Response to Referees

We thank the referees for taking the time to assess our manuscript and their useful suggestions for its improvement. Hereafter we answer all the comments.

Anonymous Referee #1

<u>General Comments</u>

The authors present an overview of in-situ measurements of aerosol optical properties made during the DACCIWA aircraft measurement campaign over the coastal region of Southern West Africa in summer 2016. The region has a varied aerosol environment, in which they sampled mineral dust aerosol, biomass burning aerosol, and anthropogenic pollution. Along with the meteorological situation, instruments on the aircraft measured the aerosol size and vertical distributions, and the aerosol scattering, absorption and extinction coefficients. From this information the authors further derived the aerosol scattering and absorption Ångström exponents, the complex refractive index, the single scattering albedo (SSA), the mass extinction efficiency, and the asymmetry parameter. Using this information, the authors describe the vertical distribution of the aerosols with respect to their optical properties, as well as showing that the SSA of the measured biomass burning aerosols is primarily a function of the imaginary part of the refractive index rather than of the size distribution. The derived SSA values of the biomass burning aerosols appear to be particularly low compared to other measurements from other regions of the world.

This is a very straightforward paper, and I appreciate that the scope of this work is primarily concerned with presenting the measurements and providing an interpretation of their significance. Within this scope, I would say that this is a successful paper, and hence I only have a few minor comments to make.

<u>Specific comments</u>

Figure 1: I assume that the black lines are the flight tracks in background conditions?

Black lines are the tracks of all the flights performed during the field campaign. They include flight tracks in background conditions as well as those in conditions that were not included in our classification due to various reasons (aerosol properties varying during a SLR, instruments not working properly,…).

We have modified Figure 1 to show the selected flight tracks in background conditions. We have also added the location of vertical profiles as requested by Referee #2.
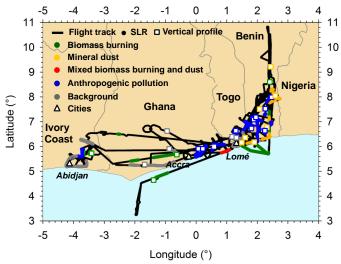

Figure 1. Tracks of the 15 flights analyzed in this study. The colors indicate aircraft flight sampling layers dominated by biomass burning (green), mineral dust (orange), mixed dust-biomass

burning (red), anthropogenic pollution (blue) and background particles (grey) from both vertical profiles (squares) and straight and level runs (SLRs; thick lines).

Section 2.2: as a qualitative comparison, I wonder if it would help to include one of these satellite images (MODIS or SEVIRI) over SWA with one of the flight tracks? This may be useful context to add and may aid the reader's understanding of the regional environment. Maybe such a figure may be more appropriate with respect to Section 2.3.2, and it may also be useful to have an image for each of the aerosol types.

During the summer monsoon, large areas of southern West Africa and Atlantic Ocean are covered by clouds presenting a high surface albedo, making it challenging to observe aerosol properly from satellite observations. When analyzing MODIS or SEVIRI images, we were unable to follow the back-trajectories of dust or biomass burning plumes due to too many cloudy satellite pixels.

Section 2.3.2: might it also be helpful to tabulate (briefly) the aerosol classification specifications? This may be helpful for quick reference when the reader wants to re mind themselves of how the aerosols are classified in the later sections and figures.

The figure below has been added in the paper to illustrate how the aerosols were classified.
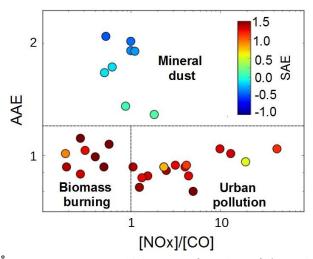

Figure 2. Absorption Ångström Exponent (AAE) as a function of the ratio NOx to CO. The markers are colored according to the Scattering Ångström Exponent (SAE). Classification of mineral dust, biomass burning and urban pollution particles has been added to the figure.

