Response to referee comments and suggestions on acp-2019-775 by Holanda et al.

Response format description:

Black text shows the original referee comment, blue text shows the authors response, and red text shows quoted manuscript text. Changes to the manuscript text are shown as *italicized and underlined*. We used bracketed comment numbers for referee comments (e.g., [R2.1]) and author's responses (e.g., [A2.1]). Line numbers refer to the discussion/review manuscript.

Anonymous Referee #1

Received: 21 October 2019

General comment:

The current manuscript discusses observations during a single research flight of two biomass burning layers off- and on-shore of Brazil, where the upper layer was characterized by very high black carbon (BC) number concentrations while the lower layer by a much lower BC concentration. Using back trajectory calculations, the high BC layer was identified as originating from the African coast. Using this finding, the authors examined the long term data collected at the ATTO site to identify that the early part of the Amazonian dry season tends to be influenced by the long-range transport of the African biomass burning emissions while the late dry season is dominated by local burning. One of the core findings of this paper is that the SSA (at 637 nm) during the early portion of the dry season (i.e., influenced by the African plume) is nominally 0.85 while local BB emissions are found to be nominally 0.9. This finding suggests that optical and microphysical changes with the African BB plume occur during transport. The information and analysis contained in this manuscript is certainly worthy of publication, but, as discussed below, additional analysis that could be done with available data needs to be done to help explain underscore these findings. Therefore, it is recommended that the manuscript be revised and resubmitted.

Author response: We thank Referee #1 for the critical feedback and constructive suggestions. We address the individual comments below.

[R1.1] I read this manuscript with great excitement as long-range transport of biomass burning emission is a subject area that has not received much study. However, upon reading this manuscript I felt cheated. Combining the aircraft data with the long-term ATTO dataset, the authors make a very compelling argument that Amazonia is indeed influenced by African fires, but do present any further data analysis that might provide the community with a deeper understanding of what processes might be present. In the abstract, the authors correctly point out that the "microphysical properties, spatiotemporal distribution and long-range transport" BC aerosols are "not well constrained". But as highlighted below, the authors seemingly have the data to help probe this, but did not analyze that data and, in turn, are missing the opportunity to further strengthen their manuscript.

[A1.1] We understand your concern and considered it carefully. With this longer and more general response, we would like to address your comments on "missed opportunities" expressed in the referee comments R1.1 to R1.6. Subsequently, the comments R1.2 to R1.6 are further addressed individually and in more detail.

For a better understanding of the atmospheric and climatic relevance of BC, a detailed analysis of its spatiotemporal distribution in the atmosphere, on one hand, and its microphysical properties, on the other hand, is needed. Both aspects were probed during the ACRIDICON-CHUVA campaign, which yielded a large data set with manifold interesting findings (lots of "nuggets of gold", quote from R1.6).

The problem we have dealt with quite intensively was how to portion the manifold BC-related results from the large ACRIDICON-CHUVA data set best into 'compact and coherent publishable units'. Let us clarify one key aspect here at the beginning already that may have contributed to the referee's concern(s): We did not 'forget' the BC microphysics, size distributions, aerosol chemical information, etc. but rather decided to address all these results in another paper as a dedicated follow-up study (currently being prepared). In fact, in the first drafts of the present manuscript, we had an extended section on BC/aerosol microphysical properties included.

However, we noticed quite soon that the paper became way too long and hardly readable. In this context, Referee #2 seems actually very concerned that even the current form of this paper is too long (see R.2.1).

A few more thoughts on our rationale regarding 'compact and coherent publishable units': Our intention has been to split the results on BC and related aerosol aspects into several manuscripts, being (more or less) split along the large blocks of "BC spatiotemporal distribution in the atmosphere" and "BC microphysical properties". Evidently, the present study belongs to the first block and specifically deals with the layered transport of BB smoke from southern Africa to South America, which was observed during the only ACRIDICON fight (AC19) over the ocean. In order to systematically address the BC microphysical properties, also the other flights (probing several contrasting conditions) have to be included. So if we had addressed detailed BC/aerosol microphysics just within the scope of flight AC19, the study would be inherently incomplete, since some very interesting microphysical aspects would be missing that result from the comparison of contrasting flights and flight patterns. In contrast, if the microphysics from other flights (beyond AC19) would be included here, the manuscript would become way too long. So in the end, we decided to focus the present manuscript just on AC19 with one main focal point, which is the description of the layered transatlantic transport, its properties in terms of profiles and transport dynamics, as well as an estimate of its significance on the Amazon Basin in general.

As mentioned in the beginning, we considered the referee's comment(s) carefully. We came to the conclusion that certain aspects of the manuscript could/should indeed be changed and improved to account for the referee's concerns:

- We clarified several statements on the aim and scope of the study as outlined below. Hopefully this avoids any impression that the manuscript (i.e., the introduction) promises more than the results part actually covers.
- Second, we added several further results from the instruments UHSAS and C-ToF-AMS (as requested by the referee in R1.2 to R1.6) to discussed selected aspects of aerosol size distributions and chemical composition, which indeed helped to improve the quality of

the manuscript. We think that this addresses several of the referee's comments. For details, please refer to our responses to R1.2 to R1.6.

