Response to Referee number 1

16th June 2021

The authors would like to thank Referee no. 1 very much for her/his expertise, precise, detailed and very valuable comments to further improve and clarify the MS. We have considered all recommendations and made the appropriate alterations in all cases. We also accomplished some further smaller corrections. Our specific responses are as follows, while the textual modifications amended to the MS can be traced in its marked-up version, which is available online.

General comments

The manuscript investigates cloud droplet activation properties of aerosol particles, not really cloud droplet activation (there are studies that differentiate the real cloud droplet population from cloud interstitial particles, and studies that aim to related the cloud droplet population to below-cloud aerosol population) . To avoid any potential confusion, it is important to make this difference in the paper. I therefore strongly suggest modifying the title of the paper into something like "Cloud activation properties of aerosol particles in…". The same concerns wording on lines 9, 67 and 78.

Response 1. The title and related text at the specified locations were changed to conform to the plausible arguments of the Referee.

I appreciate the detailed description of methods used in this investigation. However, there are couple of minor issues related to this section. First, the motivation for the criterion introduced on 198-203 should be improved. What is the real purpose of selecting this ratio, and why to select the value of 70% for this ratio? Second, section 2.3 is not really about modeling, but about using existing mathematical formula. Therefore, the title of that section should be modified into something like "2.3 Calculation of particle hygroscopicity".

Response 2. The main motivation of the comparative exercise, namely to validate the $N_{CCN,1.0\%}$ data by the DMPS measurements was better emphasized. The basic assumption for this validation is that most particles activate at an S of 1.0%. This is evidently not fulfilled over time intervals when the contribution of very small particles (e.g. with diameters <30 nm) is considerable since they do not activate. These data should be excluded from the comparison. The original exclusion criteria – namely that the $N_{6–30}/N_{6–1000}$ ratios are a) to be smaller than 10% or b) to be between 10 and 20% – worked perfectly in remote and regional locations. However, in our case of an urban environment, they jointly resulted in a relative number of only 2% of all DMPS data on a yearly scale. This very small portion is casued by the large contributions of

high-temperature emissions in cities and by new particle formation events, which both result typically in particles with a diameter <30 nm. The former situation often occurs in cities, and, therefore, we had to devote more attention to this issue than at regional or remote locations. We think that the representativity of any conclusion for the whole data set based on such a limited (2%) number of the data could be statistically questioned. Instead, we introduced a new criterium as a compromise between the larger relative number of cases and still constrained contributions of non-activating particles. The limit value of 70% was determined as a compromise in this sense in a pragmatic manner. We are aware that alternative values could have also been set. The paragraph was substantially rearranged and modified at several places to express all this more clearly and directly. As far as the title of the section 2.3 is concerned, we changed it to the formulation suggested by the Referee.

Detailed comments

The statement on lines 19-20 (They were related to the size-dependent chemical composition and external mixtures of aerosol particles) gives the wrong impression that this paper measured the aerosol composition and mixing state. It is very true the size-dependent chemical composition and external mixtures of aerosol particles are the most likely explanation for the observations made in this paper. So it should more clearly stated that this is the most likely explanation rather than a real finding of this paper.

Response 3. The sentence under the consideration was removed from the abstract to avoid any possible misunderstanding.

Lines 30-32: Written like it is now, it is not clear what is the result from this particular work (low kappa values in an urban site) and what is based on studies made by others (lower kappa values in regional or remote locations). Please modify.

Response 4. The sentence was reformulated to separate our fidings from the earlier results in an unambigous way.

Lines 43-47: The text is not quite consistent with itself. Since S is affected not only by the updraft velocity but also by sink of water vapor (existing cloud droplet population determined by CCN), I would recommend writing "Different updraft velocities in clouds, together with existing cloud droplet population that depend on CCN concentrations, result in different Ss…".

Response 5. The original statement involved only the main governing property, which is the cloud dynamics, as it was inicated in the sentence. We readily extended the discussion with further possibilities as suggested.

Line 62: This is unclearly written. Maybe one could write "…interactions at S values typical for atmospheric conditions and…"?

Response 6. The suggestion was implemented.

Lines 79-81: I would modify the writing a bit: "Specifically, we will report, … various Ss, in order to determine…".

Response 7. The formulation was modified to: Specifically, we report, discuss, explain and interpret here… .

Lines 426-427: One of the very first studies showing that the minimum diameters of aerosol particles able activate into a cloud droplet is typically well below 100 nm in a remote environment was that of Komppula et al. (2005, J. Geophys. Res., 110, D06204, doi:10.1029/2004JD005200). It might be worth mentioning that paper here.

Response 8. We readily added and cited the reference.

Line 460: Please explicitly write what dependency you refer to here. I assume this refers to the observed slope of the S vs. particle diameter relation in Figure 3 that is different from the theoretical slope for a particle population with a size-independent chemical composition. A reader might not catch this because it requires returning to the information given in the previous paragraph.

Response 9. The text was extended to express explicitely that we discussed here the dependency of the deviation of the experimentaly determined ($d_{c,eff}$, S_c) data points from the calculated line for the simulated global continental mean κ as a function of $d_{c,eff}$, which was shown in Fig. 3.

Lines 515-516: I do not fully understand this statement. Does it refer to different seasonal behavior of the S=0.1% curve as compared to those of other values of S? The following text (lines 555-558) is also somewhat difficult to understand.

Response 10. The related sentence was reformulated and extended to clarify that the statement refered only to one of the properties in Fig. 5 namely to the $d_{c,eff}$ (Fig. 5c). We also modified and corrected the following two sentences to make them more comprehensible and fluent.

Line 630: ..at lower sizes?

Response 11. The whole sentence was reworded. Firstly, the limiting diameter was specified better as ca.130 nm, secondly and more importantly, we clarified that the rest of the sentence dealt with the difference between the general and urban hygroscopic properties and that this difference increased with decreasing size.

Lines 630-632: I do not understand how particle hygroscopic properties could depend on the particle number concentration. I suppose the authors mean something else here, but it is written in a confusion manner.

Response 12. The sentence was extensively modified to express that we meant the CCN concentrations at various *S*s and activation fractions here.

Table 3: The table caption should explicitly tell that the unit of the numbers given in the table is "nm".

Response 13. The heading of Table 3 was extended by: …in units of nm… .

Imre Salma
corresponding author