We thank Reviewer #2 for his/her positive, constructive and detailed comments, which we will account for in the revised manuscript. Our point-by-point responses to the issues raised by the reviewer are below.

Scientific questions:

I have reservations to word "coating" used in this study, simply I do not see clear experimental evidence, to believe that authors are able to coat silica by mixing it with salt, sugar or protein in water, without any special treatment – high temperatures, oxidation or similar. Even though the authors claim they used hydrophilic fumed silica.

We agree that the term "coating" is not an accurate description of our mixed particles. We will modify the revised manuscript accordingly, using the term "mixture" instead of "coating".

Specific comments

Page 6, mid paragraph about CCNc, it would be good to mention what sheath and sample flows of CCNc were used during the experiment.

We will modify the revised manuscript by adding the following text to the the paragraph in question: "Humidified sheath flow (454 cm^3 min^{-1}) surrounds the sample flow (45.4 cm^3 min^{-1}) in the CCN column to hold it in the centre of the column in the region of maximum supersaturation. The ratio of the flows was thus 1 part of sample air to 10 parts of sheath air and the total flowrate was 500 cm^3 min^{-1}. "

Page 7 eq. 3, is not it enough to make reference only to original source? Why to reference to four manuscripts? Similarly through the whole manuscript.

This is a good point. We will modify the revised manuscript accordingly.

Page 13 starting line 361, authors discuss the use of previously reported kappa parameter from literature, listed in Table 1. Why they do not use their own, as showed in Fig.7? Or were the experimentally obtained kappa values exactly the same as reported in literature? Would be beneficial to see activation curves as a function of mobility diameter for pure compounds, since in Fig 8 the activation curves for 150 nm do not reach unity in activation, especially in the case of ammonium sulphate which is used for CCNc calibration.

We used the literature values for κ of ammonium sulphate (AS) and sucrose to demonstrate the good agreement between our results and previous work on pure component activation. The κ values fitted for AS and sucrose using our experimental data would be 0.78 and 0.08. We will add this information to the revised manuscript. Here we should probably also clarify that all the activation curves that were used in further analysis were normalized, using a correction factor derived from the ammonium sulphate experiments, assuming that AS activation probability reaches unity at high supersaturations. Figs. R1-R3 show the unnormalized data on the activation curves as a function of diameter and supersaturation, as requested by the reviewer. We will clarify the normalization procedure in the revised manuscript and add also the normalized curves (which were used in the analysis) to Fig. 8.
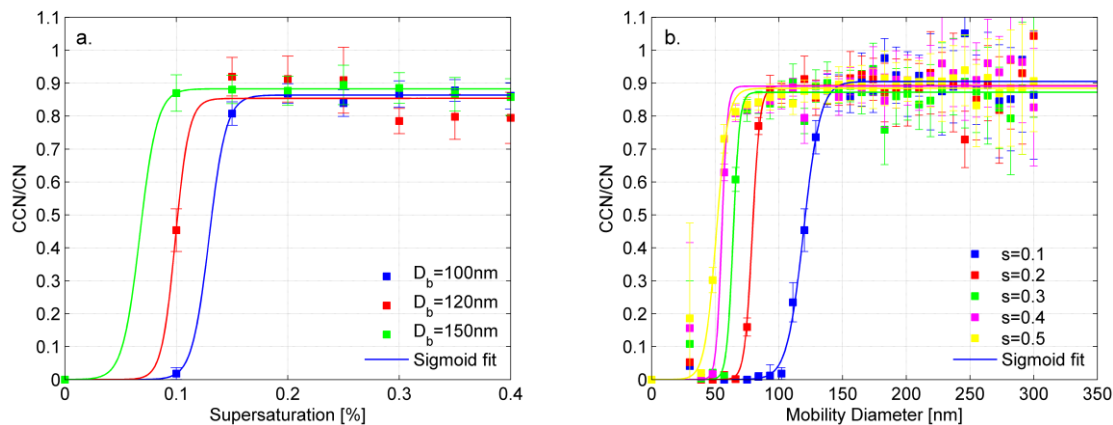
Fig. R1: a) The average activation ratio vs. supersaturation for pure (NH₄)₂SO₄ particles in different mobility diameters. b) The average activation ratio vs. mobility diameter for pure (NH₄)₂SO₄ particles in different supersaturations.
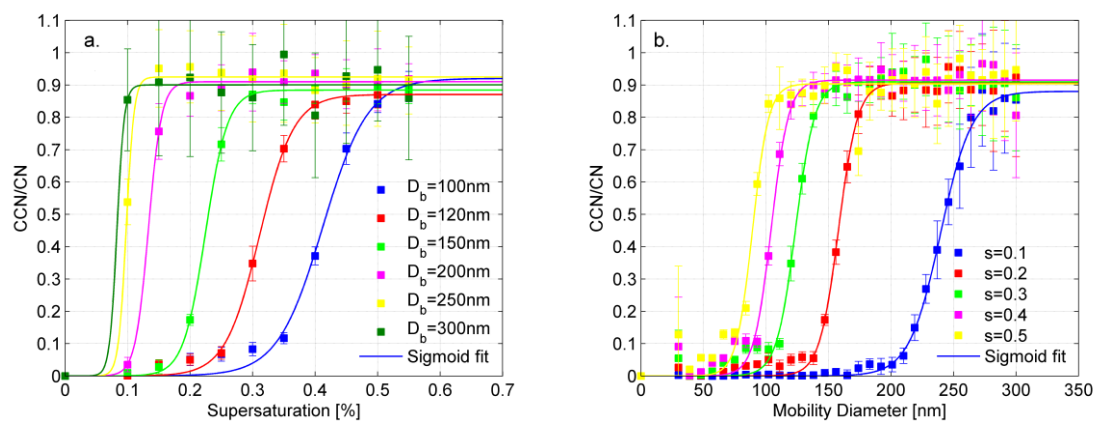

