

The reviewer's comments are bolded and italicized while our comments are in plain text.

Response to Reviewer #2

This paper presents a characterisation of an SP2 instrument that is to be used as a pre-filter for ice nucleation experiments. It presents a series of careful experiments to examine the evaporation of rBC by the incandescence laser as a function of laser power and shows that even at low laser powers an ultrafine particle population is observed in the exhaust of the instrument. TEM analysis shows that this is due to nucleation and not fragmentation. The conclusion is that a laser power of 930 nW is required for complete vaporization. The effect of black carbon on the ice nucleation of a range of standard proxies was carried out and it is shown that only when black carbon is internally mixed with the proxy does its IN efficiency change.

These conclusions therefore allow the effectiveness of black carbon particles as IN to be separated from other IN as long as the population is externally mixed. Where significant internal mixing has taken place then its use as a probe may be less unequivocal. This characterisation is important and is certainly worthy of publication in AMT. The paper concludes that the results bolster confidence in the method to separate the effects of rBC in IN experiments and cites wildfires as an important area.

I would question whether the tone should be so optimistic since significant dust is often present in wildfires and the mixing of rBC and dust in the near field of fires is not well characterised. My interpretation of the results presented here is that this is a potential shortcoming that cannot easily be overlooked nor tackled when using ambient data. This should be included in the final discussion and methods to identify its influence identified.

The paper is largely very well written in my view and the figures are clear and understandable. There are one or two places where the text could be made more readable and these are identified below.

The authors would first like to thank Reviewer #2 for his/her insightful comments, which have improved the clarity and utility of this manuscript.

The authors agree with the reviewer about the tone of the discussion. In light of this comment and a general comment by Reviewer #1, we have amended the final paragraph of Section 3.4 to now read:

“When the INP proxies are internally mixed with rBC, we do see a reduction in INP concentrations due to the SP2 laser. Thus, INPs internally mixed with rBC generally cause overestimations of the rBC contribution to INP concentrations in the SP2-CFDC. To account for this, we determined the fraction of “attached-type” particles (section 2.3). Interestingly, we find that the attached-type fraction correlates well with the number fraction of deactivated INP for the internally mixed NX-illite case. The INEs in NX-illite are refractory, and therefore will not be fully vaporized when an attached rBC particle is heated to 4000 K; thus, scattering material

would traverse SP2 laser and these particles would appear as “attached” in the SP2 analysis. From Figure 8, we see that 97% of the attached-type fraction was deactivated after exposure to the SP2 laser. This is consistent with results from Levin et al. [2014], who used a different analysis to estimate the number of attached-type particles, and found that, at minimum, 74% of mixed Aquadag®-ATD particles were deactivated as INP following exposure to the SP2 laser. Thus, for refractory INE attached to rBC, the attached-type fraction can be used to estimate the number of deactivated, rBC-containing INP.

In contrast, the attached-type fraction does not correlate with the number of deactivated INP for the internally mixed SRFA and Snomax® cases. Here, unlike NX-illite, the INEs from SRFA and Snomax® are non-refractory/heat-labile. Thus, when rBC heats to 4000 K in the SP2 laser, any attached, non-refractory INEs are completely evaporated or destroyed. To confirm this, we estimated the fraction of rBC-SRFA/Snomax® particles that were affected by the SP2-laser. During one-pot nebulization, or what we are calling the “internally mixed” scenario, some fraction of particles will be pure rBC, some fraction will be pure SRFA/Snomax®, and a final fraction will be truly internally mixed. As a conservative estimate, we assume that all particles that contain incandescing material also contain INPs. From the SP2 raw data, 96 and 76% of all Aquadag®-SRFA and Aquadag®-Snomax® particles in the “internally mixed” scenario contained incandescent material, respectively. Thus, only 96 and 76% of the particles could contain rBC that is both greater than 90 nm and physically attached to an INE. From Figure 8, the fraction of deactivated INP was 90% for “internally mixed” Aquadag®-SRFA and 69% for “internally mixed” Aquadag®-Snomax® particles. Thus, 94% of the incandescent Aquadag®-SRFA particles and 91% of the incandescent Aquadag®-Snomax® particles were deactivated following exposure to the SP2 laser. From the combined above analyses, we believe that heating rBC to its vaporization temperature of ~4000 K will destroy >90% of any physically attached INE. Therefore, while this technique cannot tell us the number of rBC INP, it can tell us the number of rBC-containing INP. Thus, we recommend using the terminology “rBC-containing contribution,” instead of the previously used “rBC contribution.”