The following clarifications on statements have been included in the last paragraph of the introduction (P4-5, L130-144): This study focusses on the transatlantic transport of African BB smoke into the Amazon Basin by combining *in-situ* aircraft observations, modeling results, and remote sensing data. *The core of this work are aircraft observations made within a defined African pollution layer upon its arrival at the South American coast during the ACRIDICON-CHUVA campaign over Amazonia in September 2014 (Wendisch et al., 2016). We focus primarily on the spatiotemporal distribution and advection dynamics of the BB smoke layers by analyzing (i) aerosol and trace gas concentration profiles,* (ii) backward trajectories and African BB source regions, (iii) the seasonality of the pollution transport, (iv) the horizontal and vertical extent of the transported layers, and (v) the convective mixing and smoke entrainment from the layers into the planetary boundary layer as they are transported from the ocean into the South American continent. *Note that a detailed characterization of the microphysical aerosol properties within the BB smoke layers (e.g., the BC core diameters and mixing state) is beyond the scope of the present work and will be the subject of a separate and extended follow-up study (Holanda et al., in preparation). As a final step of the present study, we integrate its key results into the broader picture of the long-term aerosol observations at the central Amazonian ATTO site to estimate the relevance of African pollution for the aerosol life cycle in the dry season.*

[R1.2] A quick perusal of the Wendisch et al., BAMS article reveals that the HALO payload included the two SP2s, one three-wavelength PSAP, a single wavelength PSAP, UHSAS, and a C-ToF-AMS. (This reviewer is quite surprised to learn that a nephelometer was not part of the HALO payload.) These available datasets open up additional analysis that would greatly strengthen and help elucidate properties and processes. For example, the authors could very easily combine the PSAP absorption coefficients with the SP2 to derive the mass absorption cross-sections for both layers. This would inform us about how different the optical properties are between the plumes.

[A1.2] We agree with the referee. The focal point of this manuscript is to address the first observation of highly aged African BB plume over the Amazon basin using the SP2 instrument that measures the mass of individual rBC particles. Complementary aerosol measurements onboard HALO allowed us to estimate the contribution of black carbon particles in relation to other aerosol species in terms of number and mass concentrations. With respect to optical properties of the plume, the main reason why we did not include the PSAP (3-λ) data is because the atmospheric layers were detected primarily during ascents and descents of the aircraft. In such situations, the pressure fluctuations inside the inlet prevent reliable measurements with the PSAP because the instability of the filter leads to strong artefacts. Aside from that, integration times are too short to obtain good measurement statistics for the PSAP. The second PSAP instrument (1-λ) was operating behind the CVI, that means, measuring aerosol residuals, which is not addressed in this work. With respect to the UHSAS and C-ToF-AMS data sets, please refer to comments A1.4 and A1.5, respectively.

[R1.3] Furthermore, using the SP2 the authors could probe the black carbon mixing state (e.g., coating thickness). This reviewer suspects that this analysis will reveal that the coating thickness for the African plume will be thinner than that derived for the local BB plume - which would further explain why the ATTO-site derived SSA is 0.85 for the African plume and 0.90 for the local BB plumes. Also, sticking with the SP2, why not examine the BC size distributions? What do the differences - should be they exist – tell us about the two plumes?

[A1.3] That's true – the rBC size distributions and mixing state represent key properties for a detailed aerosol microphysical study. Please refer to our response A1.1, which specifies our rationale on why this is not included in the present manuscript.

[R1.4] Continuing with this theme, the authors have the UHSAS which could be used with the SP2 to examine (and compare) the two size distributions. As referenced above, the authors cite the paucity of BC microphysical properties data, yet seemingly do not take advantage of the readily available data.

[A1.4] As suggested by the referee, we included the aerosol number size distributions, based on UHSAS data for the different layers and BB plumes discussed, as a new figure in the main text (Figure 4). As expected, clear differences in the size distributions were observed between the two BB plumes/conditions: the aged African BB is characterized by a modal diameter (d_o) of 132 nm, while the fresh Amazonian BB show a smaller d_o = 124 nm. The largest particle mean diameter was observed in the marine boundary layer (MBL) with d_o 143 nm, and the smallest, in the clean layer (CL) and lower pollution layer (LPL) with d_o = 90 nm and d_o = 105 nm, respectively. The latter is consistent with the new particle formation in regions of cloud detrainment as discussed in the results section (L480 to L486). The UHSAS results are summarized in Table 2. Moreover, we found a good agreement between the SP2 and UHSAS measurements for D_o > 200 nm, as shown in the individual particle number size distributions (PNSD) in Fig. S5. Below this threshold, the SP2 efficiency decreases significantly for the scattering signal.

