Reviewer #1

Unfortunately, the trajectory analysis does not directly compare the same altitudes between the three periods, making it difficult to determine whether the transport pathways truly are distinct between these three periods. In addition, the result that elevated smoke in period 3 is of South American origin may be a misinterpretation of the ensemble HYSPLIT trajectories.

Back trajectories were calculated based on the layers of aerosols observed by lidar, which are different from one period to another, the objective being to identify their origin. For this reason, back trajectory levels change from one period to the next. In response to the reviewer's remark, we have introduced the back trajectories associated with the 3 layers of aerosols observed by lidar in the free troposphere for the 3 periods (see the new Figure 13 where we show 9 sub-figures (3 height ranges x 3 periods) instead of 3 in the original version of the manuscript).

The major issue with the manuscript is the interpretation of the HYSPLIT trajectories, and in particular the interpretation of the result that elevated smoke from South America is of major importance for radiation over the southeast Atlantic.

This is indeed one of the important points of the article, but the general characterization of aerosol plumes and their high variability over time is also important. However, we agree that the part concerning the importance for radiation of the southeast Atlantic needs to be backed with a bit more evidences. Therefore, we have put more nuances in our conclusions.

Section 5 and Figure 11 cannot be interpreted as showing differences in transport pathways between periods, as the authors attempt to do, because the comparisons are “apples-to-oranges” in that the trajectories are initialized at very different altitudes. Even if the circulation had been perfectly steady throughout all three periods, vertical wind shear alone would lead to apparent differences using this methodology. The correct “apples-to-apples” comparison would be to compare trajectories at the same altitudes for all three periods.

One solution could be to divide Figure 11 into three figures: one with the trajectories from 1500-3000 m for all three periods, another from 3000-4500 m, and a third from 4000-6000 m. 2000 m is a fairly large vertical area to lump together, so I would further suggest subdividing from the original three altitude bins. For instance, binning by 1 km increments (1.5-2.5 km, 2.5-3.5 km, 3.5-4.5 km, 4.5-5.5 km, 5.5-6.5 km) could be more illustrative. These are all just suggestions: there are many different ways to expand the analysis to make more meaningful comparisons between the periods.

As mentioned above, we decided to calculate the back trajectories for the aerosol layers observed by the lidar. The calculations were done every 500 m on the vertical over Henties Bay in the original manuscript and are now done every 250 m in the revised version of the paper, for the sake of capture all the possible transport pathways. Figure 11 is representative in the sense that it uses the probability of air mass passages at each point and that this calculation is performed over each period for more than 500 back trajectories. This is the choice we had to make in order to synthesize the information and not present too many back trajectories. This aspect is better explained in Section 5. The reviewer will find in the new version the results over the 3 periods for the same altitude ranges: [1500 3000[, [3000 5000[and [5000 6000[m

AMSL. Original Figure 11 (now Figure 13) was therefore effectively divided into 9 sub-figures. The text has been revised accordingly.

Another concern relates to the interpretation of the trajectory ensembles. Figure 11c) iscurrently being interpreted by the authors as showing that some of the elevated smoke is coming from South America. However, there appear to be a significant number of trajectories that are limited to re-circulations around the African continent. Are all of the re-circulating trajectories from below ~5 km? If not, then the ensemble is actually telling us that it is plausible that the air came from South America but equally plausible that it re-circulated locally. In a similar vein, Figure 12 shows that South America is a plausible source for the elevated smoke observed in period 3 but certainly does not prove that this must be the source.

The reviewer is right, this aspect is poorly explained in the text and is not visible in old Figure 11. This aspect is now better described, mainly for the period P_3 when we have separated what happens under 6 km AMSL and above.

