

Dear Editor,

As a leading author of the acp-2015-330 manuscript, and on behalf of my co-authors, I would like to express my sincere gratitude for the work you have done to make sure the processing of my paper is smooth and on time. We all appreciate it. Thank you.

All of the referee comments have been addressed to the best of my ability and certain changes to the manuscript have been implemented. The responses to the referee comments have already been uploaded, and can also be found below.

Once again, thank you for your work. Let me know about the outcome of the revision process and whether any additional modifications need to be implemented.

Mikhail Paramonov

Anonymous Referee #1

Received and published: 12 July 2015

This paper provides an overall analysis of the CCNC measurements in EUCAARI sites with an emphasis to develop common features based on the data from various sites. Although there is not a lot of new science, it has improved the understanding of the characteristics of the EUCAARI datasets with a few important overall observations. It is well written and clear and I just have a few minor questions for the considerations of the authors.

Response: The authors of the current manuscript would like to sincerely thank the referee for the constructive comments, questions and suggestions. All of the comments have been carefully considered and addressed, and responses can be found below after each comment, in italics. Nomenclature symbols used in the responses are the same as in the original manuscript.

Comments

1) It is said that water activity was asked to be parameterized according to EAIM or ADDEM. What is the outcome of such parameterization? Estimation of K from composition?

Response: The sentence in question has been modified and now reads “To predict S_{eff} for instrument calibration, water activity was asked to be parameterised according to either the AIM-based model (Rose et al., 2008) or the ADDEM-model (Topping et al., 2005).” I hope this is what was meant by the comment.

2) D_c , wherever available, is suggested to be included in Table 3.

Response: While the inclusion of D_c can provide more results in the paper, actual absolute values of D_c are not discussed anywhere in the manuscript and are outside of the current scope of the paper. The decision not to include D_c in Table 3 or elsewhere in the manuscript was due to the following reasons: 1) Recalculation of D_c to correspond to the ACTRIS S_{eff} levels (page 15051, lines 13-18) is not possible for all stations/campaigns due to insufficient data, and is prone to errors due to differences between the actual and target (Table 3) S_{eff} levels. 2) The inclusion of D_c values in Table 3 or elsewhere would warrant an expansion to the discussion. As mentioned above, actual absolute D_c values are not currently discussed in this paper and are outside of the scope of the paper. 3) If D_c values are included in table 3, one may then argue for the inclusion of kappa values as well, which is, again, not possible for all locations and is not discussed in the manuscript. The discussion about the absolute D_c and kappa values for each location can be found in original publications referenced in Table 2. I hope the referee sees why values of D_c and kappa have not been included in Table 3.

3) Figure 4 and 5 are key results of the paper. While it is understood that A increases as S_{eff} increases, it is less clear why the data are not in a sigmoidal shape. Pls explain.

Response: The sigmoidal shape of the activation curve is expected when A is examined as a function of particle size for any one given supersaturation S_{eff} . Figures 4 and 5 examine A as a function of supersaturation S_{eff} , not particle size. In this case, even at very low S_{eff} levels activated fraction A is not zero and rapidly increases as S_{eff} increases. As pointed out by the r values in Table 4, the utilised fitting procedure fits the data very well for the studied S_{eff} range.

4) The discussions of the results are categorized based on the groupings as a result of Figure 4 and 5. While this is useful in a European context, it may be useful to the general audience if there are discussions in grouping of CCN results at low S_{eff} and high S_{eff} . It is expected that at high S_{eff} , particles are easily activated and hence the total CN concentrations would play a dominant role in the total CCNC. At low S_{eff} , hygroscopicity and size distributions may be more important. Maybe some discussions with an expanded Table 3 can give more insights on the characteristics of the CCNC results. For example, it is interesting to know the sensitivity of CCNC to Kappa under different conditions.

Response: The author is correct when suggesting that the discussion about the results can be presented in several different ways, including that of grouping CCN results by Seff levels. The referee does, however, also acknowledge that the way it is presented in the manuscript currently is useful in the European context – something the paper tries to achieve since it deals with the EUCAARI community. I am not sure if rearranging the discussion at this stage would improve the quality of the manuscript. I would also like to draw the referee's attention to the fact that I do briefly discuss which effects are important at which Seff levels in the first paragraph of section 3.1 (page 15051, lines 23-26 and page 15052, lines 1-8). I hope this is sufficient.

