**Review of the manuscript entitled „In-cloud measurements highlight the role of aerosol hygroscopicity in cloud droplet formation " from Olli Väisänen et al.**

This study investigates the role of aerosol hygroscopicity on its ability to form cloud droplets. Many previous studies have addressed the roles played by particle size and particle hygroscopicity on the ability of such particles to act as cloud condensation nuclei (CCN). However, this was for the most part done based on theoretical considerations, laboratory studies and/or simulated droplet formation on ambient aerosol in CCN counters. This study is one of very few studies that directly investigated the activation of aerosol into the droplets of atmospheric clouds as a function of their size and hygroscopicity. State-of-the-art experimental methods were applied to independently determine the hygroscopicity distributions of the total aerosol and the interstitial aerosol, based on which they could clearly show that the less hygroscopic particles are much less efficient in forming cloud droplets compared to the more hygroscopic particles of equal size. While this essentially confirms expectations, it is still a very valuable result as it directly affects how local/regional emissions contribute to cloud droplet number and to what extent the less hygroscopic particles can be processed by clouds.

The manuscript is generally well written, concise and within the scope of ACP. The data analysis approaches seem appropriate for the most part. However, in two cases I am not sure whether the results are internally consistent. Besides, one figure, which is essentially just an internal consistency check of the data analysis approaches, comes as if it was an independent result (see major comments). The "minor comments" are largely just meant to clarify e few things, to improve the notation in equations in order to avoid ambiguities and to provide several ideas for additional analyses. I do not expect that the latter should all be addressed in a comprehensive manner (nor would I expect exhaustive rebuttals for most of them).

In conclusion, I recommend this interesting and relevant manuscript for publication in ACP after the most relevant comments have been addressed by the authors.

**Major comments:**

1) P.8, l.21-22:
   "Typically, the activation efficiencies calculated by Eq. (9) ($f_{act,GF>0.80}$) appeared somewhat larger than the DMPS derived values ($f_{act,DMPS}$)." – I claim that this is impossible! It must be caused by errors in the calculations (e.g. from inconsistent choice of GF-PDF normalization approach and integration of the GF-PDFs in Equation 9 or from comparing at different times). Let me explain: "GF>0.80" means that you integrate over the whole GF-range of the GF-PDF. The fact that the GF-PDF, as used in Equation 9, must be normalized to unit area, directly implies that, for "GF>0.80", Equation 9 simplifies to
   $f_{act,GF>0.80}(D_p)= ( dN_{tot}(D_p)/dlogD_p – dN_{int}(D_p)/dlogD_p ) / dN_{tot}(D_p)/dlogD_p$
   and this is nothing else than how I would define $f_{act,DMPS}(D_p)$. Or in other words: there is no more HTDMA derived information left in $f_{act,GF>0.80}(D_p)$, only DMPS-derived information. Therefore the poor time resolution or other potential issues with the HTDMA measurement cannot influence the result.
   Please reconsider your calculations and add the results from all other available measurements to Table 1 too, if there was really a mistake in the previous approach.

2) Table 1:
Shouldn't the following equality hold for the data shown in Table 1?
$f_{act,GF>0.80}(D_p) = f_{GF<1.25}(D_p) * f_{act,GF<1.25}(D_p) + (1 - f_{GF<1.25}(D_p))* f_{act,GF>1.25}(D_p)$
(simply use Equation 9 to get this). It does seem to be fulfilled for some columns of Table 1 but not for others. Please check your data or clarify the manuscript if I misinterpreted the meaning of Equation 9 or $f_{act}$ or so.