Figure 5, labels: "wavelength"
Done

Anonymous Referee #2

This paper describes the aerosol physical and optical properties observed during the DACCIWA campaign. The paper is based on in-situ measurements performed a-board the ATR-42. The paper is well written and the topic fits the ACP's scope. However, I have many remarks and some needs to be taken into account before this paper could be published in ACP.

Major comments

The title of this manuscript. I believe the title refers to the BB aerosols that were observed with really low SSA in comparison to all previous results. However, this part is described only on the 23th page. I would strongly recommend changing it to "aerosol optical properties overview during the DACCIWA campaign" or something similar.

The title has been changed to: *"Overview of aerosol optical properties over southern West Africa from DACCIWA aircraft measurements"*

First major comments about the Mie code used for this study. So you stated few lines (P8 L237-245) about how you used the Mie code to retrieve the refractive index of aerosols (reverse method) based on their physical and optical properties. First of all you are using a Mie code for dust and BB aerosols. In fact you measure the asymmetry factor (which is not that good with the TSI nephelometer) and found out that the particles are far from spherical. You never stated the possible issues with it. Then you never say a word for the mixing state you used. I found a word about it later when you say that the mixing was external which was consistent with previous observations. Have you tested the case with internal mixing (either core shell or just coating) ? Moreover, what is the acceptable difference between measurements and calculation to stop the iterative process (5%, 10% more ???)? Is it just iterative process with an a priori k and n or it an optimal estimation method that will avoid any local minimum? Later on (P20-21, L548-556), you used the Mie code again but this time I believe not in a reverse mode. So you entered a k and n along with the size distribution and you get optical properties that you used to calculate a SSA. Which k and did you used? In Africa, there are many sources of dust and BB aerosols associated with many different k and n... How these calculations made with a Mie code, for spherical particles, prove that your aerosols are externally mixed? You need here to prove it with a spheroidal code, for different mixing state.

We have chosen to use a Mie code to calculate the aerosol optical parameters by assuming the particles are spherical. First, because the particle shape was not measured during the field campaign and it would be very hazardous to associate a shape to the aerosols, especially since aerosols from different sources were sampled at different times after emission. Second, this facilitates a quantitative comparison with past data, since a large majority of them used this simplification. Third, because climate models usually use Mie theory due its computational efficiency and applicability to radiative transfer models.

To explain this choice we added the following text in Section 2.3.1: *"Optical calculations were performed using Mie theory, implying a sphericity assumption, because it facilitates a quantitative comparison with past data, mostly using this simplification and because most climate models assume spherical properties."*

Concerning the aerosol mixing state, the application of Mie theory in our retrieval procedure to calculate aerosol optical parameters requires information about the size distribution, refractive index and particle density. These input parameters refer to the properties of the overall particle population within the aerosol. There is no assumption in the aerosol mixing state. By contrast, in climate models, the aerosol optical parameters are calculated from multiple particle types with

different chemical composition and apply a mixing rule to calculate the refractive index of the aerosol from the refractive indices of the individual component species. Therefore, the mixing state of the aerosol must be addressed for climate modelling to calculate aerosol radiative forcing. Currently, the most common assumption made in climate models, which is also the easiest from a computational perspective, is that the aerosols are externally mixed. This is the reason why we assessed whether the assumption of external mixing allow to reproduce the observed optical properties in section 4.1. To test internal mixing assumption would need to know to what degree the aerosol population is internally-mixed. In fact, there are an enormous number of degrees of freedom in the aerosol mixing state. Even within a single distribution mode there are a prohibitively large number of combinations involved in mixing the constituents. At one extreme the aerosols can be internally mixed as a homogeneous material reflecting the chemical and physical average of all the contributing components. At another extreme, each aerosol component is physically separated from the other components creating an external mixture of chemically pure modes. The real mixing state can be expected to lie somewhere in between these two extremes, with aerosol in an external mode that is or is not an internal mixture of components. Therefore, in the absence of information on mixing state and chemical composition of individual particles, we cannot test internal mixing assumption.
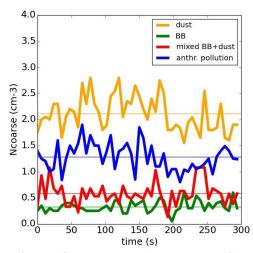
We have added more information in the paper about the choice of the refractive indices used in the Mie code in section 4.1: *"The refractive indices at 550 nm were assumed to be 1.52-0.002i and 1.60-0.040i for dust and anthropogenic pollution particles, respectively, which are the mean values deduced from the data inversion procedure."*

