Reviewer #2

The argument that the South American aerosol layer is "distinct" is unconvincing and the conclusion that it is of significant importance for climate modeling is premature. Yet, aerosol transported from South America to Africa is still interesting enough to highlight, even without this. Better to be very straightforward about what is and is not known about it, to inform and motivate future study.

Agreed, we have tamed down some of our previous conclusions to account for this. For instance, the back trajectories analysis has been revised for the sake of clarity and we have also added Figure 2b to highlight the likely contribution of the South American aerosol layer.

Additionally, some lack of clarity makes it difficult to understand the details of the lidar retrieval; in particular the derivation of the lidar ratio, and the associated uncertainties. The lidar retrieval is subject to large uncertainties because of the underdetermined nature of the problem, but there is a lack of useful information about the size of the uncertainties or their impact on the conclusions.

The processing of the mean lidar profiles has been clarified in Appendix A. The main uncertainty sources have been presented and assessed: the statistical noise due to the shot noise and the atmospheric variability during the average time, and the uncertainty linked with the lidar ratio.

119 "between about 1 and 4 hours". Actually, several of these averages are 7 or 8 hours long, so this should be indicated correctly. Even 4 hours seems like quite a long time, considering the variability of aerosol in the region. Was there any need for filtering or any analysis to check for stationarity?

There is a low signal-to-noise ratio for individual profiles, especially during the day. For this reason, but also because the vertical structure of the atmosphere does not vary significantly, we have performed some averages over several hours. These averages were very constrained by the presence of clouds as explained in the text. The temporal variability of lidar profiles is now illustrated in Figure A2.

Section 2.3 The Bruneau et al. 2015 paper referenced at 135 is missing from the bibliography. I think I know which paper you mean, though, and it says this instrument has HSRL capability at 355 nm. If so, why does it require the retrieval procedure that's used for the ground-based elastic backscatter lidar? Even if I'm wrong about the HSRL, the appendix says that the lidar has a 355 nm channel. So then why is the 355 nm data not shown in figures 6b and 8b? (if it's available, it would be very helpful there!)

Yes, LNG indeed has HSR capability at 355 nm. Nevertheless, this channel did not work during the campaign and the 355 nm channel was very noisy and unusable. We use only the 532 channel. We have added the sentence in Section 2.3: "We only use the 532 nm channel because the high level of noise of the 355 nm channel". Hence, the lidar operated has a simple backscatter Rayleigh-Mie lidar. The reference to Bruneau et al. (2015) has been added.

Section 3. I found the organization of section 3 hard to follow. It goes back and forth multiple times between different subjects. Please consider reorganizing to make one paragraph about the overall AOT trend, one comparing P1 to P2, and another separate paragraph comparing P2 to P3.

We agree that this section is difficult to read because the elements are not grouped together. We have reviewed its structure and divided it into two parts for clarity: "3.1 Identification of periods from the total AOT" and "3.2 Aerosol vertical profiles". We have removed the MODIS figure that did not belong in this paragraph. However, we have added Figure 2b, which allows us to better monitor the temporal evolution of AOTs at 355 nm and better highlight the correspondences with AOTs at 550 nm.

215-218 statement that lidar ratios in the free troposphere "suggest the presence of terrigenous aerosols mixed with smoke. This is coherent with the polluted dust type inferred from the CALIOP observations." Is this circular reasoning? The Appendix suggests that only the lidar ratio in the PBL is retrieved and that the lidar ratio in the free troposphere is taken directly from the inferred lidar ratio in CALIPSO or CATS.

Agree, the sentence is poorly formulated and has been removed.

The presence of dust in the smoke layer isn't very consistent with the AEROCLO-Sa ground-based lidar measurements in Figure 3, which shows quite small values of the particle depolarization ratio within the smoke plume for all profiles.

It's not so small. In the presence of pollution aerosols, the PDR is rather between 1 and 2% because they are generally hydrophilic. For biomass burning aerosols, they are often mixed with dust that may have been raised by pyro-convection process. Values of ~ 5 to 10% are therefore likely for aerosol mixtures of biomass burning and dust aerosols.

Even if I'm misunderstanding how the constrained retrieval works on the ground based lidar, I believe it's likely that the retrieved lidar ratio in the free troposphere must have a large degree of uncertainty which probably precludes making a distinction between smoke and polluted dust, whereas the uncertainty on the particle depolarization ratio is likely to be quite reasonable. Does this conform to your analysis of systematic errors and uncertainties?

We have had a discussion on this uncertainty in the Appendix and now calculated the error due to the LR on the aerosol extinction coefficient (and AOT) and the particle depolarisation ratio (PDR). In addition, statistical noises have been added to the lidar profiles in Figures 3 to 7.