5) The use of N50 and N100 as the basis for calculating A to reduce the variations of the results is interesting. It would be useful if there can be more discussions on how these observations can be generalized. For example, it appears that these general trends happen when there is an abundance of particles smaller than 50nm, which are not easily activated. The difference (ratio) in A100 and A50 is rather constant at Seff of larger than 0.4% or so, which implies that the N50/N100 ratio of these sites are pretty constant.

Response: The generalisation of observations based on A100 and A50 currently cannot be performed adequately, I'm afraid, due to a small number of datasets for which N100 and N50 data were available (as seen by the number of curves in Fig. 5). This is most certainly one of the aspects of the paper that could be investigated in more detail had more data been available. One could also consider using e.g. A10 and A25 to investigate the effect of size distribution on A and to reduce the variation of results. At the current state, the manuscript tries to draw the reader's attention to the fact that A_{xx} can and, in principle, should be used together with normally-derived A for a more accurate comparison and for investigating the effect of size distribution.

6) There are discussions on the presence (and absence) of diurnal patterns of aerosol hygroscopicity at different sites and in different seasons. Can one argue that the lack of a diurnal pattern indicates the role of long range transport? Local meteorological effects and photochemical activities, which would lead to diurnal changes, did not happen.

Response: For the size range of aerosol particles discussed in this paper (>50 nm), long-range transport is probably always the dominant source of the particles. At the same time, the most likely reason for the absence of a diurnal pattern of CCN activation and hygroscopic properties is simply that processes occurring on a diurnal scale are too slow/short to affect D_c and κ significantly. The observed and discussed diurnal patterns are most probably not a question of long-range transport, but rather of time scales.

7) The statement in conclusion "that in most cases the size distribution and its variation have a larger effect on the NCCN than the particle hygroscopicity and its variation with size" seems valid, especially for the sites shown in Figure 5. I would be interested to see more evidence for the other sites, especially the non-European sites, in this study.

Response: It is absolutely true that an expanded discussion about the effect of size distribution and its variation on Nccn would improve the paper, especially if non-European sites are included. In this paper we simply did not have more data to take the paper there.

Thank you very much, again, for taking the time to read, comment and, therefore, improve the paper.

Anonymous Referee #2

Received and published: 30 July 2015

General comments:

This paper is well written, and provides a clear synthesis of measurements of CCNC performed across various EUCAARI sites. Due to the importance of reducing current uncertainties surrounding the impact of aerosol indirect effects on climate the paper is within the scope of ACP. Whilst there is limited new science presented, the paper does provide an overview of the characteristics of CCN across a wide range of aerosol environments. The description of the measurements sites is clear, and the explanation of the observational strategies and measurement techniques are mostly clear; both of which are described adequately. However, the description of how the data has been treated in the context of the conclusions presented is less clear, and needs further clarifying. I recommend publication after the following comments and minor corrections outlined in the following are addressed:

Response: The authors of the current manuscript would like to sincerely thank the referee for the constructive comments, criticism and suggestions. All of the comments have been carefully considered and addressed, and responses can be found below after each comment, in italics. Nomenclature symbols used in the responses are the same as in the original manuscript.

Comments:

1.) The key conclusions of the paper stem from the results presented in Fig. 4, however, how the curves presented in Fig. 4 are obtained is unclear and needs clarifying. Fig.4 should be reproducible by anyone with the raw data.

Response: The points of this first comment are addresses individually below. It should be noted that we had access to the raw data only for a handful of stations (e.g. Hyytiälä). Other locations provided already averaged station/campaign values (e.g. all University of Manchester datasets). This is mentioned on page 15050, lines 10-13.

Specifically:

Please provide some justification on the choice of the functional form of the fitting used to the observed CCN spectrums with respect to other power laws documented in detail in the literature e.g. Sotiropoulou et al., 2006.

Response: The justification of the fitting used in Figure 4 for the activation curves is the excellent Pearson product-moment correlation coefficient r for the overwhelming majority of datasets (Table 4). For the Seff range studied in this paper (below 1.3%), the functional form of the used fitting fits the data very well. It should be noted that the suggested reference by Sotiropoulou et al., 2006 deals with a) N_{ccn} as a function of Seff and b) a much wider Seff range. Figure 4 in the manuscript examines A (not N_{ccn}) as a function of Seff and for a narrower Seff range. I was unable to find a reference with a similar figure/activation curve fitting for a similar Seff range. If the referee has a suggestion for an appropriate reference, it would be greatly appreciated.

