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Interactive Comment

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.

S. S. Gunthe et al.

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We thank Dr. Roberts for constructive comments and suggestions. Detailed responses to the individual comments are given below.

Comment 1:

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 scientific community. 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 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 is 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).

Response:

Rain events occurred on most days of the campaign; the median daily amount of precipitation was higher during the focus period at the end of the campaign (~17 mm d-1; 6-12 March 2008, Sect. 3.1.2) than during the rest of the campaign (~9 mm d-1). This 9, S2918-S2931, 2009

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information will be added in the revised manuscript (Sect. 2.1). The questions concerning aerosol composition have been addressed and partly answered in our manuscript, and we will add further information as detailed below:

p. 3825, l. 14: "As shown in Fig. 5, the effective hygroscopicity of accumulation mode particles was substantially lower during the focus period than during the rest of the campaign ($^{3}0\%$ at D 2 200 nm), whereas little difference was observed for Aitken mode particles (D 50 - 90 nm). At S=0.10% not only kappa but also MAF was lower during the focus period (Table 1), indicating a higher proportion of externally mixed CCN-inactive particles at D_200 nm. These and other aspects of aerosol composition and mixing state will be discussed below (Sects. 3.3 and 4) and in follow-up studies (Pöschl et al., 2009b)."

Electron microscopic investigations of filter samples collected during AMAZE-08 (Sinha et al., 2009) suggest that the contribution of fungal spores and similar primary biological particles in the relevant size range (100-300 nm) were negligibly small (<5%), but other types of solid carbonaceous particles and mineral dust particles were more frequently detected ($^10-20\%$ of particle number in the range of 100-300 nm) and may have been CCN-inactive at S = 0.1%. As mentioned in our manuscript and above, these and other aspects of aerosol composition and mixing state will be further investigated in collaboration with the other partners of AMAZE-08, but they go beyond the scope of this paper. As far as we can tell for now, fungal spores are most likely not relevant for the composition and CCN activity of aerosol particles < 300 nm.

With regard to explaining the decrease in kappa_p by an increase in AMS-organic mass fractions for the 200 nm particles during the "Focus Period" the answer is "yes", as explained in our manuscript and below:

p. 3835, I. 5: "Figure 13 shows the time series of organic and sulfate mass fractions derived from integral AMS measurements, with the organic mass fraction varying mostly in the range of 0.65 - 0.95. Comparison with Fig. 4b reveals that the highest

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organic mass fractions, coincided with the lowest kappa values observed at S=0.10%, i.e. for accumulation mode particles with D \sim 200 nm. Especially during the focus period the organic mass fraction was much higher and less variable than during the rest of the campaign (0.91 +/- 0.03 vs. 0.76 +/- 0.28). On the other hand, elevated sulfate mass fractions, in particular during late February coincided with the highest kappa values observed for accumulation mode particles. In Fig. 14 the effective hygroscopicity parameters derived from the CCN efficiency spectra measured at S=0.10% (kappa_a) are plotted against the organic mass fractions (Xm,org) determined by integral and size resolved AMS measurements, respectively. The close correlation between the integral composition data and the size dependent hygroscopicity parameter (R2=0.81, Fig. 14a) can be explained by the coincidence of the CCN activation diameter at S=0.10% with the maximum of the aerosol mass size distribution (\sim 200 nm, Fig. 12a). The lower correlation coefficient obtained with the size resolved AMS data (R2=0.66, Fig. 14b) is a result of the lower signal-to-noise of these data under the very low concentration conditions of this campaign."

The relevant technical details are specified in the methods section, p. 3822, I.5: "For scatter plots and correlations, the non-size-resolved AMS data were averaged over the time intervals of the CCN measurements (~35 min per CCN efficiency spectrum; 1 - 6 AMS data points per CCN data point). With regard to the size resolved AMS data, the CCN measurement data were averaged over the AMS averaging intervals (~6 h per AMS size distribution; 1-5 CCN data points per AMS data point), and the AMS size distributions were integrated around the midpoint diameters of the CCN efficiency spectra (Da+/-kappa_a)."

Size-resolved AMS data confirm that the higher integral organic mass fractions observed during the focus period ($^{\circ}0.9$) were also reflected by higher size-resolved mass fractions at particle diameters around $^{\circ}200$ nm ($^{\circ}0.9$) relative to the rest of the campaign ($^{\circ}0.7$ integral and size-resolved around $^{\circ}200$ nm). The consistency of size-resolved data for $^{\circ}200$ nm particles and integral AMS measurement data is also demonstrated by the

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similar correlations and trend lines shown in Figs. 14a and 14b.

Comment 2:

It is 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 authors' statement that the supersaturation values published in Roberts et al. and Andreae et al. are \sim 50% lower than the reported nominal values? As mentioned by the reviewer, 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.

Response:

Our statement concerning the supersaturation levels of the CCN measurements reported in Roberts et al. (2001, 2002) and Andreae et al. (2004) is based on the technical note of Frank et al. (2007), who calibrated the supersaturation in the applied CCN counter (static thermal gradient chamber). However, since this issue is not crucial for our study, we intend to remove the questioned statement from the revised manuscript.