220 "being remobilized by pyroconvection and mixed with BBA before being transported aloft". How do you know these details about the origin of the dust? What evidence is there of pyroconvection?

We observe well mixed layers whose PDR remains substantially constant and above what is expected from pure biomass burning aerosols. One explanation is the simultaneous injection of dust during intense fires. This is only a hypothesis at his stage.

217 What does the citation Flamant et al. 1998 refer to here? Is this a typo? (Also, the paper is missing from the bibliography).

This refer to the LR value for marine aerosols. It is now made clearer in the text. The reference has been added.

232 "LR values in the FT evolving from 55 to 70 sr". I believe the lidar ratio in the free troposphere is assigned based on aerosol type that is somehow inferred from the CALIPSO and CATS aerosol type inferences. It would be better, then, to directly refer to a change in inferred aerosol type rather than an evolution of the lidar ratio, which implies more quantitative detail than is actually available.

Agreed, as explained in the text the LRs are derived from the CALIPSO and CATS aerosol typing. The aerosol typing is clearly identified in the text with the LR.

238 "LR values observed in the PBL". Please be specific about which retrieval you mean. The combined lidar-sunphotometer retrieval or the sunphotometer only retrieval from the previous sentence?

This is now better specified in the text. We have added the aerosol typing in brackets after the LR value. The aerosol typing is also given further in the text.

239 Again the presence of dust would be expected to be accompanied by an elevated particle depolarization ratio which is not evident.

As we explain, it is not pure dust particles, but a mix with other types of aerosols. The LR is then weighted against the ration between dust and other particles.

245-247 The relatively high particle depolarization ratio in the altitude region between the smoke plume and the PBL seems unlikely to be significant compared to its uncertainty. The systematic error of particle depolarization ratio increases dramatically for low backscatter ratios (see, e.g. Freudenthaler et al. 2016, Burton et al. 2015). If you have done a calculation of the expected systematic uncertainty and believe these to be significant, then that should be included to support your interpretation.

The uncertainties have been calculated and added on the profiles. As expected, they are generally more important when the AEC is low, but this is also a function of the signal to noise ratio. We reviewed the discussions in the Appendix and in the Section 3.2, which were indeed not enough clear.

Table 3 What is the number after the plus-and-minus sign?

This is the statistical noise due to the detection and the natural variability of the atmosphere. This is now indicated.

Table 3 Why is the lidar ratio consistently only a few different values? In the PBL, the retrieval is described as a constrained retrieval, so I expected it to be able to take a continuous range of values.

Our choice was to remain within the range of values proposed for CALIOP and CATS and these values allow us to find with enough precision the AOTs derived from passive instruments. With the uncertainties about the LR, little more can be expected.

Figure 3 Add error bars? I expect significant amount of variability due to the very long averaging and also a significant amount of systematic uncertainty. Even if only a coarse estimate of systematic uncertainty can be included, this would be valuable for interpreting the significance of features in the profiles.

The error bars have been added in Figures 3-7. They show that our discussion is appropriate.

316ff. Somewhere in this paragraph please state the distance between the profiles.

The distance between the 2 profiles (~100 km) is now mentioned in the text.

318 "operate at different wavelengths". Table A1 indicates that LNG also operates at 355 nm. Can you show 355 nm retrievals from LNG as well? It might help establish that there really is correspondence between the airborne and ground-based retrieved extinction and give better support that the differences are not due to the wavelength.

The LNG detection did not work properly at 355 nm during AEROCLO-sA. Hence, we have not used the data. Nevertheless, we agree that this is not mentioned in the manuscript, Table A1 just providing an overview of the LNG system. In the revised version of the manuscript, we are now mentioning that the LNG detection at 355 nm was not working nominally (Section 2.3) and hence the data not included in the analysis.

324 "the difference can be explained". There are multiple factors affecting this difference, including the approximately four hour time difference between the profiles, which has not been addressed. Any comment about the role of the time difference? In any case, soften the statmeent. Maybe "More important is the regional scale circulation"

We have added this information together with that on the wavelength difference, in the form: "there is a 4-hour difference between the aircraft profiles and the mean profile over Henties Bay". We also have softened the statement by using "may' instead of "can".

328-329. Does the CAMS model also show this difference between the aerosols over the land and ocean? You previously made an apparently conflicting point that CAMS indicates homogeneity in the aerosol around Henties Bay.

No, the CAMS model does not show this difference in the structure of the aerosol profiles between the land and the coastline.