The data from which the fit was performed should be described more clearly in the text, and the measurement points should be overlain on the curves for each station. Was a fitting performed on annual means for each station or from the raw observations and then averaged to get annual activation curves?

Response: Page 15053, the beginning of the section 3.2 now includes the sentence: "Each activation curve in Figure 4 is based on the arithmetic mean values of A calculated from all available data for each station for each Seff level." This was done either by me (for raw datasets) or by the data providers (for station/campaign averages). In its current state, Figure 4 is already a

fairly complex figure. The inclusion of the data points will further complicate and clutter the figure, warranting the expansion of the legend and additional discussion. It is hoped that the added text describing the data in the figure coupled with high r values in Table 4 are sufficient to support the conclusions related to Figure 4.

Assuming the data points stem from Table 3 (this needs clarifying), one would see these point lying exactly on top of the curves for all stations in which the correlation coefficient (r) (Table 4.) is 1.0. A correlation coefficient of 1.0 is surprising in the context of the shape of the curves with respect to observations from previous studies. Overlaying the observations used would clarify this, since it is clear from Fig. 5 that observations do not follow a perfectly smooth curve.

Response: Table 3 contains CCN number concentrations as a function of Seff, while Figure 4 presents A as a function of Seff. I am, therefore, not sure the clarification of what the referee would like to see. As mentioned previously, I was unable to find relevant references about A as a function of Seff for the studied Seff range to support the choice of the fitting procedure. And, again, the overlaying the observations means the inclusion of over 100 data points to an already busy figure. I have carefully looked at the data and rerun the computations to double-check and confirm the correctness of the Figure 4 and Table 4.

It is shown that a more stable dependence of A on S is found when employing N50/N100 in Fig.5 compared to Fig. 4, however, in Fig.5 a log-y-axis is employed. The comparison should be made using the same axis as this is minimising the differences between the stations compared to Fig. 4. In Fig. 5 the activated fraction can be >1. This is assumed to be because $D_c < D(N50, N100)$, however, some clarification in the text would be beneficial. In addition, since see values >1 for N50 does this not suggest the use of N100 is redundant?

Response: The utilisation of a linear-y-axis in Figure 5 leads to exactly the same results – curves in Fig. 5 are closer to each other than in Fig. 4. Activated fractions over unity logically stem from critical diameters being less than 50/100 nm, as rightly pointed out by the referee. The discussion on pages 15055-15056 coupled with Figure 5 should make it rather evident why some A values in Figure 5 are above unity.

2.) The description of the methodology used to obtain size-dependent and independent kappa within the contexts of the results and conclusions presented needs clarifying. In particular, in the abstract:

“In a boreal environment the assumption of a size-independent k can lead to a potentially substantial overestimation of NCCN at S levels above 0.6 %; similar is true for other locations where k was found to increase with size.”

*Response: No size-independent kappa values were provided, calculated or discussed in this manuscript. The sentence above deals only with the **assumption** of a size-independent kappa for one particular dataset (Hyytiälä). How this conclusion was reached is described on page 15058, lines 4-8. The procedure for calculating size-dependent kappa values is described in the methodology section, page 15051, lines 5-9.*

Specifically:

In section 3.3 it is mentioned that kappa was provided, but more detailed explanation of the differences in how it was calculated is required with respect to the calculation of a size-independent kappa and conclusions presented. Linking text to K_{calc} in table 2 would be beneficial.

Response: Again, no size-independent kappa values were calculated in this work. In order to obtain the conclusion presented in the abstract we calculated Nccn with D_c when $K_{acc} = K_{Aitken}$ and compared it to Nccn calculated with D_c when $K_{acc} \neq K_{Aitken}$. A sentence has been added to the first

paragraph on page 15058: “Nccn was calculated using the median annual size distribution and Dc calculated with size-dependent and the assumed size-independent κ values.”. I hope it is clearer now how this conclusion was reached. I am not sure why the referee mentions κ_{calc} here, since κ_{calc} is the same as κ , except that Dc_{calc} used to calculate κ_{calc} stems from non size-segregated CCNC measurements (combining CCNC with DMPS/SMPS as described on page 15050, lines 27-28 and page 15051, lines 1-9). The procedure for calculating κ and κ_{calc} is identical, the difference is in how Dc was obtained.