Even if a more rigorous evaluation of the trajectories does confirm that there are meaningful circulation differences between the three periods (which is extremely likely) and that the elevated plume in period 3 is very likely of South American origin (which I am open to but more skeptical of), the significance of these results is either inflated or incompletely explained. In the conclusion, the idea that there may be some influence from elevated South American smoke is deemed "of paramount importance" for the region in the context of aerosol-radiation and aerosol-cloud interactions. However, even accepting the hypothesis that the elevated smoke is South American in origin, it still only comprised ~10% of the column loading of aerosol and is too high in altitude to plausibly influence low cloud microphysics or have relevant semi-direct effects (unless high clouds occur more frequently over Henties Bay than I assume). Given the pre-existing uncertainties in aerosol loading, vertical distribution, and optical properties over the region, I find it hard to argue that an occasional 10-15% contribution to the direct radiative effect is "fundamental."

Yes, the contribution of aerosols from South America should not represent more than 10-15% of the total aerosol load for mainly 2 days, on 6 and 7 September. We agree that they will not contribute significantly to interactions with low-level clouds and we have not presented it in this way. This aspect is now better specified in the text where Figure 2b was introduced, which gives the temporal evolution of AOTs at 355 nm per layer. The aerosols in the layer [5000 6000[m will nevertheless likely have a radiative impact that has yet to be correctly estimated, but this is not the subject of this paper. It is also necessary to see how often this kind of phenomenon can occur, in relationship with cut-off lows. In order to comply with the referee's comment, we have agreed to be more cautious in the conclusions regarding the "fundamental" aspect of the direct radiative effect related to aerosols from South America.

Page 2, Line 38: What metric are you using to determine that southern Africa is the "most important source" of biomass burning aerosol?

Our metric is the AOT. This has been now specified.

Page 2, Lines 49-51: There is now an accumulation of evidence from the LASIC (e.g., Zuidema et al. 2018, GRL) and ORACLES (e.g., Diamond et al. 2018, ACP) campaigns that the MBL in the southeast Atlantic often contains quite a bit of smoke. In particular, the ORACLES campaign in September 2016 observed some very smoke-polluted MBLs not far from the Namibian coast. The MBL-FT dichotomy in this sentence is a bit oversimplified.

The references have been added and we have added the possible presence of BBA within the MBL.

Page 2, Line 53: The papers from Costantino & Bréon do not show that clouds in this region are particularly sensitive to aerosol increases — indeed, their aerosol cloud interaction parameter estimates are well within the range of the other literature they cite. They do show an apparent inverse relationship between cloud effective radius and aerosol index when smoke is near cloud tops, which is consistent with a widespread Twomey effect. If the point here is more that marine cloud radiative properties should be particularly sensitive to aerosol increases, the paper from Oreopoulos & Platnick (2008) cited below may be a more appropriate reference, among other suitable choices.

The reference has been changed. Thank you for suggesting this.

Page 2, Line 56-60: The vertical distribution is also incredibly important for indirect effects, not only semi-direct effects. Without contact between the plume bottom and cloud tops, smoke cannot entrain into the MBL and influence cloud microphysical properties. The Costantino & Bréon papers would be good to cite here. Diamond et al. (2018) and Painemal et al. (2014) may also be relevant.

Agree. We have added this information. Thank you for suggesting this.

Page 2, Line 72: What does "aerosol activation" mean in this context?

It is not the "aerosol activation" but the activation of their sources. The term "activation" has been replaced by "emissions".

Page 3, Line 89: Namib desert, not "Namibia" desert

The correction has been done.

Page 3, Line 91: Please define "ALS."

The information has been done.

Page 3, Line 93: Please define "LEANDRE."

LEANDRE is now defined.

Page 3, Table 1: Please explain what the "X" and "-" symbols mean in the table caption.

X was meant to indicate when coincident AERONET and lidar were available. X has been replaced by "yes" or "No".

Page 5, Figure 1: In the caption mention that the Henties Bay and Walvis Bay locations are marked by orange dots.

Thanks for picking this up. This has been added.

Page 8, Line 200: How does the non-colocation of the MODIS area average and Henties Bay affect your results, if at all?

The accuracy of MODIS data is much better over the sea, we have insisted on this in both Section 2.4.2 and Section 3.1. Henties Bay is a coastal site influenced by sea breeze, so we have considered it more representative to take an average of the AOTs offshore the site. Local aerosol production can affect the measurements, especially in the event of dusts being lifted. Nevertheless, from observations of depolarization lidar, we have seen very few such situations.