We have also added more information about the iterative process developed to calculate aerosol optical parameters in section 2.3.1: *"The retrieval algorithm consists of iteratively varying the real part of the complex refractive index (m) from 1.33 to 1.60 and the imaginary part of the complex refractive index (k) from 0.000 to 0.080 in steps of resolution of 0.001. m and k were fixed when the difference between calculated values of σ_{scat} and σ_{abs} and measurements was below 1%."*

Number concentration of coarse particles. The authors are showing number concentrations of particles from 1-5um. These concentrations are measured from the OPC-GRIMM. The concentrations are between 0 and 2.5 cm-3. Could you provide the reader the noise of this instrument regarding these large particles. Is it significant ?

The advantages of an OPC is that (1) it can count particle numbers very accurately when the concentration is low; (2) it has very good signal to noise ratios for large particles (>1μm). The figure below shows the time evolution of particle concentration from 1-5μm measured by the OPC-GRIMM for different aerosol plumes sampled during straight levelled runs. You can see that the signal to noise ratio was higher than 3 for all the aerosol plumes, which make the instrument well suited to quantify particle concentration in this size range.

We have added this information in section 2.3.1.: *"The signal to noise ratio of the OPC for particles in this size range was higher than 3, which makes the instrument well suited to quantify variations in N_{coarse}."*

Temporal variation of particle number concentration measured in the size ranged 1-5µm by the GRIMM-OPC during different SLRs.

During the field campaign, the ATR-42 performed vertical profiles at the beginning of each flight. These are used in Figure 3. On Figure 1, you highlighted the BB, dust and pollution plumes. How far are the vertical profiles from the plume observed ? I wonder how the variability of the vertical profile average is due to the distance to the actual plume. I had to wait until Figure 6 (P21) to have information about altitudes at which you observed the different plumes. At P14 L366-370, P15 L398-399, you are showing that aerosols, within a layer located below 2.5km, are a efficiently mixed with anthropogenic emissions. Then on Figure 6 you are trying to interpret the optical variations throughout the vertical profile. From my understanding, if you are interested in "pure" BB or Dust particles than you should only show/interpret vertical profiles from 2.5km up to 4km.

The location of vertical profiles has been added in Figure 1. You can see that the vertical profiles were located near the straight leveled runs.
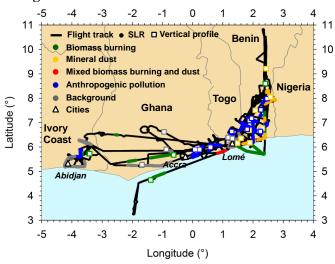

Figure 1. Tracks of the 15 flights analyzed in this study. The colors indicate aircraft flight sampling layers dominated by biomass burning (green), mineral dust (orange), mixed dust-biomass burning (red), anthropogenic pollution (blue) and background particles (grey) from both vertical profiles (squares) and straight and level runs (SLRs; thick lines).

Data presented in Figure 6 (now Figure 7 in the update manuscript) are based on measurements during SLR. This has been clarified in the paper at the beginning of section 4.1.: *"We exclusively*

consider measurements acquired during SLRs, since only during these phases the whole set of aerosol optical properties were measured."

Minor remarks

Page 2 L37 : I think something is missing to introduce properly the BB aerosols or just to skip a line here.

The sentence has been rewritten as follows: "*For the layers dominated by the biomass burning particles, aerosol particles were significantly more light absorbing than those previously measured in other areas (e.g. Amazonia, North America) with SSA ranging from 0.71 to 0.77 at 550 nm.***"**

Page 2 L68 : It's usually called MEE that stands for Mass Extinction Efficiency and its units is m2/g not g/cm3. In the table2's caption you even called it extinction mass efficiency. Please be consistent throughout the manuscript. By the way how did you measure the mass concentration of aerosols ? Did you assume a density for each type ? And did you assume the same density for each mode ?