3) Figure 3 and first paragraph of Section 3.4:
The comparison of cloud droplet predictions made with considering the hygroscopic mixing state or with assuming internal mixture (and using $\kappa_{avg}$) is useful, as it confirms previous results on the sensitivity of CCN predictions to simplified treatment of mixing state for the effect of mixing state of total cloud droplet concentration in real clouds.
By contrast, the comparison of HTDMA+$DMPS_{tot}$+$S_{c,eff}$-predictions of cloud droplet number concentration with those derived from the $DMPS_{tot}$ minus $DMPS_{int}$ measurements is meaningless because it is circular argumentation! Equation 8 was first used to infer the $S_{c,eff}$ from the HTDMA, $DMPS_{tot}$ and $DMPS_{int}$ measurements. The $S_{c,eff}$ obtained in this manner was then used, along with the HTDMA + $DMPS_{tot}$ data, for cloud droplet prediction to be compared with those derived from $DMPS_{tot}$ minus $DMPS_{int}$ measurements as shown in Figure 3. Consequently, this analysis has nothing to do with a closure study, instead it is simply an internal consistency check of the approach to infer $S_{c,eff}$ and to predict cloud droplet number. Therefore, this part of the figure (i.e. the red data points) has to be removed and the discussion needs to be adapted.
Nevertheless, it is worth to have a closer look into Figure 3. The fact that the grey points have higher correlation and slope closer to unity than the red points is unexpected. If the cloud droplet prediction is so insensitive to assuming internal mixing state, why is then the "internal consistency check" of inferring and re-applying the effective supersaturation from/to the HTDMA measurements so poor (referring to slope and particularly the scatter of the red points)? Is this related to improper treatment of activation plateau values that differ from unity (see separate comment below concerning this potential issue)? Or is the minimization approach applied to infer the effective supersaturation (Equation 8) not suitable? Other reasons?

**Minor comments:**

4) Abstract:
Well written but for one missing element: The range of supersaturations occurring in the  is not mentioned. This would be worthwhile as they appear to be rather low, which increases the sensitivity of CCN number concentration to hygroscopicity/composition compared to high supersaturation.

5) P.3, l.26:
It might be worthwhile to mention how the visibility was measured.

6) Sect. 2.3:
A $PM_{1.0}$ impactor was used in the interstitial inlet to remove the activated cloud droplets just leaving the interstitial aerosol behind. However, the mode of none-activated droplets in stable equilibrium within the cloud may potentially extend to diameters larger than 1 µm, if the peak supersaturation at cloud formation was very low (the lower the peak supersaturation, the larger the activation cut-off diameter, the larger the maximal diameter of non-activated interstitial

particles). In such a case, the largest and most hygroscopic particles of the true interstitial aerosol in the cloud would be missing in the aerosol sample measured behind the interstitial inlet. Hammer et al. (2014) address this issue in more detail. Can you exclude this potential artefact based on supporting data such as droplet size distributions or estimates of the maximal possible equilibrium droplet size for the clouds formed with the lowest peak supersaturation?
Hammer, E., Gysel, M., Roberts, G. C., Elias, T., Hofer, J., Hoyle, C. R., Bukowiecki, N., Dupont, J.-C., Burnet, F., Baltensperger, U., and Weingartner, E.: Size-dependent particle activation properties in fog during the ParisFog 2012/13 field campaign. *Atmos. Chem. Phys.*, **14**, 10517-10533, doi:10.5194/acp-14-10517-2014, 2014.

7) Sect. 2.4:
The residence time between humidifier and DMA is provided (2s). The residence time within the humidifier might also be of interest, possibly even including a brief remark on the reasons for choosing a rather short or rather long residence time.

8) Equation 2 and other instances in Sect. 2.5 and possibly the rest of the manuscript:
This equation (Köhler-equation) describes the equilibrium water vapour saturation ratio ($S_{eq}$) as a function of particle hygroscopicity, diameter, etc. By contrast, the critical saturation ratio ($S_c$) is the maximum of the Köhler curve described with Equation 2. You are using the same symbol for these two different quantities. This needs to be fixed.

9) Line just below Equation 3, "…where $V_s$ and $V_w$ are the soluble and water volumes…":
In general, $V_s$ is the volume of the whole dry particle, not just the soluble components. Insoluble fractions, if present, are accounted for with the κ-value.

10) Equation 5: This analytical solution for the critical saturation ratio contains mathematical approximations which become increasingly inaccurate with decreasing critical saturation ratio. Therefore, this equation should only be used for qualitative purposes, whereas a numerical solution of the Köhler equation must be implemented for quantitative purposes.

