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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.

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We thank referee #2 for her/his positive review and recommendation that the study should be published in ACP after "some small modifications". The comments by referee #2 have been very constructive to improve several aspects of our study. The comments and our answers are listed below.

[2.1] Referee comment: I suggest that the period be referred as "full seasonal cycle"

instead of "almost one year".

Author Response: Agreed. We changed this on page 9 in line 35.

[2.2] Referee comment: On page 11. section 3.2, line 21, we can read: "A close look reveals a gap be-tween the activation curves for S = 0.47 % and S = 0.29 %, which corresponds to a jump in kappa(S,Da) (discussed below)." I could not see this gap! If we look closer the picture, we also can see that the inter-vals level used in supersaturation inside the CCNC jumps from ∼0.05% to ∼0.15%, which can explain the gap on the featured curves. So, what authors claim to correspond to a jump in hygroscopicities is, in fact, a result from the measurement. Is that right?

Author Response: We agree – our statement that the referee cited is indeed nonsense. We changed the corresponding section from:

"A close look reveals a gap between the activation curves for S = 0.47 % and S = 0.29 %, which corre-sponds to a jump in κ(S,Da) (discussed below). Moreover, the gap relates – in a way – to the bimodal size distribution and the characteristic Hoppel minimum (at 97 nm for the annual mean size distribution, see Table 2) between Aitken and accumulation mode, as S = 0.47 % represents the onset of significant activation in the Aitken mode size range."

to:

"The step from the activation curves at S = 0.47 % to S = 0.29 % relates to the position of the characteristic Hoppel minimum (at 97 nm for the annual mean size distribution, see Table 2) between Aitken and accumulation mode in the bimodal size distribution. Thus, the step to S = 0.47 % represents the onset of significant activation in the Aitken mode size range."

[2.3] Referee comment: Page 14: "Comparing the seasonal kappa(S,Da) size distributions in Fig. 6, it is obvious that the (seasonally averaged) kappaAit values in the Aitken mode size range are surprisingly stable between 0.13 and 0.14 throughout the whole

year." This was already said at beginning of page 12 and also at page 10 (line 15). It was said three times in the text (and presented on table 1 too) that there is not an appreciable variation of hygroscopicity. Please, verify it. So, is Figure 7 really needed?

Author Response: The referee asks if "Figure 7 is really needed". Based on the context of the comment we assume that "Figure 7" is a typo and that the referee was referring to Fig. 6. Figure 6 is one of the key figures in this study and we think that it should not be omitted. We are convinced that it is justified to mention the small variation of κAit multiple times since this one of the key observations that supports our argumentation.

[2.4] Referee comment: The parameterization of CCN spectra with constants (Twomey parameteriza-tion) has been used in many studies, most of them for short term observations. Though simple to carry out, it does not take into account any variation in the CCN loading, as was said in the text. It seems obvious to me, that the use of annual average for the constant used on the CCN spectra would result in overestimation of CCN concentration during the wet season, and in underestimation during the dry season. I would be more interesting if you could provide the constants for each season, instead of that for the whole year. Then the current section 3.5.3, as it is now, more weakens rather than strengthens the present work. Consider removing Figure 11.

Author Response: The referee brings up a valid point. To implement his comment we made several modifications in the manuscript.

First, we conducted a seasonally resolved CCN prediction based on the Twomey and erf fit functions. The corresponding results have been added to Fig. 11.

Second, we added two further tables (as Table 6 and 7) into the manuscript, which summarize the Twomey and erf fit parameters for the annually average and seasonally resolved cases.

Third, we added the results from the seasonally resolved Twomey and erf fits to the overview Table 3.

Fourth, we modified the corresponding text section in Sect. 3.5.3 from:

"Figure 11a and b show the corresponding NCCN,p(S) versus NCCN(S) scatter plots. In general, parametrizations based on CCN spectra yield a mean state based on average concentrations (see fit parameters in Fig. 10) and ignore the temporal variability of the aerosol concentrations (Martins et al., 2009; Rose et al., 2010; Jurányi et al., 2011). On closer inspection, Table 3 shows that the erf fit allows somewhat better predictions (deviation of power law fit about 227 % versus 215 % for erf fit), which can be explained by the fact that the erf fit presents the experimental data more appropriately (compare Fig. 10). Overall, however, the power law fit and the erf fit approaches give rather poor correlations, due to the missing representation of the aerosol's temporal variability, which is an inherent limitation of the CCN spectra parametrization. It can be concluded that this parametrization requires a minimum of aerosol input data (i.e., only the parameters of the corresponding fit function), which explains its wide distribution in various modelling attempts. However, Fig. 10 and Table 3 show that this simplicity is clearly at the expense of the prediction accuracy."

to:

