

Interactive comment on “Use of the Single Particle Soot Photometer (SP2) as a pre-filter for ice nucleation measurements: Effect of particle mixing state and determination of SP2 conditions to fully vaporize refractory black carbon” by Gregory P. Schill et al.

Anonymous Referee #1

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* General comments

The authors report on a set of laboratory measurements to investigate the use of a single particle soot photometer (SP2) as a pre-filter for ice nucleation measurements, in order to determine the concentration of rBC-containing particles that act as ice nucleating particles (INPs). This technique has been applied by some of the authors in previous studies. The present study extends on this work by investigating the condi-

C1

tions required to fully vaporize Aquadag BC in an SP2, and by examining the effect of the SP2 laser on internal and external mixtures of INP proxies and Aquadag. The topic of the paper is interesting and suitable for AMT. Besides from the relevance of INPs, the results are also of interest to Laser-Induced Incandescence studies. The experimental design and measurements are both good quality. For the most part the manuscript is well-written and the results well presented. However, I have a number of comments that I think must be addressed before the manuscript is considered for publication in AMT.

One of the most important results I learned from this study is that the technique in question can't be used on internal mixtures of INPs and BC. The SP2 laser deactivates some of the INPs and it is not simple to work out exactly how much. This is a somewhat negative result as it severely limits the applicability of the technique. But it is still an important point that I believe requires more discussion. Most ambient BC samples contain a sizeable fraction of internally mixed BC. Is my conclusion premature, and do the authors think the technique can still be used to determine the concentration of rBC-containing particles that act as INPs in these situations, albeit with an increased measurement uncertainty? If not, under what conditions can the technique be safely applied?

What I presume to be a smaller peak of doubly-charged particles appears below 200 nm in Figs. 2 and 3. I would expect it to show up at 250 nm. What's going on here?

A number of results are concluded from the rBC-containing contribution to N_{INPs} values shown in Fig. 8. I found these results quite difficult to comprehend within this framework. Perhaps this is just me, but I would encourage the authors to think about how they could more effectively communicate their main results. One suggestion for the pure proxies result is given below in my specific comments. I would also suggest the authors at least write out somewhere in the manuscript how the rBC-containing contribution to N_{INPs} is calculated, to facilitate understanding of the discussions based around this parameter.

C2

* Specific comments

P1, L20: 'Fluence' should be changed to 'power'. Fluence is energy per area, and a power value is given in the square brackets.

P2, L21: 'Direct' repeated in the final part of this sentence.

P5, L26: What were the typical concentrations in the tank, to indicate timescale of coagulation.

P6, L13: Extra 'is then' in this sentence.

P7, L3: Minor point but this comparison does not hold up exactly. In pulsed LII, particles are generally heated to below their vaporisation temperature by choice, to ensure they don't lose mass due to evaporation. To account for this, particle temperatures are typically monitored by 2-color pyrometry, and thus the particles are not required to have 'uniform detection efficiency'. A more recent and relevant reference for pulsed LII might be Michelsen et al., 2015.

P7, L22: What sheath:sample air flow rate ratio was used for these measurements.

P8, L6: Could sintering of Aquadag fragments possibly also cause the change in microstructure from graphitic to amorphous?

P8, L9: ATD has not been defined previously in the manuscript.

P8, L35: 0.002% of what? The total particle mass?

P9, L10: Does Fig. 7 show CFDC measurements for when the SP2 laser was on or off? To more easily show that the SP2 laser minimally affected the ice nucleation ability of the pure proxies, would it be possible to plot both SP2 laser on and off measurements on this Fig.? I believe this would be easier to understand than the way this point is currently made, which is through the rBC containing contributions to N_INPs of the pure proxies shown in Fig. 8.

C3

P9, L21: Missing word between 'determined' and 'for'. E.g. '...previously determined value for ...'

P9, L38: This conclusion is stated too strongly. Only 2 of the 3 cases show the laser did not affect INP efficacy - I rather consider the SRFA case as an example when it did affect it - and only a limited range of experiments have been conducted (1 type of incandescing particle, 1 external mixture of each type).

P10, L2: Please provide some justification for this 30% threshold. I don't imagine this represents the random uncertainty in taking the difference between two low INP concentration measurements, since one might also obtain negative fractions of similar magnitude, and the pure proxy measurements show this doesn't seem to be the case.

P10, L5: Could it also be the case that the attached-type particle measure introduced in section 2.3 is not sensitive enough? Since this measure does not seem to have been validated against independent measurements or Moteki's method I don't think this can be ruled out.

P10, L7: Taking this point even further, can it be that since the SRFA was coating the Aquadag and not just attached to it, the SRFA material evaporated more or less completely, removing all possible INPs. A possible suggestion for a future experiment could be given: comparison of the rBC-containing contributions of internal mixtures generated by the current mixed-solution method and by coagulating the two particle types.

P10, L9: Typo here 'INE' instead of 'INP'. Also, this sentence seems to say exactly the same thing as the one before it.

P10, L36: Care must be taken not to over-generalize here. Only Aquadag particles have been investigated in this study. Other types of BC will absorb varying amounts of energy from the SP2 laser based on their imaginary refractive index, and it is not clear homogenous nucleation will always result from vaporized BC.

C4

P11, L23: The big caveat here is that this determination will not work if a sizeable fraction of INPs are internally mixed with BC, which is the case for burns and wildfires. Therefore, I do not agree with this statement.

P17, Fig. 3: Please indicate in the caption that particle concentrations were kept constant at 600 cm^{-3}

P18, Fig. 4: Please indicate in the caption what sheath:sample air flow rate ratio was used for these measurements.

P21 and 22, Figs. 7 and 8: Please provide an explanation here or in the main text of exactly what the error bars represent.

P22, Fig. 8: X-axis label missing (even if one can still kind of figure out what is shown).

* References

Michelsen, H. A., Schulz, C., Smallwood, G. J. and Will, S.: Laser-induced incandescence: Particulate diagnostics for combustion, atmospheric, and industrial applications, *Progress in Energy and Combustion Science*, 51, 2–48, doi:10.1016/j.pecs.2015.07.001, 2015.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2017-389, 2017.