11) Equation 5 and line just below:
This is now the critical saturation ratio. It should be explained what it is – in contrast to the equilibrium saturation ratio appearing in Equation 2 – and the symbol should be defined.

12) Equations 6 and 7:
It could be mentioned that the approach and equations used to predict the CCN number concentration from particle number size distribution (from SMPS) and GF-PDFs (from HTDMA) is identical to the approach introduced by Kammermann et al., 2010b (cf. their Equations 2&3).

13) P.6, l. 5-8:
This step also involves interpolation in time (besides inter-/extrapolation in size).

14) P.6, l. 11ff:
"Nonetheless, it has to be noted that above 200 nm, hygroscopicity is quite rarely a limiting factor and the most crucial activation characteristics are dependent on particle properties between the ~80 nm and ~200 nm sizes." – Isn't this statement somewhat in conflict with your result that a substantial portion of the non-hygroscopic particles remains interstitial (at the largest diameter covered by your measurements)? Therefore you need the additional argument

that the number fraction of non-hygroscopic particles depends only weakly on size across the size range relevant in this context.

15) P.6, l.13ff:
"Secondly, the method assumes that the subsaturated hygroscopicities are representative for supersaturated conditions. Such an assumption is not always totally valid and discrepancies between the two saturation regimes have been reported based on laboratory and field experiments…" – The study by Jurányi et al. (2013) could also be referenced here, as one of the examples that found very good closure between sub- and supersaturated regimes for externally mixed urban aerosol.

16) P6., l.19ff:
Comments on the approach to estimate the effective peak supersaturation (PS: I'd suggest $S_{c,eff}$ rather than $S_{eff}$ as symbol):

a. I suggest to start with a brief explanation of the concept behind estimating $S_{c,eff}$, possibly also referring to Hammer et al. (2014). This would be likely be helpful for the "average" reader of this manuscript.

b. If entrainment occurs or in the case of partially/fully glaciated clouds the plateau value of $f_{act,DMPS}$, i.e. the value $f_{act,DMPS}$ takes at large diameters at which also the non-hygroscopic particle activate, may be substantially smaller than unity (e.g. Fig. 3 in Verheggen et al., 2007). The minimization approach given in Equation 8 would cause a bias for such a scenario. – Did you observe evidence for entrainment and/or glaciation or did the plateau value of $f_{act,DMPS}$ always reach unity?
Verheggen, B., Cozic, J., Weingartner, E., Bower, K., Mertes, S., Connolly, P., Gallagher, M., Flynn, M., Choularton, T., and Baltensperger, U.: Aerosol partitioning between the interstitial and the condensed phase in mixed-phase clouds. *J. Geophys. Res.*, **112**, D23202, doi:10.1029/2007JD008714, 2007.

c. Our experience from similar measurements at the Jungfraujoch research station is that the diameter range across which $f_{act,DMPS}$ increases from 0 to 1 is much broader than can be explained with the heterogeneity of the aerosol in terms of mixing state/GF-PDF (the JFJ-aerosol is rather internally mixed). This indicates that the width of $f_{act,DMPS}$ is mainly driven by heterogeneity of $S_{c,eff}$ on small spatial scales due to e.g. turbulence. What does it look like in your case (the aerosol at Puijo is obviously much more externally mixed than that observed at the Jungfraujoch)? Can the shape of $f_{act,DMPS}(D_p)$ be explained with the external mixing alone? This question possibly goes beyond the main focus of this paper, but it might still be worth looking at it. You have the data at hand and could possibly produce a supplementary figure using representative examples).

d. An alternative approach would be to fit something like a sigmoid curve into $f_{act,DMPS}(D_p)$ to obtain an effective activation cut-off diameter (half rise). Inserting this cut-off diameter and $\kappa_{avg}(D_p)$ into Köhler theory then provides $S_{c,eff}$ under the assumption of internal mixing. I would expect that these values are very similar to those obtained with your approach accounting for external mixture. If not, you should comment on the fact

that proper treatment of mixing state is crucial for inferring $S_{c,eff}$ when dealing with clouds formed on externally mixed aerosol.

e. Did you define $f_{act,DMPS}(D_p)$?