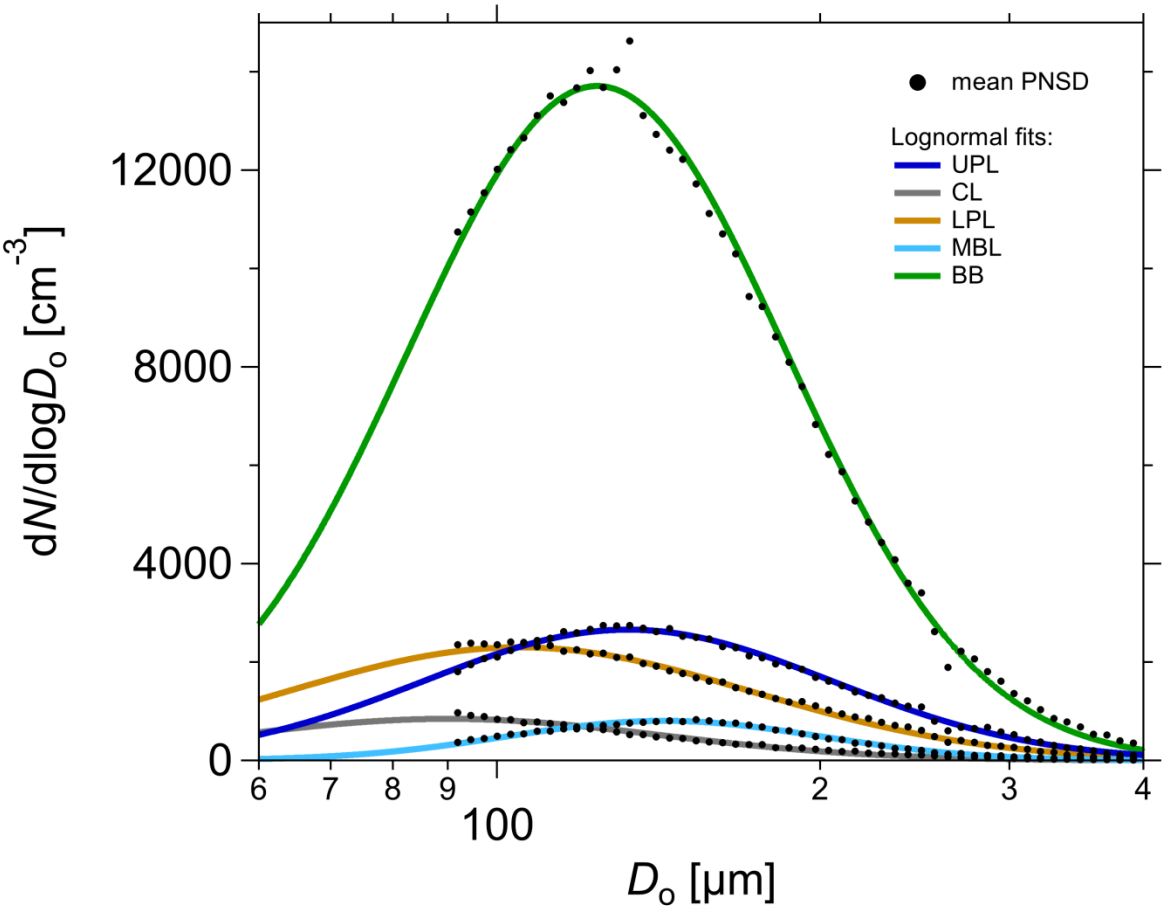

Figure 4. *Particle number size distributions (PNSD) measured by the UHSAS for UPL, CL, LPL and MBL, as defined in Fig. 3, and the fresh BB plume probed during AC19 (see Fig. 1). The data points (black dots) are fitted by lognormal functions between 90 and 500 nm (Heintzenberg, 1994).*

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Table 2. *Fit parameters of UHSAS size distributions (Fig. 4) for the different layers/plumes probed during AC19. A log-normal function (Heintzenberg, 1994) was used to fit a one-modal size distribution to the mean data points:* $\frac{dN}{d\,ln\,d_p} = \frac{A}{\sqrt{2\pi}\,ln\,\sigma_g}\,exp\left(-\frac{(ln\,d_p - ln\,d_0)^2}{2\,ln(\sigma_g)^2}\right)$

	UPL	**CL**	**LPL**	**MBL**	**BB**
A	2920	970	2890	680	13930
d_0 *(nm)*	132	90	105	143	124
σ_g	1.55	1.58	1.65	1.40	1.50
R^2	1.00	0.99	1.00	1.00	1.00