We have changed MEC by MEE in the text.

Information about the mass densities of each aerosol type used to calculate aerosol mass concentration and henceforth MEC are already provided on P8 in L261-264.

P3 L73-75: Why do you introduce remote sensing data that you will never ever talk again ?

Our intention is to provide a comprehensive overview of aerosol optical properties over SWA for the climate modelling and the remote sensing communities. From the remote sensing perspective, in-situ data constitutes independent verification of satellite data products and can be used to assess the performance of inversion algorithms. For known TOA reflectance, the retrieval of AOD is very sensitive to aerosol size distribution and refractive index. Improper assumptions used in the algorithm regarding such key parameters may lead to a spurious AOD long-term trend in the regions under influence of absorbing particles due to the regional biases. This is the reason why we introduce remote sensing measurements in the introduction and we invite the remote sensing community to use this dataset in the conclusion.

P3 L112 remove the 's' at aerosol

Done

P 5 When DACCIWA took place (month, year) ? What season was it (apparently during the monsoon period)?

The campaign period is already presented before on P4.

P6 L 176 : the CPC MARIE model needs to be described here. There is no paper associated with this instrument and I was unable to find any specifications on internet. Is it working with a flow regulator or a critical orifice? How is it counting in comparison to any TSI CPC ? You used the CPC measurements to infer the Nfine but you never say anything about a comparison, in term of number concentration, with the OPC, UHSAS or the SMPS. As this instrument is not (yet?) considered as a reference, you cannot use it like a reference. You could use the calibration data for the size distribution (PSL and oil) to prove that this instrument was measuring the same number concentrations than the OPC.

The international standard method defined by the European Committee for Standardization for determining the particle number concentration is based on a CPC (CEN/TS 16976:2016). This instrument is considered as a reference for ambient aerosol monitoring for many years. The CPC Marie allows measuring number concentration of particle with a size above 10-15 nm

(depending on the temperature conditions). It is structurally the same as a TSI CPC3010 except that there are more controls on the condensation and saturator temperatures and thermal dissipation has been improved. Thus, its measurement behavior is comparable to a TSI CPC3010 (Mertes, Schröder, and Wiedensohler, 1995; Russell et al., 1996; Wiedensohler et al., 1997). OPC, UHSAS and SMPS are measuring particle size distribution in limited size ranges (0.3-32μm for OPC, 0.1-1 μm for UHSAS during DACCIWA, 0.02-0.5 μm for SMPS). Given the relative importance of the smallest particles to the total concentration, the CPC concentration can be considered as the best estimation of the total particle concentration sampled.

We have added in the text the following references that provide a description and validation of the CPC Marie, as well as its intercomparison with other CPCs: *Mertes, Schröder, and Wiedensohler, 1995; Russell et al., 1996; Wiedensohler et al., 1997.*

Concerning the calibration experiment suggested by Referee #2, artificially produced PSL (or oil) particles would generate a mixture of air/water/PSL(oil) mixture with elevated concentration of ultrafine particles. These small particles cannot be fully detected by the OPC, UHSAS or SMPS instruments, so direct comparisons between instruments won't be relevant for this experiment. Otherwise, Marie CPC and SMPS CPC have been inter-compared in the laboratory at low pressure with concentration differences lower than 10%.

P6 L 187 Replace 525 by 550nm.
Done

P10 section 2.3.2: A figure showing the AAE versus the SAE would have been appreciated here.
The new figure below has been added in the paper.

Figure 2. Absorption Ångström Exponent (AAE) as a function of the ratio NOx to CO. The markers are colored according to the Scattering Ångström Exponent (SAE). Classification of mineral dust, biomass burning and urban pollution particles has been added to the figure.

P10 L282 : remove the 's' at the end of theses
Done

P11 L 301 : I believe that the background is also without the detectable influence of Dust and BB as well as anthropogenic emissions.
Done

P13 L334: The AEJ is a synoptic flow. It is always present over West Africa at a specific location (between 10-20 ∘N between 2-6km). The ATR flew between 3-11∘N. Therefore, I don't see how you would have been able to observe it and why you would have been able to observe it for specific cases and not for the others.