17) P.6, l.26ff:
First, this paragraph belongs above the paragraph describing how you infer $S_{c,eff}$, as this is still about predicting CCN number concentration from HTDMA data, if I got that right. Second, you may have to explain how you obtain $\kappa_{avg}(D_p)$.

18) Sect. 3.1:
Some at least partially glaciated clouds wouldn't be surprising if ambient temperature was sometimes below zero (minimum was -9.7 °C).

19) Figure 1, bottom row:
GF-PDFs shown here are normalized to unit area. I wonder whether it would be instructive to add an extra row (or replace the current bottom row) with a version in which you re-normalize the GF-PDFs as follows. For the total aerosol, multiply the GF-PDF that is already normalized to unit area with $dN_{tot}/dlogD_p(D_p)$. And equivalently for the interstitial aerosol. The area between the curves representing the total and interstitial GF-PDFs for equal size would then directly correspond to the activated particles. Furthermore, normalized in this manner, the bottom row of Figure 1 would then be are more close graphical representation of what you calculate with Equation 9. You could even add an extra row of panels that shows $f_{act,GF}$ for each diameter and GF-resolved, i.e. as a function of GF rather than integrated over a GF range.

20) P.7, l.20:
Was the shift of the more hygroscopic mode towards larger GF with increasing particles size less or more than what can be explained by the size dependence of the GF imposed by the Kelvin effect?

21) P.7, l21ff:
The observations by Laborde et al. (2013) in Paris revealed even a little more detail. There was very clear evidence that particles from fresh traffic emissions appeared mainly at GF≈1.0, whereas particles from wood burning appeared mainly at GF≈1.1, together forming the "non-hygroscopic" mode in the HTDMA. I have added this detailed remark because the GF-PDFs shown in the bottom row of your Figure 1 for particles with diameters of 120 nm and 150 nm seem to provide evidence that the cloud droplet active fraction differs slightly between GF≈1.0 and GF≈1.1. Could you confirm this or is this difference within uncertainty?

22) Equation 9:
$N_{tot}(D_p)$ should be replaced by $dN_{tot}(D_p)/dlogD_p$, shouldn't it? And so for $N_{int}(D_p)$? – You define $N_{tot}$ as: "…where $N_{tot}$ and $N_{int}$ are the total and interstitial number concentrations…". This rather rather sounds as if $N_{tot}(D_p)$ was representing a cumulative number concentration, which would be wrong in Equation 9 (as I understand the purpose of Equation 9).
PS: you could of course also choose $dN_{tot}(D_p)/dD_p$ instead of $dN_{tot}(D_p)/dlogD_p$ as the factor in between those two eventually cancels out.
Besides: I recommend adding another line to Equation 9, in which you rearrange it as follows:

$f_{act,GF1<GF<GF2}(D_p, GF_1, GF_2)= ( dN_{tot}(D_p)/dlogD_p * f_{tot,GF1<GF<GF2}(D_p) – dN_{int}(D_p)/dlogD_p * f_{int,GF1<GF<GF2}(D_p) ) / dN_{tot}(D_p)/dlogD_p * f_{tot,GF1<GF<GF2}(D_p)$

where $f_{tot,GF1<GF<GF2}(D_p)$ is the number fraction of particles (total aerosol) with dry diameter $D_p$ and GF between GF1 and GF2 (and equivalent for the interstitial particles). This addition should help in understanding the meaning of Equation 9.

23) P.9, l.3ff:
"The most interesting remark concerns the difference between the low and high hygroscopicity particles at 120 and 150 nm. While the activation efficiency of total aerosol and more hygroscopic particles increases with size, the less hygroscopic particle mode remains almost non-activated." – The "size dependence" mentioned in this statement is distracting from the main message. In my view Table 1 already captures the central and very nice results of your study, which is: "...., the cloud droplet activated fraction of the less hygroscopic particles is much smaller than that of the more hygroscopic particles of equal size…which confirms that cloud droplet activation critically depends on particle hygroscopicity for particle sizes for all sizes in the range of the droplet activation cut-off…." The very nice thing is that you showed this, which is expected based theory and hygroscopicity-resolved HTDMA-CCN closure studies, for the activation of atmospheric aerosols in atmospheric clouds. Personally I would focus on this, i.e. comparing less versus more hygroscopic at equal size, and address size dependence in the next paragraph.