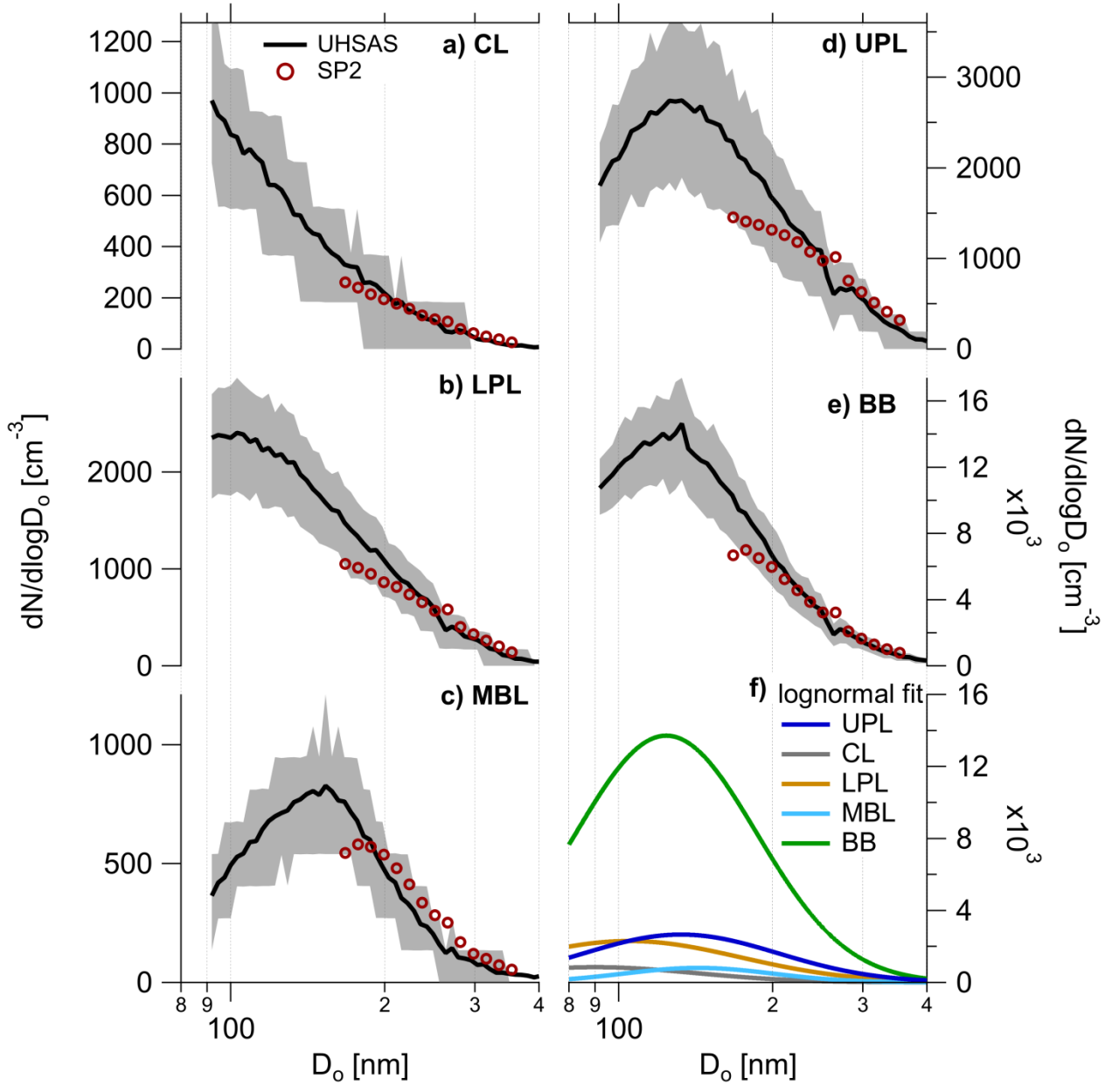

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Figure S5. *Particle number size distributions (median and interquartile range) derived from the UHSAS and SP2 (rBC + SC) for the* ***(a)*** *CL,* ***(b)*** *LPL,* ***(c)*** *MPL,* ***(d)*** *UPL and* ***(e)*** *fresh BB plume probed during flight AC19. Panel* ***(f)*** *shows the curve fits of the UHSAS data points.*

170 After including Fig.4 and Fig S5 in the present study, the following changes were necessary in the results section:

(L369-376): In the atmospheric column, f_{vol} reaches its minimum of 16 ± 9 % within the UPL and generally shows a similar profile as f_{fine}, indicating a rather aged plume (Grieshop et al., 2009; Zhou et al., 2017). *The particle number size distributions of the UPL aerosol – in comparison to the LPL, CL, MBL, and fresh BB aerosols probed during AC19 – are shown in Fig. 4 and summarized in Table 2. A modal diameter of 132 nm was observed for the UPL aerosol, whereas the fresh BB aerosol showed a clearly smaller modal diameter of 124 nm. Further note that the modal diameter in the UPL is smaller than the 220 nm observed directly off the African coast (Weinzierl et al., 2006).*

(L446-447): One interesting aspect of the LPL is that the ultrafine fraction accounts for about half of the aerosol number concentration *(d_0 = 105 nm, see PNSD in Fig. 4).*

(L454-456): In the MBL (with its top at ~600 m asl), the total and accumulation mode particle concentrations are somewhat lower than in the layers aloft ($N_{CN,20}$ = 420 ± 160 cm^{-3} and N_{acc} = 230 ± 50 cm^{-3}) *and present large diameters (d_0 = 143 nm).*

(L465-467): We further found $N_{CN,20}$ = 500 ± 60 cm^{-3}, which is comparable to N_{CN} = 500 cm^{-3} in another CL as reported by Hobbs (2003). *Within the CL, the aerosol size distribution is substantially shifted towards the Aitken mode (d_0 = 90 nm, Fig. 4).*

NOTE: By including the UHSAS data into the analysis, we have to use a new filter for defining the fresh BB plume in order to get a more characteristic and well defined size distribution. Therefore, in order to keep the "BB" definition consistent in all the analyses, data points in Figs. 5 and S6 have also been updated.

[R1.5] Sticking with the theme of missing an opportunity, the authors chose to center their discussion on number concentration: higher number fraction of BC particles in the African layer versus the measured number fraction in the local layer. The availability of the C-ToF-AMS, along with the SP2, enables the authors to derive a more meaningful mass fraction of BC in both layers. Additionally, the AMS pro-

vides a measurement datastream that can provide aerosol composition, yet is not taken advantage of. (It is interesting to note that the authors make a passing reference to the AMS on Page 13, line 424 where they cite the sulfate content concentration.) A plume advecting for 10 days from the African shore to the South American shore should exhibit differences with that observed for a localized ("fresh") plume.

[A1.5] We agree (see also A1.1). Aerosol chemical components internally or externally mixed with the rBC cores are an important factor influencing the particles' hygroscopicity and radiative effects. As requested, we included composite plots with the rBC mass fractions (with respect to the total mass detected by the SP2+AMS) for each of the BB plumes as a new figure in the main text (Fig. 6). We also included the LPL, CL, and MBL compositions for comparison. Note that the different layers could only be sampled with the C-ToF-AMS during the inshore intercepts. During the offshore section of AC19, the C-ToF-AMS was measuring aerosol residuals through the CVI inlet and could not be used in our analysis. Therefore, the statistics related to the C-ToF-AMS data are reduced due to the lower time resolution of the instrument (30 seconds) and to the use of only-inshore profiles. Despite the limited statistics, we found clear differences in the aerosol mass fractions between the different air mass. These new results were included in the manuscript as outlined below.

