Response to the referee #2

Manuscript: **M. L. Pöhlker et al., Long-term observations of cloud condensation nuclei over the Amazon rain forest – Part 2: Variability and characteristics of biomass burning, long-range transport, and pristine rain forest aerosols**, ACP-2017-847)

We appreciate the very thorough and helpful comments by Referee #2, which have been considered carefully and helped to improve the quality of our manuscript. The referees' comments and our responses are outlined in detail below:

[2.1] Referee comment: The meaning of "near-pristine" is not very well defined. In section 2.3 the definition of NP excludes periods with concentrations of EBC that surpass a certain threshold to identify "the cleanest aerosol conditions" (p. 6, l. 22). Clean aerosol conditions and pristine conditions are however not the same. "Pristine" hints towards a natural aerosol state. Before anthropogenic emissions natural aerosol conditions would have included LRT of Saharan dust and natural forest fire emissions occasionally. Hence, defining "near-pristine" as the absence of any other natural aerosols and only taking into account the rain forest generated particles is not representative of the variability of pristine conditions. Calling them "near"-pristine does not help, because this is with reference to the unavoidable anthropogenic influences. From how the "near-pristine" conditions are identified and described, I understand that they are meant to reflect unperturbed aerosol conditions that are dominated by the Amazon rain forest as particle
and precursor source. It is also evident from page 12, l. 39 that the authors hope to provide data for the preindustrial-like Amazonian atmosphere with the near-pristine cases. In this context it is even more important to acknowledge that clean cases exclude other conditions as mentioned above that were however present in the preindustrial time. For that reason, I urge the authors to rename their "near-pristine" classification into "clean" conditions as this reflects the actual definition much better, "near-pristine" is misleading. As more and more work is being done to extract preindustrial-like information in various environments it is important to be clear about the terminology.

Author Response: We understand the referee's concerns and tried to clarify the corresponding aspects. Please refer to our detailed response to comment **[2.2]**, which is closely linked to comment **[2.1]**.

[2.2] Referee comment: I am also surprised that "near-pristine" periods are only defined via EBC. Why did the authors not include CO and ozone mixing ratios or back trajectories which are all at hand? If particles were removed through precipitation, EBC can be low, while CO is elevated and hence the air mass cannot be classified as "near-pristine" or rather clean, even though the focus is on aerosol particles.

Author Response: We revised our analysis on pristine and near-pristine conditions at ATTO very carefully and extended the corresponding analysis. Besides black carbon, we have now taken CO, total aerosol concentrations, and backward trajectories into account. Based on these markers we defined four different filters to approximate pristine conditions to the best of our instrumental capabilities. We agree with the referee that the wording of measured or modelled (near-)pristine conditions is crucially important as this topic is quite complex. Therefore, we precisely define what we consider as pristine states at ATTO. Moreover we discuss the corresponding limitations of our definitions. The former Sect. 2.3 (now Sect. 2.7) has been extended substantially and now discusses the role and relevance of pristine conditions at ATTO as follows:

"**2.7 Definition of empirically pristine rain forest aerosol conditions at ATTO**

The term pristine is bound to a pre-human reference state with prevailing natural atmospheric conditions, in the absence of any anthropogenic influences. Andreae (2007) pointed out that in the present-day atmosphere "there are no places where we can expect to find truly pristine conditions". This is particularly true with respect to long-lived trace gases, such as CO_2 and CH_4, which have accumulated in the atmosphere due to man-made activities. However, also the aerosol abundance and composition is substantially perturbed by anthropogenic emissions. This also includes aerosols at remote locations, which are altered to varying degrees by a globally pervasive background pollution. It has been controversially debated if and to what extent certain marine and remote continental locations still approximate pristine atmospheric conditions (e.g., Andreae, 2009; Martin et al., 2010b; Pöschl et al., 2010; Chi et al., 2013; Hamilton et al., 2014). Frequently, conditions with low anthropogenic influences are characterized as 'clean' or 'near-pristine', although these terms are rather differently and often vaguely defined. The discussion is inherently difficult since truly pristine conditions are not available any more to quantify how close the present-day atmosphere still gets to its original pre-human state. Accordingly, truly pristine conditions remain hypothetical and any degree of a near-pristine state has to be defined indirectly with respect to the absence of man-made emissions.

In the Amazon Basin, aerosol conditions during most of the wet season are comparatively clean with low – though detectable – concentrations of background pollution. During the multi-months wet season period that can overall be characterized as near-pristine, certain episodes can be identified where pollution concentrations drop even further and below analytical detection limits. Under these conditions, the aerosol composition can be regarded as empirically undistinguishable from a pristine state. The present study uses these episodes to operationally define a pristine state of the aerosol composition and properties in a tropical rain forest environment, which we call empirically pristine rain forest (*PR*) aerosol.

