Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Long-term observations of atmospheric aerosol, cloud condensation nuclei concentration and hygroscopicity in the Amazon rain forest – Part 1: Size-resolved characterization and new model parameterizations for CCN prediction" *by* Mira L. Pöhlker et al.

Mira L. Pöhlker et al.

m.pohlker@mpic.de

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We thank Referee #1 for the pertinent comments and suggestions that have helped us to improve the quality of our manuscript. The referees' comments and our responses are outlined in detail below:

[1.1] Referee comment: In section 1.2 the authors discuss the hydrological regime of the Amazon forest. Onset and end of the rainy season in the central Amazon Basin,

where the authors conducted their measurements, show the largest variations compared to other parts of the basin. Satellite-based outgoing longwave radiation (OLR) measurements and the low-level wind field show that in the central Amazon, onset is associated with anomalous anticyclones and enhanced trade winds in the Atlantic. In addition, there is an apparent association between sea surface temperature anomalies in the tropical Atlantic and Pacific and the onset and end of the rainy season in the central Amazon, in that a warm Pacific and cold Atlantic result in a delayed onset and early withdrawal. It is not clear in the text to what extent the clean (wet) and polluted (dry) seasons of the studied period (March 2014 to February 2015) are being impacted by large-scale atmospheric circulations, especially the temperatures of the Pacific and Atlantic and El Nino-Southern Oscillation.

Author Response: We agree that discussing (potential) anomalies in the hydrological regime for the studied period in 2014/15 due to teleconnections with the ocean surface temperatures would strengthen the paper. Therefore, we modified and adjusted the following parts in the paper:

First, we changed the footnote number 1 on page 3 from:

"Note that this definition of the seasons in the central Amazon is oriented on the seasonality in aerosol sources and prevalence rather than the meteorological conditions. For example, the 'meteorological wet season' typically has its core period in February (maximum in precipitation), whereas the 'pollution-defined wet season' has its core period in April/May (e.g., minimum in CO and BC concentrations) (Andreae et al, 2015)."

to:

"The Amazonian seasons are mostly defined meteorologically with respect to precipitation data (Fu et al., 2001; Fernandes et al., 2015). Note that we use in this study a slightly different definition of the seasons in the central Amazon based on meteorological and aerosol data to emphasize the seasonality in aerosol sources and prevalence. For example, the 'meteorological wet season' typically has its core period in February

(maximum in precipitation), whereas the 'pollution-defined wet season' has its core period in April/May (e.g., minimum in CO and BC concentrations) (Andreae et al, 2015)."

Second, we added the following text section on page 10 in line 5:

"Figure 1a presents precipitation data from satellite and in situ measurements at the ATTO site to illustrate the meteorological seasonality for the measurement period. The precipitation rates in the Amazon Basin can show pronounced anomalies due to teleconnections with the Atlantic and/or Pacific sea surface temperatures (SST) (Fu et al., 2001; Fernandes et al., 2015). The most prominent example here is the El Niño-Southern Oscillation (ENSO) and its various impacts on the Amazonian ecosystem (e.g., Asner et al., 2000; Ronchail et al., 2002). For the measurement period, the Oceanic Niño Index (ONI) ranged between -0.4 and 0.6 °C, confirming that only towards the end of the measurement period a slightly positive anomaly was observed. In Fig. 1a, satellite data from the tropical rainfall measurement mission (TRMM) are presented for the area around the ATTO site. The TRMM data is provided for an extended time period (Jan 1998 until June 2016) and, in comparison, for the CCN measurement period (Mar 2014 until Feb 2015). This comparison shows that the 2014/15 precipitation rates do not deviate substantially from the 18-year average and, thus, further confirms that the measurement period can be regarded as a 'typical' year with 'typical' seasons and no pronounced hydrological anomalies."

Third, precipitation data has been added to Fig. 1 to illustrate the absence of potential hydrological anomalies in the measurement period.