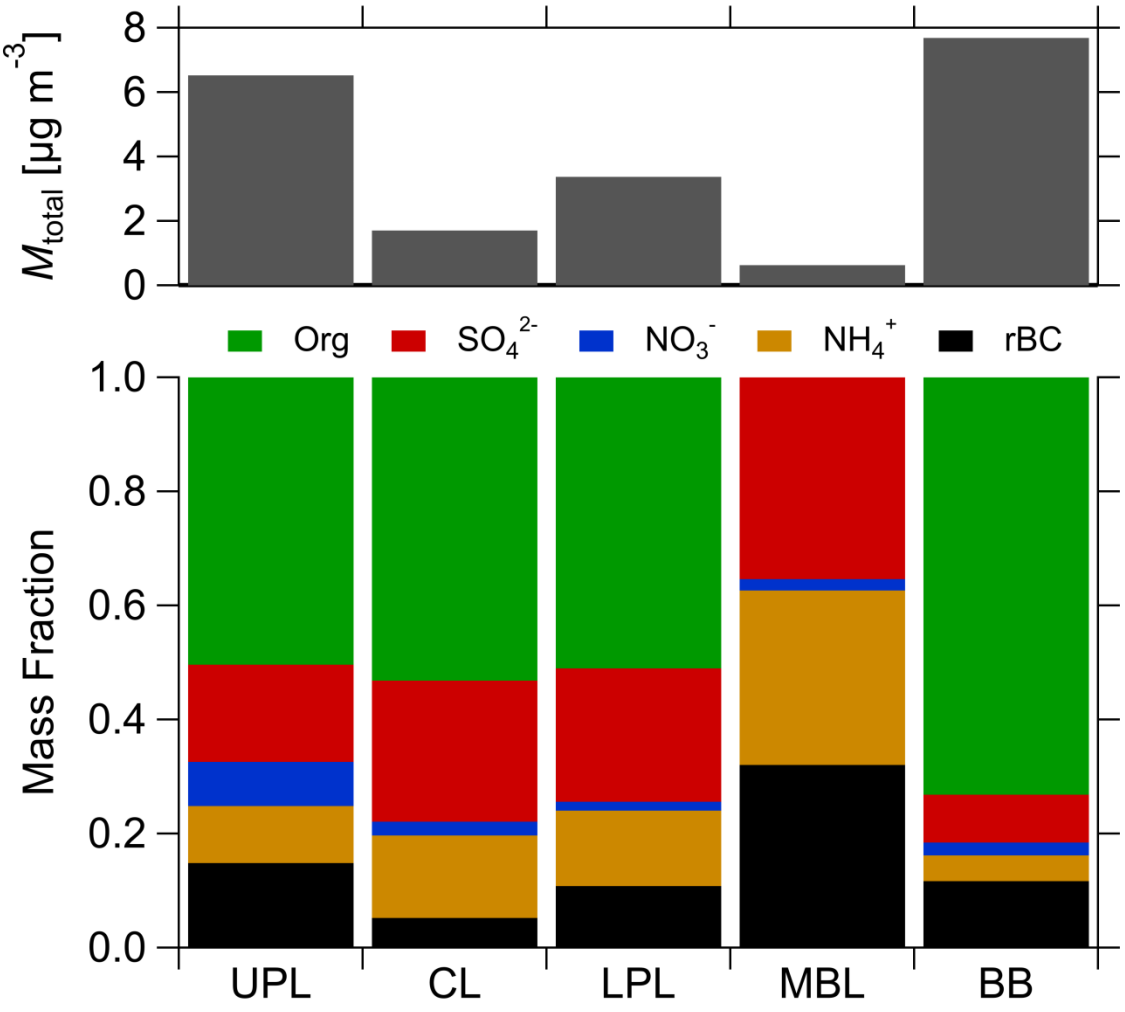

Figure 6. *Cumulative mass concentrations of non-refractory submicrometer species (i.e., organic (Org), sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+)) and rBC* ***(top)****; and mass fractions of the respective species to the total mass ($M_{total} = M_{Org} + M_{SO4} + M_{NO3} + M_{NH4} + M_{rBC}$) in the UPL, CL, LPL, and MBL, as defined in Fig. 3, and the fresh BB probed during AC19 (see Fig. 1)* ***(bottom)****. Note that no C-ToF-AMS data were available from 17:27 to 19:05 UTC during the offshore section of the flight AC19 and, therefore, a reduced number of measurements points is included in the averages. The concentration of organics was below the detection limit in the MBL.*

Further, we analyzed the photochemical aging of the organic material, which is presented in the supplementary material (Figure S7).