24) P.9, l.11ff:
"Here, the residual aerosol-properties were estimated indirectly by using the hourly averaged total and interstitial GF-PDFs and their actual number concentrations." – Which factors did you apply to the normalized GF-PDFs, $dN_{tot}(D_p)/dlogD_p$ and $dN_{int}(D_p)/dlogD_p$ for the total and interstitial inlets or did you revert the normalization factor of the GF-PDFs with the normalization factor that had been applied? I would believe that the former is the better choice, if the total and interstitial DMPS measurements are corrected such that they are identical for out of cloud measurements. However, this is just a subtlety.
PS: Applying the number of counts of the HTDMA raw measurements would be "wrong" because the detection probability in the HTDMA is GF-dependent for a fixed dry size. However, again just a small but still systematic bias.

25) P.9, l.10-17: why do you not discuss the size dependence of the activated fractions of the more and of the less hygroscopic particles in this paragraph? There are good reasons for how they depend on size and why there is hardly any difference between total and interstitial inlet at the smallest covered size.

26) Figure 1:
According the legend in the bottom row of Figure 1 the difference of the average GF between total and interstitial inlet is 0.04 for the two dry diameters 80 nm and 120 nm. However, the difference between total and interstitial seems to be much larger for 120 nm compared to 80 nm when looking at the bottom row of Figure 1. Please check carefully and adapt the figure and discussion on P.9 l.10-17 if needed.

27) P.9, l.18 ff:
"To our knowledge, this is one of the very few studies characterizing the hygroscopic properties of different in-cloud aerosol populations." – there might exist some CCN based literature on this topic; you could check for authors like U. Pöschl and D. Rose.
Concerning chemical composition: you could check for SP2-based studies by J. Schroder et al. This might potentially link in to the behaviour of the less-hygroscopic particles.

28) Figure 2 and associated discussion:
Relevant analysis, however, somewhat incomplete. Equation 5 tells us that the three parameters $S_c$, $\kappa$ and $D_{50}$ are related to each other. Therefore, the relation between the three of them should be reflected in the analysis, figures and discussion. Some thoughts on this:

a. The dependence of $D_{50}$ on $\kappa$ could be the result of cross-correlation rather than causality. You should confirm that $\kappa$ and $S_c$ are not correlated to make your result stronger. This is definitely required before you make the statement at the end of Section 3.3.

b. Color code: the most relevant information I seem to learn from the colour code is that the variability of $\kappa$ is for the most part driven by the variability of the number fraction of less and more hygroscopic particles rather than the variability of the respective mean GFs of these two modes. Correct? This would be better seen from a scatter plot of $\kappa$ versus $f_{GF<1.25}$.

c. Fit curve: the fit curve can be quantitatively interpreted, i.e. it provides you an estimate about the $S_{c,eff}$ "averaged" over the whole data set. Is this value consistent with your other analyses of $S_{c,eff}$? Caveat: Equation 5 is an approximation, which is likely not accurate for the rather low critical supersaturations you are dealing with. ➔ see next comment.

d. I suggest you include multiple theoretical lines in Figure 2 that show $D_{50}$ versus $\kappa$ for different $S_c$ (based on unbiased numerical solutions rather than the approximate Equation 5). These theoretical lines might possibly save you the trouble of including a fit curve. Additionally you should choose the $S_{c,eff}$ as colour scale for the data points (you can have multiple versions with different colour scales if you like to keep your old colour scale too). This will give you a more complete picture on the influence of $\kappa$, $S_{c,eff}$ and also "measurement noise" on the variability of $D_{50}$.

e. You should also create a figure in which you swap the roles of $\kappa$ and $S_{c,eff}$, i.e. you plot $D_{50}$ vs $S_{c,eff}$ and choose $\kappa_{avg}$ (I'd say for 120 nm or 150 nm or a value interpolated to the mean D50) as colour scale (theoretical lines should also be added). How does it compare with the figure suggested above?

f. The outlier in Figure 2: is it an outlier in the sense of "cannot be explained" or do you have independent evidence that the very high $D_{50}$ could possibly be caused by exceptionally low supersaturation (you cannot use $S_{c,eff}$. to argue as $S_{c,eff}$ is inferred from D50)?

