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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Effect of photochemical aging on the ice nucleation properties of diesel and wood burning particles" *by* C. Chou et al.

Anonymous Referee #1

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This study reports on the ice nucleation efficiencies of soot particles stemming from two types of Diesel combustion engines and biomass burning particles generated from a modern log wood burner for temperatures from -30 to -40°C. Photochemical processing of these particles was achieved by irradiation of the gas and particle sample and by introduction of α-pinene. Ice nucleation experiments in the deposition and condensation freezing modes were conducted with the Portable Ice Nucleation Chamber. Wood burning particles seem to be more efficient ice nuclei (IN) inducing ice formation at -40 and -35°C whereas soot particles initiated ice nucleation only at -40°C. It is found that photochemical processing did not affect observed ice nucleation behavior significantly compared to non-aged particles. Only introduction of α-pinene and presence of irradiation, altered soot particles in such a way that ice nucleation was enhanced at -35°C. The authors suggest that the increased particle size due to condensation of

α-pinene oxidation products onto the soot particles may be the responsible factor in the observed enhanced ice nucleation efficacy rather than changes in the OC (organic carbon) to BC (black carbon) ratio.

This manuscript represents work on anthropogenic derived organic particles and their potential to affect the ice phase under mixed-phase cloud conditions. The study fits well within the scope of ACP. However, I have a couple of minor issues the authors should address before publication of this manuscript can be recommended. In particular, I find the conclusion that the particle size is the governing factor to explain observed enhanced ice nucleation efficiencies and not the OC to BC ratio or other parameters not entirely convincing.

P. 14699, l. 28: I believe it should be “supersaturated” vapour phase?

P. 14700: The review by Karcher is mentioned as a reference for ice nucleation experiments performed on soot particles. However, there are more recent publications which also should be mentioned such as Koehler et al., PCCP, 2009 and Friedman et al., JGR, 2011.

P. 14702, l. 16-20: How does dilution affect the condensed and gas phase species. Here I am referring to studies by the Donahue and Robinson groups showing that dilution can lead to enhanced volatilization. Please discuss these issues.

P. 14703, l. 7-13: The caveat should be mentioned that actual size and surface areas of sampled particles may deviate significantly from derived mobility diameter due to the fractal geometry of soot particles. Also, more details on how OC to BC ratios have been derived should be given. It should be clearly stated that polydisperse aerosol are introduced into PINC. What are the employed aerosol number concentrations?

P. 14705, l. 23: Does this indicate that a potential coating was not detected by the SMPS system? Please clarify.

P. 14705, l. 27: It seems odd that suddenly Fig. 7 is mentioned without having dis-

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cussed Fig. 6. Please change figure numbers according to the sequence as they appear in the text. This affects all remaining figures.

P. 14707, section 3.3: I think more is needed to convince the reader that an increase in size is the governing parameter triggering ice nucleation. An increase in particle diameter from 130 to 180 nm enhanced the ice nucleation efficiency shifting the activated fraction at 0.001 from about 113% to 104% RH. Can this be explained by nucleation theory? Showing the actucal change in aerosol size distribution might be beneficial. Does a doubling of the available surface area result in such a decrease in RH? This of course assumes spherical particles which most likely is incorrect.

I am not sure what we are learning from derived OC to BC ratios in terms of ice nucleation. I assume these are bulk ratios? For interpretation of ice nucleation it would be beneficial to know the functional groups at the particle surface. What kinds of low volatile gas phase products from oxidation of α-pinene are expected that potentially condense on the particles? How might those differ from oxidation products of the combustion generated VOCs? More discussion has to be given to rule out a "chemistry effect" on ice nucleation.

How do photochemically aged and organic (due to α-pinene oxidation) coated soot particles studied here compare to previously investigated organic coated particles such as in Friedman et al., JGR, 2011, Mohler et al., Env. Res. Lett., 2008, and Knopf et al., GRL, 2010? E.g. the latter study indicates condensation freezing for OC coated particles at similar conditions as for the soot particles coated by oxidation products of α-pinene.