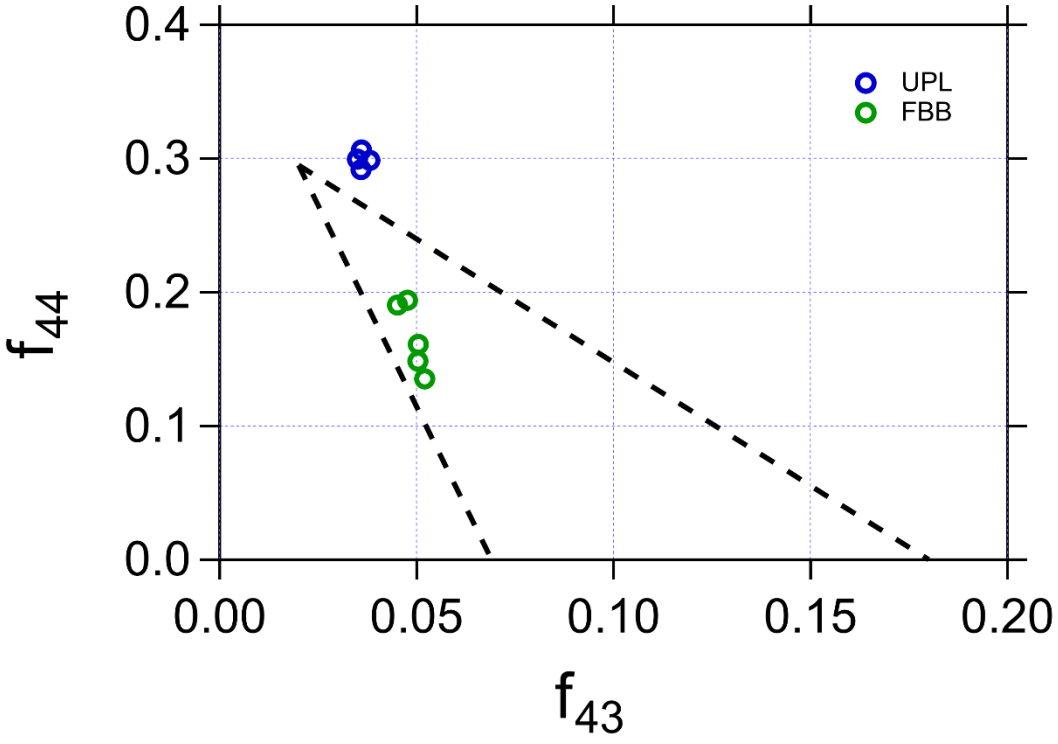

Figure S7. *Scatterplot of the ratios f_{43} (m/z 43 to total organic signal) against f_{44} (m/z 44 to total organic signal) expressing the photochemical aging of the organic aerosol measured by the C-ToF-AMS. The blue and green markers correspond to measurements within the UPL and fresh BB, respectively. The signal at m/z 44 relates mostly to CO_2^+ ions and the m/z 43 signal to $C_2H_3O^+$ ions. The triangular region (dashed lines) in the f_{44} vs. f_{43} space defines the boundaries within which most of the organic aerosol was found in previous studies and can be used as a guide to characterize oxidized organic components: data in the upper left represent more oxidized organics vs. the less oxidized organics in the lower right (Ng et al., 2010; Schulz et al., 2018).*

By including the C-ToF-AMS data in the present study, the following changes were necessary:

1. We include a brief paragraph in the methodology describing the instrumentation used (L213-217): *A compact time-of-flight aerosol mass spectrometer (C-ToF-AMS, Aerodyne Research, Inc., Billerica, MA, USA) measured the mass concentration of four chemical species (i.e., organics, sulfate, nitrate, and ammonium) of the submicrometer aerosol with a time resolution of 30 seconds (Drewnick et al., 2005; Schulz et al., 2018). A complete description of the instrument and its operation during the ACRIDICON-CHUVA campaign is given in Schulz et al. (2018) and Andreae et al. (2018).*

2. In the results section, we include the following paragraph (L389-410): *In terms of absolute mass concentrations, rBC within the UPL, with M_{rBC} = 1.0 ± 0.4 µg m^{-3} (ranging from 0.5 to 2 µg m^{-3}), approaches the highest BC levels observed at ATTO (M_{BCe} up to 2.5 µg m^{-3}; Pöhlker et al., 2018; Saturno et al., 2018b). Figure 6 shows the fractions of rBC mass relative to the other main constituents of the submicrometer aerosol (M_{total} = non-refractory + rBC) in the UPL in comparison to the CL, LPL, MBL, and fresh BB values. Organic matter – comprising co-emitted primary as well as secondarily formed organics– accounts for the dominant mass fractions in all layers, with $f_{org,M}$ ≈ 50 % in the UPL, CL and, LPL, and as much as 73 % in the fresh BB plume. Generally, the dominance of organic matter is in agreement with previous studies performed at different locations and seasons in the Amazon region (e.g. Brito et al., 2014; Chen et al., 2015; Fuzzi et al., 2005; Martin et al., 2010, 2017; de Sá et al., 2019; Schneider et al., 2011; Schulz et al., 2018; Shrivastava et al., 2019; Talbot et al., 1990). For example, in the southwestern region of the Amazon, which is heavily impacted by BB, organics account for $f_{org,M}$ > 90% in the dry season (Brito et al.2014). Note that the thermal stability of some organic species and tar balls in BB plumes can lead to an underestimation of the $f_{org,M}$ measured by the C-ToF-AMS (Adachi et al., 2018). Further, the organic matter in the UPL is significantly more oxidized than the fresh BB smoke, as shown in Fig. S7. This can be associated with the long aging times and the elevated O_3 mixing ratio in the UPL (Fig. 3h) (Martin et al., 2017). The rBC mass fractions account for $f_{rBC,M}$ = 15 % in the UPL and $f_{rBC,M}$ = 12% in the BB plume. A clear difference was observed for the mass fractions of the inorganic constituents sulfate (SO_4^{2-}), ammonium (NH_4^+), and nitrate (NO_3^-), which in sum account for $f_{inorg,M}$ = 35 % in the UPL and $f_{inorg,M}$ = 16 % in the BB*

plume. The increased $f_{inorg,M}$ in the UPL can probably be explained by aging-related condensation of the secondarily formed species SO_4^{2-}, NH_4^+, and NO_3^-. On the other hand, the lower $f_{org,M}$ in the UPL compared to the fresh Amazonian BB is related to the evaporation of organics due to fragmentation during the aging over the Atlantic.

3. We updated the values presented in the following sentence (L441-443): This possibility is supported by the relatively high sulfate content of the aerosol in this layer, which at an average value of *0.79* ± 0.02 µg m^{-3} accounts for *23%* of total aerosol mass concentration *(Fig. 6)*.