The selection of a robust marker is crucial for the assessment of the *PR* state. In previous studies, the concentrations of BC, CO, and CN as well as the air mass origin by means of BTs have been used as corresponding markers at other locations, however all of them are characterized by certain limitations (Hamilton et al., 2014 and reference therein). For the present study, we explored the suitability of BC, CO, CN, and BTs as well as combinations of them as *PR* markers. Their strengths and limitations can be summarized as follows:

- BT-based *PR* filter: BTs help to identify periods with comparatively clean air mass advection due to fact that those BTs, which reach furthest to the northeast across the South American continent, overpass the largest fraction of uninhibited and, thus, untouched forest regions (C. Pöhlker et al., 2018). Accordingly, a BT filter allows to exclude local and regional sources of anthropogenic pollution. However, BTs do not allow to reliably filter out long-range transport aerosols, particularly from African sources. Accordingly, BTs are not suitable as a stand-alone *PR* filter, however, they are useful in combination with other *PR* markers as outlined below.
- CN-based *PR* filter: The total particle concentration *per se* is not a good filter for clean or (near-)pristine states since natural aerosol conditions range from very low (e.g., N_{CN} in Antarctica reaching down to 10 cm^{-3}; Fiebig et al., 2014) to very high concentrations (e.g., N_{CN} in the upper troposphere reaching above 10 000 cm^{-3}; Andreae et al., 2018). Accordingly, N_{CN} is also not a good stand-alone *PR* filter. However, with respect to the 'typical' Amazonian aerosol concentrations in the boundary layer (i.e., few hundreds up to few thousands of particle per cm^3), the total particle concentrations provides at least a helpful reference level for the extent of pollution.
- BC-based definition of *PR*, called PR_{BC}: BC represents a unique indicator for combustion aerosol particles, which in the Amazon Basin almost entirely originate from anthropogenic sources (i.e., fossil fuel and biomass burning in South American and Africa). Accordingly, we defined PR_{BC} episodes by means of the absence of detectable BC_e. Petzold and Schönlinner

(2004) determined the detection limit of the MAAP "as the minimum resolvable absorbance" by considering "the variability of blank filter optical properties". The detection limit corresponds to the resulting mean absorbance of the blank filter + 3 times the standard deviation (std) resulting in an absorption coefficient of 0.132 Mm^{-1} for 30-min averages. The M_{BCe} was calculated by using mass absorption cross sections (MAC) retrieved by fitting MAAP absorption coefficients at 637 nm and single particle soot photometer (SP2) rBC mass measurements during the different seasons as explained in Saturno et al. (2017a). Using the MAC values measured at ATTO (MAC for the wet season: 11.4 ± 1.2 $m^2\ g^{-1}$; MAC for the dry season: 12.3 ± 1.3 $m^2\ g^{-1}$), the conversion to M_{BCe} corresponds to 0.011 – 0.012 $\mu g\ m^{-3}$. Note that this threshold would be higher if a "traditional" MAC value of 6.6 $m^2\ g^{-1}$ were used to calculate M_{BCe} (~0.019 $\mu g\ m^{-3}$). Here, we selected a threshold concentration of M_{BCe}^{*} = 0.01 $\mu g\ m^{-3}$, when the ATTO-specific MAC values are taken into account. Accordingly, PR_{BC} periods fulfill the criterion $M_{BCe} < M_{BCe}^{*}$ for ≥6 h (after applying a 5-point running average to the 1 h M_{BCe} data).

- CO-based definition of *PR*, called PR_{CO}: The BC approach has the potential drawback that it may overemphasize periods with strong precipitation, since heavy rain removes the BC/aerosol content in actually polluted air masses irrespective of the gas phase composition (see Hamilton et al., 2014). Accordingly, the PR_{CO} filter is based on the gas phase combustion marker, CO. In our definition, PR_{CO} conditions prevailed during periods, in which the ATTO c_{CO} concentrations dropped below the monthly background CO levels at the following reference stations (https://www.esrl.noaa.gov/gmd/dv/site/CPT.html, last access 07 Apr 2018): Whenever the ATTO c_{CO} data was associated with northern hemispheric BTs (i.e., NE or ENE BT clusters, Fig. S1) and dropped below the average of the monthly CO levels obtained at the three background stations at Ragged Point in Barbados (RPB), Assekrem in Algeria (ASK), and Izaña in Cape Verde (IZO), the episode was flagged as PR_{CO}. Analogously, whenever the ATTO c_{CO} data was associated with southern hemispheric BTs (i.e., E and ESE BT clusters) and dropped below the monthly CO levels obtained at the background station on Ascension Island (ACS), the episode was also flagged as PR_{CO}. Note that the time series of monthly c_{CO} levels from the reference stations was linearly interpolated to hourly values for the comparison with ATTO c_{CO} data. The BTs from the northern hemisphere (i.e., NE and ENE BT clusters) account for ~70 % of all BTs during the wet season. Among these ~70 %, ~30 % can be attributed to the NE BT clusters. The locations of the CO background stations, the wet season ATTO BT ensemble, as well as the hemispheric CO distribution is shown in Fig. S2.

