

Interactive comment on “A synthesis of cloud condensation nuclei counter (CCNC) measurements within the EUCAARI network” by M. Paramonov et al.

Anonymous Referee #2

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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

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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:

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.

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.

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?

Assuming the data points stem from Table 3 (this needs clarifying), one would see these points 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.

It is shown that a more stable dependence of A on S is found when employing N_{50}/N_{100} 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 $Dc < D(N_{50}, N_{100})$, however, some clarification

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in the text would be beneficial. In addition, since see values >1 for N50 does this not suggest the use of N100 is redundant?

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.”

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.

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 D_c values) expected from Table 3.

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).

3.) The critical diameter (D_c) 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 D_c 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}

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in table 2 would be beneficial.

4.) Previous studies have shown that N_{ccn}/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.

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.

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

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

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

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.

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?

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.

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- 8.) Section 2.2, p15049, line 5: Author surname lon > Lon?
- 9.) Section 2.3, p15050, line 11: Please clarify which stations were submitted in which temporal format in the associated table.
- 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.
- 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.
- 12.) Section 2.3, p15051, line 20: "respectively, assuming a size independent k" Refer to equation (see comment 10).
- 13.) Section 2.3, p15050, line 27: "For some of the polydisperse datasets" Which? & why were only these used?
- 14.) Section 2.3, p15051, line 9: "typically leads to an overestimation" Please clarify why and by how much?
- 15.) Section 3.1: p15052, line 1: "total particle number concentrations", please expand i.e. 100% activate at this supersaturation.
- 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.
- 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

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fit.

- 18.) Section 3.2: p15053, line 17: "except those for...": please justify why these sites were excluded?
- 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.
- 20.) Section 3.2: p15053, line 21: "Therefore, the average total number concentration Ncn 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.
- 21.) Section 3.2: p15053, line 20: "on an annual basis": Please clarify whether referring to averages here from annual data.
- 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 Ncn, if so is there a hypothesis as to why these are always cancelling?
- 23.) Section 3.2: p15055, line 17: "seasonal variability, which, as can be seen": Please clarify where this 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?
- 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

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below this size do not". Not necessarily true - only the case in the presence of sufficient water vapour. Please amend.

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

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

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.

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.

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 N_{cn} perhaps.

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

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

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

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Fig 3.

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.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 15039, 2015.

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