Page 8, Line 205: Move "only" to after "aerosols are."

The correction has been done, the section 3 has been modified.

Page 9, Lines 251-254: This period of disagreement between the observations and CAMS may be worth exploring further. What is the circulation like then? Does it seem like the FT air is being sourced from a non-biomass burning affected area, or is there perhaps loss of aerosol occurring (e.g., precipitation scavenging) that CAMS may not be capturing?

First, we highlight that no precipitation event was recorded during the field campaign, so that we can exclude any CAMS misrepresentation of wet deposition processes. Similarly, the contribution from non-biomass aerosol can be excluded as well, because CAMS simulates very low dust AOT during the campaign, with only a peak on 3-5 Sep. We highlight that CAMS total AOT is essentially organic matter AOT. We also highlight that mid-tropospheric circulation was characterised by: 1) on 2 Sep, a low pressure system localised off-shore of Henties Bay, juxtaposed to a high pressure system localised over South Africa, resulting in a small river of smoke descending along the coast (see figure); 2) on 7-8 Sep, an elongated high pressure dominating over the continent, resulting in a channelling of the smoke from north-west. Therefore, we point out that, given the features of the smoke transport over the Henties Bay region, even small differences in the simulation of the weather conditions could lead to substantial differences in AOT for specific locations, especially when AOT values are rather low. These different aspects have been added in Section 3.1.

Page 9, Line 265: Why do you refer to the observations as biased with respect to CAMS? Couldn't the CAMS value be off? It might be helpful to explain why either estimate may be different than "truth."

Thank you for pointing this out. The sentence is misleading in this form, and it has been rephrased. We also expand the discussion on the discrepancies between CAMS and observations, so that now the paragraph reads:

" These discrepancies may be also explained by the coarse spatio-temporal sampling of the model, which is insufficient to highlight the sharp variation in AOT due to a very localized channelization during these 3 days. Note that no significant precipitation event was recorded during the field campaign, so that we can exclude any CAMS misrepresentation of wet deposition processes. Otherwise, CAMS simulations show that the AOT is essentially due to organic matter (i.e. biomass burning aerosols), the contribution from non-biomass aerosol can then be excluded as well. Hence, given the features of the smoke transport over the Henties Bay region, even small differences in the simulation of the weather conditions could lead to substantial differences in AOT for specific locations, especially when AOT values are rather low. On 2 September a minimum in AOT is observed by the sun photometer which is not reproduced by CAMS simulations (even though a local minimum in the CAMS AOT can be seen). During this day, mid-tropospheric circulation was characterised by a low-pressure system localised offshore Henties Bay, juxtaposed to a high-pressure system localised over South Africa, resulting in a small river of smoke descending along the coast. On 7-8 September, the sun photometer- and MODIS-derived AOTs are larger than the one computed from CAMS. This could be related to the presence of unscreened optically thin clouds such as the ones observed in the ground-based lidar data on 8 September (Figure A2d) and/or to the heterogeneity of the meteorological field. Indeed, on 7-8 September, an elongated high pressure dominating over the continent, resulting in a channelling of the smoke from north-west.".

Page 9, Table 3: It would be helpful to explain what the uncertainty range is in the caption. Also, for the profiles encompassing a long period of time, how much of the uncertainty in average value comes from remote sensing uncertainties versus real variability over the time period?

This discussion has been added in the Appendix A. The uncertainty includes both the detection noise and the natural atmospheric variability.

Page 10, Table 3: How are these profiles divided? For instance, why are the 22/08 profiles divided at 1607?

As explained in the text, we average the profiles on the time laps without cloud cover. Hence, they are not systematically located at the same times. We try to keep as much information as possible for each day.

Pages 12-13, Figures 3-4: I don't understand why the figures are divided in this manner. Also, not all potential profiles from Table 3 appear to be plotted (e.g., it seems like there is GBL data for 22/08 from 1608-2400 that is not included). It might make more sense to divide this into a number of separate figures for each period.

Following the reviewer's remark, we have divided the figures by period in order to improve visibility (see Figure 3-7). We have now included all mean available profiles available from Henties Bay.