29) P.10, l.28-29:
"…the estimated peak supersaturations… …they provide some valuable information about the in-cloud conditions…". – The droplet activation happens at the initial stages of cloud formation.

30) Last paragraph of Section 3.4 (comparison of supersaturations with literature):
Hammer et al. (2014) reported a systematic difference in observed peak supersaturations for the two prevalent wind directions, which could be explained by differences in the orographic forcing (steep vs gentle mountain slopes). What are the cloud formation mechanisms for the clouds probed at Puijo (and Pallas)? Are the lower peak supersaturations at those two sites possibly related to weaker orographic forcing compared to the Puy de Dôme and Jungfraujoch sites?

31) Figure 4 and associated discussion:
The susceptibility of cloud droplet concentration to hygroscopicity can be quite asymmetric with respect to increase vs decrease of $\kappa$ (see e.g. Figure 8 in Juranyi et al., 2010, or other studies that did similar sensitivity analyses for CCN number concentrations). Instead of just considering the case "no less hygroscopic particles at all" (➔ higher $\kappa$), you could additionally consider the case "no more hygroscopic particles at all" (➔ lower $\kappa$) for the sensitivity analysis presented in your Figure 4 and Table 2.
Jurányi, Z., Gysel, M., Weingartner, E., DeCarlo, P. F., Kammermann, L., and Baltensperger, U.: Measured and modelled cloud condensation nuclei number concentration at the high alpine site Jungfraujoch. *Atmos. Chem. Phys.*, **10**, 7891-7906, doi:10.5194/acp-10-7891-2010, 2010.
PS: further down in the manuscript it became clear why you specifically look at positive deviations. You could try to clarify this earlier.

32) Concerning difference of the activation behaviour of the two modes:
As the aerosol at Puijo appears to have two rather well separated hygroscopicity modes, and since you prove that this directly affects the cloud droplet formation ability, you could quantify the expected difference of activation cut-off diameter for these two modes, if you like. One option would be the following: from "every" HTDMA measurement you can infer $S_{c,eff}$, $\kappa_{avg,GF<1.25}$ and $\kappa_{avg,GF>1.25}$. This allows to infer $D_{50,GF<1.25}$ and $D_{50,GF>1.25}$. Plotting $D_{50,GF<1.25}$ and $D_{50,GF>1.25}$ versus $S_{c,eff}$ then gives a fair idea of the activation cut-off diameter of the two modes, which is for example relevant for the threshold size down to which the particles in either mode can undergo cloud processing under the conditions in clouds at Puijo. (The only thing you would have to think about is how to deal with the diameter depends of hygroscopicity.)

33) There is another potentially interesting question you could look at if you like: while assuming an internally mixed aerosol can provide very good estimates of the total CCN number concentration, if properly done, it will not give an accurate answer concerning the respective contributions of the background aerosol and local/regional emissions to CCN number (with the picture in mind that the less hygroscopic mode is of local/regional origin). Based on your data set you could make at least a crude estimate of how the number fraction of local/regional particles compares between total aerosol and those particles that formed cloud droplets (pulling the idea of the previous comment even a little further). Or in other words: your data set seems to imply that most particles of local/regional origin have to undergo quite some atmospheric aging processes until they start participating in cloud droplet formation, doesn't it?

34) P.11, l.26-27:
"Understandably, by suppressing the size-dependent variations in chemical composition, the activation curves become steeper and the D50s decrease." – This is unclear. You only present data from a single size, so how can size dependence be supressed? To my understanding the D50s decrease because you make the particles more hygroscopic! Please clarify how you mean this.

Technical corrections:

35) P.7, l24:
In the context of HTDMA measurements I would speak of "non-hygroscopic" particles for GF=1.0 rather than hydrophobic. I'd rather use the latter term in the context of measurements that are sensitive to "wettability", i.e. adsorption or contact angle or similar.