With regard to the study of Roberts et al. (2002), we intend to add the following information and statement in Sect. 3.1.3 of the revised manuscript (p.3827, I. 11): Roberts et al. (2002) reported integral CCN measurements, chemical composition data and CCN closure calculations for Amazonian wet-season aerosols as observed during LBA-CLAIRE -98 (March-April 1998). Based on Köhler model sensitivity studies they showed that the observed CCN properties were consistent with average mass fractions of ~15% ammonium bisulfate and ~40% water-soluble organics. Under the assumption of kappa_AHS ~ 0.6 and kappa_org ~ 0. 1 (Sect. 3.3), this would correspond to an average effective hygroscopicity parameter of ~0.13, which is consistent with the overall average kappa_t,avg values calculated from the H-TDMA data of Vestin et al. (~0.15) and Zhou et al. (~0.11), and with the kappa_t and kappa_t,avg values deter-

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mined from our size resolved measurements ($^{\circ}0.13$ for focus period, $^{\circ}0.15$ for entire campaign).

With regard to the studies of Rissler (2006) and Vestin (2007), we intend to focus our comparison on the latter and to add the following information and statement (p. 3826, I.25): "The studies of Rissler et al. (2006) and Vestin (2007) are based on the same HTDMA measurement data sets from the LBA-SMOCC campaign. Compared to AMAZE-08 (average particle number concentration ~200 cm-3), the "wet period" of LBA-SMOCC at the beginning of the wet season (31 October - 14 November 2002) was apparently more strongly influenced by pyrogenic aerosols from local and regional sources (average particle number concentration ~800 cm-3). As discussed by Rissler et al. (2006) and Vestin et al. (2007), the data from LBA-SMOCC exhibited pronounced diel cycles, whereby the daytime values were less polluted and the afternoon hours are most relevant for the influence of aerosols on the development of convective clouds. Thus we focus our comparison on the data reported by Vestin et al. (2007) for the afternoon hours (12:00-16:00 local time). Note that the campaign average CCN and CN measurement data of AMAZE-08 exhibited no pronounced diel cycles (Fig. S6), which is consistent with the trends reported from LBA-SMOCC (i.e., less pronounced diel cycles at lower pollution levels) and confirms that the influence of local or regional pollution was very low during AMAZE-08."

Comment 3:

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.

Response:

We agree and will set the lower end of the plots in Fig. 12 to 60 nm (corresponding to

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[~]85 nm AMS vacuum aerodynamic diameter), and we will add the following information on the AMS measurement uncertainty in Sect. 2.4: "The AMS uses an aerodynamic lens system to focus the particles in a narrow beam and deliver the particles to the high-vacuum detection region. This system is capable of focusing particles in vacuum aerodynamic diameters (dva) of 30-1000 nm with size-dependent particle transmission efficiency (EL, Liu et al., 2007). In this study, we operated the AMS at a sampling pressure of 650-680 torr. Under this condition, EL is close to 100% for particles with dva [~] 100-400 nm and is greater than 20% for particles with dva [~] 50-1000 nm. For organic measurements, the estimated uncertainty is about 30% (1 μ g m-3) to 40% (0.5 μ g m-3), and it can increase to 100% for low organic loadings (0.1 μ g m-3). For sulfate measurements, the uncertainty is <10% for high loadings (0.5 μ g m-3), and about 40% for low loadings (0.05 μ g m-3). Organic and inorganic mass fractions were calculated from the AMS mass concentrations as detailed in Sect. 3.3 (Eq. 3)."

Comment 4:

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).

Response:

We agree that the smallest particles undergoing CCN activation at S=0.82% are at the low limit of the AMS detection range (~50 nm as detailed above), and that integral AMS measurement data are dominated by larger particles. With regard to the CCN activation of small particles at high S, the calculation of kappa_p from integral AMS measurements thus has to be regarded as an extrapolation and approximation.

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Fig. 15 shows that this extrapolation/approximation can lead to a substantial bias in kappa_p (average deviations up to ~77% as specified in Tab. 6 of revised manuscript). Nevertheless, our investigations also show that the kappa parameters derived from the integral AMS data enable prediction of CCN number concentrations with relative deviations <~ 20% (p. 3837, l. 26; p. 3838, l. 19):

"In spite of the positive bias and low correlation of predicted vs. measured kappa, the mean relative deviations between CCN number concentrations predicted on the basis of kappa_p and the measurement values of N_CCN,S were mostly less than 20% at S=0.19 - 0.82% (Table 4)." "Overall, the results of the present study confirm that integral AMS measurement data are useful for the prediction of CCN number concentrations from CN size distributions. Highly precise predictions, however, would require size-resolved AMS measurement data of high precision, which are difficult to obtain at the low aerosol concentration levels of pristine tropical rainforest air."