Page 14, Figure 5: The color scale here makes the figure very difficult to read. Perhaps using one that ranges from a very light color to a darker color for high AOT would both be more intuitive but also make it easier to read the wind markers.

The correction has been done.

Page 14, Line 294: What do you mean by "evolution"? That term may be misleading because you are not really showing changes over time but rather a time-space cross section following the aircraft.

Agreed. We have changed the sentence to "**Erreur ! Source du renvoi introuvable.**a shows the time-space cross section of the LNG-derived apparent aerosol backscatter coefficient (ABC) profiles at 532 nm along the Falcon 20 flight track in the morning…"

Page 15, Lines 307-309: I cannot find evidence for this statement in the Haywood et al. (2003) paper cited. Parmar et al. (2008) does establish that water vapor is emitted at fire sources along with carbonaceous species but does not claim that this could humidity an entire well-mixed continental boundary layer. Why would the high relative humidity not simply be characteristic of continental air whereas low humidity air be from subsiding tropical or midlatitude air that has been depleted of moisture via prior precipitation?

Agree, the reference to Haywood et al. (2003) has been removed.

We also agree that the RH values could be related to the characteristics of continental air masses. Nevertheless, large correlations between RH and aerosol loads between 850 and 700 hPa have been observed in this area and for the same time period (e.g. Daeconu et al., 2019). Nevertheless, we have softened our statement so to include the suggestion of the referee.

We have added 2 references:

Deaconu, L. T., Ferlay, N., Waquet, F., Peers, F., Thieuleux, F., and Goloub, P., 2019: Satellite inference of water vapor and aerosol-above-cloud combined effect on radiative budget and cloud top processes in the Southeast Atlantic Ocean, Atmos. Chem. Phys., accepted.

Clements, C. B., Potter, B. E. and Zhong, S.: In situ measurements of water vapor, heat, and CO2 fluxes within a prescribed grass fire, Int. J. Wildl. Fire, 15(3), 299–306, doi:10.1071/WF05101, 2006

Page 16, Line 342: In what way does elevated RH suggest the aerosol layer must be distinct?

A high variability of RH in the atmosphere generally means that we have different pathways of air masses. When combined with signatures on the vertical aerosol profile, it is a favourable element for the presence of different layers against altitude. However, back trajectories are required to confirm this. We agree that this is a bit of an overstatement at this stage. We have deleted the end of the sentence, i.e. "[…] which together with the AEC profile in Henties Bay, suggests the presence of a distinct aerosol layer above the main BBA layer, that is not seen in the AEC profile '1' (Figure 8b)".

Page 16, Line 362: In what way does the existence of non-negligible AEC values suggest that the aerosol has a different origin than the other aerosols observed?

Agreed. At this stage, there is not enough information for this to be concluded. Mention to the "different origin" has been removed.

Page 18, Figure 9: It would be helpful to make clear which profile corresponds to which dropsonde in Figure 8.

The correction has been done.

Page 19, Line 395: Do you mean "back-trajectory" instead of "retro-trajectory"?

Agreed, it is a mistake. The correction has been done.

Page 20, Line 447: What do you mean by "nebulosity"?

The nebulosity corresponds to the cloud cover or cloudiness. The correction has been done.

Page 22, Figure 12: I would suggest using whichever color scale you choose for Figure 5 here as well.

The colour palette has been changed.

Page 23, Lines 482-483: Couldn't high observed AOT also be due to aerosol humidification at high RH, versus thin clouds?

If there were wet aerosols, the lidar would have detected them as well. We think that they are more clouds that are not properly identified by passive detection.

Page 23, Lines 497-498: What do you mean by "trapped in the FT"?

We have changed the sentence: "…in relationship with the main transport regimes across the Atlantic Ocean".

Page 23, last paragraph in its entirety: This is very interesting, but not appropriate to introduce and discuss only in the conclusion. This could potentially be a great addition to the paper as a separate section with some new analysis of the meteorological fields to provide evidence for the claims made.

Agreed, this paragraph has been moved at the end of Section 5.