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 #2

Received: 1 Nov 2019

General comment:

This paper describes the influx of African smoke to the Amazon during the dry season and discusses the implications of this transport for the Amazon. This work is novel and of interest to the aerosol community. I do recommend this paper for publication; however, I suggest that the authors revise the current manuscript, which is too long and detailed in its current form.

We thank the referee for the critical evaluation of the manuscript and for the constructive suggestions.

Major Comments:

[R2.1] In its current form, this paper is well-written but has so many details, that I found myself losing the main point of a paragraph or an entire section. I also found that a lot of the major findings were either buried in the middle of a section or at the end of a very long discussion that could have been cut in half. I list below a few suggested areas to tighten up this paper but encourage the authors to re-evaluate their paper as a whole and determine areas that could go into the supporting information.

[A2.1] Thanks for this critical feedback. As a response to the referee comment, we have revised the manuscript carefully in order to make the text and flow of arguments (wherever possible) shorter and more concise. We think that this helped to clarify certain aspects, such as the emphasizing the key findings of the study. Note however, that your request to generally shorten the manuscript and to reduce the discussions is somewhat in conflict to the overall recommendation of referee #1, who requested to rather include further data sets and analyses that (in her/his view) are missing (refer to comments R1.1 to R1.6 by referee #1). Ultimately, we tried to account for the general criticism of both referees by (i) generally streamlining and shortening the text wherever appropriate and (ii) adding some more information to the analysis that was explicitly requested in order to strengthen the manuscript.

[R2.2] This paper has too many acronyms to keep track of. I found myself having to go back to the acronym table a lot, which made the paper difficult to read. I suggest that the authors try to reduce the number of terms and instruments discussed to focus only on the most important ones for this story and to place the rest of the measurements in the supporting information.

[A2.2] We agree and revised the entire manuscript in order to reduce the number of acronyms wherever possible/appropriate.

[R2.3] I felt that the abstract and introduction promise that the focus of the paper will be on radiative impacts but those impacts weren't as well-emphasized in the results. I suggest restructuring the conclusions section to include a brief overview of the findings then focus on the radiative impacts as an extension of their findings.

[A2.3] Actually, the radiative influence of the observed pollution layer over the Atlantic is rather a side aspect of the study. We added it, envisioning that it may be of interest to a certain fraction of the readers. Moreover, many recent studies are dedicated to estimate the radiative impacts of the African BB layer over the South Atlantic (e.g. Deaconu et al., 2019; Denjean et al., 2019; Lu et al., 2018; Mallet et al., 2019; Meyer et al., 2013). While most of these studies focus on the

eastern Atlantic Ocean, here we want to bring the attention to the change on the radiative forcing towards the western Atlantic Ocean. Moreover, in order to calculate the direct radiative forcing of aerosol particles at the top of the atmosphere (DRF-TOA), we have used the multi-year and vertically-resolved aerosol extinction coefficients derived by CALIPSO. These results integrate the third large part of the paper, which shows the remote sensing results for different longitudinal bands over the Atlantic and the calculated radiative forcing. Accordingly, we have modified the *summary and conclusions* section in order to address the referee comment (L739):

Based on the remote sensing data, we further calculate the DRF-TOA exerted by the pollution layer as a function of longitude. We found that the aging of the plume leads to a change in the DRF-TOA from a positive (warming) to a negative (cooling) effect as it moves westwards over the Atlantic.

Specific Comments:

Abstract:

[R2.4] Lines 36-40: This sentence is far too long and dense. I suggest breaking it up.

[A2.4] Thanks for pointing this out. We modified the paragraph as follows (L38-42): Black carbon (BC) aerosols are influencing the Earth's atmosphere and climate, but their microphysical properties, spatiotemporal distribution, and long-range transport are not well constrained. *This study presents airborne observations of the transatlantic transport of BC-rich African biomass burning (BB) smoke into the Amazon Basin using a single particle soot photometer (SP2) as well as several complementary techniques. We base our results on observations of aerosols and trace gases off the Brazilian coast onboard the research aircraft HALO during the ACRIDICON-CHUVA campaign in September 2014.*

[R2.5] The abstract should include a very brief clause or sentence describing the most relevant instruments used.

[A2.5] Thanks – done. Please refer to comment [2.4], where we have also addressed this point in our response.

[R2.6] Lines 46-47: I had a hard time understanding this part of your sentence.

[A2.6] We modified this sentence as follows (L47-53): *The aged smoke is characterized by a dominant accumulation mode, centered at about 130 nm, with a particle concentration of N_{acc} = 850 ± 330 cm^{-3}. The rBC particles account for ~15 % of the submicrometer aerosol mass and ~40 % of total aerosol number concentration. This corresponds to a mass concentration range from 0.5 to 2 µg m^{-3} (1^{st} to 99^{th} percentiles) and a number concentration range from 90 to 530 cm^{-3}. Along with rBC, high c_{CO} (150 ± 30 ppb) and c_{O3} (56 ± 9 ppb) mixing ratios support the biomass burning origin and pronounced photochemical aging of this layer.*

Introduction:

[R2.7] I suggest condensing the first two paragraphs down to half a paragraph. There is a lot of detail that is not necessary.

