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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Complex plant-derived organic aerosol as ice-nucleating particles – more than a sum of their parts?" *by* Isabelle Steinke et al.

Anonymous Referee #1

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The manuscript uses two different techniques to examine INP number concentrations for a number of plant and soil derived samples, namely an expansion chamber, AIDA, and a cold stage method, INSEKT. Measurements were certainly conducted under the necessary care, and the results as such are interesting. However, unfortunately, the impression arises that the text was written in a hurry, still trying to give the impression that the data are really important, and in this course, overlooking topics that should have been discussed in much more detail if claims as those made in the text are to be justified.

My main criticisms are: a) the effect of micro-organism as an INP is largely not dis-

cussed in the interpretation of the retrieved results, although for leaf litter and the agricultural samples, it cannot be excluded that this plays a role; b) the amount of plant derived INP in the atmosphere is derived without properly motivating all the parameters used in the calculation, which, however, will largely influence the atmospheric importance of these respective INP.

More specifically, it has to be said that throughout the text, there is a somewhat strange shortage of mentions of biogenic INP, which, based on their nature, have to be connected to the herein examined plant material samples. In the absence of any physical or chemical test that could have altered the respective samples (e.g., heating, treatment with H2O2), there is no information about the nature of the INPs, therefore any observed activity in the agricultural sample and the leaf litter may originate from the sample material itself or from micro-organisms connected to it (bacteria, fungi, lichen). The overall text has to be revised in connection to this issue.

Also, for all of those samples from plant-related organic compounds, the sources for the cited quantities for agricultural dust and leaf litter have to be explained in more detail. These numbers are used for a very general calculation afterwards, but the reader needs to know if the given concentrations are valid only locally or for a larger area or worldwide and in which altitudes. A mentioning of "occasionally for very strong wind events" (line 256) rather gives the impression that these values are of no general use in this context, and more information is needed. Also some mentioning of the processes that make particles from dust and leaf litter airborne should be added.

These issues and others to be found in my comments below makes a thorough revision of the text necessary before it can be accepted for publication in ACPD.

Specific comments:

line 46: "biological aerosol particles" come a bit out of nowhere, here, and it would be good to shortly mention first, that these, too, can be efficient INP (as done for the mineral dust in the previous sentence).

line 66: Your sentence “decaying plant material being one of the major sources of these macromolecules (Hill et al., 2016),” gives a somewhat wrong impression, as (from what I understood from the literature you cite here) these macromolecules may also originate from biological entities. In their abstract, it says: ”Organic INPs . . . may originate from decomposing plant material, microbial biomass, and/or the humin component of the SOM.”

line 114: It should be added that the identification of an hydrometor as ice is simply based on its size.

line 167: These individual plant-related organic compounds likely miss the contribution of micro-organisms, so it is not so astounding that they show a comparably low ice activity (“in comparison to samples from natural environments”). A discussion on this is missing completely from the text.

line 175: There is only a VERY low number of points for these "complex polysaccarides" at these higher temperatures (one, one, three, for three different ones of these samples (where the one with the three points seems to consist of two different materials, ALG and PEC)). - Particularly the three dots for ALG/PEC seem to be rather a background, as there is no change in the signal over the temperature range from 254 to 259 K, making this really look like a background issue. Interpreting this as a "weaker dependence on temperature" over-interprets what can be learned from these few data-points.

line 188 ff: The abstract of Conen et al. (2016) says: ”Together, both findings suggest that decaying leaves are a strong emission source of IN to the Arctic boundary layer.” And they examined litter consisting “of entire leaves and large fragments thereof, mainly from Betula nana and various grasses.” – I wonder how this fits to what you wrote: “. . ., leaf litter from the Arctic consisting mainly of grass leaves has been observed to show relatively low ice nucleation efficiencies (Conen et al., 2016), . . .” . Neither seems grass to be the major component of the litter examined by Conen et al. (2016) (or

did I overlook something in the Conen-study?), nor do they claim that this is a low ice nucleation efficiency. Temperature wise, data in Conen et al. (2016) only go down to -15°C, while the temperature range into which your results fall are generally below that, which has to be acknowledged when comparing values. Please revise this part of the text.

line 208-209: Is there any explanation why for the two samples discussed above large particles were not a problem and then why they might be a problem here? How else does this one sample differ from these two samples above?