The number of data points for kappa in Fig. 6 varies between the measurement stations and does not correspond to the number of supersaturation bands (and thus Dc values) expected from Table 3.

Response: The reason that the number of data points for kappa in Fig. 6 varies between the measurement stations is simply because different stations/campaigns operated CCNC at different levels and number of levels, as is evident from Table 2. If the number of Seff levels in Table 2 does not correspond to the number of data points in Figure 6 (the case only for COPS, K-pusztá and RHAMBLE datasets), no kappa values (NaNs) were provided in the submitted data for certain Seff bands. Table 3 is unrelated to Figure 6.

A clearer explanation of the methodology and figure illustrating this overestimation should be provided to clarify whether this conclusion is derived from previous studies (thus more clear citation required), or from analysis performed in this work (thus figure and clearer description of methodology required).

Response: It should be clear from page 15058, lines 4-8 that the conclusion stems from the analysis performed in this work, hence no citation. With an added sentence, the related discussion now reads: “The effect of extending the accumulation mode κ down to the Aitken mode was examined using detailed data from Hyytiälä as an example. Nccn was calculated using the median annual size distribution and Dc calculated with size-dependent and the assumed size-independent κ values. It was found that if κ of the accumulation mode is assumed to be the same for the Aitken mode, the NCCN, on average, is overestimated by 16% and 13.5% for the Seff of 0.6% and 1.0%, respectively.”. If the referee still deems this description unclear and confusing, I can add more text to this section. I am not sure, however, what kind of figure would aid in the understanding of this calculation.

3.) The critical diameter (Dc) is mentioned often in the text, and therefore should be provided in one of the tables to clarify how the data presented in the figures is provided. Specifically, some clarification in data reporting is required with respect to the figures. The 5 values of Dc corresponding to the SS bands are required in Table.3, thus, in addition some explanation in the text is required as to the number of data points in Fig.6, particularly the RHAMBLE site which has <5 data points. Linking text to D_{calc} in table 2 would be beneficial.

Response: While it may be said that Dc is mentioned often in the text, in its current state the manuscript focuses primarily on the quantities of Nccn, activated fraction and hygroscopicity, as can be seen from the sectional breakdown of the Results and Discussion section. The only detailed discussion of Dc in the later part of section 3.3 deals with the annual and diurnal variation of Dc as an indicator of hygroscopicity. The discussion about CCN hygroscopicity focuses on its dependence on particle size. Actual absolute values of Dc and kappa are outside of the current scope of the paper and are not discussed anywhere in the manuscript. The decision not to include Dc in Table 3 or elsewhere in the manuscript was due to the following reasons: 1) Recalculation of Dc to correspond to the ACTRIS Seff levels (page 15051, lines 13-18) is not possible for all stations/campaigns due to insufficient data, and is prone to errors due to differences between the actual and target (Table 3) Seff levels. 2) The inclusion of Dc values in Table 3 or elsewhere would warrant an expansion to the discussion. As mentioned above, actual absolute Dc values are not currently discussed in this paper and are outside of the scope of the paper. 3) If Dc values are included in table 3, one may then argue for the inclusion of kappa values as well, which is, again,

not possible for all locations and is not discussed in the manuscript. The discussion about the absolute D_c and $kappa$ values for each location can be found in original publications referenced in Table 2.

As mentioned in the response to the previous comment, if the number of Seff levels in Table 2 does not correspond to the number of data points in Figure 6, no $D_c/kappa$ values (NaNs) were provided in the submitted data for certain Seff bands. Recalculation to ACTRIS Seff levels was done only for Nccn and that is what is seen in Table 3.

4.) Previous studies have shown that Nccn/Na over land decreases with altitude. For the results presented herein to be relevant for climate modellers some discussion on the results in the context of altitude height and ambient relative humidity of the observations would be extremely beneficial. In addition, some additional discussion of the differences between the observations of CCN with respect to their vastly ranging altitudes would be beneficial to put the observations in context of atmospheric processes affecting the observed characteristics.