[A2.7] We agree. Since the radiative effects of the pollution plumes are not the central focus of the manuscript, we condensed the two first paragraphs into the following one (L69-89):
Biomass burning (BB) in the African and South American tropics and subtropics represents a globally significant source of atmospheric aerosol particles and trace gases (Andreae, 1991; Andreae et al., 1988; Barbosa et al., 1999; Ichoku and Ellison, 2014; Kaiser et al., 2012; Reddington et al., 2016; van der Werf et al., 2017). A major constituent of BB smoke is black carbon (BC), which is co-emitted along with organic aerosols and inorganic salts in proportions that depend on the fuel type and fire phase (Allen and Miguel, 1995; Andreae, 2019; Andreae and Merlet, 2001; Jen et al., 2019; Levin et al., 2010; Reid et al., 2005). The BC aerosol is a key component in the climate system as it significantly influences the Earth's radiative budget through the so-called direct, semi-direct, and indirect aerosol effects (Bond et al., 2013; Bou-

cher et al., 2016; Brioude et al., 2009; Koch and Del Genio, 2010; Stocker et al., 2013). Recent studies have classified BC as the second largest contributor to global warming and estimated its direct radiative forcing as high as +1.1 W m^{-2}, with 90 % uncertainty bounds spanning from +0.17 to +2.1 W m^{-2} (Bond et al., 2013 and references therein). This large uncertainty arises from our poor understanding of the BC microphysical properties and its spatiotemporal distribution in the atmosphere (Boucher et al, 2013, Andreae and Ramanathan, 2013). During their typical atmospheric lifetime of several days, BC particles undergo photochemical aging, creating internally mixed BC aerosols via the condensation of low and semi-volatile compounds, coagulation, and cloud processing (Bond et al., 2013; Cubison et al., 2011; Konovalov et al., 2017, 2019; Schwarz et al., 2008; Willis et al., 2016). 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.

[R2.8] I also suggest adding a sentence or two regarding the impact of aging on the direct effect. It has been shown in several studies that coatings can greatly alter the radiative properties of black carbon [Moffet and Prather, 2009].

[A2.8] This comment has been also addressed in the comment [2.7]. Please refer to L85 "*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.*

[R2.9] Lines 114-118 can be cut as they do not add much to the introduction.

[A2.9] We agree with that. Therefore, we have removed the first sentence completely: "Moreover, the arrival of a plume of remarkably sulfur-rich aerosol particles in the central Amazon in September 2014 has been traced back to the LRT of volcanogenic emissions from eastern Congo (Saturno et al., 2018a)". Also, we have moved the second sentence into the caption of Fig. 7 of the revised manuscript: "For general illustration, animations of the Goddard Earth Observing Model (Version 5, GEOS-5) show that aerosol particles are transported efficiently from Africa to South America and to a lesser extent from South America to Africa (Colarco et al., 2010; Yasunari et al., 2011)."

[R2.10] The sentence on lines 121-122 is not clear to me.

[A2.10] In order to make it clearer, we have substantially reduced and modified the paragraph (L106-119): *Several studies have found that the long-range transport (LRT) of long-lived species from Africa plays a major role for the Amazonian atmospheric composition. The transport of dust from distant sources into the heart of the Amazon Basin was first observed in 1977, although Africa was not identified as the source region at the time (Lawson and Winchester, 1979).* Subsequently, the plume-wise LRT of African dust and smoke during the Amazonian wet season has been well documented (Ansmann et al., 2009; Baars et al., 2011; Barkley et al., 2019; Moran-Zuloaga et al., 2018; Swap et al., 1992; Talbot et al., 1990; Wang et al., 2016). *The LRT of aerosols occurs also during the Amazonian dry season, when smoke from the intense African BB plays a substantial role.* The earliest observations of such pollution layers in the free troposphere over the Brazilian coast can be found in ozone (O_3) soundings made from Natal, on the east coast of Brazil (5.8º S, 35.2º W), where mixing ratios of ~70 ppb were measured with a maximum *in the month of* September (Kirchhoff et al., 1983; Logan and Kirchhoff, 1986). These measurements were continued over a ten-year period (1978-1988), confirming the climatological presence of a tropospheric O_3 maximum over the Brazilian coast, centered at the 500 hPa pressure level and peaking in the September-October period (Kirchhoff et al., 1991).

