et al. 2007 could be changed to that of Vaïtilingom et al. (2012) which is more complete and recent.

M. Vaïtilingom, E. Attard, N. Gaiani, M. Sancelme, L. Deguillaume, A. I. Flossmann, P. Amato, A.-M. Delort. Long-term features of cloud microbiology at the puy de Dôme (France). Atmospheric Environment, 2012, 56, 88-100.

Finally the discussion about atmospheric impacts of INMS should be completed by considering data from the literature aboutbiological IN in precipitations (snow, rain), aerosols or cloud samples. For instance Christner et al. (2008a, b) measured IN activity in precipitations on filtered samples, so such water soluble INMs might have been completely ignored. As a result the estimation of biological impact in ice nucleation process could be highly under-estimated. This is also true for modeling studies (see for instance Hoose et al., 2010) who considered only IN as whole cells, which again could underestimate the contribution of biological impact on precipitations. This list of examples is not exhaustive...

Christner, B. C., Cai, R., Morris, C. E., McCarter, K. S., Foreman, C. M., Skidmore, M. L., Montross, S. N. and Sands, D. C. Geographic, seasonal, and precipitation chemistry influence on the abundance and activity of biological ice nucleators in rain and snow, Proceedings of the National Academy of Sciences, 2008a, 105, 18854–18859.

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Christner, B. C., Morris, C. E., Foreman, C. M., Cai, R. and Sands, D. C. Ubiquity of biological ice nucleators in snowfall, Science, 2008b, 319, 1214.

C. Hoose, J. E. Kristjánsson, S. M., Burrows. How important is biological ice nucleation in clouds on a global scale? Environmental Research Letters, 2010; DOI: 10.1088/1748-9326/5/2/024009

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 24273, 2014.