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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The ice nucleation ability of one of the most abundant types of fungal spores found in the atmosphere" *by* R. Iannone et al.

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Given below are the comments from the referees and our responses. We thank the referees for carefully reading our manuscript and for their very helpful comments.

P. DeMott (Referee)

Specific Comments

Abstract

The first sentence struck me as a little too strong for the current status of understanding of biological ice nuclei as a whole. I believe that it is safe to say, at best, that biological

particles are a potentially important class of ice nuclei. I think this deserves qualification because the level of quantification of their importance at present, except by inference, is quite poor. For the same reason, I consider the statement that the results "do not appear to explain recent atmospheric measurements showing that biological particles are important ice nuclei" to similarly overstate present evidence. I suggest "participate as atmospheric ice nuclei" to replace "are important. . ."

- We agree with these suggestions and will make the following substitutions:

Replace: "Recent atmospheric measurements show that biological particles are important ice nuclei." (p. 24622, lines 2–3) with "Recent atmospheric measurements show that biological particles are a potentially important class of ice nuclei."

Replace: "...do not appear to explain recent atmospheric measurements showing that biological particles are important ice nuclei." (p. 24622, lines 13–14) with "...do not appear to explain recent atmospheric measurements showing that biological particles participate as atmospheric ice nuclei."

1. Introduction

Page 24624, lines 18-19: You might add that while number concentrations per volume of air are of first order importance, the activation spectrum (proportion active versus relevant thermodynamic conditions) is also important for establishing atmospheric relevance.

- This is a very good suggestion. We shall insert the following line:

"Furthermore, the activation spectrum (i.e., proportion of active IN against temperature and ice supersaturation) is important for establishing atmospheric relevance."

immediately following "...the order of 10(3)–10(4) m(−3) (Elbert et al., 2007)." (p. 24624, line 18).

2. Experimental

Page 24626, lines 15-16: Can you comment on whether any experiments were performed at different cooling rates and whether or not this mattered?

- Only one cooling rate, 5 K min–1, was used due to experimental constraints. Higher cooling rates were not possible with the current setup. Lower cooling rates resulted in significant mass transfer between unfrozen and frozen droplets. We will add these details to the manuscript.

3. Results and discussion

Page 24630, line 16: The authors may or may not wish to comment here on the fact that these active fractions found for fungal spores do not seem all that different than found for pure kaolinite particles of similar spherical equivalent sizes by Murray et al. (2010).

- The comparison between the work by Murray et al. and our current results is an interesting one. But we would prefer to leave out the comparison of the ice nucleation ability of fungal spores and the ice nucleation ability of mineral dust particles until a future publication.

Page 24630, lines 17 paragraph: Please clarify in words here that the data shown in Fig. 7 are either mean or median freezing temperature, whichever the case may be (terms seem to be used interchangeably in the following discussion).

- The plot of Fig. 7 does indeed use the mean freezing temperatures unlike that of the box-and-whisker plot of Fig. 8. To avoid potential confusion, the word "mean" will be inserted as "In Fig. 7, the mean heterogeneous freezing results are plotted as a function..." (p. 24630, line 17).

Page 24630, lines 24-25: To be clear, are inclusion sizes determined by tagging each drop freezing event by location and then sizing these particles, or is the inclusion size inferred from the average of all spores used in each series of freezing tests? Part of my own confusion may stem from the fact that these "heterogeneous data" data must

also be mean of median freezing temperature data. Please clarify and qualify each time the data are discussed after this point. For example, the last sentence on this page discusses an "increase in the freezing temperature," which should say "median freezing temperature."

- The inclusion sizes were determined by sizing the particles associated with each droplet. This allowed us to match size data to freezing temperatures for each droplet. The total area of all particles within each frozen droplet was determined since often the inclusions appeared as clumped spores. Then number of particles within each frozen droplet was inferred by the mean spore area, a value determined independent of the ice nucleation experiments. It was sometimes difficult to distinguish individual spores due to clumping and optical limitations in the ice nucleation experiments. The size determination experiments did not have this limitation. We do agree that the last sentence which reads "...increase in the freezing temperature..." (p. 24630, line 28) should instead read as "...increase in the median freezing temperature..." as a point of clarification.

To further clarify which aspects of temperature were intended, the following editorial changes were made.

At p. 24622, lines 8–11, we will change "However, there was a strong dependence on freezing temperature with the spore surface area of Cladosporium within a given droplet. As such, freezing temperatures for droplets containing 1–5 spores are expected to be..." to "However, there was a strong dependence between the freezing temperature and the total spore surface area of Cladosporium within a given droplet. The mean freezing temperatures for droplets containing 1–5 spores are expected to be..."

At p. 24631, lines 7–8, we will change "The dependence of spore inclusions on freezing temperature has a pronounced effect..." to "The dependence of spore inclusions on freezing temperatures has a pronounced effect..."

