We thank the reviewer for the constructive and thoughtful comments. Please find below detailed responses to each comment or question, including notations of improvements to the manuscript. Reviewer comments are in blue fonts. Changes to the text are highlighted in yellow.

General comments:

Study by Thalman et al. presents results of κ_{CCN} and mixing state derived from size resolved CCN observations collected during the GoAmazon2014/5 campaign. From κ_{CCN} and additional aerosol chemical composition measurements, the authors then estimate the hygroscopicity of the organic fraction κ_{org}. The results presented in the study, together with those presented by Pöhlker et al (2016) in another experimental site, represent the first long-term measurement of hygroscopic behaviour of aerosols in the Amazon. Overall, it is a well written manuscript on an important subject. Therefore, I recommend it for publication in ACP after minor corrections.

Specific comments:

Since chemical composition of aerosols in the study was largely dominated by the organic fraction, a discussion on the uncertainties related to limited solubility or surface active species could be relevant, even if no quantitative assessment of these uncertainties is provided.

Yes, limited solubility and reduced surface tension could impact the CCN activity and hygroscopicity of organics. As the κ value was derived from dry particle diameter and critical supersaturation assuming the surface tension of pure water, κ value reported in this manuscript represents the "apparent hygroscopicity", which includes the potential impact due to limited solubility and reduction of surface tension (Sullivan et al., 2009). We have clarified this by including the following sentence in the text:

As hygroscopicities reported in this study were derived from particle dry diameter and critical supersaturation, they represent "apparent hygroscopicity", which includes the potential impact due to the limited solubility of organics and the reduction of surface tension by surface active species (Sullivan et al., 2009).

P 21 L 431-440 Given that the importance of hygroscopicity to the aerosol CCN behavior is enhanced for κ values below 0.2, the choice of an adequate κ value to represent the Amazonian aerosol CCN behavior is important, for instance, to modeling studies. In this sense, it could be interesting to compare the presented results with κ_{HTDMA} results from campaigns previous to 2008 as well, since κ have already been calculated from the original data of some of these campaigns in other works (see, for example, Gunthe et al. (2009) and the Supplement of Sánchez Gácita et al. (2017)). In the authors opinion, differences between presented results and κ_{HTDMA} from earlier campaigns are solely due to the use of H-TDMA or CCN technique, or other factors could be important as well?

We thank the reviewer for this suggestion. We added particle hygroscopicity (κ_{HTDMA}) derived from HTDMA measurements during campaigns before 2008 in Figure 2 for comparison, including κ_{HTDMA} measured in July 2001, during a "recent biomass burning period" of the CLAIRE-2001 study (Rissler et al., 2004), and that measured from 11 September to 8 October 2002, during the dry period of the LBA-SMOCC (Rissler et al., 2006). During the above two periods, aerosols were strongly influenced by fresh local biomass burning emissions. The measurements during CLAIRE-2001 took place at Balbina, about 125 km northeast of Manaus, and the κ_{HTDMA} during the "recent biomass burning period" agrees well with κ_{CCN} derived at the T3 site for air masses with strong local biomass burning influences. In contrast, κ_{HTDMA} observed during the dry period of LBA-SMOCC is substantially lower than κ_{CCN} at T3 for local biomass burning air mass at all sizes. As LBA-SMOCC took place in the state of Rondônia in southwestern Amazonia with extensive biomass burning activities during the dry season, the difference in κ may be due to the differences in fire condition and the type of vegetation burned. Previous studies show particles sometime exhibit larger κ values for droplet activation (derived from CCN measurements under supersaturated conditions) than for particle growth (derived from particle GF under sub-saturated conditions), this could also contribute to the difference in κ values. We have updated Figure 2, and included following discussion in the text:

For the air masses with strong influence from local biomass burning, the value of κ_{CCN} and its size dependence are consistent with the κ value derived from particle growth factor measurements in July 2001, during a "recent biomass burning period" of the CLAIRE-2001 study (Rissler et al., 2004), which took place at Balbina, about 125 km northeast of Manaus. In contrast, κ values derived from particle growth factor measurements from 11 September to 8 October 2002, during the dry period of the LBA-SMOCC (Rissler et al., 2006) are substantially lower than κ_{CCN} observed at the T3 site for local biomass burning air masses at all sizes. As LBA-SMOCC took place in the state of Rondônia in southwestern Amazonia with extensive biomass burning activities during the dry season, the difference in κ could be due to the differences in fire condition and the type of vegetation burned. Previous studies show particles sometime exhibit larger κ values for droplet activation (derived from CCN measurements under supersaturated conditions) than for particle growth (derived from particle growth factor under sub-saturated conditions), this could also contribute to the differences in κ values.

Supp. P10 L172 Please provide a reference for equation S1 of supplement

The reference to the supplementary material in Mei et al. (2013) has been added, the formula as given in equation S1 converts the formula in the previous paper to the hygroscopicity dispersion instead of the critical supersaturation dispersion and hence is somewhat different.

