Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-915-RC1, 2018

Atmospheric Chemistry and Physics Discussions
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Interactive comment on "Laboratory study of the heterogeneous ice nucleation on black carbon containing aerosol" *by* Leonid Nichman et al.

Anonymous Referee #1

Received and published: 23 October 2018

*General comments: The authors conducted careful and dedicated lab experiments. The manuscript is well structured and carefully written to derive a delicate conclusion (i.e., P13L14-22). The research topic is an important addition to ACP. Though some parts seem speculative, the authors are knowledgeable in the subject, and their findings warrant future follow up studies. I support publication of this manuscript in ACP after the following comments (some are major/critical; e.g., P12 comments) are properly addressed.

*Summary: The authors studied the ice nucleation behavior of particles generated using diverse, commercially available BC materials and an ethylene combustion product in the temperature and SSi range of 217 - 235 K and 1.0 - ∼1.5, respectively. In particular, six BC materials (possessing different physicochemical properties as experimental

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Discussion paper

variables) were used to look into the relationship of their IN abilities vs. morphologies, aerosolization methods, sizes (100 and 800 nm), degrees of surface oxidation and organic surface coating types under controlled settings. Out of these variables, the morphology and a subset of surface coatings seem dominant factors altering IN propensities of BC particles according to the results presented. The reviewer finds the notable suppression of BC-IN via oxidation and the tolerance (no substantial deformation) of BC-IN to the employed particle generation methods (i.e., both wet & dry dispersion) very interesting and informative, and their findings should be shared in the IN research community and beyond.

*Specific and technical comments: P1L16: It should read "...human health, aerosol-cloud interactions, precipitation formation, and climate.". Without aerosol-cloud interactions and precipitations, there is no climatic impact.

P1L19: It should read "...formation of ice crystals in temperature and ice supersaturation conditions relevant to cirrus clouds.".

P1L21 and elsewhere applicable: ice nuclei (IN) should be replaced with ice-nucleating particles (INPs) to be consistent with the common terminology typically used in the IN research community (Vali et al., 2015, ACP; https://doi.org/10.5194/acp-15-10263-2015). The authors can use IN for the abbreviation of 'ice nucleation' (e.g., P2L7).

P1L29: The reviewer suggests the authors to report/add SSi and SSw ranges used in SPIN alongside the T range here.

P1L34: "...dependence on temperature, supersaturation condition, and...".

P1L36: "initial ice nucleation ability" → "initial formation efficiency of pristine ice crystals" The authors provided an observational hint, but not any nanoscopic evidences of oxidation altering IN ability.

P2L7-9: The reviewer suggest the authors to expand the discussion of the indirect effect of soot/BC particles here. Perhaps, the discussion of Bond et al. (2013) in Sect.

4 better fits in here?

P3L7-9: “Modeling INPs requires . . . description (Knopf et al., 2018).” → “Two common approaches to parameterize IN of atmospherically relevant particles include a stochastic . . . description (e.g., Knopf et al., 2018).”

The reviewer presumes the authors mean parameterization by modeling. There exist many more papers should be cited here.

P3L9-10: Add Connolly et al. (2009; Atmos. Chem. Phys., 9, 2805–2824, 2009) and Niemand et al. (2012; DOI: 10.1175/JAS-D-11-0249.1) in addition to Vali (2014).

P3L11: The n_s abbreviation has been already given in L9.

P3L14-15: Cite at least Murray et al. (2012, Chem. Soc. Rev, 41), Vali (1994, J. Atmos. Sci., 51) and Vali (2014, ACP, 14) for references of time-dependence approach.

P3L19: (Marcolli, 2014 and 2017; Wagner et al., 2016; Ullrich et al., 2017) should be placed at the end of L19 instead of L20.

P3L24-28 & P9L16: So what is the pore size, which influences condensation process? The reviewer wonders if the authors can be a bit more quantitative than ‘on the order of nanometers (L27)’ on this discussion.

P4L13: The reviewer suggests the authors to add SSw and SSi ranges in addition to the T range.

P6L4 & P10L24-: What could be the potential influence of acid suspension generation process on IN activity? It can be another variable, correct?