[1.2] Referee comment: In section 2.1 of the methodology, the authors describe the characteristics of the measurement site and period. Although they justified in section 1.2 their preference for defining seasonality in terms of aerosol sources rather than meteorological variables, I suggest to include the monthly rainfall of the period of study overlayed the climatology of the central Amazon. Rainfall is a good indicator of how anomalous is the period of measurements. The monthly precipitation shown in the pa-

per of Andreae et al. (2015), reported as a reference for an overview of the atmospheric conditions at the site, is incomplete for the year 2014.

Author Response: We agree that precipitation data will help to clarify the meteorological seasonality. We added to Fig. 1 precipitation data from the TRMM satellite mission and from in situ measurements at the ATTO site – see our response to comment [1.1]. Moreover, we would like to point out that the seasonality in precipitation will be discussed in more detail in part 2 of this study and, therefore, complete the picture.

[1.3] Referee comment: Page 10 (lines 14-19): Using hygroscopicity parameter as reference, the au-thors state that particles' chemical composition is stable throughout the year and the maximum in CCN concentration during the dry season is mainly related to the overall increase in aerosol concentration. In addition they considered their results as consistent with the previous result of Andreae et al. (2004) showing CCN efficiency (expressed as the ratio of CCN to CN) for the Amazonian wet and dry season aerosol. I do not particularly see condition to compare the two different studies. First, the finds of this study is unique in the sense it is the first time that we see a full year of CCN measurements in the Amazon. Therefore, there is no parallelism with the field campaigns CLAIRE-98 and SMOCC-2002, which are short campaigns. Second, they are measurement sites completely different. The only comparable CCN efficiencies are observed between SMOCC-2002 (cloud-processed smoke at altitude 2000 – 4500 m, cloud-processing might change the chemical composition and increase the hygroscopicity) and CLAIRE-98 (background ground-based measurements, naturally hygroscopic).

Author Response: Agreed. We removed the following sentence from the text:

"Furthermore, this observation is consistent with the previously reported similarity between the CCN efficiency of Amazonian wet and dry season aerosol (Andreae et al., 2004)."

[1.4] Referee comment: Page 10, lines 30-34: Even with a sparse occurrence of parti-

cles in the nuclea-tion mode, did the authors find any seasonality in the number of nucleation episodes, such as the three representative days shown by Ortega et al. (2014) in Figure 9? How do the climate and forest of the central Amazon affect the absence of new particle formation, mainly when compared to other continental background locations such as the Manitou Experimental Forest Observatory (MEFO) described by Ortega et al. (2014)?

Author Response: The referee points at an interesting aspect, which is the sparse occurrence of nucleation mode particles in the Amazon. The question whether there is any seasonality in the frequency of the nucleation mode events is not trivial to answer. It requires a reliable discrimination of event versus non-event cases and, furthermore, a systematic statistical approach to extract seasonal trends. A detailed analysis on the abundance, properties, and seasonality of the rare nucleation mode events at the ATTO site is subject of a study that is currently prepared for publication. We added the following statement into the text (page 11, line 6):

"A systematic study on the abundance, properties, and seasonality of the sparse nucleation mode bursts in the central Amazon is subject of an upcoming study."

[1.5] Referee comment: Page 11, lines 32-37 (discussion involving size dependence of hygroscopicity parameter): Why the values of hygroscopicity parameter, when averaged over the entire campaign (0.13±0.03 - Table 2 & Fig. 3) are practically constant in the Aitken mode? This is not observed in the accumulation mode. Could the differences be explained by the chemical composition or the cloud pro-cessing of the particles in the accumulation mode?

