

***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.**

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** Our responses, which start with “***”, follow each one of the reviewer’s comments, which are in *Italics*.

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 rela-

tionships describing the coupling between activable aerosol number and parameters which may define the onset of precipitation in convective clouds. 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 some particular areas 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.

** Response: We have added a reference for describing the UECE CCN counter measurement principle, construction and calibration procedure. Furthermore, we added a figure that shows the CCN spectra of both CCN counters as the result of the inter-comparison between the instruments. The figure shows that the measured CCN concentrations of one instrument are generally within the variance of the other instrument, and that the CCN concentration's dependence on supersaturation was comparable. The figure can be downloaded from the following link: http://earth.huji.ac.il/Data/File/Eyal/CCN_Spectra_BW.PNG

More minor points include the following:

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

** Response: "nucleation activity" relates to the CCN-CN (total number concentration) ratio. This is mentioned now in the text

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?

** Response: Added "semi-direct"

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

** Response: Done

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

** Response: Added a reference

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.

** Response: Omitted "best", changed to "very well"

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

** Response: CAPE (up to 5 km) between 0 and 150 J/Kg. Added to text.

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.

** Response: The stated CAPE values correspond to a fairly stable lower troposphere. CAPE values can easily exceed 2000 J/Kg in unstable cases. Therefore values lower than 100 J/Kg do not favour strong updrafts (as mentioned in the text). CAPE alone is poorly correlated with Z24 (R=0.33 for 7 data points) or with any other "microphysical" parameter. It does not have any significant effect on Z24 (p-value=0.47 in a linear regression). Surprisingly though, its effect qualifies as significant (p-value=0.026) when it is inserted together with CCN0.5 in a linear regression model, but, strangely enough, an increase in CAPE results in a decrease in Z24. We believe it is incidental.

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

** Response: Changed "that" to "2.5 μ s" to clarify

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.

** Response: There is a good match between FSSP calculated LWC and the hotwire LWC (R=0.945) for short average transit times (i.e. when there is no significant co-

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incidence) although the FSSP LWC is on average 20% lower. This could be partly because the FSSP does not count small rain drops while the hotwire does not distinguish between cloud and small rain drops and therefore gets a small water addition. The artificial widening of the droplet spectra caused by the coincidence problem, as stated in the text, can sometimes compensate for the "missing" water, but it is less correlated with the hotwire LWC. It is true that we can not fully trust the droplet spectra measured in pyro-clouds because of the coincidence problem, but we know that it is shifted towards the larger droplet size. But even despite this shift, we get smaller effective radii in these clouds in relation to the just polluted clouds (p.10164 lines 6-17). For pyro-cloud comparing purposes we use the DL which is much less sensitive to the coincidence problem (p. 10166 line 19). In any case we do not draw any quantitative conclusions based on the pyro-cloud measurements.

p10166 the authors should explain why the DL is less affected by the coincidence problem than LWC.

** Response: Ok, added the explanation.

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

** Response: There is a reference to Andreae et al., 2004. The flight scientists saw radar echoes on the aircraft radar and visible impacts of raindrops on the windshield normally when DL exceeded 24 μm during the flights in the developing convective clouds. Added this to the text.

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 Nd and Z24 over that of CCN and Z24.

** Response: One would expect Nd to have a better relationship with Z24 than CCN because Nd should take into account the updrafts. According to our dataset CCN0.5

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has a slightly higher linear correlation coefficient with Z24 than what Nd has (but both are very high). Possible reasons: Small updraft variance. The physical relationship is not necessarily linear. Nd is slightly sensitive to where exactly the aircraft penetrated the cloud (higher droplet concentration near the core). Coincidence in the FSSP has an increasing (and non linear) effect for higher concentrations. Nd is normally decreasing higher in the clouds due to droplet coalescence. We have a fairly small sample (8 coupled datapoints). All these possible reasons (there could be more of course) apparently over-compensate for the theoretical advantage that Nd has on CCN due to its sensitivity to real updrafts, leading to a slightly smaller correlation coefficient with 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

** Response: Rosenfeld and Lensky (1998) have already measured with aircraft in Indonesia robust relations between re and depth above convective cloud base. Added this to the text.

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 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.

**** Response:** Added a paragraph to section 5 and a sentence to the summary to emphasize this important remark.

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.

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

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