

***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.**

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These comments focus primarily on the major results of this article that are presented in Figures 6 and 14. In general, I agree with the referees' comments that the article needs to be refocused, but will limit my comments here to the interpretation of the results for the sake of a scientific discussion.

In figure 6, the focus period and entire campaign show similar kappa values except at the largest diameter (ca. 200 nm). The authors briefly mention the increasing trend

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in kappa, and only discuss the application of an average kappa value in comparisons to measurements elsewhere. The limitation of a globally averaged kappa is implied by one of the reviewers and in the concluding statements of this work.

By turning the problem around, there is valuable information of aerosol chemistry as a function of particle size that can be explored more thoroughly in this paper. Normally, one might expect to see an enhancement in kappa with size as larger particles are generally associated with lower critical supersaturations and are readily involved in cloud processes or chemical aging. However, the observations (change in kappa) indicate an abrupt change in aerosol chemistry and a potentially large source of organic particles during the 'Focus Period'. For example, the decrease of kappa at 200 nm from 0.21 to 0.14 (Figure 6) during the focus period could be a result of a large source of biological parties such as bacteria or spores. Elbert et al., (ACP, 2007) reports a large increase in the emissions of spores after rain events. It's not clear from the article if the 'Focus Period' was influenced by rain events, but the low concentrations of aerosols suggest this may be the case. Similar concentrations of aerosols were also observed by Roberts et al. (2000) after rain events at a site within 100 km of the observations presented here. Perhaps this change in kappa can be explained by the AMS or size distribution measurements. Figure 3 shows a good correlation between median organic and inorganic mass fractions determined by the size-resolved AMS measurements. Can the authors explain this decrease in kappa(p) by an increase in AMS-organic mass fractions for the 200 nm particles during the 'Focus Period'? As a minor note, the error analysis of the AMS-kappa calculations should be included in the figure (more on this in a later paragraph).

It's also worth noting that kappa values estimated from Roberts et al. (JGR, 2002) are ca. 0.1 – while not shown in this article, this value is similar to values reported by Zhou et al. and Rissler et al. in Figure 6. On these lines, what is the basis for the author's statement that the supersaturation values published in Roberts et al. and Andreae et al. are ~50% lower than the reported nominal values? As mentioned by the reviewer,

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the CCN closure was independently verified by Zhou's HTDMA – so one fully expects kappa values derived in the Roberts et al. (JGR, 2002) paper to be similar to the Zhou's values reported in Figure 6.

The mass distributions for the AMS measurements extend to 25 nm (Figure 12), which seems well below the size limit of the AMS. What is the lower size limit of the AMS? What are the associated errors of the measurement? There is no error analysis in Figure 12. The paper should limit its discussion and presentation of data in the figures to sizes above the detection limit of the AMS organic and inorganic mass fractions. Likewise, I doubt that the AMS data is of much use for predicting CCN concentrations at 0.82% supersaturations (Figure 11 & 15) because those CCN are associated to particles that are likely smaller than the threshold particle size of the AMS. In any case, the importance of measuring size-resolved mass fractions seems to be highlighted in this article when comparing Figures 3 and 15, where the comparison of kappa calculated by the AMS to those derived from CCN-measurements is quite good in Figure 3 (size resolved) and less-so in Figure 15 (bulk or integral).

Nonetheless, it is worth noting that these kappa values at large sizes (low supersaturations) are quite low in the Amazon compared to measurements of aerosol at a maritime/long-range transport site in the Indian Ocean (Roberts et al., PNAS, 2008); where 0.1% supersaturation (ca. 200 nm) particles had kappa values approaching that of an inorganic salt aerosol (PNAS Fig. 2). While not entirely surprising given the different regional sources of aerosol, it does present an interesting contrast with respect to cloud development between Blue Ocean and Green Ocean regimes – especially in light of a recent article that highlights the greater sensitivity of cloud development to aerosols at low supersaturations (Reutter et al., ACPD, 2009).

Figure 14 presents a relationship between the organic mass fraction and kappa values and when extrapolated to each limit, the intercepts agree with previous published results for inorganic salts and secondary organic aerosol. The authors also suggest that most of the organic fraction is secondary organic aerosol; however, in light of other

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sources of biological particles with different chemical properties (i.e., spores as discussed above), were there any observed differences in the kappa-OMF relationship during the 'Focus Period'?

Finally, the linear relationship in figure 14 is the basis for an expression for an effective kappa based on organic and inorganic fractions ($k = X(\text{OMF}) \cdot 0.1 + X(\text{SULF}) \cdot 0.6$). However, kappa is defined as a hygroscopicity factor for inorganic salts where soluble fraction plays the dominant role and where surface tension effects are negligible. As most of the particle mass (and volume) is organic, do the authors believe that surface tension effects can be neglected – especially in using kappa to infer the properties of the organic material (such as molecular weight)? This section can be strengthened with a discussion on potential surface tension effects and how they might affect the interpretation of the kappa results for the mostly organic particles in the Amazon.

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