Response: While it would certainly be of benefit to include some discussion about altitudinal differences, as suggested by the referee, at this point with the current dataset it is rather impossible to draw any meaningful conclusions about the vertical variation of CCN characteristics. The overwhelming majority of sites is within the well-mixed planetary boundary layer (PBL), so that even if the altitudes are different, altitudinal differences are negligible. As for sites in the free troposphere, the two sites of Jungfraujoch and COPS are insufficient to warrant a discussion about CCN differences with altitude. There are simply not enough data in our case to do this.

Minor comments:

1.) Introduction, p15044 line 14: make clear not only talking about total activated fraction. CCN only provides number particles that will activate at *specific* supersaturations.

Response: The sentence in question has been modified and now reads: "However, atmospheric aerosol is frequently externally mixed, with particles of different sizes exhibiting different chemical composition, and, therefore, in practice, D_c is usually estimated as the diameter at which 50% of the particles activate and grow into cloud drops at any given S ."

2.) Introduction, p15045 line 19: Define aerosol activation efficiency.

Response: The sentence in question has been modified and now reads: "While undeniably important, the effect of size distribution on NCCN and the size-resolved activated fraction (e.g. Dusek et al., 2006; Quinn et al., 2008; Morales Betancourt and Nenes, 2014) is not investigated herein, and an overview of the existing EUCAARI aerosol size distribution data can be found in Asmi et al. (2011) and Beddows et al. (2014)."

3.) Introduction, p15044, line 20: define critical supersaturation in the context of D_c and CCN observations.

Response: The sentence in question has been modified and now reads: "The effect of hygroscopicity on the activation of CCN into cloud drops has also been studied extensively, and several simplified theoretical models have been suggested to link particle composition with critical supersaturation S_c , i.e. the minimum S required for the particles of a certain size to activate into cloud drops (e.g. Svenningsson et al., 1992; Rissler et al., 2005; Khvorostyanov and Curry, 2007; Wex et al., 2007)."

4.) Introduction, p15044, line 23: k , also known as "kappa".

Response: The sentence has been altered and now reads: "One such approach is the hygroscopicity parameter κ , also known as "kappa", a unitless number describing the cloud condensation nucleus activity (Petters and Kreidenweis, 2007)."

5.) In section 2.1: Since this paper is focussed on presenting CCNC measurements, a more detailed description on how CCN is linked to different instrumentation SS bands would be beneficial and the uncertainty associated with the variability in these fixed bands during the measurement period.

Response: It is maybe not entirely clear what is meant by this comment. The discussion of how Nccn is related to Seff is found on page 15043, lines 25-29 and page 15044, lines 1-4. Potential deviations from nominal Seff levels and the calibration technique used to correct them are discussed in the second paragraph of section 2.3. Since the exact operating procedures of CCNC vary greatly among locations, it may be more useful to look at the corresponding references for each station/campaign. The sentence on page 15046 has been modified and now reads: "Typically, a CCNC operates at several different levels of Seff, most commonly ranging between 0.1 and 1.0%; the deviations from the nominal assigned Seff values can be monitored and corrected by applying a standardized calibration procedure, as described in section 2.3." I hope this is what was meant by the comment.

6.) Section 2.2: It would be beneficial to provide more information on why the 14 stations used in the paper were selected, and why certain EUCAARI stations were not included. Were these the only stations with the necessary observations?

Response: We used all the data that were provided to us by the respective responsible institutions and station representatives. If an EUCAARI station is not included in the analysis, the station simply did not provide any data. I expect this to be evident from the beginning of section 2.2, p. 15047, lines 2-3.

7.) Section 2.2, p15047, line 20: If the site is used to monitor pollution transport it is unclear how it is not affected by local pollution.

Response: The description of the Vavihill station, as stated in the text, comes from the indicated reference by Tunved et al. (2003). I suppose what is meant is that Vavihill is a background station where local sources of pollution are insignificant. However, due to its geographical location, it can capture the transport of pollution from continental Europe to the Nordic countries.

8.) Section 2.2, p15049, line 5: Author surname lon > Lon?

Response: Author's surname is lon indeed. Nothing changed.

9.) Section 2.3, p15050, line 11: Please clarify which stations were submitted in which temporal format in the associated table.

Response: Table 2 now includes an additional column indicating whether the data were submitted in the original or averaged time resolution.