Fig. 2 & P6L16: DMA should replace SEMS in Fig. 2, unless the authors measured size distribution of size-selected BC particles and are willing to present those data.

P6L18-20: Please include a discussion of why these three particular organic acids were used in this study. Atmospheric relevance? Please justify with proper citations.

P7L12: Please clarify how you estimated a geometric volume from a 2-D image.

P7L33: The reviewer presumes that SPIN was operated in the way of scanning SS from low SS to high SS at fixed T until the ice active fraction (AF) of 1% was observed. If so, please state so in the text. If other operational procedures were employed for this study, please describe and include them in the text.

L8L1: Maybe ". . ., a correction factor, Lf in Eqn. 1, (5.8) is . . ." – the authors may want to remind the reader that this correction factor is relevant to what's discussed in the previous section.

L8L16: Can the authors please explain experimental error bars in the text or figure caption more quantitatively? How did the authors estimate these uncertainties. They seem larger at colder T and lower SS. What is causing it?

P8L21: Maybe ". . . other BC particle types all exhibit heterogeneous freezing abilities below Koop line (= homogeneous freezing and water saturation line in Fig. 3)." Is this what the authors mean? The relationship between the ice nucleation onset SSi and T is known as "isoline" (see Fig. 19 of Hoose and Möhler, 2012, ACP). The authors may explain it here for the reader to understand your point properly.

P9L5-12: Very nice results.

P9L27-28: Was the particle effective density measured using atomized- or air dispersed particles? Or the authors tried both individually and confirmed no difference? The reviewer is not asking any additional measurements. Please just address what has been done.

P9L31-37: Briefly describe the shift in polarization (i.e., s1, p1) and tell the readers what it physically means in the context of your study. Elaborate a bit further with including proper references.

P10L1-7: This part is interesting – the tolerance of BC particles towards compaction is a very unique feature of BC as compared to other compositions (e.g., dust surrogate, Sullivan et al., 2010, AS&T, 44). The authors might want to address this point. The reviewer thinks that adding this only strengthens the paper.

P10L15-16 & L20-21: How does the n_s value of 800 nm ethylene combustion BC particles compare to that of 100 nm ones? The IN "efficiency" is perhaps similar? Please discuss it within this section.

P11L6-8: Speculative, but it is good as is.

P11L15: What would you suggest on how such a "through characterization" can be done? Are there any specific techniques/methods currently available for the nanoscopic surface characterization while cooling?

P11L16-27 & P12L26-27 & P13L21-22 & P1L32: What is the overall atmospheric implication of the observed results/differences depending on the type of organic coating? For instance, the enhancement of IN due to oxalic acid coting matters in what occasion/situation?

P12L1-2: Comparing AF onset of 1% to n_s is the apples-to-oranges comparison. The authors can covert 1% AF to n_s in Fig. 9 using Eqn. 1, and delineate their own "n_s isolines". For more information regarding the isoline, the authors may refer to Hiranuma et al., 2014 (Atmos. Chem. Phys., 14, 13145–13158, 2014) and/or Hoose and Möhler, 2012 (ACP). The authors might want to revise Fig. 9.

P12L7-10: Fig. 1-7 (b) of Kanji et al. (2017) represents a compilation of immersion and/or contact mode freezing data. Fig. 1-7 (a) of K17 is for the deposition data. Why do the authors compare their deposition ice nucleation data to Fig. 1-7 (b) instead of (a)?

P12L7: Why do the authors choose to estimate n_s "at the lowest measured temperatures"? If the authors wish to compare their data to K17 Fig. 1-7 (b), they should estimate the n_s values at the nearest point of water saturation (i.e., Koop line). Note that immersion freezing can be considered part of isolines (Hiranuma et al., 2014; Atmos. Chem. Phys., 14, 13145–13158, 2014). The reviewer suggests revising Fig. 10.

P12L34: The reviewer suggests adding the discussion of the atmospheric implication of what the authors found for the effect of oxidation here.

P13L18-19: See my comment for P10L15-16 & L20-21. Can the authors add the statement regarding IN “efficiency” to the IN activity statement?

P13L26-29: And the IN characterization at low Ts of the cold cirrus T regime.

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