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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The immersion freezing behavior of mineral dust particles mixed with biological substances" *by* S. Augustin-Bauditz et al.

Anonymous Referee #2

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General comments

This paper presents a study of the generation, ice nucleation ability, and modelling of an internally mixed-aerosol composed of mineral dust and water soluble biological material. The mineral dust chosen in this case (NX-illite) has been used extensively in the past as a surrogate for atmospheric mineral dusts. Many biological materials such as the Birch pollen washing water (BPWW) presented here have been previously shown to exhibit significant ice nucleation activity though few studies have examined a combination of the two. The authors use of the recently developed modified Soccer Ball Model (SBM) for parameterizing internally mixed aerosols based on parameters

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derived from the pure components is novel. However, there are several issues which need to be addressed before publication.

A significant amount of the manuscript deals with the characterization of the mixed-aerosols used in the freezing study. The authors utilize multiple techniques to characterize the mixing state of the aerosols though only two of the techniques (humidity and volatility growth factors) provide any conclusive evidence as to the mixing state. The other techniques while not contradicting the growth factor results do not provide much additional information on the mixing state. I would suggest reworking these sections to remove unnecessary details which make the paper more confusing while not adding any additional information. The SEM and EDX sections, for example, could be removed or shortened as they provides little additional information.

Secondly, the reader is left confused when going through the paper as to whether the pure particle freezing results (illite-NX and BPWW) were performed for this study or are simply reproduced from previous papers. For example, Table 3, clearly states that the SBM parameters used were taken from the literature while page 29655, lines 6-9 indicate that a different sample of BPWW was used for this study. Clarification throughout the manuscript as to which of the measurements were made for this study is necessary before publication.

Finally, the results presented here only use a single particle size (500nm) and a single coating thickness. While it is useful to show that the modified SBM is capable of predicting the freezing results of a monodisperse sample of internally mixed particles such as those presented here, additional measurements showing the model's capability with different particle sizes or relative amounts of illite and BPWW would significantly enhance the conclusion that the SBM can be used to predict the freezing behaviour of mixed aerosols as presented in the manuscript. While not essential to the publication of this manuscript, I strongly suggest expanding the laboratory results presented here.

Specific comments

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page 29641, lines 8-9 – The references here refer only to *P. syringae*. Additional references showing the ice nucleation ability of biological particles should be added.

page 29642, line 4 – Provide a reference for the size of INM (10nm).

page 29642-29643, lines 26-1 – How was the concentration of illite-NX determined from the suspension? Please add detail regarding how these measurements were performed.

page 29643, lines 2-3 – A comparison of the freezing ability of suspended illite-NX particles with those of dry generated particles is discussed though no results are presented. Are these measurements performed using size-selected particles or the full spectrum of generated aerosols? I recommend adding a figure showing the freezing comparison in the supplemental information so that the reader can see for themselves that the results are the same.

page 29643, lines 11-12 – The authors should indicate the pore size of the filters used to remove the pollen grains and comment on whether or not they expect any solid material to pass through the filters.

page 29643, lines 12-13 – Similar to above, detail should be added indicating how the Swedish birch pollen concentration was determined.

page 29644, line 13 – Since the point is made regarding doubly charged particles, the size cut (D_{50}) of the cyclone used should be provided to indicate that these particles are unlikely to be present.

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page 29646, lines 11-12 – While it is obvious why the C/Si ratio would provide some measure of the relative amounts of biological material and mineral dust in each particle, no explanation is given as to why a factor of 10 was chosen as the cutoff (ie. why a C signal greater by a factor of 10 indicates a purely biological particle and similarly why a Si signal greater by a factor of 10 indicates a purely mineral dust particle). The authors should add an explanation as to why these values were chosen. Would this technique work using different chemical tracers (eg. C/Al ratio)?

page 29647, lines 1-13 – Similar to the above comment, the authors should provide a reference or a rationale for using the intensity ratio (Na/(Na + SiO)) as a metric for biological versus mineral dust particles. Additionally, results presented in the supplemental information suggest that many pure BPWW particles have intensity ratios between the chosen cutoffs of 0.1–0.65. Justification should be provided for the chosen cutoffs.