The seasonality in the frequency of occurrence, *f*, of the PR_{BC} and PR_{CO} filters is generally very similar, as shown in Fig. 1: The first PR_{BC} and PR_{CO} episodes occur in February. Their highest *f* is reached around the second half of April and the first half of May. Afterwards, *f* decreases steeply and *PR* conditions occur only occasionally in June and July. Overall, PR_{BC} and PR_{CO} episodes overlap for ~25 % of the total time that is flagged by at least one of the two filters. Since both approaches are associated with certain limitations and uncertainties, we further analyzed two combinations of PR_{BC} and PR_{CO}: the union of both sets ($PR_{BC \cup CO}$) and the intersection of both sets ($PR_{BC \cap CO}$) as shown in Fig. 1. For the in-depth analysis of aerosol and CCN properties in this work, we chose the $PR_{BC \cap CO}$ filter, which we consider as most strict and, thus, robust since the criteria of both filters have to be fulfilled.

With respect to the *PR* filters, the following aspects are worth mentioning: *PR* episodes at ATTO exclusively occur during the wet season due to a combination of two effects: First, the ATTO site is very remote and the characteristic wet season air mass advection occurs mostly over uninhabited areas (see C. Pöhlker et al., 2018). Second, the high precipitation rates entail strong scavenging and, thus, relatively short aerosol particle lifetimes, which reduces the long-range transport of the background pollution aerosol load. In contrast, dry season *PR* episodes have almost never been observed at ATTO due to the extensive biomass burning emissions in South America and

Africa in combination with low scavenging rates and, thus, long atmospheric aerosol life times. This is relevant since wet vs. dry season conditions have likely been associated with different atmospheric states, for example with respect to VOC concentrations and aerosol populations as well as photochemical conditions. While wet season *PR* states are still experimentally accessible as outlined in this work, the dry season *PR* state appears to be entirely swamped by pollution.

The primary filters are based on the combustion markers BC and CO, which do not allow a discrimination between wild vs. man-made fires. Accordingly, the contribution of wild fire emissions, which were part of an originally pristine atmosphere in the Amazon, are erroneously filtered out. Generally, wild fires in the tropical Amazonian forests are rare events due to the fact that the dense and moist canopies – if unperturbed – effectively maintain a fire-immune state (e.g., Cochrane, 2003; Nepstad et al., 2008). Nevertheless, wild fires play a certain, although minor, role for the ATTO observations, since the wet season BTs cover the Guianan savanna ecoregions that account for ~8 % of the ATTO footprint region (Olson et al., 2001; C. Pöhlker et al., 2018). Within these regions, the infrequent occurrence of wild fires is part of the savanna-specific fire regime (de Carvalho and Mustin, 2017). Figure S3 illustrates the relevance of wild fires for ATTO under wet season conditions.

Moreover, Saharan dust as well as marine aerosols from the Atlantic Ocean are advected towards the ATTO region via wet season LRT plumes, which are most frequent in February and March (Moran-Zuloaga et al., 2017). Saharan dust and marine aerosols can also be considered as part of an original pristine atmospheric state in the Amazon. However, all LRT plumes arriving at ATTO are filtered out by the PR_{BC} and PR_{CO} approaches due to the fact virtually all LRT plumes contain a substantial fraction of smoke from mostly man-made fires in West Africa (Moran-Zuloaga et al., 2017). Accordingly, the role of the present-day LRT plumes arriving at ATTO has to be differentiated carefully as they represent a (partly internal) mixture of natural and anthropogenic aerosols."

The following figures have been revised according to the updated analysis and definition of empirically pristine rain forest (*PR*) aerosol conditions at ATTO: Fig. 1, Fig. 3, Fig. 5, Fig. 6, and Fig. 11. The following figures have been newly added to the supplement file in the course of the *PR* analysis: Fig. S2, Fig. S3, Fig. S4, Fig. S5, Fig. S6, Fig. S7, Fig. S8, and Fig. S9.

[2.3] Referee comment: The results of the paper would be even more informative if the authors provided the fraction in time throughout which the four different conditions are present. How often do clean conditions prevail? How often the mixed-polluted? It would be very informative if these simple statistics could be added.

Author Response: Thank you for the suggestion. We have added the requested statistics in a dedicated new paragraph in Sect. 1.2 as follows:

"Second, we will analyze the following four case studies that represent characteristic events and conditions in the central Amazon region:

- During certain wet season episodes, when no tracers of pollution aerosols are detectable any more, the aerosol population can be regarded as empirically not distinguishable from pristine, i.e., completely unpolluted rain forest conditions. This empirically pristine state of the rain forest (*PR*) aerosol prevails during 10 to 40 days per year (depending on *PR* definition, see Sect. 2.7).
- Long-range transport (*LRT*) aerosol advection during the wet season brings Saharan dust, African biomass burning smoke, as well as marine aerosol particles from the transatlantic passage. The *LRT* case study represents conditions that prevail between 50 and 60 days per year (see Moran-Zuloaga et al., 2017).