Fig. R2: a) The average activation ratio vs. supersaturation for pure sucrose particles in different mobility diameters. b) The average activation ratio vs. mobility diameter for pure sucrose particles in different supersaturations.
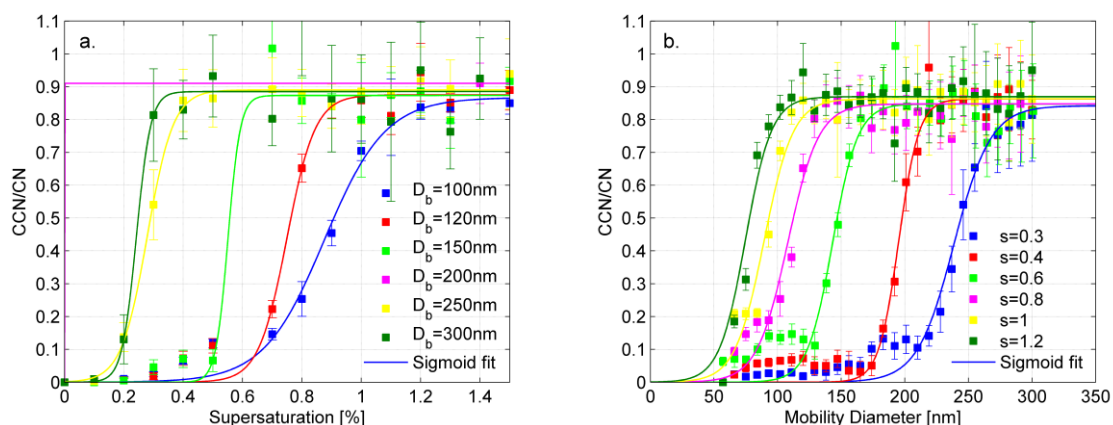

Fig. R3: a) The average activation ratio vs. supersaturation for pure BSA particles in different mobility diameters. b) The average activation ratio vs. mobility diameter for pure BSA particles in different supersaturations.

Page 15 line 411, I see this part as very problematic. In Kumar et al (2011b) you can find that they were using several samples of real mineral dust collected in several locations. I have no doubts that their samples undergo certain natural process of aging and use of shell-core model in their case is appropriate. However, this is not the case of this study, making water solution/dispersion of fumed silica and ammonium sulphate (max 25% mass fraction) will not lead to any coating. If authors have different opinion I would like to see any proof of that. For example preparing the dispersion, in the same way as in this work, of aquadac (colloidal graphite) and 70 % mass fraction of AS leads only to about 10% of coated particles, the rest is externally mixed. In the case of BC containing particles you can find out the shell thickness using single particle soot photometer (SP2-DMT, CO, USA), in the case of this study I understand that such a kind of analysis is very difficult. On the other hand the presence of externally mixed particles should be visible from activation spectra (CCN/CN vs Dp) as activated fraction plateaus at a value that is different from unity. I would recommend authors broader discussion with justification for usage of shell-core model or simply omit this part. I am not sure if sucrose and BSA are doing better job in coating than AS.

We fully agree that our mixed particles are not exactly coated, and, as also stated above, we will removed the term "coating" when referring to our mixed particles throughout the text in the revised manuscript. Indeed, there is a wide range of compositions present at a given size range, as also shown in our Figs. 8-11 for all the soluble compounds. However, we also agree that if two distinct aerosol populations (e.g. "mixed" and "pure" particles) we present as a simple external mixture we should see two plateaus in the CCN activation curve – which we don't. Instead, we see a relatively shallow CCN/CN vs. supersaturation curve (see Fig. 8), supporting the hypothesis of a continuous distribution of soluble vs. insoluble volume/mass fractions in the particles – as also indicated by the results shown in Figs. 9-12 in the manuscript. We tried to highlight this in the sentence describing these figures beginning with: "The small contribution of the adsorption term to the theoretical predictions combined with the shallow activation ratio curves (see Fig. 8) suggest that the reason for the apparent discrepancy between the theoretical and the observed critical supersaturations is a non-constant distribution of the soluble material with varying particle size." Also, while the model by Kumar et al. (2011b) was indeed originally introduced for fresh dust coated by a layer of soluble salt after aging, which is not the case for our experiments, it is currently the only readily applicable theoretical formation, and we believe it does give a reasonable estimate on the potential importance of the adsorption as compared with the bulk solubility of the mixed particles. We will clarify this and discuss the limitations of our approach more thoroughly in the revised manuscript.

Page 17 sentence starting on line 498: Similarly as the previous comment, "Our mixed particles : : : representing an aerosol population with various degree of aging in atmosphere." I have hard time to agree with such conclusion. I would encourage the authors to provide any arguments to support use of word "aging". Fig 9-11 shows directly disagreement with shell-core model.

We agree with you. We will remove this sentence and modify the revised manuscript accordingly.

Fig. 3 would be nice to see some statistical measure of the fits, e.g. coefficient of determination.

We will add the R^2 values for the fits to Fig. 3 in the revised manuscript (see below).

In Fig. 3a: $R^2 = 0.992$

In Fig. 3b: R^2=0.978 & 0.997

In Fig. 3c: R^2=0.991