Author Response: The referee points at an interesting aspect. The origin and nature of Aitken mode particles in the Amazon Basin still raises a number of open questions. In a recent study, Wang et al. (2016) propose that Aitken mode particles originate from nucleation in the free troposphere and are frequently injected into the boundary layer by down-drafted air masses in connection with strong rain. Pöschl et al. (2010)

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showed in the context of the AMAZE-08 campaign that the Aitken mode size range almost exclusively consists of organic constituents, whereas the accumulation mode contains a certain amount of sulfates (and probably also other inorganic ingredients) beside its dominant organic fraction. However, there is so far only sparse information on the chemical composition of Aitken mode particles available. In this sense the data in our study is unique since it confirms for a long time period that the Aitken mode aerosol population consists of almost entirely organic constituents (indirectly via the hygroscopicity properties). Moreover, we find that the accumulation mode showing elevated hygroscopicity is in agreement with observations by Pöschl et al. (2010). If and how cloud processing influences the distinct differences in chemical composition of Aitken and accumulation modes is beyond the scope of this paper and will be subject of future studies. Some further information on the abundance and hygroscopicity of Aitken and accumulation mode particles for specific events will be addressed in the part 2 paper of this study (M. L. Pöhlker et al., 2016).

[1.6] Referee comment: Page 13, lines 29-30: There are studies suggesting that aerosols from biomass burning is an ingredient to invigorate convective clouds. This is based on the fact that aerosols have a major impact on the microphysics of continental mixed-phase convective clouds. In addition to the solar heating suggested by the authors, could the aerosol effect also be a plausible explanation for the small Hoppel minimum and high cloud peak supersaturation in the dry season?

Author Response: We agree and added the corresponding statement on page 13 line 30 from:

"A plausible explanation for the comparably small DH and high Scloud(DH,κ) in the dry season could be the invigorated updraft regimes due to stronger solar heating."

to:

"A plausible explanation for the comparatively small DH and high Scloud(DH,κ) in the dry season could be invigorated updraft regimes in the convective clouds. This invig-

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oration could be caused by the stronger solar heating during the dry season and/or the increased aerosol load under bio-mass burning impacted conditions, as suggested previously (Andreae et al., 2004; Rosenfeld et al., 2008)."

[1.7] Referee comment: Page 14, lines 28-32: The inclusion of the diurnal cycle of NCN, similar to Fig. 7, could enrich the discussion on the non detectable diurnal trend in the hygroscopicity parameter.

Author Response: We agree. The diurnal cycle of the total aerosol number concentration has been included in Fig. 7.

Furthermore, we added the following statement on page 14 in line 32:

"For comparison, the diurnal cycles in NCN concentration have been added to Fig. 7, which con-firm the absence of strong diurnal variations in the aerosol population."

[1.8] Referee comment: Page 34, Table 1: Hygroscopicity parameter is calculated as 0.13 ± 0.03 for both supersaturation 0.47% and 1.10%. However, looking at Fig. 01c, the parameter seem to be more dispersed at 1.10% than at 0.47% throughout the year. Is it correct a same std of 0.03 for both hygroscopic parameters?

Author Response: The same standard deviation of 0.03 for both cases is correct. Please note that the error bars in Fig. 1c represent the experimental error in $\kappa(S,Da)$, derived from the experimental error in S. The $\kappa(S,Da)$ values for every S reported in Table 1 are given as mean $\pm$ one standard deviation.

[1.9] Referee comment: Technical corrections: Page 10 (lines 8, 9 and 10) and wherever in the text: Please standardize the symbol for critical diameter Da(S) Page 17 and 19: It may be better to define a symbol for "width" in equations 7 and 8, considering its potential use in future articles.

Author Response: Done. We standardized all symbols of Da(S) (replacing the Da). Moreover, we followed the referees suggestion and replace "width0" in the context of CCN spectra and "width1" in the context of CCN efficiency spectra by "w0" and "w1"

throughout the text.

[1.10] Referee comment: Page 39: Fig. 1 is very useful to inform about seasonal trends in time series. However a plot of the diurnal cycle is missing for better understanding.

Author Response: In the new version of Fig. 8 the diurnal cycles in κmean and NCN, resolved by seasons, are shown. More specific diurnal cycles of aerosol concentration in defined size ranges will be subject of upcoming studies.

[1.11] Referee comment: Page 41: Change "Aiken" to Aitken in the legend of Fig. 3.