P. 14708, l. 8: Please provide references for this last statement.

P. 14708, section 3.4: In general I agree with this discussion. However, the Koop et al. (2000) homogeneous freezing curve depends strongly on particle size. Uncertainties in particle size will shift the curve accordingly (larger droplets shift the curve towards observations). Also, Koop (2004) states an uncertainty of $\pm$2.5% RH for higher temper-

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atures. All this combined would make the occurrence of homogeneous ice nucleation more likely but deposition ice nucleation cannot be ruled out either.

P. 14709, section 3.5: Comparison of ice nucleation by investigated organic particles with different mineral dust species lacks the information of particle size and number and total particulate surface area available. Some of the authors of this study have previously shown that changes in particle size and available total particle surface area affect ice nucleation. For a fair comparison these information should be provided.

P. 14709, l. 25: This sentence implies that photochemical aging also involves the formation of sulfuric acid coatings?

P. 14709, l. 7-11: As mentioned above, I would be more careful in stating that size is the governing factor determining ice nucleation.

Technical corrections:

P. 14702, l. 6: "an" EURO2. . ..

P. 14704, l. 25: I believe a word is missing after "aged" such as "ones"?

P. 14705, l. 18: Delete one occurrence of "in the same".

P. 14705, l. 21: I suggest a hyphen between "non" and "aged".

P. 14708, l. 18: Please add "aqueous" in front of "ammonium".

Figures in general: Titles in figures such as "Ice Nucleation of EURO-2. . ." should be avoided. These titles can be given in caption as first statement, followed by figure description.

P. 14719, Fig. 4: red line should be magenta in color.

P. 14720, figure caption Fig. 4: Definition of magenta line can be taken from Fig. 2. Last sentence should be changed to "The dotted line represents the homogeneous freezing threshold for 200 nm supercooled aqueous ammonium sulphate droplets based on the parameterizations of Koop et al. (2000).

P. 14722, Fig. 6: Dotted line is narrow dotted instead of wide dotted as in previous figures. Common lines can be referred to Fig. 2.

P. 14723, Fig. 7: Panel a: activated fraction is in arbitrary units? Please give panel indicator in figures. Change RH_{ice} to RH_i as in text. Please change RH_i scale to ticks every 5%. Panel b: I assume the blue arrow should be red and vice versa? Mention experiment temperature in figure or caption.

P. 14724, 14725, Fig. 8, 9: OC:BC ratio is in arbitrary units? Use decimal point instead of comma for y-legends. Please give units for "Time".

P. 14726, Fig. 10: Dotted line inconsistent with previous figures. Give exemplary uncertainties for cited data points. Correct "DeMott". What does it mean "from a different IN chamber"? Is this an instrument?

References:

Koehler, K. A., P. J. DeMott, S. M. Kreidenweis, O. B. Popovicheva, M. D. Petters, C. M. Carrico, E. D. Kireeva, T. D. Khokhlovac, and N. K. Shonijac (2009), Cloud condensation nuclei and ice nucleation activity of hydrophobic and hydrophilic soot particles, Phys. Chem Chem. Phys., 11, 7906-7920, doi:10.1039/b905334b

Friedman, B., Kulkarni, G., Beranek, J., Zelenyuk, A., Thornton, J., and Cziczo, D.: Ice nucleation and droplet formation by bare and coated soot particles, J. Geophys. Res., 116, D17203, doi:10.1029/2011JD015999, 2011. 14708

Mohler, O., S. Benz, H. Saathof, M. Schnaiter, R. Wagner, J. Schneider, S. Walter, V. Ebert, and S. Wagner (2008), The effect of organic coating on the heterogeneous ice nucleation efficiency of mineral dust aerosols, Environ. Res. Lett., 3 (2), 14251435

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Knopf, D. A., B. Wang, A. Laskin, R. C. Moffet, and M. K. Gilles (2010), Heterogeneous nucleation of ice on anthropogenic organic particles collected in Mexico City, Geophys. Res. Lett., 37, L11, 803, doi:10.1029/2010GL043362

Koop, T.: Homogeneous ice nucleation in water and aqueous solutions, Z. Phys. Chem., 218, 1231–1258, 2004.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 14697, 2012.