Additional information about the applicability of integral AMS data for the prediction of kappa_p and N_CCN,S will be added in the revised manuscript (p.3837, I.21): "The mean relative deviations and bias between kappa_ and kappa_a are summarized in Tab. 6. The strong positive bias of kappa_p at high S (up to +77%) is due to the enhanced organic mass fraction in small particles and could be corrected on the basis of the average AMS size distribution data (Figs. 3 and 11b), but further processing of the AMS data would go beyond the scope of this study. In spite of the positive bias and low correlation of predicted vs. measured kappa (Fig. 14, Tab. 6), the mean relative deviations between CCN number concentrations predicted on the basis of kappa_p and the measurement values of NCCN,S were mostly less than 20% at S = 0.19-0.82% (Tab.4), reconfirming the relatively low sensitivity of NCCN,S against kappa as discussed above (Sect. 3.2)."

Comment 5:

Nonetheless, it is worth noting that these kappa values at large sizes (low supersat-

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urations) 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).

Response:

We agree and intend to include the following statement and reference in the revised manuscript (end of Sect. 3.1.2): "During the interactive public discussion of this manuscript, Roberts (2009) pointed out that the kappa values observed at S = 0.1% (D \sim 200 nm) during the pristine focus period were by a factor of \sim 4 lower than the kappa values observed at remote maritime locations for accumulation mode particles of comparable size (kappa \sim 0.6, equivalent to pure ammonium sulfate; Roberts et al., 2008). In view of the enhanced sensitivity of cloud droplet formation to aerosol hygroscopicity at low supersaturation (updraft limited regime of CCN activation; Reutter et al., 2009), this may be a potentially important difference between "blue ocean" and "green ocean" regimes (Andreae et al., 2004; Roberts, 2009)."

Comment 6:

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 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"?

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Response:

Electron microscopic investigations of filter samples collected during AMAZE-08 (Sinha et al., manuscript in preparation) suggest that the contribution of fungal spores and similar primary biological particles in the relevant size range (100-300 nm) were negligibly small (<<5%), but other types of solid carbonaceous particles and mineral dust particles were more frequently detected (~10-20% of particle number in the range of 100-300 nm) and may have been CCN-inactive at S = 0.1%. As mentioned in our manuscript and above, these and other aspects of aerosol composition and mixing state will be further investigated in collaboration with the other partners of AMAZE-08, but they go beyond the scope of this paper. As far as we can tell for now, fungal spores and other primary biological particles are most likely not relevant for the composition and CCN activity of aerosol particles < 300 nm. With regard to the correlation between the focus period and the rest of the campaign. Note that most of the data points with low kappa and high OMF were from the focus period and most of the data points with higher kappa and lower OMF were from the rest of the campaign.

Comment 7:

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(OMF)*0.1 + X(SULF)*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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The kappa values derived from CCN measurement data through Köhler model calculations assuming the surface tension of pure water have to be regarded as "effective hygroscopicity parameters" that account not only for the reduction of water activity of the solute ("effective Raoult parameters") but also for surface tension effects (Petters and Kreidenweis, 2007; Rose et al., 2008b; Mikhailov et al., 2009; Pöschl et al., 2009a). We intend to clarify this aspect and include the above statement in the revised version of our manuscript (Sect. 2.2).

The observed linear correlation between kappa and organic mass fraction suggests that a value of kappa_org = 0.1 effectively accounts for the influence of organics on the CCN activity of the investigated aerosol particles, including both the reduction of water activity and/or the suppression of surface tension regardless of their relative importance. Only for the estimation of an effective molar mass of the organics did we regard kappa_org as an "effective Raoult parameter" (p. 3836, I. 4): "Under the assumption that the surface tension of the aqueous droplet formed upon CCN activation equals the surface tension of pure water, the effective hygroscopicity parameter kappa_org can be interpreted as an "effective Raoult parameter", i.e. as an effective molar density of soluble molecules or ions in the dry organic material normalized by the molar density of water molecules in liquid water (rho_w/M_w \sim 55 mol L - 1, Pöschl et al., 2009a):"

In analogy to Roberts et al. (2002), we tend to think that surface tension effects are likely to be of limited importance for Amazonian aerosols, but we have no information about the actual surfactant activity of the organic fraction of the investigated aerosol particles. According to the relative sensitivities of surface tension and kappa when interpreted as an effective Raoult parameter (Kreidenweis et al., 2009; Mikhailov et al., 2009; Pöschl et al., 2009a), a decrease of surface tension by ~10% would be compensated by a decrease of the effective Raoult parameter by ~30%, and the latter would correspond to a ~30% increase of the effective molar mass or a ~30% decrease of the density of the organic material. In any case, the effective hygroscopicity parameter kappa_org ~ 0.10 as defined above (Sect. 2.2 and references therein) accounts for

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both the Raoult effect related to molar mass and density of the solute and for the Kelvin effect related to surfactant activity of the solute, regardless of their relative importance. The applicability of this approach is confirmed by the close linear correlation between kappa and Xm,org.

We intend to clarify this aspect and include the above statement in the revised version of our manuscript (Sect. 3.3).

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