Author Response: Done. This has been changed:

[1.12] Referee comment: Page 43: CCN activation curve at supersaturation of 0.47% shows strange values in the plots of the Fig. 5, including values of NCCN/NCN above 1.0.

Author Response: Agreed. To clarify this aspect, we added the following statement to Sect. 2.3:

"Throughout this study, we observed a slight systematic deviation of the results for the supersaturation S = 0.47 %. This effect can be seen for example in MAF(0.47%) values exceeding unity in Fig. 1 and NCCN(0.47%,D)/NCN(D) values exceeding unity in Fig. 5. The effect persists even after applying all aforementioned corrections to the data and is most pronounced during the dry season. Yet, we did not find any evidence of this data being erroneous, we decided to keep it in the study."

References:

Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M., and Silva-Dias, M. A. F.: Smoking rain clouds over the Amazon, Science, 303, 1337-1342, 10.1126/science.1092779, 2004.

Asner, G. P., Townsend, A. R., and Braswell, B. H.: Satellite observation of El Nino effects on Amazon forest phenology and productivity, Geophys. Res. Lett., 27, 981-

984, 10.1029/1999gl011113, 2000.

Fernandes, K., Giannini, A., Verchot, L., Baethgen, W., and Pinedo-Vasquez, M.: Decadal covariability of Atlantic SSTs and western Amazon dry-season hydroclimate in observations and CMIP5 simulations, Geophys. Res. Lett., 42, 6793-6801, 10.1002/2015gl063911, 2015.

Fu, R., Dickinson, R. E., Chen, M. X., and Wang, H.: How do tropical sea surface temperatures influence the seasonal distribution of precipitation in the equatorial Amazon?, Journal of Climate, 14, 4003-4026, 10.1175/1520-0442(2001)014<4003:hdtsst>2.0.co;2, 2001.

Pöhlker, M. L., Pöhlker, C., Klimach, T., Hrabe de Angelis, I., Barbosa, H. M. J., Brito, J., Carbone, S., Chi, X., Cheng, Y., Ditas, F., Ditz, R., Gunthe, S. S., Kesselmeier, J., Könemann, T., Lavric, J. V., Martin, S. T., Moran, D., Rose, D., Saturno, J., Su, H., Thalman, R., Walter, D., Wang, J., Wolff, S., Artaxo, P., Andreae, M. O., and Pöschl, U.: Long-term observations of cloud condensation nuclei in the Amazon rain forest - Part 2: Ultrafine particle bursts, biomass burning and long range transport events in prep., 2016.

Pöschl, U., Martin, S. T., Sinha, B., Chen, Q., Gunthe, S. S., Huffman, J. A., Borrmann, S., Farmer, D. K., Garland, R. M., Helas, G., Jimenez, J. L., King, S. M., Manzi, A., Mikhailov, E., Pauliquevis, T., Petters, M. D., Prenni, A. J., Roldin, P., Rose, D., Schneider, J., Su, H., Zorn, S. R., Artaxo, P., and Andreae, M. O.: Rainforest Aerosols as Biogenic Nuclei of Clouds and Precipitation in the Amazon, Science, 329, 1513-1516, 10.1126/science.1191056, 2010.

Ronchail, J., Cochonneau, G., Molinier, M., Guyot, J. L., Chaves, A. G. D., Guimaraes, V., and de Oliveira, E.: Interannual rainfall variability in the Amazon basin and sea-surface temperatures in the equatorial Pacific and the tropical Atlantic Oceans, International Journal of Climatology, 22, 1663-1686, 10.1002/joc.815, 2002.

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Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A., and Andreae, M. O.: Flood or drought: How do aerosols affect precipitation?, Science, 321, 1309-1313, 10.1126/science.1160606, 2008.