10.) Section 2.3, p15050, line 23: "can potentially affect some of the conclusions" This needs expanding, at least in brief, and in addition some discussion regarding the uncertainty in the parameterisations themselves.

Response: Indeed, deviations from the nominal Seff values can potentially affect some of the conclusions presented in the paper. However, exactly what kind of effects these deviations can cause depends on the sign and the magnitude of the deviation, other aspects of the instrumental setup, calibration procedure and the provided and calculated parameters. Since I do not specify and actually do not know anything about these deviations, I would refrain from expanding this part since I cannot be certain about which of my conclusions can be affected by these deviations. The sentence about the water activity parameterisations has been modified and now reads: "To predict Seff for instrument calibration, water activity was asked to be parameterised according to either the AIM-based model (Rose et al., 2008) or the ADDEM-model (Topping et al., 2005); both of these

models can be considered as accurate sources of water activity data, and the discussion about their associated uncertainties can be found in the corresponding references.”

11.) Section 2.3: Please provide equation by which size-independent kappa was calculated for future reference since kappa forms an important aspect of discussion in this study.

Response: As has been already addressed in Major Comment #2 (see above), no size-independent kappa values were calculated in this manuscript. The procedure for calculating size-dependent kappa values is described in the methodology section, page 15051, lines 5-9. I have included the equation by which kappa values were calculated following the first paragraph on page 15051.

12.) Section 2.3, p15051, line 20: “respectively, assuming a size independent k” Refer to equation (see comment 10).

*Response: Again, the sentence above deals only with the **assumption** of a size-independent kappa for one particular dataset (Hyytiälä). The kappa values that are discussed throughout the manuscript are size-dependent (presented as a function of size), and the procedure for calculating them is described in the methodology section, page 15051, lines 5-9. Equation added.*

13.) Section 2.3, p15050, line 27: “For some of the polydisperse datasets” Which? & why were only these used?

Response: Similarly to comment #6 above, this refers to sites where DMPS/SMPS data were actually available. I have modified the beginning of the sentence and it now reads: “For some of the polydisperse datasets, where available, Differential/Scanning Mobility Particle Sizer...”. Which datasets these actually are can be easily seen from Table 2.

14.) Section 2.3, p15051, line 9: “typically leads to an overestimation” Please clarify why and by how much?

Response: The sentence in question has been modified and now reads: “Due to the surface tension of actual cloud droplets being lower than that of pure water droplets (Facchini et al., 2000), this assumption, although commonly used, typically leads to an overestimation of the Nccn (Kammermann et al., 2010b).” Within the intent and the scope of the current paper, the exact magnitude of such overestimation is trivial.

15.) Section 3.1: p15052, line 1: “total particle number concentrations”, please expand i.e. 100% activate at this supersaturation.

Response: It is unclear what is meant by this comment. The Seff of 1.0% does not result in the activation of all (100%) aerosol particles, but only in a certain and large fraction of them. At this Seff level, however, the effects of size distribution and hygroscopicity are minimised.

16.) Section 3.1: p15053, line 5: “under the clean and convective conditions: : :” This is unclear, in the context of the following discussion since Pallas/Cabauw experience significantly different Ncn. Therefore, the discussion at end of this section needs clarifying for accessibility. Also some discussion of the concentrations with respect to the altitude of the observations would be beneficial, or clarification as to why this is not given more attention.

Response: The sentence “It has been reported that, although under the clean and convective conditions ambient Sc may reach as high as 1.0%, in the polluted boundary layer Sc usually remains below 0.3% (Ditas et al., 2012; Hammer et al., 2014; Hudson and Noble, 2014).” is a general statement, not necessarily about any particular location. When I discuss the assumed implications of this statement with respect to Pallas and Cabauw, I do not talk about the absolute

values of N_{ccn} or N_{cn} , rather about the fraction of aerosol that would activate into cloud droplets (page 15053, lines 8-10).

As to why there is no discussion about the altitudinal differences in CCN characteristics, please, refer to the comment #4 addressed on page 4 of this document.

17.) Section 3.2: Confidence bounds in the form of prediction bound bars for each individual measurement site would be beneficial in the context of interpreting the overall fit.

Response: While individual prediction bound bars would be of interest, the reason for not including them, again, goes back to the issue of an already busy figure becoming more cluttered. Of course, error bars (standard deviation values) would show the individual variability of A; however, for the current purpose I would like to draw the focus towards the similarity among locations rather than the individual variability, which is, of course, present as well.