4. Accordingly, a few modifications followed up in the conclusions section (L721-727):
The plume was dominated by aerosol particles in the accumulation mode size range (N_{acc} = 850 ± 330 cm^{-3}), peaking at ~130 nm diameter, and consisting mostly of particles containing non-volatile material. Remarkably, rBC particles appeared to be a dominant species, with mean number and mass concentrations of N_{rBC} = 280 ± 110 cm^{-3} and M_{rBC} = 1.0 ± 0.4 µg m^{-3}, respectively. This accounts for ~40 % of total aerosol number and 15 % of the submicrometer aerosol mass concentrations. The UPL also shows high mass fractions of organics (50 %), sulfate (17 %), ammonium (8 %) and nitrate (10 %).

[R1.6] It is because of the available datasets outlined in this paragraph and the one before it, that this reviewer felt cheated in that so much more could be derived from the aircraft dataset and thus provide a much more complete story. It is recommended that the authors mine their data for more nuggets of gold that are surly there.

[A1.6] We understand your criticism since the ACRIDICON-CHUVA data set is rich and unique, and therefore we tried to address most of your suggestions that fit the scope of this study. We tried to find the right balance in order to have a clear and comprehensive story as outlined in A1.1. For the discussion of other aspects of this data set, please look to our papers in preparation, which will be submitted to ACP.

Other specific issues:

[R1.7] Page 2, line 46. The authors state that the BC particle number fraction is ~ 40%. I think this is wrong and that the value is closer to 30%. Using number concentrations reported on page 12 (lines 371, 372, and 383, N_CN,20 = 970, N_acc = 850, and N_rBC = 280 particles/cc) the fractions I derive are 0.29 (using N_CN,20) and 0.33 (using N_acc). As a rough check, derived values from figure 4 support the 30% fraction vs 40%.

[A1.7] Within individual plumes, we consider the enhancement ratio $\Delta N_{rBC}/\Delta N_{CN,20}$ to be more meaningful than the fraction $N_{rBC}/N_{CN,20}$. Moreover, N_{rBC} equals 0 does not imply that $N_{CN,20}$ is also 0. For avoiding confusion between enhancement ratio and fraction, we clarified this in section 2.2 (L224-228):

The rBC enhancement ratio relative to CO ($En_{RBC,M} = \Delta M_{rBC}/\Delta c_{CO}$, where Δ is the difference between the concentration of the species in the plume and in the background atmosphere) was obtained by applying a bivariate fit to the rBC and CO correlation within individual pollution plumes. *Analogously, CCN and rBC enhancement ratios relative to total CPC particle counts ($\Delta N_{CCN,0.5}/\Delta N_{CN,20}$ and $\Delta N_{rBC}/\Delta N_{CN,20}$) were obtained by applying a bivariate fit between the respective quantities.*

For consistency, we include a panel in Fig. 5 for estimating the activated fraction $\Delta N_{CCN0.5}/\Delta N_{CN,20}$ for both the UPL and BB plumes using the regression method. Note that, from comment 1.4, we used a new flag for determining the BB interval, and therefore, the data points have been updated in Fig. 5.

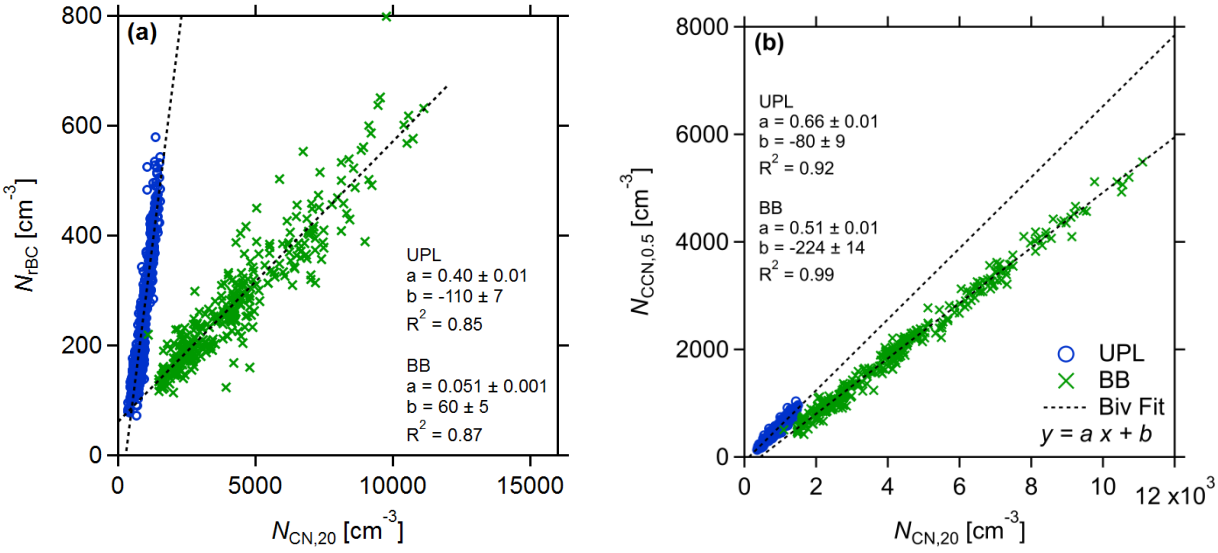

Figure 5. *Correlation between **(a)** rBC particle number concentrations (N_{rBC}) and total aerosol ($N_{CN,20}$); and between **(b)** CCN at S = 0.5 % ($N_{CCN,0.5}$) and total aerosol ($N_{CN,20}$) in the UPL (blue) and in the fresh biomass burning plume (green). The dashed lines are bivariate linear regressions applied to the data sets.*

Following the modifications in Fig. 5, we have changed the sentences:

(L377-379): *The CCN concentrations at S = 0.5 %, $N_{CCN,0.5}$, show a maximum within the UPL with $N_{CCN,0.5}$ = 560 ± 180 cm^{-3} as well as a high CCN fraction, $f_{CCN,0.5}$ = 56 ± 9 % (Fig. 3d).*