Methods:

[R2.11] I strongly suggest reducing the text in this section. For example, the gas phase data does not play a major role compared to the aerosol data. I suggest mentioning what was measured in the gas phase then directing the reader to the SI for more details on the instrumentation. This will also reduce the number of acronyms that the reader must memorize.

[A2.11] We understand your point, but have a somewhat opposing opinion here. In our view, every aspect that is prominently shown and discussed in the results or discussion part should also have an appropriate visibility in the methods section. We regard it as more confusing than helpful to distribute the information on methods and data analysis between main text and supplement. We critically checked the trace gas paragraph, did few improvements here and there, and finally came to the conclusion that this section is concise and rather easy to understand. Therefore, we prefer to keep the paragraph in the section 2.1, which is dedicated to describe the HALO data sets that we made use of.

[R2.12] I also suggest placing details about AIRS data and AERONET in the SI. The AIRS and AERONET data were not as integral to the study as other methods.

[A2.12] For the same reason stated in comment [2.11], we prefer to kept this information in section 2.4 of the methodology. However, we have substantially reduced the length and content of these paragraphs by removing (too) detailed information about remote sensing techniques, which are not the central focus of the study. Therefore, the following changes were made in the section (L254-268):

2.4. Satellite and ground-based remote sensing

In this study, we used the vertically-resolved extinction coefficients (LIDAR Level 2 Version 3 Aerosol Profile product with 5 km horizontal resolution) of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar system, onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite (Winker et al., 2009). The CALIPSO al-

gorithms detect and classify aerosol layers based on their observed physical and optical properties into the subclasses: polluted continental, biomass burning (smoke), desert dust, polluted dust, clean continental, and marine aerosol (Omar et al., 2009).

To obtain CO concentrations between the 400 and 600 hPa pressure levels, we used the Atmospheric Infrared Sounder (AIRS) onboard the NASA Aqua satellite available from the Giovanni online data system (https://giovanni.gsfc.nasa.gov/giovanni/, last access on 13 June 2019). Daily averages of aerosol optical depth (AOD) at 550 nm with original grid resolution of 1^o x 1^o was obtained from Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol products from the NASA Terra and Aqua satellites (Remer et al., 2005). Finally, AOD at 500 nm (level 2.0) was obtained by direct sun measurements in Ascension Island (7.976^o S, 14.415^o W), using the CIMEL sunphotometer of the AErosol RObotic NETwork (AERONET, https://aeronet.gsfc.nasa.gov/, last access 12 Mar 2019) (Holben et al., 1998).

We further streamlined the section 2.1 (L147-156):

2.1. The ACRIDICON-CHUVA campaign

The data presented here were obtained during flight AC19 of the ACRIDICON-CHUVA aircraft campaign (Machado et al., 2017; Wendisch et al., 2016), which took place over the Atlantic Ocean and the Amazon Basin on 30 September 2014. The main objective of ACRIDICON-CHUVA was to study the interactions between aerosol particles, deep convective clouds, and atmospheric radiation using a broad set of instruments for airborne observations of aerosol physical and chemical properties, trace gases, radiation, and cloud. The measurements were conducted onboard the German HALO (High Altitude and LOng range) research aircraft, operated by the German Aerospace Center (DLR), covering a wide geographic area of the Amazon Basin and probing different pollution states by means of highly resolved atmospheric profiles (altitudes up to 15 km).

Results:

[R2.13] I suggest placing Figure 2 in the SI.

[A2.13] Evidently, this figure is not really essential for the key findings of the paper, however, it also does not really disturb the flow of reading as it is rather 'light'. We think that figure gives the reader a good visual impression of the pollution layer and, thus, aids the perception of the numbers presented in the manuscript. It further visually underlines how pronounced the layers actually are. Since the supplementary material typically receives significantly less attention, we prefer to keep Fig. 2 in the main text.

[R2.14] Sections 3 and 3.1 are too long and detailed. This information dilutes the major findings regarding the transport of biomass burning and its impact on the optical and cloud nucleating properties of Amazonian aerosol. I suggest condensing the material from 330-394 down to a paragraph if possible and placing much of this information in the SI.

[A2.14] The main aspect of the study is the *in situ* observation of the pollution layer at the South American coast. A unique aspect of our study is the broad spectrum of techniques in the aircraft that were available to probe the layer in great detail. Accordingly, a systematic and detailed summary of the key numbers, concentrations, and properties is the foundation for other aspects of the analysis. Accordingly, we strongly prefer to keep some aspects as detailed as it is. However, after revising the section, we performed some changes in order to make it more concise:

We moved the following sentence to the caption of Fig. S3 of the revised manuscript: The sounding shows similar tropospheric stratification as presented in Fig. 3. The first layer (top around 1000 m) is associated with the boundary layer, the second (top around 3200 m) is related to the shallow clouds top and the third one (around 5000 m) is the large scale inversion.