The average climatological position of the AEJ varies during the summer between 10°N in June and 15°N in July. However, because it is a synoptic feature, the position of the AEJ core varies significantly from day to day, the DACCIWA period being no exception as shown by Knippertz et al., 2017 using ERA-I reanalysis data. In their Figure 8, Knippertz et al. (2017) show that the position of the AEJ on 2 July is centered at 4°N and on 10 July at 24°N. This is due to the many disturbances sweeping through southern West Africa during the monsoon season, which impact the location and the intensity of the AEJ core.

It is also worth pointing out the AEJ is a broad feature, generally confined to a width of 5°–10° of latitude as seen in reanalyses, but also in the airborne observations made using dropsondes along north-south transects across southern West Africa during JET 2000 (Tompkins et al., 2005) and AMMA in 2006 (Flamant et al., 2009). In the later study, the southern fringe of the AEJ is observed to reach the latitude of the SWA coast around mid-July.

Therefore, due to the important day to day variability of its mean position, but also of this latitudinal width, we can assess that observations made with the ATR between 3–11°N were indeed acquired in regions impacted by the AEJ and under a variety of AEJ-forcing conditions.

We have added a sentence about the latitudinal variation of the AEJ: "*Above 2.5 km amsl, the wind speed increases in the urban pollution and dust cases and the wind remains easterly, indicating the presence of the African easterly jet with its core typically farther north over the Sahel (Figure 8 in Knippertz et al., 2017 for the latitudinal variations of the African easterly jet during the DACCIWA field phase)."*

P13 L 335 : there are two dots after Sahel.
Done

P13 L356-357 : According to the Nfine vertical profile, the BB plume is observed from 2.5 – 4km. Your sentence isn't clear for me. Is this between the surface up to 4km or as suggested by the Nfine vertical profile ?
We have clarified in the paper that we refer to the BB plumes transported above 1.5 km asl: "*The biomass burning plume extends from 1.5 to 5 km amsl and is associated with transport from the southwest in a layer of enhanced wind speed just below 4 km amsl as discussed above."*

Figure 4 : I think you could do something in this figure to show the altitude of each SLR. You never mentioned that the different lines in Figure 4b were associated to all the plumes observed. Maybe the line width of each line could be coded as a function of the altitude? Obviously for dust plume one SD is significantly different from the others. Why ?
We have modified the figure to show the altitude of the aerosol sampled.
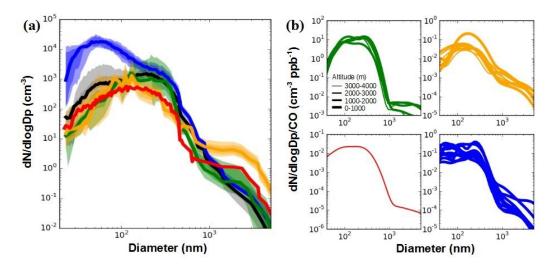
Figure 5. (a) Statistical analysis of number size distributions with colored areas representing the 3th, 25th, 75th and 97th percentiles of the data and (b) number size distributions normalized to CO for plumes dominated by biomass burning (green), dust (orange), mixed dust-biomass burning (red), anthropogenic pollution (blue) and background particles (black). In panel b, the line thickness is scaled by the altitude of the aerosol plume.

P15 L 388 : remove 'the' in front of plumes
Done

P15 L 392-393 : This is impossible to see that on Figure 4. We could see something on Figure 4b but it could also be result of the CO concentration…
Colors in the figure have been changed to enable a better observation of variability in particle concentration (see figure above).

L 395 – 396 : We must trust you about the fact that the smaller particles are observed within the lower dust plume…
We have added in the text that this observation can be seen in the previous Figure 3.

