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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contribution of pollen to atmospheric ice nuclei concentrations" *by* J. D. Hader et al.

Anonymous Referee #2

Received and published: 29 January 2014

General comments

The above mentioned article presents recent ice nuclei (IN) measurements during the pollen season 2013 in Raleigh, North Carolina, USA. Ambient samples were collected using a swirling aerosol collector and rainwater collection for analysis with a droplet freezing array. The authors investigate possible correlations between pollen concentration and ambient IN spectra obtained from experiments to compare with previous bioaerosol studies concerning this topic. Additional reference experiments with local long leave pine pollen as a major source and ATD are presented to support their technique.

Assuming an abundance of pollen in the ambient sample, such a clear correlation could

not be observed. Furthermore a source of pollen multiplication processes is suggested to be unlikely for this study. The authors conclude the contribution of pollen on a global scale to be relatively low compared to background signals in such IN experiments. A low atmospheric relevance of pollen as IN is expected to be due to the low pollen number concentrations, the seasonality and the vertical distribution in the atmosphere.

The present paper addresses the need to investigate the atmospheric relevance of pollen as natural ice nuclei, which is of increasing interest in the community. Beside the novel experiments on ambient pollen as potential IN in the immersion mode, I question the procedure used to test the hypothesis the authors put forward. The reason for this concern is the processing of the ambient samples prior to measurement (for details see below). Thus, I only recommend this paper for publication in Atmospheric Chemistry and Physics after the following remarks are included:

Specific comments

Abstract:

Line 2-5: Do you suggest that IN active macromolecules are being dispersed during drying as well?

Lines 6-8: The 2013 pollen season
How do you define the local pollen season (peak number concentration, pollen species,...)?

Lines 24-26: Please explain the self-regulated feedback cycle between the atmosphere and biosphere via the release of cloud forming particles.

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Line 4: How does a change of the cooling rate affect the experiment and why was this cooling rate chosen?

Line 13: Better to explicitly define 'median [freezing] temperature' and add the word

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'freezing' between 'median' and 'temperature' for consistency.

Line 25: Please add that the larger droplets freeze at higher temperatures because of the increase in ice nucleation active sites.

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Line 6-8: Can you rule out that the ice activation of pollen is via macromolecules as suggested by Pummer et al., 2012 and Augustin et al., 2013 if you restrict yourself to sampling above 200nm?

Line 13; Table 1: Sample periods are summarized in Table 1, where the reader can find sampling at different conditions and also variations in the diurnal time of sampling. In contrast to this, the ambient pollen concentrations are reported as 24h average pollen concentration. Can you exclude diurnal variations in pollen concentrations?

Line 17-20:
If you freeze and thaw the samples, could that cause pre-activation? Or could that destroy the viable biological particles?

Line 22-25: The authors describe the process of filtering and re-suspending rainwater samples for which a 0.2μm filter has been used and the filtrate removed before analysis. This is in contrast to previous immersion freezing studies on pollen (Pummer et al., 2012; Augustin et al., 2013) where the IN active particles were found in the filtrate of commercial pollen grains and therefore did not contain the whole pollen grains. How can the authors conclude that no correlation between pollen and IN concentration is found when excluding parts of the sample which might contain potential IN?

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Line 1-2: How did you estimate the re-suspended fraction?

Line 21-24: Only particles with diameter $>$10μm are considered as pollen here. Multiplication processes (release of IN active particles e.g. by rupturing) are not taken into account to estimate the pollen concentration. Is the pollen grain number concentration

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thus identical to the particle concentration that has to be taken into account to estimate its IN ability? How do the authors conclude this? Can you rule out a possible influence of other IN?

Page 31682
Varying cooling rates....only leads to a shift of a few degrees K in the population median observed freezing temperature is a lot. Please justify why this is negligible.

Page 31688
Line 9: '...at least one nucleation site per grain...' is only true for Birch pollen measured by Pummer et al., 2012 shown in Figure 6, but not for any other pollen measurement. Please correct

Line 11: 'IN activity of pine pollen .... are well within the range of results obtained from previous studies,...'
Name the studies which you are referring to as Figure 6 does not contain reference data for pine pollen.

Line 20: How can the authors conclude from Figure 6 the 'generic pollen grain' induces ice formation at T$<$-20°C? This only seems to hold true for your measurements and thus should be stated as such.

Page 31693
Line 22-25: The authors conclude that 'pollen only accounted for a fraction of the observed IN concentration'. What else do the authors expect to be activating as IN in their samples?

Page 31696
Line 12: The authors should explain why the observed IN burst in their study is based on biological origin without chemical analysis or size distribution measurements that could distinguish larger bioparticles from other sources (e.g. mineral dust).
Furthermore, I think direct comparison with results from the BEACHON-RoMBAS cam-

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paign is difficult, because this campaign was conducted in a different season and climate zone (as mentioned by the authors), which can result in different vegetation and therefore resulting in different pollen species.

Technical corrections

Droplet size is often given in volume. A droplet diameter would be helpful for comparison with previous publications on immersion and droplet freezing techniques (e.g. Pummer et al., 2012; Augustin et al., 2013; Diehl et al., 2002), which are referred to in the paper.

Page 31675
Line 22: The phrase 'pollen particles' should be replaced by 'pollen grains' as it refers to the whole grain without bursting or multiplication processes taking into account.

Page 31692
Line 25: Change 'been' to 'be'.

Figures:

In general I would like to suggest color coding for the figures to help the eye of the reader. Additionally, the authors should check figures for consistency (e.g. x-axis labelling for 'Freezing temperature' and Temperature' as well as 'IN' and 'IN concentration'). Details concerning the figures are as follows:

Figure 3
Due to the number of measurement points it would be helpful to color code the figure to support the reader in finding the main result of this figure (e.g. filtered vs. unfiltered experiments; nanodrops vs. picodrops). This statement applies to the following figures as well. The 'solid black line' mentioned in the caption in reality is 'grey'.

Figure 4
The top edge of the figure is cut and the label 'homogeneous freezing of pure water' is

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not completely visible.

Figure 5
Font and symbol size should be magnified in both panels.

Figure 6
I suggest to color the different data points.

Figure 7
Color coding the three y-axes would be much easier for the eye and would make labelling in the plot redundant.

Figure 8
The figure is too small and not readable. Please also indicate the unit (exponent) for IN number per sample in the graphs y-axis labels directly instead of in the caption only. Think of maybe combining all the daily data in one plot.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 31673, 2013.

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