- Biomass burning (*BB*) smoke from man-made forest fires in the various deforestation hotspots in the basin influences the atmospheric state at ATTO almost permanently during the dry season and for extended episodes during the transition periods (>100 days per year) (Saturno et al., 2017a). The *BB* case study in this work analyzes large deforestation fires in the southeastern basin, whose smoke reached ATTO after few days of atmospheric processing. Accordingly, the *BB* case study characterizes the typical conditions of aged smoke influencing the atmospheric state at ATTO.
- Mixed pollution (*MPOL*) from African *LRT* and local/regional fires represents a frequent aerosol scenario at ATTO (Saturno et al., 2017a). The advected African aerosols mainly comprise biomass and fossil fuel combustions emissions, although the exact composition of these dry season *LRT* plumes is still poorly analyzed. The *MPOL* case study focusses on a period when African volcanogenic aerosols were advected to ATTO – an event that has been well documented in Saturno et al., 2017b. We selected this episode since the microphysical properties of the volcanogenic aerosol are characteristic enough to discriminate them from the local/regional smoke emissions. Accordingly, the alternating pattern of *LRT* vs. local/regional pollution can be clearly resolved for the *MPOL* period. However, note that volcanogenic plumes are comparatively rare events, whereas African combustion emissions, which are much harder to discriminate from the local/regional emissions, are a more common scenario. Accordingly, the *MPOL* case study is an example for a complex aerosol mixture due to alternating African vs. local/region influences during the dry season."

[2.4] Referee comment: The paper is well written, however there many qualifiers and redundancies in the text. In the technical comments section some instances are mentioned, but it would be helpful if the authors worked through the manuscript again to remove those. Additionally, the use of some terms is rather unusual. The term "pulses" should be replaced by "events". Also the precise meaning of "aerosol cycling" needs to be explained. Is cloud processing meant, or atmospheric processing? Please use the exact description. The word "trend" is frequently used in contexts, where no trends can be observed. The meaning is rather "variability, curve shape etc.". Below this is pointed out in detail.

Author Response: We agree with the referee and appreciate the hints. We have addressed the semantic aspects brought up to clarify our statements as requested. Furthermore, we worked through the entire manuscript and removed unnecessary qualifiers and redundancies. Details are outlined in the technical comment section below.

[2.5] Referee comment: Please go through the references again. Often XY et al. yyyyb comes before "a".

Author Response: Has been corrected.

[2.6] Referee comment: p.2, l. 14: At this point it is not clear what the efficiency spectrum is, a brief explanation is required.

Author Response: We have implemented the following explanation for the CCN efficiency spectrum into the corresponding sentence: "… (CCN divided by aerosol number concentration plotted against water vapor supersaturation) …".

[2.7] Referee comment: p.2, l. 27: the reference to volcanic emissions is confusing without any explanation, either provide one or do not mention volcanic influence here.

Author Response: We have revised the abstract to make it clearer and more concise in this (and various other aspects). The relevant sentence here have been changed to:

> "Mixed pollution (*MPOL*) conditions with a superposition of African and Amazonian aerosol emissions during the dry season. During the *MPOL* episode presented here as a case study, we observed African aerosols with a broad monomodal distribution ($D \approx 130$ nm, $N \approx 1300$ cm^{-3}), with high sulfate mass fractions from volcanic sources (~ 20 %), and correspondingly high hygroscopicity ($\kappa_{<100nm} \approx 0.14$, $\kappa_{>100nm} \approx 0.22$) that were periodically mixed with fresh smoke from nearby fires ($D \approx 110$ nm, $N_{acc} \approx 2800$ cm^{-3}) with an organic-dominated composition and sharply decreased hygroscopicity ($\kappa_{<150nm} \approx 0.10$, $\kappa_{>150nm} \approx 0.20$)."

[2.8] Referee comment: p.2, l. 30f: No D_ait is provided for the size distributions, but kappa_ait is given. Be consistent in providing all information.

Author Response: The broad size distribution of the case study *MPOL* (previously called MixPol) did not allow a meaningful bimodal fit. Accordingly we used a monomodal fit to describe the distribution. Accordingly only one modal diameter is reported. However, since kappa shows a size dependence we reported kappa for the size range >100 nm and <100 nm. For clarification we placed κ_{Ait} by $\kappa_{<100nm}$ and κ_{acc} by $\kappa_{>100nm}$. Moreover, we edited the relevant text sections throughout the manuscript and Table 2 to clarify that only a monomodal fit is reasonable for the *MPOL* case study.

[2.9] Referee comment: p.2, l. 33: Why is the sensitivity of the CCN population towards changing supersaturation not mentioned for the mixed pollution cases?

Author Response: The abstract has been revised completely. The sensitivity towards *S* for all cases is not outlined in the abstract any more. Instead, the following statement has been added:

> "The measurement results suggest that CCN activation and droplet formation in convective clouds are mostly aerosol-limited under *PR* and *LRT* conditions and updraft limited under *BB* and *MPOL* conditions."

[2.10] Referee comment: p. 3, l. 36: "multi-month trends" are not trends but rather changes or variability across the time period, also l. 38, not diurnal "trends" rather "diurnal cycle"

Author Response: We have replaced "trends" by "variability" as suggested.

[2.11] Referee comment: p. 4, l. 1: Include info on what is the relevant size range?

Author Response: We have removed "CCN-relevant size range" to avoid confusion here.