18.) Section 3.2: p15053, line 17: “except those for: : :”: please justify why these sites were excluded?

Response: The sentence in question has been modified and now reads: “Included in the figure is the overall fit shown with prediction bounds (95% confidence level) based on most of the activation curves, except the outlying ones of Finokalia, COPS, Jungfraujoch and Pallas A, B and C.”

19.) Section 3.2: p15053, line 19: “no discernible difference in how A responds”. Fig. 3 does show discernible differences; please clarify this statement in text with some numbers from the figure.

Response: I suppose referee means Figure 4, not 3. What I mean when I say “no discernible differences” is that if I were to include the error bars for each activation curve, it would be very obvious that there is no apparent difference in how A responds to S for most of the locations. And instead of cluttering the figure further with error bars, I included the prediction bounds of the overall fit for better visual representation.

20.) Section 3.2: p15053, line 21: “Therefore, the average total number concentration N_{cn} alone is sufficient in order to roughly estimate: : :” Please provide some examples in terms of calculated numbers as evidence here, or better, a figure to back up this *main* conclusion.

Response: The following sentences have been inserted into the first paragraph of section 3.2: “The appropriateness of the overall fit for estimating N_{ccn} based on N_{cn} alone was investigated for the whole Hyytiälä dataset, by comparing the N_{ccn} measured by the CCNC with the N_{ccn} calculated using the N_{cn} and the overall fit presented in Table 4. Such a comparison revealed that for Hyytiälä the overall fit leads to an annual median overestimation of NCCN of 49, 41, 33, 17 and 2% for the Seff levels of 0.1, 0.2, 0.3, 0.5 and 1.0%, respectively.”

21.) Section 3.2: p15053, line 20: “on an annual basis”: Please clarify whether referring to averages here from annual data.

Response: A sentence has been added to the first paragraph of the section 3.2: “Each activation curve in Figure 4 is based on the arithmetic mean values of A calculated from all available data for each station for each Seff level.”

22.) Section 3.2: p15055, line 13: “seem to result in no apparent difference in the fraction of the aerosol that activates into cloud drops”. This paragraph needs clarification as to why, is this supposedly due to cancellation affects between k and N_{cn} , if so is there a hypothesis as to why these are always cancelling?

Response: At this stage, the speculation as to why we observe no differences among many locations depicted in Fig. 4 is beyond the scope of this paper. Especially so since we do not have enough data for enough locations to support such speculations. The goal here is to present the result and to draw the reader's attention to this it.

23.) Section 3.2: p15055, line 17: "seasonal variability, which, as can be seen": Please clarify where this can be seen.

Response: The same sentence from which the quote above is taken includes the reference to Fig. 4. That is where the seasonal variability can be seen.

24.) Section 3.2: p15055, line 21: "does not capture the variability on shorter time scales": The implications of this needs to be expanded upon in the conclusion and discussion with respect to the main results presented (Fig. 4). How useful are these results in the context of global climate modelling? For instance, can we expect to see the same stable dependence of A on S if the analysis was repeated for seasonal averages?

Response: When talking about Figure 4 and its results, I specifically talk about annual mean values of A and Nccn everywhere in the paper: in the abstract, discussion and conclusion. This is done exactly for the reasons to draw the reader's attention to the fact that it does not represent the variability on shorter time scales, as the sentence in question says. Same page 15055, lines 15-18 has a sentence where I draw the reader's attention to the example of Pallas campaigns, where seasonal variability, as seen in Fig. 4, is very evident. Page 15061, lines 3-4 also states that the linear fit I suggest to use for estimating Nccn can be used for estimating annual mean Nccn.

25.) 15044 line 9-11: "For internally mixed polydisperse aerosol particles, this diameter indicates that all particles above this size activate into cloud drops, and all particles below this size do not". Not necessarily true - only the case in the presence of sufficient water vapour. Please amend.

Response: The previous sentence, where I define D_c , starts with "At any given $S...$ ", where S is supersaturation. The next sentence (the one in question) is obviously tied to the previous one. I have modified the sentence to read: "For internally mixed polydisperse aerosol particles, this diameter indicates that in the presence of a sufficient amount of water vapour all particles above this size activate into cloud drops, and all particles below this size do not."