P 15 L 401-406: Again you stated that below 2.5 km all the aerosol are mixed. You observed 3 dust plumes below 3km. I assume that these three samples where associated with the small particles. The BB plumes where observed above ∼2.2km. I'm not convinced that the location has anything to do but mainly the altitude… Most of the BB plumes have been observed close to big cities (Abidjan, Accra, Lome).
As written in the paragraph, BB and dust plumes were sampled at approximatively the same altitude. The lowest BB plumes was observed at 2.2 km asl., which do not make a big difference with the dust plume sampled at 2.0 km asl. Since pollution plumes were exported up to 2.5 km asl., the altitude is probably not the reason of the different mixing with pollution plumes.

P15 L417-419 : The accumulation mode exceeds the amount of large particle. What is "large particle" here ? As there are no information about the mode the authors are referring to, it's hard to fallow. There is probably a need for a table that describe all the modes observed for each aerosol type. Anyway, is that surprising in number concentration?
"Larger particles" has been replaced by "particles centered at 230 nm" in the text for clarity.

It is already explained in the paragraph that the mode centered at 100 nm is surprising for aged biomass burning, when comparing with the literature.

P16 L 423. In this part you are talking about a phenomenon observed by colleagues during two specific days. Do they observe it during other days ? Could it happen during other days ? As you are not explaining the general mechanism there is no way for us to infer if this could have happen during the whole campaign. So when did you observed all plumes ? Was it during these specific days ? If not how do you know that this is always the case between the surface and 2.5km ?
We have removed the sentence referring to papers of Flamant et al. (2018) and Deroubaix et al. (2019).

P16 L430 : So the same dusty mode was observed during several campaigns over West Africa. From this observation you conclude that dust does not efficiently mix with other particles. Could you please explain ?
The coarse mode of the dust plume is expected to be impacted by the mixing with other particles in case of an internal mixing, which should somewhat increase the particle size. We have clarified it in the manuscript: *"The super-micron mode of the dust plume is expected to be impacted by the mixing with other particles in case of an internal mixing, which should somewhat increase the particle size".*

P16 L435: Then, within the anthropogenic pollution plumes you observed a coarse mode. As the sites are nearby the coast you assume that it could be sea salt. Do you have any evidence for that? Could it be dust or road dust or pollens or any other large particles???
Analysis of the spectral dependence of aerosol optical properties allows for interpretation of aerosol chemical composition. As stated in sections 2.3.1 and 2.3.2, mineral dust particles have the unique ability to absorb light strongly in the blue to ultraviolet spectrum. The presence of dust particles in aerosol plumes is thus expected to increase AAE above 1. We measured AAE in the range 0.7-1 in anthropogenic pollution plumes, which suggests negligible contribution of mineral dust in these plumes. This result is not surprising since anthropogenic pollution plumes originated from the south while the source region of dust was located in the north east when looking at the backtrajectories of air masses (Figure 2). For anthropogenic pollution plumes being sampled near the coast and below 0.5 km amsl, mixing of sea salt in pollution plumes is the most relevant interpretation for the presence of a coarse mode.
We have added the following explanation in the manuscript: *"We measured AAE in the range 0.7-1 in anthropogenic pollution plumes (Figure 2), which suggests negligible contribution of mineral dust in these plumes. This coarse mode has most likely significant contributions from sea salt particles, as plumes arriving from the cities were transported at low altitude over the ocean (Fig. 3)."*

P17 L458 : The strongest spectral dependence of SSA is observed for the lowest absorption plume. Well from the Figure 5b this is not the case. Indeed, the largest SSA values for the BB aerosol at 440nm (Around 0.78) show a quite strong SSA at 660nm (around 0.74) and therefore quite a low spectral dependencies. The largest spectral depend are observed for both 'middle' SSA values at 440nm. Could you then remove the part right after that suggest similar results with the literature that are no more consistent with your results?
The referee is right. We have removed this part.

P18 L477 : I found it really odd for you to compare small SSA spectral dependencies observed over African cities using in-situ measurements with literature using AERONET measurements or even with plumes observed over European cities. The principle (column integrated versus in situ) of measurements as well as the pollutants (European polluted plumes are clearly different from African polluted plumes) have to be a bit similar to gain information. In this case arguing that what you observed is entirely different from these studies is not convincing.