Wang, J., Krejci, R., Giangrande, S., Kuang, C., Barbosa, H. M. J., Brito, J., Carbone, S., Chi, X., Comstock, J., Ditas, F., Lavric, J., Manninen, H. E., Mei, F., Moran-Zuloaga, D., Pöhlker, C., Pöhlker, M. L., Saturno, J., Schmid, B., Souza, R. A. F., Springston, S. R., Tomlinson, J. M., Toto, T., Walter, D., Wimmer, D., Smith, J. N., Kulmala, M., Machado, L. A. T., Artaxo, P., Andreae, M. O., Petäjä, T., and Martin, S. T.: Amazon boundary layer aerosol concentration sustained by vertical transport during rainfall, Nature, in press, 2016.
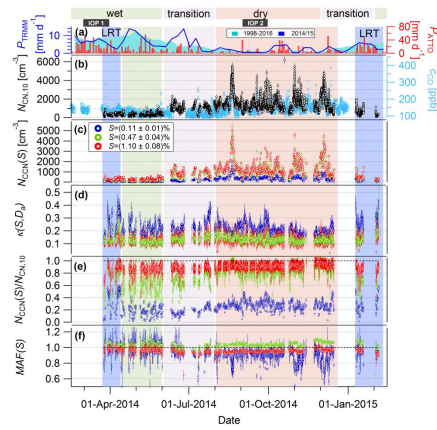
Figure 1. Seasonal trends in time series of precipitation rate P, total aerosol concentration $N_{CN,10}$, carbon monoxide mole fraction (c_{CO}), and CCN key parameters for three selected supersaturations S for entire measurement period (shown in original time resolution). (a) Precipitation rates from tropical rainfall measuring mission (TRMM) P_{TRMM} and *in situ* measurements at the ATTO site P_{ATTO}. The P_{TRMM} seasonal cycles are derived from an area upwind of the ATTO site (W 59.5°, N 2.4°, W 54.0°, S 3.5°), covering a long-term period from 1 Jan 1998 until 30 June 2016, on one hand, and the period of the CCN measurements from 1 Mar 2014 until 28 Feb 2015, on the other hand. (b) Time series of pollution tracers $N_{CN,10}$ and c_{CO}. (c) CCN concentrations $N_{CCN}(S)$, (d) hygroscopicity parameter $\kappa(S,D_a)$, (e) CCN efficiencies $N_{CCN}(S)/N_{CN,10}$, and (f) maximum activated fraction $MAF(S)$. Three different types of shading represent: (i) the seasonality in the Amazon atmosphere according to Andreae et al. (2015) (wet *versus* dry seasons with transition periods, illustrated in top of graph), (ii) periods of IOP1 and IOP2 during GoAmazon2014/5, (iii) seasonal periods of interest in context of the present study as defined in Sect. 3.3 (shading in background of time series).
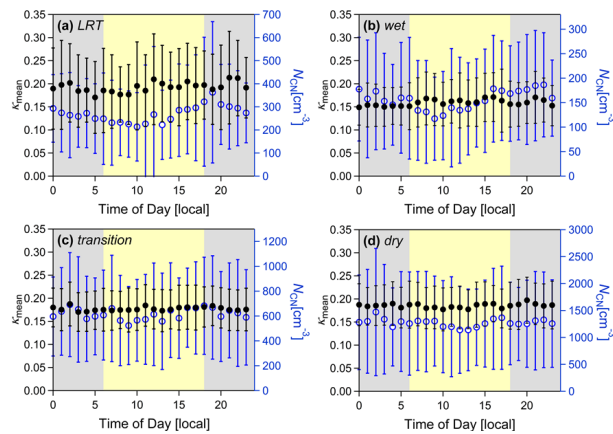
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Figure 7. Diurnal cycles in hygroscopicity parameter κmean and total aerosol number concentration NCN subdivided into seasonal periods of interest as specified in Sect. 3.3. No diurnal trend is detectable throughout the year. Note that range of one standard deviation of κmean around mean is surprisingly small given that long seasonal time periods and data from all S levels have been averaged. Only perceptible difference is larger scattering during period with LRT influence (a). Grey and yellow shading indicates night and day.

Fig. 2.