Supp. P10 L174-175 The use of equation S2 of supplement was suggested by Petters and Kreidenweis (2007) for κ > 0:2. Considering that for most conditions in this study κ_{CCN} was below this threshold, perhaps an iterative approach on the original κ-Köhler equation (see, for example, Carrico 2008) would have been more adequate. A sensitivity test on a small fraction of the data would be desirable to show that uncertainties derived from the use of this simplified approach are indeed low or negligible.

We thank the reviewer for this suggestion. We carried out the sensitivity study using an iterative approach based on the original κ-Köhler equation as suggested by the reviewer. The relative and absolute uncertainties in κ_{CCN} derived using equation S2 are shown in Fig 1A and 1B below for each particle diameter. The lower κ_{CCN} limit of each curve represents 1st percentile of κ_{CCN} values derived at the corresponding particle diameter. The results show that the relatively uncertainty is less than 7% for 99% of the data at all particle diameters. Furthermore, the main conclusions of the manuscript are based on κ_{CCN} derived at 112, 142, and 171 nm (e.g., Figures 3-12), and the relative uncertainty is less than 4.5% for 99% of data at the three sizes. Therefore, the uncertainties derived from the use of the simplified approach are low and negligible.

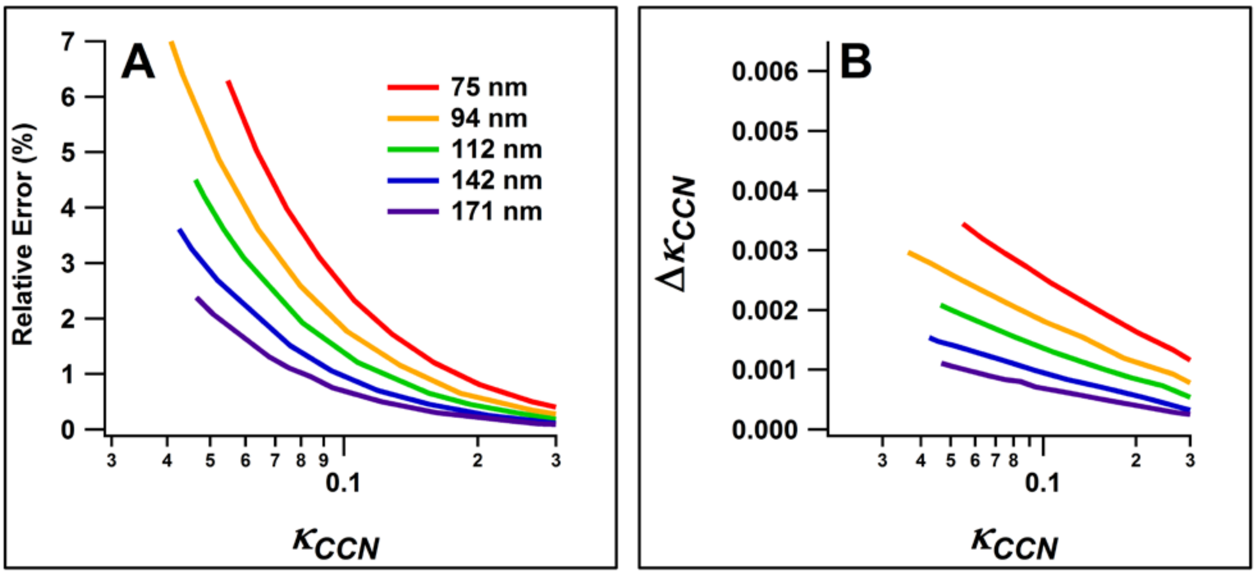

Figure 1: (a) Relative and (b) absolute uncertainties of κ_{CCN} derived using equation S2 at each of the particle diameters. The lower κ_{CCN} limit of the curves represents the 1st percentile of the κ_{CCN} value at corresponding particle diameter.

References

Mei, F., Setyan, A., Zhang, Q., and Wang, J.: CCN activity of organic aerosols observed downwind of urban emissions during CARES, Atmospheric Chemistry and Physics, 13, 12155-12169, 2013.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961-1971, 2007.

Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L. V., and Artaxo, P.: Physical properties of the sub-micrometer aerosol over the Amazon rain forest during the wet-to-dry season transition - comparison of modeled and measured CCN concentrations, Atmospheric Chemistry and Physics, 4, 2119-2143, 2004.

Rissler, J., Vestin, A., Swietlicki, E., Fisch, G., Zhou, J., Artaxo, P., and Andreae, M. O.: Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia, Atmos. Chem. Phys., 6, 471-491, 2006.

Sullivan, R. C., Moore, M. J. K., Petters, M. D., Kreidenweis, S. M., Roberts, G. C., and Prather, K. A.: Effect of chemical mixing state on the hygroscopicity and cloud nucleation properties of calcium mineral dust particles, Atmos. Chem. Phys., 9, 3303-3316, 2009.