AERONET has been extensively compared with various types of airborne in situ airborne measurements for a range of different aerosol types including urban pollution particles and the Africa continent. These were recently summarized in Andrews et al. (2017). They confirm previously reported comparison that the large majority of SSA comparisons for AOD> 0.2 are within the reported in-situ SSA uncertainty bounds, while AERONET inversions tend to overestimate SSA for low AOD. All AERONET measurements that included the AOD > 0.2 constraint have shown that SSA systematically decrease with increasing wavelength for a range of different urban pollution plumes across the world (Dubovick et al., 2002; Shin et al., 2019). However, in the light that no AERONET or in-situ measurements are available for comparison in other African cities, we have nuanced the sentence: *"However, the flat spectral dependence of SSA appears to be anomalous for anthropogenic pollution aerosols, as SSA has been shown to decrease with increasing wavelength for a range of different urban pollution plumes over European, American and Asian cities (Dubovick et al., 2002; Di Biagio et al., 2016; Shin et al., 2019)."*

P18 L 481 : During the NAMMA campaign, Omar et al. compare the extinction coefficients from the LIDAR and from the in situ measurements (assuming a dust LIDAR ratio). They found that the extinction was really close and then that no large aerosol was lost within the inlet. I'm sure you could do something similar here to estimate the losses for all cases.

This paragraph was unclear. We wanted to highlight the difficulty to compare our results with previous measurements because the measurement of SSA is highly dependent on the extent to which the coarse mode is measured behind the aerosol sampling inlet. For instance, Ryder et al. (2013) found an overestimation of SSA in dust plumes when the SSA is measured behind inlet during the FENNEC campaign. In our case, the impact of the Community Aerosol Inlet on the measured optical properties has been calculated for dust aerosols in Denjean et al. (2016). We found that the absolute error associated with SSA, g and MEE due to the aircraft inlet is in the range covered by the measurement uncertainties. However, different aerosol inlet systems were used during previous field campaigns (cut-off diameter of 5µm during DACCIWA and 2.5µm for Weinzierl et al. (2011) during SAMUM-2 for instance), which can lead to discrepancies when comparing SSA from different campaigns.

The paragraph has been changed as follows: *"The magnitude of SSA increased at the three wavelengths when dust events occurred. Large variations in SSA were obtained with values ranging from 0.76–0.92 at 440 nm, 0.81–0.94 at 550 nm and 0.81–0.97 at 660 nm. The measurement of SSA is highly dependent on the extent to which the coarse mode is measured behind the aerosol sampling inlet. Denjean et al. (2016) found that the absolute error associated with SSA, g and MEE of dust aerosols due to the CAI inlet is in the range covered by the measurement uncertainties. However, different aerosol inlet systems were used during previous field campaign, which makes comparison of our results with previous measurements difficult."*

P18 L484 : 'measurements are comparable to each other'. So I'd like to disagree with you. It depends on the event right ? Some events will be associated with uplifting of large particles and therefore the

inlet cut off may play a major role OR some events could also be associated with uplifting of smaller particles and then the inlet cut-off won't be an issue anymore.

As explained in the previous comment, the paragraph was unclear and has been rewritten.

P19 clarify MEE versus MEC vs extinction mass efficiency vs extinction mass coefficient!

We have exchanged MEC by MEE in the text.

P19: Another Mie test… So now you used a Mie code to estimate the impact of each aerosol mode ? Could you please explain what you did and how you did it ? Ân We found MEC to be positively correlated with SAE (not shown) with measurement wavelengths (450–660 nm), which agrees with Mie theory. Âz Again, I don't understand the sentence. Are you talking about measured MEE vs measured SAE having the same Ân correlation Âz than the calculated one ? What about a straight comparison of SAE/MEE calculated and observed ? And how this Ân correlation Âz is relevant for your analysis ?

This is not another Mie test; this is the result of the iterative process that provides SSA, MEE and g. The aim of the sentence is to explain some of the variability in MEE for a given aerosol type. SAE being a good proxy for variation of size distribution for a given aerosol chemical composition, it was expected that MEE correlates with SAE.