(L384-386): *The ratio $\Delta N_{rBC}/\Delta N_{CN,20} \approx$ 40 % in the UPL is much higher than $\Delta N_{rBC}/\Delta N_{CN,20} \approx$ 5 % in the fresh BB plume (Fig. 5a).*

(L410-416): *Note that, despite the higher $\Delta N_{rBC}/\Delta N_{CN,20}$ in the UPL compared to the fresh BB (Fig. 5a), the UPL shows higher CCN activated fraction ($\Delta N_{CCN,0.5}/\Delta N_{CN,20}$ = 66 %, Fig. 5b). The high CCN efficiency is likely due to internal mixing of rBC with sulfate, nitrate, and highly oxygenated organic aerosol. These findings, in combination with the UPL's large geographic*

extent, suggests that it represents an aerosol and CCN reservoir of particular significance for the Amazonian cloud cycling and rainfall formation – i.e., cloud droplet formation and growth.

(L725-727): Despite the large fraction of rBC, the aerosol in the UPL appeared to be very CCN efficient due to internal mixing of rBC with sulfate, nitrate, and oxygenated organic aerosol, with *~70 %* of particles activated at S = 0.5%.

[R1.8] Page 3, Line 73 -75. The authors are reminded that semi-transparent coating also alter BC optical properties.

[A1.8] For clarity, we have added the word "semi-transparent" to the sentence (L85-89, see also comment A2.7): *The formation of non-absorbing or semi-transparent coatings on the BC cores changes the particles' optical properties (Fuller et al., 1999; Moffet and Prather, 2009; Pokhrel et al., 2017; Schnaiter, 2005; Zhang et al., 2015) as well as their ability to act as cloud condensation nuclei (CCN) (Laborde et al., 2013; Liu et al., 2017; Tritscher et al., 2011), which influences their atmospheric transport and lifetime.*

[R1.9] Page 5, line 138-139. The authors refer to the African latter as pollution, yet their discussion centers on biomass burning. Pollution tends to suggest an anthropogenic contribution. It is suggested that the authors that consider changes "pollution" to biomass burning or something that describes the layer as containing BB emissions.

[A1.9] Please refer to [A1.1] where we have also addressed this comment.

[R1.10] Page 7, 209- 211. The authors might want to examine the paper by Adachi et al., (AST, 52, 46-56, 2018) where these authors present data from a mid-latitude wildfire that exhibit thermal stability well above the 250 C employed in the TD. How would increased thermal stability alter conclusions derived in the present analysis?

[A1.10] The study by Adachi et al. (2018) discusses how aerosol particles react to high temperatures (up to 600°C) based on ambient samples collected from agricultural biomass burning. Specifically, they show that some organic species or tar balls do not completely vaporize after being heated to 600°C. From the data set presented in our study, we can expect some bias between the CPC counts after the thermodenuder (which heats our aerosol samples up to 250°C) and the AMS, which relies on vaporizing aerosol particles for measuring the mass spectra. The thermal stability of organics in BB plumes could be responsible for the minimum in the volatile fraction (f_{vol}) observed within the UPL, which coincides with a minimum in the ultrafine fraction (f_{fine}). Moreover, the AMS results can underestimate the concentration of organics. However, this won't interfere with the main conclusions of the present study. In order to make it clear for the reader, we have included the following sentence (L401-403): *Note that the thermal stability of some organic species and tar balls in BB plumes can lead to an underestimation of the $f_{org,M}$ measured by the C-ToF-AMS (Adachi et al., 2018).*

[R1.11] Page 8, line 250-252. In deriving the EnRbc ratio, is the CO background corrected? I assume so, but this should be explicitly stated.

[A1.11] Yes. For the long-term measurements at the ATTO site, we used weekly 5th percentiles of CO and BCe measurements as background values, which were subsequently used for calculating the daily EnR_{BCe} (Fig. 12g of the revised manuscript). In order to make it clear, we have added:

(L249): *The 5th percentiles of the BC_e and CO measurements of the corresponding week were used as background values.*

[R1.12] Page 11, line 337. Please be consistent with respect to referencing black carbon. Within the SP2 community, the "black carbon" that is detected by this instrument is referred to as rBC (refractory black carbon). Indeed, in many places the authors refer to "rBC". To help minimize confusion, please be consistent and use only one term.

[A1.12] Thanks for noting that. We have used the term rBC only when referring to SP2 results, and BC when generally speaking about black carbon. We have further modified this paragraph in response to the Referee #2, please also refer to comment R2.14.

[R1.13] Page 12, 389- 390. The authors write "The high frBC further agrees with the pronounced brownish color of the visually observable layer in Fig. 2." This is a very misleading sentence. The brownish color could easily be due to brown carbon (BrC). Which, given the fact that the HALO payload included a three-wavelength PSAP, could be readily calculated via the Angstrom absorption exponent (AAE).

[A1.13] To avoid confusion, we changed the sentence to (L386): *Visually, the dark color of the layer observable in Fig. 2 corresponds with the high rBC fraction.*

[R1.14] Page 21, line 678-681. As highlighted above, the HALO 3-wavelength PSAP would provide insight into the presence of BrC.

[A1.14] Unfortunately, the PSAP data set could not be included on this study, as outlined in A1.2. But we will keep it in mind for further studies including the ACRIDICON-CHUVA data set. Please also refer to comment [A1.1].

[R1.15] Page 37, Table 2. Please explicitly cite the wavelength used to derive the SSA (637 nm).

[A1.15] Done.