[2.12] Referee comment: p. 4, l. 7: please provide a reference for "previous studies"

Author Response: The companion part 1 paper contains a literature synthesis section with all previous CCN studies in the Amazon region. Thus, we have added the following reference:

"… (see M. Pöhlker et al., 2016 and references therein) …".

[2.13] Referee comment: p.4, l. 13f: Provide information on what the relevant supersaturation range is for the ATTO region. Regarding the introduction, I would have expected a short paragraph that introduces the relevant literature on the ATTO site on which this study builds. After reading the abstract one expects to learn at least more about air mass transport towards ATTO in the introduction. Please introduce a short paragraph on this.

Author Response: We have added the following statement in the introduction (page 3) to specify the relevant supersaturation range in the ATTO region:

> "The relevant peak *S* levels in the Amazon are assumed to range from ~0.1 % to ~1 % (e.g., Andreae, 2009; Reutter et al., 2009; Pöschl et al., 2010; C. Pöhlker et al., 2012; Farmer et al., 2015; M. Pöhlker et al., 2016). Moreover, a recent study suggests that also substantially higher *S* (>> 1 %) can be reached in deep convective clouds under certain conditions (Fan et al., 2018). However, a systematic and quantitative assessment of relevant peak *S* distributions in the Amazonian atmosphere is still lacking (see discussion in Sect. 3.9)."

Moreover, the following section on relevant supersaturation levels has been included in the discussion in Sect. 3.9:

> "The peak supersaturation and its variability is an essential parameter to understand aerosol-cloud interactions. However, only spares quantitative information on the actually relevant *S* range is available. The spectra in Fig. 11 are plotted for a broad *S* range from 0.001 to 20 %. According to Reutter et al. (2009), this range can be generally subdivided into very low ($S < 0.1$ %), low ($S < 0.2$ %), transitional (0.2 % < S < 0.5 %), and high ($S > 0.5$ %) supersaturation regimes. Furthermore, a recent study by Fan et al. (2018) suggests that under certain conditions even very high supersaturations (S >> 1 %) can be reached in deep convective clouds. One approach to actually quantify ATTO-relevant average peak *S* at cloud base, $S_{cloud}(D_H,\kappa)$, uses the position of the Hoppel minimum, D_H, as outlined in Hoppel et al. (1986) and Krüger et al. (2014). According to results in our companion part 1 paper and the present study (see Table 2), ATTO-relevant $S_{cloud}(D_H,\kappa)$ values range from ~0.15 % to ~0.34 %. This $S_{cloud}(D_H,\kappa)$ range is shown in as a '*S* landmark' in Fig. 11 for orientation."

In terms of characteristic air mass transport to ATTO, we have added the following paragraph in Sect. along with relevant references:

> "To explore essential biogeochemical processes, such as aerosol-cloud interactions, in the Amazon rain forest, the Amazon Tall Tower Observatory (ATTO) has been established in 2010/11 (for details see Andreae et al., 2015). The central Amazon Basin is characterized by a pronounced seasonality in atmospheric composition in response to the north-south oscillation of the intertropical convergence zone. During the wet season, the ATTO site receives comparatively clear air masses of marine origin from the northeast that travel over mostly untouched rain forest areas, whereas during the dry season strongly polluted air masses are advected from the southeast, originating from numerous fires in the Amazon's arc of deforestation (for details see C. Pöhlker et al., 2018). Detailed information on characteristic differences in atmospheric state and processes for the contrasting wet vs. dry season conditions in the ATTO region can be found in a number of recent studies (e.g., Nölscher et al., 2016; M. Pöhlker et al., 2016; Moran-Zuloaga et al., 2017; Saturno et al., 2017a, Yañez-Serrano et al., 2017)."

[2.14] Referee comment: Why do the authors invent a new abbreviation for equivalent black carbon? The standard is "EBC" (Petzold et al., 2013). Please use this one.

Author Response: The abbreviation BC_e has not been invented here, but has been used in numerous aerosol studies from the Amazon region and beyond (e.g., Andreae and Gelencser, 2006; Brito et al., 2014; Andreae et al., 2015; Moran-Zuloaga et al., 2017; Saturno et al., 2017). Obviously, different nomenclatures are co-existing here. In this particularly case, we prefer to keep BC_e in order to be consistent at least with the studies on Amazonian aerosols.

[2.15] Referee comment: The equations are not numbered. There is no need to repeat Eq. 1 without ammonium. It suffices to describe the change in the text as it is trivial.

Author Response: Numbers have been added to the equations. Moreover, the repeated equation has been deleted as suggested.

[2.16] Referee comment: p. 5, l. 32: Generally the phrase is "sth. corresponds to" not "with". Please replace all cases throughout the manuscript.

Author Response: Agree. We found 6 sentence, in which "with" has been replaced by "to".

[2.17] Referee comment: In the methodology section, information on how the CCNC was operated is needed. Even though this is described in the Part 1 paper, readers should get the essential information here (e.g., monodisperse mode, duration of a scan, covered range of diameters).