page 29649, lines 1-4 – No mention is given of the range of expected coating thicknesses. While the growth factors for pure illite paritcles are quite narrow in distribution, the growth factors for the pure BPWW and the hygroscopic growth factor for the mixed particles is broader. This would result in a range of κ values as well as a range of possible coating thicknesses. The authors should provide an estimate of the spread in coating thicknesses used.

page 29649, lines 11-13 – An indication of the number of individual particles observed for the determination should be given to indicate the statistical validity of the statement made here.

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page 29651, lines 6-8 – All of the methods used to characterize the particles do not indicate that the particles contain both biological material and mineral dust. The SEM and EDX results could not identify any biological material on the mixed particles. This statement should be rewritten to make this clear.

page 29654, lines 3-4 – The results presented here and in Figure 4 indicate that the pure illite-NX and pure BPWW particles do not reach an $f_{\text{ice}} = 1$ at the lowest temperatures measured. Results for the illite-BPWW mixed particles attain $f_{\text{ice}} = 1$ below 38°C which the authors indicate is the onset of homogeneous freezing. Why was the homogeneous onset not observed for the pure particles? Were measurements not made at these temperatures or are the homogeneous results removed from the dataset presented? This should be mentioned in the manuscript.

page 29656, lines 2-4 – It is unclear here if measurements were made with polydisperse illite-NX particles and the value of $\lambda_{\text{illite}}(D_p)$ was determined in the present study or if this is taken from the literature.

page 29656, line 4 – The word "resulting" suggests that the values of μ_θ and σ_θ are determined from the value of λ_{illite} while the caption to Table 3 indicates that the values of μ_θ and σ_θ are taken from Augustin-Bauditz et al. 2014. Please clarify which measurements are made in the present study and which are taken from the literature.

page 29658, line 4 – The authors present the values of λ used for the illite-BPWW mixed particles. The specific values of λ used for both the illite-NX and BPWW pure particles fits as well as the completely dissolved BPWW case should be included as a comparison for the reader.

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Technical corrections

page 29640, line 11 – “INUIT” should be defined in the manuscript.

page 29641, line 12 – Remove “e.g.,”

page 29641, line 25 – Define “INP”.

page 29646, line 15 – Replace “illit” with “illite”.

page 29649, line 27 – This was presented as C/Si on page 29646.

page 29651, line 8 – Remove comma after “both”.

page 29651, line 20 – Replace “straight” with “solid”.

page 29651, line 21 – Replace “In the next section, it will be described” with “The next section will describe”.

page 29652, line 18 – Delete second “a”.

page 29652, line 20 – Integration limits in equation (2) should be $\int_0^\pi$ for the first term and $\int_\pi^\infty$ for the third term.

page 29655, lines 3 & 5 – The dash before λ_α makes it appear as a negative symbol.

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This should be removed.

page 29655, line 7-9 – This sentence should be rewritten for clarity.

page 29657, line 3 – No comma after “both”.

page 29657, line 13-25 – Multiple instances of “case a” or “panel c” the a, b, and c should be identified in brackets as done in Figure 5 to make it easier to read.

page 29658, line 4 – Remove dash before λ_α.

page 29658, line 14 – Remove commas after “both” and “material”.

page 29658, line 24 – This sentence should be rewritten for clarity.

page 29664, Table 1 – The authors should indicate in the caption or the text that the fits for the grown factors determined are log-normal distributions. Additionally, the spread should be defined as either the standard deviation or the variance of the log-normal distribution.

page 29668, Figure 2 – “Left part” and “Right part” should be replaced with “Panel a” and “Panel b”.

page 29670, Figure 4 – Replace “an mobility diameter” with “a mobility diameter”.

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Supplemental information – Provide detailed captions for the figure and table. The histograms should have a labelled vertical axis. Table headers should be presented in English.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 29639, 2015.