"Figure 11a and b show the corresponding NCCN,p(S) versus NCCN(S) scatter plots based on the annual mean CCN spectrum, using the Twomey and erf fits. In general, parametrizations based on CCN spectra yield a mean state based on average concentrations (see fit parameters in Fig. 10 as well as Table 5 and 6) and ignore the temporal variability of the aerosol's abundance (Martins et al., 2009; Rose et al., 2010; Jurányi et al., 2011). Table 3 shows that the erf fit allows somewhat better predictions (e.g., deviation of power law fit about 227 % versus 215 % for erf fit in case of annual mean and 80 % versus 75 % for the seasonally resolved case), which can be explained by the fact that the erf fit presents the experimental data more appropriately (compare with Fig. 10). Overall, however, the power law and erf fit approaches give rather poor correlations due to the missing representation of the aerosol's temporal variability. This is particularly obvious for the annual mean case since the total aerosol

abundance varies significantly between wet and dry season conditions. Accordingly, the CCN spectra parametrization, which operates with constants, predictably underestimates the dry season conditions and overestimates the wet season conditions. In addition to the analytical fit approaches for the annual mean spectrum (Fig. 11a and b) we conducted an analogous CCN prediction based on seasonally resolved CCN spectra (Fig. 11c and d). The prediction accuracy clearly improves (e.g., deviation of erf fit for annual mean case equals 215 % versus 75 % for seasonally resolved case; see Table 3). Figure 11 illustrates that the prediction accuracy of parametrizations, which rely on analytical fit functions of CCN spectra (i.e., Twomey, erf, and related functions), improves with decreasing variability of the aerosol population (e.g., for shorter periods with less variable aerosol properties). However, the missing representation of the aerosol's temporal variability remains to be an inherent limitation of the CCN spectra parametrization. It can be concluded that this parametrization requires a minimum of aerosol input data (i.e., only the parameters of the corresponding fit function), which explains its wide distribution in various modelling attempts. However, Fig. 11 and Table 3 show that this simplicity is clearly at the expense of the prediction accuracy."

[2.5] Referee comment: Technical corrections: The text begins expressing supersaturation by "super-saturation S". Then it changes to "S", then to "S levels". Is it correct? Please check it.

Author Response: This is correct. On page 7 in line 11, we introduced the symbol S for supersaturation. Throughout the text we then only refer to "S" or to "S levels", which is synonymously used for "supersaturations".

References:

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Martins, J. A., Dias, M., and Goncalves, F. L. T.: Impact of biomass burning aerosols on precipitation in the Amazon: A modeling case study, Journal of Geophysical Research-Atmospheres, 114, 19, 10.1029/2007jd009587, 2009.

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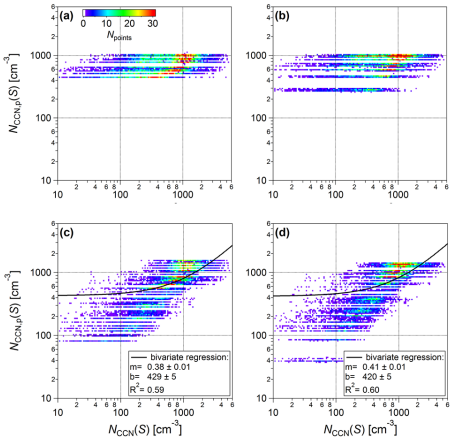

Figure 11. Predicted versus measured CCN number concentrations based on the classical Twomey power law fit (a and c) and an alternative error function fit (b and d). The top row (a and b) represents the annually averaged cases, whereas the bottom row (c and d) represents parametrizations based on seasonally resolved CCN spectra. Both predictions are based exclusively on the corresponding average fit functions (i.e., the annually averaged CCN spectra in Fig. 10 and seasonally averaged CCN spectra, as specified in Table 6 and 7) without considering time resolved aerosol parameters. The color code shows the number of data point falling into the pixel area, following Jurányi et al. (2011). Predicted and measured CCN concentrations deviate significantly, showing the inherent limitations of the CCN spectra approach. For the annually averaged data (a and b) no meaningful bivariate regression fit could be obtained.

Fig. 1.

Table 6. Twomey fit parameters describing CCN spectra $N_{CCN}(S)$ *versus* S as parametrization input data (compare Fig. 10 and 11a,c). Fit parameters are provided for annually averaged CCN spectra and resolved by seasons.

time period	N_{CCN} (1%) [cm^{-3}]	K	R^2
all	998±60	0.36±0.04	0.88
wet season	289±7	0.57±0.03	0.98
LRT period	378±9	0.38±0.03	0.94
transition	970±40	0.49 ±0.05	0.94
dry season	1469±78	0.36 ±0.06	0.86

Fig. 2.

Table 7. Erf fit parameters describing CCN spectra $N_{CCN}(S)$ *versus* S as parametrization input data (compare Fig. 10 and 11b,d). Fit parameters are provided for annually averaged CCN spectra and resolved by seasons.

time period	A [cm^{-3}]	S_0 [%]	w_0	R^2
all	1067±22	0.07±0.01	2.1±0.1	0.99
wet season	340±30	0.08±0.01	2.9 ±0.2	0.97
LRT period	532±72	0.04±0.01	4.5±1.0	0.98
transition	1180±37	0.07±0.01	3.0 ±0.2	0.99
dry season	1430±24	0.07±0.01	1.8 ±0.1	0.99

Fig. 3.