Author Response: Agree. We have added the following explanation:

> "Briefly, size-resolved CCN measurements were conducted using a continuous-flow streamwise thermal gradient CCN counter (model CCN-100, DMT, Longmont, CO, USA) in combination with a differential mobility analyzer (model M, Grimm Aerosol Technik, Ainring, Germany) and a condensation particle counter (Grimm Aerosol Technik). The DMA-selected size range spans from 20 to 245 nm. The analyzed supersaturation range spans from 0.11 to 1.10 %. A complete measurement cycle with scanning of all particle diameters and supersaturations took ~ 4.5 h."

[2.18] Referee comment: p. 6, l. 10: What was done with a2? Was there an assumption or was it fitted?

Author Response: In the discussion paper, this aspect was indeed incorrect - thanks for pointing this out! We have reworked the erf fitting procedure in Sect. 2.2 and eliminated the error. The revised version of Sect. 2.2. reads as follows:

> "The CCN efficiency spectra parameterization as introduced in part 1 plays a key role in the present manuscript. Note that we slightly revised and improved the fitting procedure from the companion part 1 paper. The main change implies that the fits are now forced through zero, which is physically more plausible and makes the erf fit parametrization more applicable for modelling studies. The single-error function (erf) fit (mode = 1) is represented by the following function

$$\frac{N_{CCN}(S)}{N_{CN,10}} = \frac{a_1}{2} + \frac{a_1}{2} \cdot erf\left(\frac{ln\frac{S}{S_1}}{w_1}\right) \qquad (3)$$

with a_1 as prefactor, S_1 as the supersaturation, at which half of the maximum activated fraction (*MAF*) of the aerosol particles acts as CCN (e.g., 50 % for $a_1 = 1$), and w_1 as the width of the erf fit. To simplify the fitting procedure, $a_1 = 1$ was assumed. For $a_1 = 1$ the erf converges against unity, corresponding to an activation of all particles as CCN at high *S*, which is adequate in most cases. Analogously, the double-erf fit (mode = 2) is represented by the function

$$\frac{N_{CCN}(S)}{N_{CN,10}} = \frac{a_1}{2} + \frac{a_2}{2} \cdot erf\left(\frac{ln\frac{S}{S_1}}{w_1}\right) + \left(\frac{a_1 - a_2}{2}\right) \cdot erf\left(\frac{ln\frac{S}{S_2}}{w_2}\right) \qquad (4)$$

with the index 1 and 2 specifying the variables for both modes. To simplify the fitting procedure, $a_1 = 1$ was assumed.

Note further that in part 1 we tested different reference aerosol number concentrations, $N_{CN,Dcut}$ (e.g., $N_{CN,10}$ and $N_{CN,50}$), for the CCN efficiency spectra parametrization. In this study, we use only one reference concentration for clarity – namely $N_{CN,10}$. The choice of $N_{CN,10}$ can be explained by the fact that it is experimentally rather easily accessible (e.g., via condensation particle counter, CPC, measurements), whereas reference concentrations such as $N_{CN,50}$ require more elaborated experimental setups (e.g., scanning mobility particle sizer, SMPS, data)."

[2.19] Referee comment: p. 7, l. 6: Are the 1000 m above sea level or above ground? If a.s.l. how high above the measurement site?

Author Response: 1000 m refers to above ground. We have added "(above ground level)" to specify this aspect.

[2.20] Referee comment: p. 9, l. 31: Does this statement refer to the Amazon or is it a general statement?

Author Response: This is a general statement. We specified it by adding "at ATTO and further locations worldwide".

[2.21] Referee comment: There is no need for footnotes. They can all be included in the main text which is already quite detailed. It is not apparent why some details are moved to footnotes. Specifically on p. 10, delete the 2nd sentence in the footnote, because it is a repetition.

Author Response: We have revised and streamlined the footnotes to remove unnecessary repetitions.

[2.22] Referee comment: p. 11, first paragraph: What about the influence of more local anthropogenic emissions?

Author Response: Please refer to our response to comment [1.2], which addresses this questions.

[2.23] Referee comment: p. 16, l. 4: What is meant by "perception"? Please clarify.

Author Response: To clarify this aspect, we have replaced the sentence:

"This perception of CCN efficiency spectra stands in certain analogy to a concept introduced by Reutter et al. (2009) …"

by:

"The CCN efficiency spectra can be linked to a concept introduced by Reutter et al. (2009) …"

[2.24] Referee comment: p. 18, l. 19f: Is the fraction of long-range transported mineral dust and sea salt really large? Please include the values in the text: What is the fraction of supermicron dust and salt in mass or number concentration?

Author Response: The influence of dust and seas salt can be remarkably strong during certain episodes. In several relevant sections of the text, we added and emphasized the reference to a recent ATTO study by Moran-Zuloaga et al., 2017, which provides a detailed discussion of the role and relevance of long-range transported dust and sea salt in the Amazonian atmosphere.

[2.25] Referee comment: p. 20, l. 21: How can the ratios of 26 and 49 be consistent? That's a factor of 2.