335-336 "the structure of [profile 2] being coherent with the ground-based AEC profile". Not convincingly so. Also, this statement is apparently contradicted at line 347. Is this a typo?

Thanks for picking this up. Indeed, this is a typo. The sentence has been corrected.

340-341 I'm confused by the indiation of 20% RH above the BBA layer. First, is this referring to 9b or 9a? Either way, I see that it doesn't drop dramatically to zero like 8b, but I do not see 20% RH for any part of the profile above the strongest gradient (top of the BBA) and even the highest value I do see (more like 10%?) is only for a few hundred meters.

Agreed. This is a mistake, the RH above the BBA does not reach 20%, and indeed more like 10% (at most) for a few hundred meters. We are referring to Figure 9b, i.e. the profiles from the dropsonde released closest to Henties Bay. The sentence now reads:

"The maximum RH in the FT is ~55% and observed near the top of the BBA layer (Figure 11b), while small RH values (less than 10%) are seen above ~6 km AMSL It is worth noting the presence of a slightly enhanced RH layer between 5.5 and 6 km AMSL.".

341-342 "suggests the presence of a distinct aerosol layer". Is the key point that there is aerosol present? In which case, I think the lidar profile itself (assuming the signature of the aerosol is significant above the retrieval uncertainty and not just an artifact of the retrieval) is evidence of aerosol and the RH is mostly irrelevant. Or is the key here that the aerosol is "distinct". I don't understand how either the extinction profile or the relative humidity indicate that the aerosol is physically distinct from the African BBA layer.

Agreed. This is a bit of an overstatement. 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)".

344 I don't see that there is much agreement in the "structure" of the PBL. You might just say more simply that the apparent height of the PBL in the aerosol agrees with the location of the gradient in RH.

Agreed. The sentence has been modified accordingly.

353-354 Not clear what "distinct aerosol layer above the main BBA layer" means. Please specify the altitudes you're talking about. To me it looks like the main BBA layer extends to higher altitude in the Henties Bay uplooking lidar profile and in profile (1), while (2) shows a lower top for that layer. I don't see it as a separate, secondary layer. I can easily agree if you say "additional aerosol above 5 km" which is more unambiguous.

Agreed. This part was removed to avoid repetition as the information about the additional aerosols above 5 km has been added earlier in the section where the features in the upper part of the BBA layer are discussed with respect to the profile in Henties Bay.

360 "related to ... hygroscopic growth ... particularly below the BBA layer where RH is high in '2'". Again, I'm having some trouble following the roundabout wording here, but I take "below the BBA layer where RH is high" to mean "below 1 km". Yet there is no enhancement of extinction in profile '2' below 1 km. So, either this statement is incorrect, or the wording is so confusing that the meaning has been lost for at least one reader.

Agreed. This part is rather unclear and has been removed.

361 How robust are the estimates of the AOT of the layer above 5 km to the uncertainties in the lidar retrieval?

The temporal evolution of the AOT between 5 and 6 km AMSL is now plotted in Figure 2b with the error bars. We have also added an assessment of the uncertainty due to the knowledge of the lidar ratio in Appendix A.

362 "suggest that the aerosol may have a different origin". Again I do not understand what evidence supports this statement.

Agreed. Mention to the "different origin" has been removed.

392-393 "RH values ... may be an indication of the transport of BBA from a different origin." Again, I don't understand why the RH values should be taken as evidence of a different origin. I believe you have stronger evidence in the back-trajectories. It would be better not to state this conclusion until it has actually been supported. At most, for this sentence, I would say something like that the difference in RH is an indicator that the meterology has changed (if indeed that is what you believe) and that that will be shown in the next section.

We agree and we have modified the sentence: "…which is an indication that the meteorology has changed and by this way that the origin of air masses may be different.".

416 "are transported very rapidly" I don't understand how to interpret the figure to conclude that the aerosols are transported rapidly. Since the trajectories are more tightly distributed around the point of origin here than in the other panels, and yet all 3 panels show back-trajectories for the same 6-day duration, I infer that in this panel they moved more slowly.

Yes, we have removed "very rapidly".

423 "They correspond to air masses arriving above 5000 m AMSL over Henties Bay". Please show this by splitting 11(c) into panels above and below 5000 m. Since you treat this point as important, it should be shown explicitly.

We have now added the back trajectories for the 3 altitude intervals and each period: [1500 3000[, [3000 5000[and [5000 6000[to better highlight the differences.

Figure 11 caption. What does "normalized occurrence" mean (normalized to what)? Does this imply that the scale does not translate to the same number of back-trajectory points in each of the three panels?