At p. 24634, lines 7–9, we will change “However, there was a strong dependence on freezing temperature with the spore surface area of Cladosporium for a given droplet. As such, freezing temperatures for droplets containing 1–5 spores are expected to be...” to “However, there was a strong dependence between the freezing temperature and the total spore surface area of Cladosporium for a given droplet. As such, mean freezing temperatures for droplets containing 1–5 spores are expected to be...”

Page 24632, lines 19-20: I may be cutting hairs here perhaps, but it was suggested by DeMott and Prenni that warmer temperature IN "could be" biological particles, a somewhat different assertion than proposing that biological particles do explain these IN.

- This is correct. To avoid misleading the reader, the sentence “It has been proposed that the effective IN at these temperatures are biological particles...” (p. 24632, lines 19–20) will be replaced with “It has been proposed that the effective IN at these temperatures could be biological particles...”

Page 24632, lines 22-23: How was the proportion of spores active warmer than -15C estimated ("we can expect. . .")? The fraction seems clearly less than observable directly with the experimental setup.

- We estimated this based on a linear extrapolation of the data shown in Figure 6b to warmer temperatures. A value of <0.1% for this temperature range appears to be a conservative estimate based on the data. However, to be more conservative, we will change <0.1% to <0.5%. The latter value was measured directly in our experiments.

Page 24632, lines 25-28: You might clarify that the estimate of number concentrations is an estimate for the boundary layer, and you might state exactly how much lower these concentrations are compared to IN numbers typically present at -15C (e.g., 100 to 1000 times lower).

- This question refers to the last paragraph on Page 24632. Question #2 by Referee

#2 also relates to this paragraph. To address both reviewers’ questions, this paragraph will be changed to the following:

Above -15°C, many mineral dusts become ineffective IN, although some field experiments show that some mineral dust particles are effective ice nuclei as warm as -5.2 to -8.8°C (Sassen et al., 2003). It has been suggested that the most important carbonaceous particles acting as ice nuclei above -15°C may be biological particles (DeMott and Prenni, 2010). Our data suggests that, at temperatures above -15°C, Cladosporium spores are not likely an important IN species in the atmosphere. Over this temperature range, we can expect that less than 0.5% will nucleate ice as a very conservative estimate from our data. Assuming that concentrations of fungal spores are on the order of $\sim$10 L(-1) in the atmosphere (Elbert et al., 2007), based on measurements in the boundary layer, and as an upper limit we assume that 50% of all spores are from the genus Cladosporium, it is estimated that the number of IN from Cladosporium spores is significantly less than $\sim$0.025 L(-1). This value is a factor of approximately 4 to 800 smaller than the number of IN observed in the atmosphere at temperatures around -15°C (DeMott et al., 2010).”

The following citation will be added to the references section:

Sassen, K., DeMott, P. J., Prospero, J. M., Poellot, M. R.: Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results, Geophys. Res. Lett., 30, 1633, doi:10.1029/2003GL017371, 2003.

Page 24633, lines 14-15: Should this say 0.001 percent or 0.001 as a fraction? The lowest measurement value shown in Fig. 6 is a fraction of about 0.004, so again I wonder about the 0.001 percent value and how it was estimated. Also, be careful about attributing any results to temperatures for which no data have been collected.

- The percentage sign is indeed a mistake here. The value should simply read as 10(–3). As mentioned, above 10(–3) was based on a linear extrapolation of our data to warmer temperatures. To be more conservative, we will replace 10(–3) with 0.5%, which is measured directly in our experiments.

Page 24633, lines 22: Should this read greater than or equal to 700 nm for the residual aerosol sizes not assessed by the MS? Also, by the end of this section I began to wonder a little bit about the point of comparison to two studies that had limited detection capabilities for the primary mode-size of single spores. Is the point that the atmospheric studies might reflect the action of some other type of biological particles, just not spores? The point should be made clearer in any case.

- The less than or equal to sign is indeed an error. The comments made by this referee concern the last two paragraphs on p. 24633. Question 3 from Referee #2 also asked questions about these paragraphs. To address these questions we have rewritten the last two paragraphs on page 24633. The revised paragraphs are as follows:

In a recent study by Pratt et al. (2009), ice residuals collected in situ from cloud particles at -31 to $-34°C$ contained a significant fraction of biological material. There was, however, a notable size cutoff: ice residual particles >700 nm were not admitted to their MS instrument (for identification of biological markers within individual particles; a total of 46 particles were examined). The number of intact spores with aerodynamic diameters less than 700 nm in the atmosphere is likely small (Hameed and Khodr, 2001; Reponen et al., 2001; Jung et al., 2009a; Fröhlich-Nowoisky et al., 2009). Hence, some other biological material, besides intact fungal spores, must have been responsible for the observations by Pratt et al. (2009).