Author Response: Emission ratios such as $\Delta N_{CN}/\Delta c_{CO}$ and $\Delta N_{CCN}(S)/\Delta c_{CO}$ typically reveal quite some variability as a function of fuel type and burning phase (flaming vs. smoldering). Accordingly, emission ratios are characteristic within certain ranges - for example $\Delta N_{CN}/\Delta c_{CO}$ = 30 +/- 15 cm^{-3} ppb^{-1} for vegetation fires. With respect to these uncertainties, the results by Kuhn et al. (2010) ($\Delta N_{CCN}(0.6\ \%)/\Delta c_{CO}$ = 26 cm^{-3} ppb^{-1} for biomass burning) and our results ($\Delta N_{CCN}(0.61\ \%)/\Delta c_{CO}$ = 18 cm^{-3} ppb^{-1} also for biomass burning) are indeed quite consistent. To clarify this statement, we removed the results for urban plumes ($\Delta N_{CCN}(0.6\ \%)/\Delta c_{CO}$ = 49 cm^{-3} ppb^{-1}) by Kuhn et al., (2010), which are only peripheral information here and may lead to confusion. Moreover, we added "(i.e., $\Delta N_{CCN}(0.61\ \%)/\Delta c_{CO}$ = 17.9 ± 1.3 cm^{-3} ppb^{-1}, see Table S4)" for clarification.

[2.26] Referee comment: p. 21, l. 32: Define aerosol "cycling"

Author Response: We have removed the entire sentence "Ultimately, these observations are discussed in the light of the related CCN cycling." as it is not essential here and may lead to confusion.

[2.27] Referee comment: p. 22, l. 29: The African volcanic emission appear out of nowhere. Were they never observed before at ATTO? Why can the authors be so sure that they have observed volcanic emissions?

Author Response: A recent study by Saturno et al., 2017b ("b" in the revised version) analyzes the African volcanogenic plume that impacted ATTO during Sep 2014 in details. The reported evidence for volcanogenic influence on the aerosol composition in the central Amazon is quite convincing. We have added the reference to this study in the corresponding section of the paper for clarification.

[2.28] Referee comment: p. 24, l. 35: The here defined NP conditions are very selective because they describe only clean conditions. They are hence not an approximation of a preindustrial state, but represent only a potential fraction of the preindustrial atmosphere in that region.

Author Response: See our response to comment **[2.2]**.

[2.29] Referee comment: p.2, l. 24: What do you mean by "CCN cycling in relation to aerosol-cloud interactions"?

Author Response: The abstract has been revised and the corresponding statement remove for clarity.

[2.30] Referee comment: p.3, l. 20: change to "Ndb ranges from few hundred: : : for clean to 1000 and : : : for polluted conditions"

Author Response: Has been changed.

[2.31] Referee comment: p. 3, l. 32: delete "of this endeavor"

Author Response: Deleted.

[2.32] Referee comment: p. 4, l. 3: D_a has not been defined yet.

Author Response: We added "with D_a as midpoint activation diameter" to define it.

[2.33] Referee comment: p. 4, l. 10f: Delete the last sentence of the paragraph, this information is not really needed and the paper can benefit from being shortened.

Author Response: Done.

[2.34] Referee comment: p. 4, l. 7: Delete " As a particularly: : :concept". This is a qualifier that does not help the reader to understand the efficiency spectra better.

Author Response: Deleted.

[2.35] Referee comment: p.4, l. 29f: delete "particularly interesting"

Author Response: Deleted.

[2.36] Referee comment: p. 4, l. 38: What is an "efficient CCN prediction?" Is this method particularly fast or do you mean effective?

Author Response: "efficient" has been replaced by "effective". Thanks.

[2.37] Referee comment: p. 5, l. 19: delete "predicted"

Author Response: Deleted.

[2.38] Referee comment: P. 9, l. 4: Spell out Mar

Author Response: Done

[2.39] Referee comment: p. 9, l. 7f: The sentence beginning with “First, the: : :” is not a grammatically correct sentence.

Author Response: True. The sentence has been modified to ” First, P_{TRMM} data represents the area-averaged precipitation rate in the ROI_{ATTO} (see Fig. S1).”

[2.40] Referee comment: p.9, l. 17: a reference is missing

Author Response: The following reference has been added: “(e.g., Moran-Zuloaga et al., 2017)”.

[2.41] Referee comment: p. 9, l. 28: replace “trend” by “behavior”

Author Response: Replaced.

[2.42] Referee comment: p. 9, l. 32: Shorten this paragraph: “Figure 1 provides: : : sections will focus on detailed aerosol and CCN characteristics in wet: : : year 2014.” Delete the rest as is has been mentioned already.

Author Response: Modified as suggested.

[2.43] Referee comment: p. 10, l. 28: delete “in the context of this study”.

Author Response: Deleted.

[2.44] Referee comment: p. 11, l. 10: replace “trend” with “observations”

Author Response: Done.

[2.45] Referee comment: p. 11, l. 13 ff: delete this sentence, it has already been said.

Author Response: Deleted.

[2.47] Referee comment: p. 11, l. 31: replace “in more detail of” with “on”

Author Response: Done.

[2.48] Referee comment: p. 12, l. 1: replace “pendant” with “counterpart”

Author Response: Replaced.

