

***Interactive comment on* “Robust relations between CCN and the vertical evolution of cloud drop size distribution in deep convective clouds” by E. Freud et al.**

**Anonymous Referee #3**

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Reviewer comment on: Robust relations between CCN and the vertical evolution of cloud drop size distribution in deep convective clouds, Freud et al.

The effects of aerosol input into convective clouds on the precipitation level and the dependence of the development of the cloud droplet size with cloud depth on input pollution levels are areas of critical importance to our understanding of anthropogenic influence on the indirect effect. The current study makes compelling arguments for robust relationships describing the coupling between activable aerosol number and parameters which may define the onset of precipitation in convective clouds.

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The motivation for the work ensures that it is in scope of Atmospheric Chemistry and Physics and the rigour of aspects of the study means that it is worthy of publication.

However, there are is one particular area which require significant attention and further clarification / justification as well as some more minor amendments:

The quantitative aspect of the first major goal relies entirely on CCN measurements made from the UECE aircraft. The temporal and spatial correspondance between aerosol and droplet measurements is sufficient justification for wanting to use these measurements. However, to assess the validity of their use in the analysis, much more information is required than is provided in the current manuscript. A great level of detail is provided in the discussion of the coincidence errors in droplet spectrum measurements by FSSP. These have been previously well documented in the literature and are possibly not required in such detail. In contrast, the CCN measurements were poorly described. The UECE instrument is described as a "DH Associates CCN counter". Firstly, no reference is made to literature describing the instrument principle, construction or calibration procedure. If no reference is made, the required details must be provided in the current manuscript. Furthermore, twice (p10160 & p10162) it is simply stated that the UECE and INPE instruments were intercompared. No data is provided for this intercomparison. To assess the accuracy and validity of use of the measurement data, the intercalibration must be tabulated or illustrated in terms of both number and supersaturation if possible. It is of concern that "there were unexplained large differences between the UECE and INPE CCN measurements for the same days and regions" (p10160). Only the INPE instrument had independent salt calibration and errors have only been quantified for this instrument. What was the frequency of the calibrations. How did the instruments compare before and after the discrepancy? Had the calibration of the INPE instrument drifted? I am not questioning the validity of the approach, but a better instrument description, measurement evaluation and error analysis are required to convince me of the results presented in figures 9 and 10 and the associated text.

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More minor points include the following:

p10157 "40-60% nucleation activity" - presumably this relates to the entire averaged size distribution. This should be clarified

p10157 the stabilisation of the troposphere by aerosol longwave absorption is referred to as a direct effect. Is it not more fashionable to refer to this effect, along with potential cloud burnoff by absorptive layers, as semi-direct and reserve "direct" for scattering?

p10158 the CCN efficiency for natural biogenic and manmade pyrogenic aerosols is quite similar. This should be quantified to be a useful statement.

p10158 is there evidence for the enhanced penetration of smoky deep clouds into the stratosphere? If so, a reference would be useful.

p10159, line 4. If the word "best" is to be used, the other parameters against which the CCN concentration at 0.5% SS has been rated should be named.

p10160, line 2. What were the criteria used to define the "relatively" similar thermodynamic conditions?

p10162 I have no intuitive feel for the range of CAPE values stated. Nonetheless, has the data been examined to establish whether there is a trend towards changes in microphysics that might be expected with increasing updraught velocity with the slightly increasing CAPE values? i.e. is there a significant contribution, even if not the dominant one.

p10164 the sentence beginning The half-second... is ambiguous. It is not clear to what the word "that" refers.

p10165 - 10166 the very significant broadening of the FSSP spectra inferred to explain the match with the hotwire LWC is quite disturbing. It calls into question the usefulness of the FSSP in pyro clouds, since the broadening cannot be consistently assumed to cancel out the coincidence errors.

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p10166 the authors should explain why the D\_L is less affected by the coincidence problem than LWC.

p10166 the authors should also explain how the value of 24 microns for the threshold D\_L was determined for all regimes

p10168 I'd suggest that the strong relation between CCN and Z24 is not as convincing as the lack of an improvement in the relationship between N\_d and Z24 over that of CCN and Z24.

p10172 the last sentence suggests that effective radius is robust enough for remote sensing applications in any location. The measurements only suggest this for the current dataset - this cannot be generalised away from the western Amazon in the current script

The final comment I have concerns the discussion of the reason behind the effective radius relationship to LWC variation due to entrainment in different absolute effective radius regimes (sections 5 and summary). This is really just a question of balance in the discussion. Undoubtedly the three processes described in the text and illustrated in figure 14 will contribute greatly to the evolution of the effective radius on mixing of an adiabatic air parcel with one of lower supersaturation. In addition, as correctly stated the processes will contribute to the effective radius evolution by different relative amounts according to the air parcel's new state (and the new aerosol distribution). However, I'm not too sure how useful it is to frame the hypothesis inferring that these are the only processes occurring based on the current dataset. Will not the interaction between sampling frequency and small scale mixing of parcels of different histories (i.e. microparcel) play a significant role in relationship between effective radius and LWC? That the instantaneous evaporation assumed in previous considerations of inhomogeneous mixing is too extreme may well be true, but sampling will average across a range of turbulence scales, a range of adiabatic fractions and a range of mixed air parcels of different evaporation, coalescence and dilution histories. The apparent independence

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of effective radius and LWC at large effective radius may have a significant contribution from the changing effects of sample averaging across the parcels of changing relative contributions of the processes (which occur on different timescales due to different droplet populations). I don't suggest that the hypothesis is not useful, but that future studies should try to disentangle sampling frequency and mixing scale interaction as well as the contributing physical processes.

I believe that the observed relationships are justification enough for the publication (irrespective of whether the hypothesis can be justified or validated) provided the descriptions of the CCN instrument, measurement and calibration are made in much more detail such that the use of the data are convincing.

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Interactive comment on Atmos. Chem. Phys. Discuss., 5, 10155, 2005.

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