The back trajectories have been calculated with a vertical step of 250 m. Hence, the number of back trajectories is different against the altitude intervals but stays relevant (> 500 back trajectories). The normalization is performed with respect to the total number of pixels for a horizontal resolution of 0.5-1° (this information has been added in the figure captions). The use of normalized variables is that they are comparable provided that the number of samples is large enough, which is the case here.

502 "of paramount importance" should be toned down. "Highlighting" the transported aerosol is interesting and worthwhile but not of "paramount importance" until it is shown to be climatically significant and not already captured in climate models.

Agree, we have reviewed the sentence: "Highlighting the transport of BBA from South America and its likely advection on top of the BBA layers originating from Angola and northeast Namibia may be climatically significant but not already captured un climate models in this region of the globe, where the feedback of aerosols and clouds on the radiative balance of the Earth system is still poorly known.".

750 Define "apparent backscatter coefficient", preferably using an equation. Not knowing for sure what this is is making it hard for me to follow the rest of the section.

This is like the total attenuated backscatter coefficient and the equation is always given in Royer et al. (2010). We have added both the definition and reference.

751-752 "must follow the slope of the molecular bacskcattering". This is not well explained. Won't there be attenuation of the signal by air molecules and aerosol at lower levels? (wheras the molecular backscattering from ERA5 is not affected)?

Agree, there is obviously an attenuation, but the molecular slope is not affected when there are no aerosols. It is therefore a strong element, often used by instrument designers, to check the alignment of lidar far-field. This is what we use regularly to check the alignments of the lidars that we develop. The use of Era5 is interesting because there is little error on the temperature and pressure profiles with these reanalyses which assimilate the IASI radiances. These profiles are not affected by atmospheric transmission.

759-760 "the error remains below 2-3%" Which error remains below 2-3%? The error in the molecular backscatter, or the aerosol backscatter derived from it, or the aerosol extinction derived from it?

This is only the error dur to the variability of the molecular density. This point has been better explained.

779 "uncertainty sources are exhaustively quantified". Given that you have access to a methodology for quantifying the uncertainty, it should definitely be quantified for the data presented in this paper. Depending on how big the retrieval uncertainites are, some of your conclusions can be affected, as discussed elsewhere in this review, so it's not just an academic exercise.

The uncertainties due to the statistical noise have been added on each vertical profile in the main part of the paper. The bias linked to the LR have been computed and illustrated by two representative cases in the Appendix.

781-782 "using aerosol typing determined from the CALIOP and CATS measure ments". How to you obtain the free-troposphere aerosol type and lidar ratio for cases where there is no satellite overpass in table 1? How do you get them for cases where CALIPSO or CATS or both infer multiple types in different pixels in the region (like the 31 August case, for instance).

When there is no CALIOP or CATS overpasses, we take the LR values of the nearest day. We have added the sentence: When there is no CALIOP or CATS overpasses we take the value of LR of the nearest day also considering the shape of the AEC profile and the origin of air masses using back trajectories. Note that the CALIOP and CATS typing is very much in agreement.

795-798 It seems like a stretch to infer from just a few individual cases of transported smoke from a totally different source as presented by Muller et al. 2007 that the lidar ratios for dust and smoke at 355 nm are the same as 532 nm. Is this the only relevant paper?

To our knowledge, this is the only one article that uses a multi-wavelength lidar over a long enough period to deduce statistically convincing results.

800 "matches best the AOT from the sun photometer". At the lidar wavelength?

Yes, we have specified the wavelength.

812 "uncertainties of 2% on the PDR". Do these uncertainties include sources of systematic error? Doesn't the uncertainty level depend strongly on the amount of aerosol? So, is there a minimum aerosol amount to get the quoted 2% error?

Yes, the uncertainty on the PDR depends on the value of the AEC. We have reviewed this part by taking two representative examples in the Appendix.

Minor comments

52 "as also mixes". Should this be "and also mixes"?

The correction has been done.

91 says the ground-based lidar is an ALS 300, but in the appendix it says ALS 450. Is there a difference between these two designations?

No, they are very similar. The correction has been done.

93 please indicate what type of lidar the LNG lidar is.

The LNG usage mode has been specified.

98 UTC is "coordinated universal time"

It is the Universal Time Count as explained in the text. It is like the definition of the reviewer.

154-155 a better reference for CALIOP 4.10 typing would be Kim et al. 2018

The reference has been added.

175-176 "The standard deviation on the AOT" should be "the uncertainty in the AOT".

The correction has been done.

205 change word order "when aerosols are only observed"

The correction has been done.

209 refers to lines highlighted in green in the table. I don't see highlighting. Does this need to be reworded?

It is a mistake, the correction has been done.