[2.49] Referee comment: p. 12, l. 13: Do you mean N_cn10? There are a couple of other instances where “10” has been omitted.

Author Response: Has been corrected.

[2.50] Referee comment: p. 12, l. 28: delete “interesting” this qualifier is not needed.

Author Response: Deleted.

[2.51] Referee comment: p. 13, l. 16: What is meant by aerosol cycling here?

Author Response: “Cycling” has been replaced by “variability”.

[2.52] Referee comment: p. 14, l. 39: delete “mode”

Author Response: Has been deleted.

[2.53] Referee comment: p. 15, l. 14: replace “ranged” with “was”, and its “contributions”

Author Response: Done.

[2.54] Referee comment: p. 15, l. 29: delete “solid”

Author Response: Done

[2.55] Referee comment: p. 15, l. 35: write just “a tool”

Author Response: Done.

[2.56] Referee comment: p. 16, l. 34f: remove “as a particularly instructive example

Author Response: Removed.

[2.57] Referee comment: p. 17, l. 11: shouldn’t the units of SW_in the figures be W per square meter?

Author Response: Yes – has been changed.

[2.58] Referee comment: p. 17, l. 24: A “LRT pulse” does not make sense, replace by “LRT event”

Author Response: This has been changed: “pulses” has been replaced by “events”.

[2.59] Referee comment: p. 17, l. 28: remove “exactly”

Author Response: Removed.

[2.60] Referee comment: p. 18, l. 10: Do the authors refer to natural or anthropogenic fires, or both?

Author Response: The fires in Africa are to significant extent of anthropogenic origin. This clarify this aspects, we added the following sentence: “Note that the African fires in West Africa are to a large extent agriculture-related and, thus, man-made (e.g., Barbosa et al., 1999; Capes et al., 2008).”

[2.61] Referee comment: p. 19, l. 20: remove “fortunate”

Author Response: Removed.

[2.62] Referee comment: p. 19, l. 23: remove “rather”

Author Response: Removed.

[2.63] Referee comment: p. 20, l. 27: remove the sentence beginning with “It: : :”. This has already been said.

Author Response: Has been removed.

[2.64] Referee comment: p. 21, l. 28: Delete this sentence, redundancy.

Author Response: Deleted.

[2.65] Referee comment: p. 21, l. 31: delete “pulses”

Author Response: Deleted.

[2.66] Referee comment: p. 22, l. 24: delete “such as industrial emissions, : : :River”, This has already been said.

Author Response: Deleted.

[2.67] Referee comment: p. 22, l. 33: delete “far””

Author Response: Done.

[2.68] Referee comment: p. 23, l. 34f: shorten to “CCN efficiency spectra serve as CCN signatures.”

Author Response: Has been shortened.

[2.69] Referee comment: p. 23, l. 38: delete “trends” because trends are not discussed in this paper.

Author Response: Deleted.

[2.70] Referee comment: p. 23, l, 39: Why “could”? Can they or can they not?

Author Response: “Could” has been replaced by “can”.

[2.71] Referee comment: p. 24, l. 1: it seems that D_H has not been introduced.

Author Response: We have added “… uses the position of the Hoppel minimum, D_H, …” to define D_H.

[2.72] Referee comment: p. 24, l. 12f: replace “trends” with “curve shapes”, and again with “shapes”

Author Response: Done.

[2.73] Referee comment: p. 24, l. 17: replace “pulses” with “events”

Author Response: Done.

[2.74] Referee comment: p. 24, l. 38: replace “pulses” with “events”

Author Response: Done.

[2.75] Referee comment: p. 25, l. 9: delete “very”

Author Response: Deleted.

[2.76] Referee comment: p. 25, l. 15f: “growth as well as to enhance the : : :” otherwise the sentence is grammatically not correct.

Author Response: Thanks – has been corrected.

[2.77] Referee comment: p. 25, l. 33: Be more specific regarding what is meant with cycling.

Author Response: We agree – "aerosol cycling and cloud properties" has been replaced by "aerosol-cloud interactions".

[2.78] Referee comment: p. 26, l. 5 delete "instructive"

Author Response: Deleted.

[2.79] Referee comment: p. 26, l. 14: delete "very" and "highly"

Author Response: Both have been deleted.

[2.80] Referee comment: p. 26, l. 22: "The aerosol particles are composed of: : :"

Author Response: "comprise" has been replaced by "are composed of".

[2.81] Referee comment: p. 26, l. 31: "on event-basis" instead of "pulse-wise"

Author Response: Has been changed.

[2.82] Referee comment: p. 27, l. 11: "mixed" instead of "superimposed"

Author Response: Has been changed.

[2.83] Referee comment: p. 27, l. 25: "behaviors" instead of "trends"

Author Response: "differences, similarities and trends" has been replaced by "observations".

[2.84] Referee comment: p. 27, l. 25 f: The sentence starting with "The array of : : :." is cryptic for saying that there exists large variability, rephrase.

Author Response: That is true. The sentence has been removed in the course of a major revision of the summary and conclusion section.

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