212-213: Why should there not also be a contribution from micro-organisms (= biological particles) in the leaf litter sample? After all, originally it was leaves from which Pseudomonas syringae was derived (Maki 1974). While the temperature of the signal’s increase does not suggest the influence of a bacteria such as P. syringae, the presence of other micro-organisms could still be responsible for this increase. Testing with e.g., heat or acids or other methods could have increased the understanding.

line 248/249 and line 254 ff: The explanation on how INP concentrations were obtained, i.e., the atmospheric concentrations assumed for leaf litter and agricultural soil dust, needs to be given first, before referring to the derived INP concentrations shown in Fig. 3. Also, it should be discussed in more detail that these indeed are upper boundaries (you mention “very strong wind erosion events”). And also, please motivate the aerosol surface area (1 m2/g). This would need to be the surface area ascribed to leaf litter or to the dust, exclusively. Is this a reasonable value? This needs to be discussed. Also: the mass concentrations: are they to be expected over larger areas, or were these just measured near sources. This is important if you want to make statements on atmospheric wide INP concentrations and the importance of leaf litter and agricultural soil dust in this context.

line 265: Could it be that this sentence is based on using unrealistically high atmospheric concentrations of materials that were examined and then using this as an argument for the importance of the present study? For enabling your readers to judge that, motivate the 1 m2/g that you used above, and the mass concentrations of leaf litter and dust, as said above.

line 267: The “Section 4: Conclusion” comes a bit abrupt and contains statements that should either be given in a discussion part or otherwise earlier – giving new points of discussion (as the different sources that may emit organic particles or the global extent of the areas that may contribute) is not something that should appear in the conclusions for the first time.

line 277: Why were INSEKT data not also used to obtain atmospheric INP concentrations? As you say here, the measurements with this instrument were done to get results on a wider temperature range, but then they are not used in the further evaluation. Maybe it could also be seen if AIDA or INSEKT give the more trustworthy data? At least this could be discussed when comparing to the data from literature.

line 282-284: This sentence might have to be revised if the used values for mass concentration and aerosol surface area are revised or cannot be well justified.

line 284-286: Here it is again not clear why macromolecules from micro-organisms were not considered, as they provide an obvious explanation.

Figure 3: As also already mentioned above, I wonder if you want to imply here that the atmospheric INP mostly come from leaf litter and agricultural dust? There is a lot of literature around that ascribes ice activity at temperatures of roughly < -20°C to desert dust, so I wonder if you really want to challenge this. If you feel your data is strong enough to do that, come up with a good justification. If you don’t trust your derived concentrations so much, make it clearer in the text that you present the absolute possible maximum and that likely values from the lower end of the ranges you give or even blow are more likely.

Technical comments:

line 51-52: Check the style of the citations (no brackets should be used in a bracket) Table 1: Somehow a “555” shows up at the right side of the table. Probably a line number gone wild? Figure 3: “.” is missing at the end of the caption.

Literature:

Conen, F., E. Stopelli, and L. Zimmermann (2016), Clues that decaying leaves enrich Arctic air with ice nucleating particles, Atmos. Environ., 129, 91-94, doi:10.1016/j.atmosenv.2016.01.027.

Hill, T. C. J., P. J. DeMott, Y. Tobo, J. Froehlich-Nowoisky, B. F. Moffett, G. D. Franc, and S. M. Kreidenweis (2016), Sources of organic ice nucleating particles in soils, Atmos. Chem. Phys., 16(11), 7195-7211, doi:10.5194/acp-16-7195-2016. Maki, L. R., E. L. Galyan, M.-M. Changchi, and D. R. Caldwell (1974), Ice nucleation induced by Pseudomonas-Syringae, Appl. Microbiol., 28(3), 456-459.

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