210 "averaged AOT of ~0.15" Indicate which instrument or model this refers to.

This referred to Figure 2, the correction has been done.

213 reference to Angstrom exponent. Which instrument or model?

This is from the sunphotometer, the information has been added.

227-228 "between the 2 periods". Between P1 and P2?

Yes, the correction has been done.

234 add reference for sun photometer retrievals of lidar ratio

The reference to Dubovik et al. (2000) has been added.

250 "match perfectly" is an overstatement. Please reword. It's better to be quantitative anyway.

The correction has been done.

256 "positively biased" reword to avoid "biased" and use a more neutral phrase like "larger than". You've given reasons why either the model or the observations (or both) could be incorrect in this case.

Right, the correction has been done: "the sun photometer- and MODIS-derived AOTs are larger than the one computed from CAMS".

Table 3 caption, UAL is used here where elsewhere it is described by the label FT. Is the difference signficant? If not, please pick just one.

Yes, it is different. The free troposphere is not only the aerosol layer. Nevertheless to avoid confusion we now used AFT for Aerosol layer within the free troposphere.

Table 3 column "AE" please indicate which instrument this is from.

The information is already given in the table caption.

Figure 2 It would be easier to understand the x-axis if tick marks were an integer number

of days instead of 1.5 days.

Yes, the correction has been done. We also have added the temporal evolution of the AOT for each altitude interval in the free troposphere.

292 "where" should be "were"

The correction has been done.

294 I believe this is probably attenuated backscatter coefficient rather than aerosol backscatter coefficient. This is an important distinction.

Yes, the correction has been done.

308 remove "wood". I think it's more likely grasses and crops here than wood, but either way it's speculation. "Combustion" is sufficient.

Ok, the correction has been done.

323 "apparition" should be "appearance"

The correction has been done.

334 "offshore airborne lidar measurements from Henties Bay". Replace with "the profile west of Henties Bay" for clarity.

The correction has been done.

351 "below the base of the BBA layer observed further south". Please simply state the altitude for easier reading.

The altitude has been added.

352 "The RH above the top of the BBA layer". Again, it would be helpful to be more specific about what altitude and which RH profile you are referring to, for easier reading. Figure 6

Yes, we have rephrased this part in a sake of clarity: "Figure 9, the RH sharply increases close to the BBA layer, which together with the AEC profile in Henties Bay suggests the presence of a distinct aerosol layer above 4.5-5 km AMSL, that is not seen Figure 8b in the AEC profile '1', but seen over Henties Bay."

386 add "at Henties Bay" for clarity

We added it.

408 "Illustrating" not "corroborating" (this isn't independent evidence)

The correction has been done.

412 "between the 3 regimes" perhaps rather than "periods" since the different regimes include different choice of ending altitude as well as choice of ending date.

"Period" is better adapted because the transport regimes are very variable in time and what explains the differences on the lidar profiles involves the transport regime but also the origin of the air masses. We have revised our explanation in considering the evolution of the back trajectories.

412 similarly "For the layers observed during P1" for the start of the next sentence, since you are only showing back-trajectories corresponding to observed aerosol layers.

The same answer as the previous one.

414 "consistent" not "coherent"

The correction has been done.

484 "illustrate" or "show", not "evidence"

The correction has been done.

497 probably delete "trapped" or reword to make the meaning clearer

"trapped" has been deleted.

498 I don't understand the meaning of "in link with the dominant transport patterns of lifted aerosols". Please reword.

We have reworded the sentence: "in relation to the main transport regimes across the Atlantic Ocean".

504 replace "fundamental" with "necessary". Fundamental has a connotation of primary importance in addition to meaning "necessary" and therefore sounds like an overstatement.

The correction has been done.

Table A1 add post-processing temporal resolution

The information has been added.

Replace section title with "Overlap correction" or something similar, since this is only one of many aspects of calibration but the only one discussed.

Yes, the main correction is about the lidar overlap and we have added "Overlap correction" in the title of the section: "Overlap correction and rightness of lidar profiles".

761 "average lidar profiles".

The correction has been done.

766 replace "perfect coincidence" with "very good agreement"

The correction has been done.

777 replace "exogeneous" with "external"

The correction has been done.

798 replace "standard deviation of LRs derived from spaceborne lidars" with "expected uncertainty in LRs for spaceborne lidars". Lidar ratio is not dervied from CALIOP and CATS; they do not measure or retrieve lidar ratio. It's an input, not an output, of the retrieval.

The correction has been done.

802 replace "coherent" with "consistent"

The correction has been done.

807 replace "inverse" with "invert"

The correction has been done.