Interactive comment on “Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity” by S. S. Gunthe et al.

Anonymous Referee #1

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This manuscript combines measurements of CCN with size distributions and chemistry to derive hygroscopicity parameters for Amazonia aerosols and demonstrates a unique method of calculating the kappa hygroscopicity parameter using inorganic and organic contributions measured by an AMS. The manuscript presents a thorough analysis and the efforts to quantify the errors associated with the measurements are commendable. Nonetheless, the manuscript seems present all of the analyses instead of synthesizing the results for the reader. If my comments can be successfully addressed, I recommend the manuscript for publication. My major concerns are outlined in the paragraphs...
In this work, the authors clearly show that once an average chemical effect (or hygroscopicity factor) has been determined (by the AMS, for example), then aerosol particle number and size are the major predictors for the variability. This is further corroborated by Fig. 15, which shows relatively poor agreement between effective hygroscopicity parameters determined from integral AMS measurements and those determined by the CCN measurements. Subsequently, the conclusions drawn by the authors (P3814; L5 & P3840; L24) should be reworded to reflect these results more accurately, rather than simply stating that particle number and size are the major predictors for the CCN variability. In reading the manuscript, it was not clear if the inorganic and organic fractions were based solely on AMS measurements or if there were additional measurements of aerosol mass to account for the refractory contributions not measured by the AMS. Presumably if the total mass were measured (as a function of size), then $k_p$ predictions would improve.

The authors need to clarify the statement that supersaturation values in publications by Roberts and Andreae are 50% lower than reported nominal values (P3830; L18-24). This statement is likely incorrect as Roberts et al. conducted closure experiments where independent calculations of CCN spectra agree with the measured CCN values. In addition, the references to Roberts et al. should be included in the section 'Comparisons with other studies'.

Several examples of extra analyses include presentation of 2-parameter and 3-parameter CDFs (and ‘a’ & ‘t’ subscripts), N20 and N30 concentrations, three observation periods, and a supplementary discussion that compares various single parameter calculations. While quantifying the sensitivity of different parameters is necessary, I suggest describing the differences between the approaches (if they are relevant) and present one parameter throughout the text for consistency. The 2 & 3-parameter CDFs are in good agreement and certainly within the error of the measurements (P3818; L18-25 & Fig 3); so it is not necessary to present both $k_a$ and $k_t$ throughout the en-
tire manuscript. I also suggest using N30 as the reference for number concentrations throughout the text and drop parameters using N20. As stated by the authors, smaller particles will not activate at the supersaturations measured here. It’s also worth noting in the manuscript that 30 nm is the limiting case for a pure ammonium sulfate particle at ~0.8% supersaturation.

The authors also need to update their reference list and check the order of the figures. One reference is cited without a title (Chen et al.) and the manuscript cites a number of articles that are in preparation or are not easily accessible (user manual and conference abstract). The reference (Frank et al., 2007) is also incorrectly cited (APCD vs. ACP). In addition, the figures are not in sequential order in the text and some of them are not even referenced in the manuscript. Editing the manuscript’s style should not be required by the reviewers, but these oversights detract from its message.

Specific comments to the manuscript are listed below:

P3814; L10: The authors should use caution in extrapolating results from the Amazon to rainforests in Africa which have different sources of aerosols (Artaxo, 1995; particularly, with respect to the much larger contribution of dust in Africa).

P3814; L24: replace _absorb_ with _uptake_

P3817; L5: Cite Rose et al., ACP, 2008 as well.

P3817; L22+: Maximum difference of temperature between the optical particle counter and the temperature of the bottom of the column should be stated as this is particularly important for the measurements at low supersaturations.

Tables 1a and 1b contain the same information and need not be repeated. I suggest condensing Table 1b (keeping the error analysis, but removing ’a’ or ’t’ and columns related to N20) and removing Table 1a.

P3827; L1-8: The authors should reference their choice of hygroscopic thresholds (or describe how they determined the values of these thresholds).
P3829; L9: The statement that coupling remote sensing measurements and kappa values to predict CCN concentrations is tenuous as the size distributions from satellite measurements are not detailed enough to predict CCN concentrations within the range presented in this manuscript. This sentence should be removed.

P3831; L13+: This classical power law approximation is not physically-based and should in general be avoided. I suggest simply reporting the values for comparison to older literature rather expanding a new section to include a modified power law approach.

P3833; L12: replace _little higher_ with a quantitative value

P3852; Table 3: What is significant about the time period ’Rest of the campaign’? It does not seem to present any new information here.

P3853; Table 4: The differences between $k=0.147$ and $k=0.149$ are insignificant; both columns are not necessary.

P3860; Fig. 5: Figure 5 is not referenced. Remove figure or combine with Figure 3.

P3865; Fig 10: Remove figure (not referenced). Verify order and reference for all figures.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 3811, 2009.