26.) Section 3.2: p15055, line 25: "lower limit of Ncn unavailable" Why? This was surely reported along with instrumentation for each station.

Response: Unfortunately, no, it wasn't. The initial request for the data submission concerned only the CCNC data, nothing else. We never specifically asked for size distribution data or lower/upper limits of Ncn

27.) Section 3.2: On the motivation of use of N50/N100 due to lack of Dmin information: There is no discussion of potential impact of presence or lack of, of Dmax information on N50/N100 analysis (although it is expected to be of smaller consequence due to very low concentrations of particles $>D_{max}$).

Response: Indeed. D_{max} is not expected to affect Ncn to any degree that would warrant its inclusion/discussion in the text.

28.) Section 3.2: Discussion on aerosol modes with respect to activation: The paper would benefit by including aerosol modal information where available to aid analysis. Please state how N50/N100 was obtained for annual data, is this the average over a whole year? If so the bounds must be substantial.

Response: While, this is an excellent suggestion, unfortunately, aerosol modal information is not available for enough locations/datasets to warrant its inclusion into the discussion. N50/N100 data were obtained from corresponding DMPS/SMPS data (p. 15051, lines 10-11), and the calculated A50/A100 were then averaged for the whole dataset for each Seff level.

29.) Section 4: p15060, line 26: “particle number is often dominated by the Aitken mode particles”: Would benefit from some additional text related to CCN limited regimes as previously discussed in literature, e.g. Reutter et al., 2009 with respect to observed aerosol concentrations in the different modes and shape of CCN spectra.

Response: CCN limited regimes are typically discussed with respect to the activation of CCN into cloud droplets under ambient conditions (updraft velocities and supersaturation levels) and in ambient environments, as is the case in the suggested reference by Reutters et al. (2009). The current manuscript does not directly deal with ambient cloud droplets and ambient levels of water vapour supersaturation, making it somewhat unclear as to how the discussion about the CCN limited regimes would benefit the current manuscript.

Tables/Figures:

Table 1: Please add whether ground based or flight observations for ease of accessibility of station information. Is this table ordered in any particular way, if not would be clearer to rank by Ncn perhaps.

Response: I am confused as to why the referee thinks any of the discussed observations were flights observations. None of the discussed observations were performed by flying, they are not mentioned anywhere, and Section 2.2 describes each measurement station/campaign sufficiently. Table 1 does not include Ncn info, so not sure why I would rank Table 1 entries based on Ncn. The stations/campaigns are listed in the same order in Tables 1 and 2, and are loosely grouped first for long-term datasets and then by the data provider.

Table 3: Please clarify in the text how the averaging was performed for the different sites: Median, mean?

Response: Sentence on page 15051, lines 13-15 has been modified to read: “All Nccn concentrations were averaged for each site for each Seff level and then recalculated to correspond to the target Seff levels suggested by the Aerosols, Clouds and Trace gases Research InfraStructure (ACTRIS) Network: 0.1, 0.2, 0.3, 0.5 and 1.0%.”.

Figure 1: All measurement stations should be provided on global map (as well as subset smaller EU map).

Response: Figure 1 corrected as per suggestion.

Fig. 3: Where is Pallas B? It should be clarified in text why included in Fig. 4 but not Fig 3.

Response: Figure 3 only contains datasets for which the extrapolation of Nccn to ACTRIS Seff levels was appropriate (see missing values in Table 3). Figure 4 deals with activated fraction for actual Seff levels at which the measurements were performed.

References

Reutter, P., Su, H., Trentmann, J., Simmel, M., Rose, D., Gunthe, S. S., Wernli, H., Andreae, M. O., and Pöschl, U.: Aerosol- and updraft-limited regimes of cloud droplet formation: influence of particle number, size and hygroscopicity on the activation of cloud condensation nuclei (CCN), *Atmos. Chem. Phys.*, 9, 7067-7080, doi:10.5194/acp-9-7067-2009, 2009.

Sotiropoulou, R.-E. P., J. Medina, and A. Nenes (2006), CCN predictions: Is theory

sufficient for assessments of the indirect effect? *Geophys. Res. Lett.*, 33, L05816, doi:10.1029/2005GL025148.

Thank you for an interesting paper.

Response: You are very welcome! Thank you so much for taking the time to comment and, therefore, improve this paper!