It might be worth commenting in the discussion on whether the nucleation arises from the organic matter evaporating and then re-nucleating as well as the core. Whilst this is not important in your experiment since the particles were nearly all composed of rBC but for particles with significant coating, such as biomass burning particles it could a much bigger effect. One might also expect significant condensational growth of the nucleated particles in these conditions. Given the conclusions are focused on the use of the instrument for investigating the IN effectiveness of biomass burning this is worth including.

The authors agree that organic matter coating an rBC core would evaporate, and could re-nucleate or condense on nucleated particles. This, in theory, could affect the ice nucleation effectiveness of externally mixed INP. In general, however, it is not thought that non-biologically sourced, liquid organics would affect the ice nucleation of insoluble particles. Interestingly, a coating of amorphous elemental carbon may affect externally mixed INP, but we saw no evidence of this from the externally mixed experiments. Nonetheless, we have added the following statement to Section 3.4:

“Finally, condensation of evaporated organics or vaporized rBC may coat and deactivate externally mixed INPs; while this is a lesser concern for non-biologically sourced liquid organics (Prenni et al., 2009; Schill et al., 2016), a coating of solid, amorphous carbon could cause a significant reduction in ice nucleation ability.”

Page 3 line 36: This line seems to alternate between singular and plural (finesses/face)

Agree. “Finesses” has now been changed to “finesse.”

Page 4 lines 5-6: s in figure and S in text

The authors thank the reviewer for carefully reading the manuscript for typos. In this case, however, the “s” in the equation is actually a capital “S.”

Page 4 line 25: The description of the positions is not clear. Is this the time delay from the first point the signal passes the reference threshold? If so, which detector? This needs to be clear.

The authors agree that the description of the positions could be clarified. The sentence in question has been changed to the following:

“To approximate the number of attached-type particles, we take the difference between the scattering peak’s half-decay position and incandescent peak’s half-decay positions”

Page 4: line 28: depend(a)nt dependent

Thank you. Corrected.

Page 6 line 7: A lesson learned from bitter experience I suspect

This was indeed a lesson learned from experience, and hopefully one that is useful for others replicating this technique.

Page 7 line 4: “of at least At”

Thank you. “At” was changed to “at.”

Figure 3: caption state the laser power used

The authors agree that this adds clarity to Figure 3. We added the following sentence to the figure caption:

“The absolute SP2 laser power was 1290 nW/(220-nm PSL).”

Page 9 lines 24-27: I do not feel that “rBC containing contribution” in figure 8 is well described at all. What is “the effect” on laser power? and what does the scale on the x axis of the figure represent? This needs clarification.

Upon review, the authors agree that the results in Figure 8 is difficult to comprehend because there are several key pieces of information. As suggested by the reviewer, we have removed the pure proxies from Figure 8, and added the laser’s effect on the ice nucleation properties of each pure proxy into Figure 7. We now feel both that Figures 7 and 8 effectively communicate their main results.

Additionally, the meaning of the x-axis changes depending on the symbols in the legend. The authors, upon reviewing, agree that it is nonetheless confusing not to label the axis. Thus, we have changed the x-axis label to “Contribution or Fraction”

Page 10 line 16 of (the) SP2 laser

Corrected. Thank you.

Page 10 line 26: particles

Corrected. Thank you.

Author’s Response References

Prezzi, A. J., Petters, M. D., Faulhaber, A., Carrico, C. M., Ziemann, P. J., Kreidenweis, S. M., & DeMott, P. J. (2009). Heterogeneous ice nucleation measurements of secondary organic aerosol generated from ozonolysis of alkenes. *Geophysical Research Letters*, 36(6), 1–5. <https://doi.org/10.1029/2008GL036957>

Schill, G. P., Jathar, S. H., Kodros, J. K., Levin, E. J. T., Galang, A. M., Friedman, B., et al. (2016). Ice nucleating particle emissions from photochemically-aged diesel and biodiesel exhaust. *Geophysical Research Letters*, n/a-n/a. <https://doi.org/10.1002/2016GL069529>