(L331-348): *The flight track of AC19 followed the direction of the Amazon River from Manaus towards the coast and included cloud-profiling maneuvers over the Atlantic Ocean (Fig. 1). A*

remarkable observation during AC19 was the strong stratification of the troposphere over the ocean with vertically well-defined and horizontally extended layers, with varying degrees of pollution (Table 1). Based on contrasting aerosol concentrations, size ranges, and composition, we distinguished an upper and a lower pollution layer (UPL and LPL) with a horizontal clean air mass layer (CL) in between. The layers were discernible visually from the aircraft cockpit (Fig. 2).

In this study, we present the tropospheric stratification for the lowest 5 km of the atmosphere, focusing primarily on aerosol and trace gas properties within the UPL and contrast them with the properties of the CL, LPL and the marine boundary layer (MBL). Aerosol properties in the upper troposphere during ACRIDICON-CHUVA have been characterized in previous studies (Andreae et al., 2018; Schulz et al., 2018). Upon ascent and descent, the UPL was probed six times at offshore locations[1], right before it reached the South American continent, and two times onshore ~200-400 km from the coast line (blue squares in Fig. 1). The eight UPL penetrations were several hundred kilometers apart from each other, underlining the large horizontal extent of the layer. Later on the route back to Manaus airport, we observed an active fire plume northwest of Belém (green square in Fig. 1, photo of plume in Fig. S3). This plume was probed at ~1 km above the fire and is expected to be only a few minutes old. Below, selected aerosol properties in this local, fresh BB plume are contrasted with the UPL aerosol properties.

(L356-358): Generally, the profile of θ indicates rather stable conditions along the entire profile with the UPL being centered at ~3.5 km altitude. *For comparison, radiosonde profiles at Belém airport for the same day as flight AC19 are shown Figure S4.*

[R2.15] Lines 442-443: It is not necessary to explain that O3 is a secondary pollutant produced photochemically in BB plumes.

[1] Note that we count two passages through the layer over the Amazon River delta as offshore.

[A2.15] In response to your comment, we shortened and streamlined the entire paragraph substantially, as follows (L425-428): *The ozone as secondary pollutant also presents a maximum within the UPL (c_{O3} = 56 ± 9 ppb) and appears to be anti-correlated with NO (c_{NO} = 0.10 ± 0.02 ppb). Therefore, the fact that O_3 and NO_y (c_{NOy} = 2.5 ± 0.8 ppb) are strongly enhanced in the pollution layers, reflects the photochemical age of the plume.*

[R2.16] Lines 447-467: This paragraph could go in a discussion section.

[A2.16] All the discussions in this manuscript have been placed along with the results. For consistency, we would prefer to keep in this section the discussion on further airborne observations of BB layers near the African coast. This paragraph is also a link to the next section where we look more in detail the evolution of BB layers over the South Atlantic Ocean.

[R2.17] Lines 470-473 belong in the introduction.

[A2.17] We moved the sentence to the introduction section (L90) as suggested: The Amazonian atmosphere is strongly influenced by the yearly *north-south oscillation of the intertropical convergence zone (ITCZ) (Andreae et al., 2012; Martin et al., 2010; Pöhlker et al., 2019),* which causes a pronounced seasonality in aerosol concentrations (e.g., BC and CCN) and other aerosol properties (e.g., single scattering albedo) (Roberts et al., 2001; Roberts, 2003; Martin et al., 2010; Artaxo et al., 2013; Rizzo et al., 2013; Andreae et al., 2015; Pöhlker et al., 2016; Saturno et al., 2018b).

[R2.18] The authors should note that the methodology shown in Figure 7 is similar to the methods used in [Barkley et al., 2019] to identify similar plumes.

[A2.18] We included (L562): *More examples of similar layers over the Atlantic Ocean in September 2016 are shown and discussed in Barkley et al. (2019).*

[R2.19] Lines 591-606 are really important for understanding the implications of your work. I strongly suggest either giving this paragraph its own section or placing it in a discussion section.

[A2.19] Please refer to comment A2.3. The DRF-TOA is rather a side aspect and is an extension of what we can get from satellite derived aerosol extinction coefficients.

References:

Barkley, A. E., J. M. Prospero, N. Mahowald, D. S. Hamilton, N. Mahowald, K. J. Popendorf, A. M. Oehlert, A. Pourmand, A. Gatineau, K. Panechou-Pulcherie, P. Blackwelder, and C. J. Gaston (2019), African biomass burning is a substantial source of phosphorus deposition to the Amazon, Tropical Atlantic Ocean, and Southern Ocean, Proceedings of the National Academy of Sciences of the United States of America, DOI: 10.1073/pnas.1906091116.

Moffet, R. C., and K. A. Prather (2009), In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates, Proceedings of the National Academy of Sciences of the United States of America, 106(29), 11872-11877.