A recent study concerning biological IN in the wet season over the Amazon rainforest has demonstrated that the level of atmospheric IN can be predicted through measurements of a combination of mineral dust and biological particles (Prenni et al., 2009). To explain their data, Prenni et al. had assumed that the sampled biological particles could induce ice nucleation with an efficiency of $\sim$0.2 for temperatures between -18 and $-31°C$. At temperatures above approximately $-25°C$, biological particles appeared to dominate. The size range of IN measured by Prenni et al. was $\sim$1.3 μm in

aerodynamic diameter. Some species of fungi can produce spores in this size range, but the fraction of Cladosporium spores in this size range is very small. In addition, for Cladosporium, less than 0.5% of the droplets were observed to freeze at temperatures above $-25°C$ according to Fig. 6. Hence, Cladosporium spores cannot explain the observations by Prenni et al. Some other type of biological material must have also been active as ice nuclei in these studies.

Editorial comments

Introduction, page 24622, line 25: I suggest stating that freezing occurs “initially” by heterogeneous nucleation, to distinguish the fact that freezing in clouds includes other secondary processes not involving nucleation.

- We agree with this recommended change and so the word “initially” will be inserted as “...freezing occurs initially by heterogeneous nucleation...” (p. 24622, line 25).

Introduction, page 24624, line 27: Please explain the meaning of the terminology "passively launched."

- This indeed requires some explanation. Will modify the statement “The spores of Cladosporium are passively launched and they have mean aerodynamic...” (p. 24624, line 27) with this: “The spores of Cladosporium are passively launched (i.e, separated from the mycelium via wind currents) and they have mean aerodynamic...”

Results and discussion, page 24629, lines 19-20: “The freezing results for pure water droplets are consistent with results expected for homogeneous freezing.” The sentence is repetitive with statements made earlier in this paragraph and is probably not necessary. In the prior sentence I suggest clarifying as “classical homogeneous nucleation theory.”

- The sentence “The freezing results for pure water droplets are consistent with results expected for homogeneous freezing.” (p. 24629, lines 19–20) will be removed as it does essentially repeat the earlier phrase. Also, “...temperatures expected for homogeneous freezing.” (p. 24629, line 15) will be replaced with “...temperatures expected when considering classical homogeneous nucleation theory.”

Experimental, Page 24631, last sentence: Phillips

- “Philips” will be changed to the correct spelling as “Phillips.”

Anonymous Referee #2 Received and published: 3 January 2011

I do have 3 points that I would like the authors to consider in their revised manuscript.

1. I echo Prof. DeMott’s “general specific comment” regarding the abstract: “The first sentence struck me as a little too strong for the current status of understanding of biological ice nuclei as a whole. I believe that it is safe to say, at best, that biological particles are a potentially important class of ice nuclei. I think this deserves qualification because the level of quantification of their importance at present, except by inference, is quite poor.” Indeed, I do not know of a comprehensive study that shows biological particles to be of high abundance either in the atmosphere in general or in ice forming particles specifically. There are several recent publications on this topic (correctly referenced here) but, to repeat the above term, all appear to “infer” as opposed to “show”. Since this publication largely details a “negative result” (these abundant biological particles don’t appear active participants in ice formation) I think this result needs to be made more clear and not only the abstract but the paper in general needs to make it clear that biological material as ice nucleators is not a fate accompli.

- We agree and will make editorial changes as recommended above regarding the potentiality of biological particles as efficient IN.

2. Following up on this point the Introduction seems to suggest biological material is the only possible ice nucleator above -15 deg C. I was surprised that publications that have seen mineral dust form ice clouds in this range were not included. Specifically absent is Sassen et al., Saharan dust storms and indirect aerosol effects on clouds: CRYSTAL-FACE results, GRL 2003 (which found mineral dust acting to form ice at -5

deg C) and references therein.

- This is a good point raised by the referee. To address this point we will change the first paragraph in the Atmospheric Implications section. See details above.

3. I found several parts of the paragraph starting “In a recent study by Pratt et al. (2009). . .” on page 24633 to be confusing and in need of a rewrite. (1) After reading this reference I think I understand that only ice nucleating particles from 140–700 nanometers were considered and these from ice crystals only greater than 7 micrometers (an upper limit was not given in that paper which appears an omission). A CFDC flow chamber with a 1.2 micron limit was mentioned in that paper but it did not appear this was involved in the attempted identification of biological ice nucleators using a mass spec (MS) instrument. I believe the authors need to check their wording: I don’t think the 1.2 micron cut size was important and I think the MS instrument looked at particles smaller (not larger) than 700 nanometers. (2) Also after reading this publication I was rather surprised it only considered something like 40 total particles (some 10 identified as biological) collected over some 20 minutes on one fall day in one location in the US. I think this paucity of data should be mentioned here since the authors spend such a large part of their Atmospheric Implications section discussing it.

- We will rewrite this paragraph to address both referees’ comments (see above).

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 24621, 2010.