Review of "Cloud Droplet Activity Changes of Soot Aerosol Upon Smog Chamber Ageing" by Wittbom et al.

General Comments:

The authors present measurements of photochemically aged diesel exhaust and flame soot particles sampled from a Teflon smog chamber using a CCN counter, an SP-AMS, and an APM. It is shown that while POA and BC aerosol exhibit no CCN activity (s<2%) prior to the initiation of UV-induced photochemistry, the condensation of SOA onto the particles rapidly transitions them to more CCN-active particles over atmospheric aging timescales of only a few hours. Given the global climatic importance of BC-containing aerosols, it is important that we understand how these particles evolve in the atmosphere and affect clouds. As such, this study is important and highly relevant for ACP. In general, the manuscript is well-written and the results are interesting and thoroughly explained; the latter so much so that some of the explanations become repetitious and some attempt to streamline these in the revised manuscript would be appreciated. I have only a few comments that the authors should address prior to publication.

I also note that the manuscript has been submitted to a special issue entitled: "Interactions between climate change and the Cryosphere", which seems like a strange choice for this paper given its focus on laboratory soot in a smog chamber. While BC is thought to be an especially important climate forcing agent in the Arctic, it is not clear how this particular study relates at all to the cryosphere.

Specific Comments:

The experimental and instrumentation sections should come before the theory and modeling sections to give better context for the analysis. For example, in Equation 12, I was left wondering how you distinguished between POA and BC until I got to section 4.1 and realized that the AMS is actually an SP-AMS.

Pg. 8867, Line 1: Specify what SFCA ramp times were used.

Pg. 8868, Lines 9-15: More discussion of the SP-AMS is warranted. If I understand correctly, the vaporizer/filament are always on while the laser is only on for a continuous 5 minute period, once per hour. What is the reason for operating the laser for 5 minutes only? What collection efficiencies were assumed for both the conventional vaporizer and the new SP-AMS laser ablation technique? Was the fragmentation table altered to account for increased gas-phase CO_2 from the diesel engine? Is the laser ablation/ionization only used to detect BC or is it also used to look at organic coatings?

Similarly, more details on the DMA-TD-APM operation would be useful. For example, what transfer function, rotation, voltage ranges were used. I'm assuming that the peak of the mass distribution was used with d_ve in Equation 11 or was a more complex inversion used?

The dual, size-resolved CCN operation is nicely described.

Pg. 8870, Line 1: I don't understand how the soot primary particle diameter affects the CCN activity since this refers to the sizes of the spherules making up the soot agglomerate rather than the overall particle effective diameter that feeds into Kohler theory.

Pg. 8871, 2-3: This discussion should be pulled out of the supplement and included in the main text. It should also be mentioned earlier when discussing the instrument rather than in the results section.

Figure 1: This schematic should be updated to reflect the complete experimental setup described in the text. For example, the second CCNC and the upstream DMA should be added.

Figure 9 and in the text: I don't understand the role of the activation sites, which seem to imply a kinetic-based CCN activation theory rather than the traditional, well-established thermodynamically-driven activation theory. How are the TEM images used to distinguish hygroscopic or hydrophobic parts of the soot? Typically, the overall soluble volume fraction is what is important for CCN activation, and knowing how this soluble volume is distributed within each particle is less critical since other factors/uncertainties may come into play in determining the humidified aerosol morphology (e.g., liquid organic and water surface forces).

Minor Comments:

Pg. 8853, Line 18: Strike "for"

Pg. 8854, Line 9: Change "acquire" to "require"

Pg. 8854, Line 22-23: This is not a sentence.

Pg. 8854, Line 27: By mass? By atoms? Please specify.

Pg. 8857, Line 18: Change "fore" to "for"

Figures: The yellow lines and points are difficult to distinguish. Please consider darkening or changing that color to make it easier to read.