To clarify this, we have rewritten the sentences as follows: *"MEE is heavily influenced by the mass concentrations in the accumulation mode where the aerosol is optically more efficient in extinguishing radiation. We found MEE to be positively correlated with SAE (not shown), which was expected because of the dependence of MEE on particle size".*

P19 L 509-511: So the total and back-scattering coefficients have an error associated with the measurement principle. Did you correct the data fallowing Anderson and Ogren recommendations? What are the error associated with the measurements and then with the g calculation? Moreover, the g is also affected by the morphology of the particle.

Data were corrected for truncation error using a Mie code in the data inversion procedure that takes into account the aerosol size distribution. This correction is more accurate than the Anderson and Ogren recommendations which are based on single sample. This was already written in Table 1 and Appendix 2, but we have added the following sentence in section 2.1. : *"The particle scattering coefficients (σ_{scat}) at 450, 550 and 635 nm were measured using a AURORA3000 3-wavelength nephelometer and corrected for angular truncator error in the data inversion procedure using a Mie code (i.e. Section 2.3.1 and Appendix 2)."*

Concerning the aerosol morphology, please look at our response above in major comments.

P20: Is this part 4 based on single SLR analysis ?

Yes, it is now specified in the text: *"We exclusively consider measurements acquired during SLRs, since only during these phases the whole set of aerosol optical properties were measured."*

P20 L530 – 532 : On figure 6c there are no dots below 1500m and I cannot sea the NOx decrease that you are talking about.

The sentence has been removed.

P20 L 532 : You qualify it as a strong SSA variation (0.8 to 0.95) but since page 15 you told us that below 2.5km all the layers are mixed together. From the number concentration profile there are almost no dust at that altitude (below 1cm-3).

We clearly mentioned previously that large variations in the concentration of fine particles (sections 3.1 and 3.2) and SSA (section 3.3) were observed in dust plumes. Concerning the concentration, only small number concentration of dust particles will contribute significantly to the aerosol mass concentration and extinction coefficient, and consequently to the aerosol radiative effect. You can see that effect in Fig 3a, which shows that the extinction coefficient increased from 20 to 80 Mm^{-1} when dust was transported at 2.5 km asl over the region.

P21 L564 : Án Figure 6 indicates markedly different processes affecting optical properties of biomass burning aerosols. Âz Which processes are highlighted by this figure ? To me, all BB profiles on Figure 6 are notably stable.
The sentence has been removed.

P21 L569 : Ân remote location Âz . Please define it. For me the anthropogenic plumes were also observed in remote location, don't you think ?
We have removed the part of the sentence about the location of BB.

P21 L564 to P22 L 578 : I don't really understand the need for this paragraph. There are no new information and we also have to wait for further explanations.
The important message of this paragraph is that BB plumes were not affected by pollution emissions. This interpretation is based on Fig. 6 showing the vertical distribution of SSA, SAE and NOx concentrations. So we think it is very important to keep this discussion here. However, we agree that the end of the paragraph can be moved in section 4.2 and it has been done.

P22 L589 – 591 Ân We did not find any correlation between the values of SSA and their spectral dependence, which suggests that the variability in SSA cannot be attributed to different contributions of marine aerosol in pollution plumes. Âz So I'm not sure I do understand this statement. A correlation between SSA values and their spectral dependency is evidence that SS are not contributing much?
We have removed this sentence.

P22 L595 – 597 "The high absorbing properties (SSA□0.81 at 550nm) of background aerosols is consistent with being a mixture of aged biomass burning and Atlantic marine aerosol. Âz Please explain. And Ân Moreover SSA of background aerosol was lower than previously reported over the Southern Atlantic (Ascension Island) outside the fire season in Central Africa (Zuidem al., 2018), which supports this conclusion. Âz Now I'm missing something. You are comparing measurements performed over a small island, far from any anthropogenic sources or any natural sources of absorbing particles, to your results and found that aerosols you observed are more absorbing. Is that surprising at all ? Does that prove that you observed BB aerosols or rather anthropogenic aerosols ?
Concerning the interpretation of the low SSA in background plumes, we have completed the sentence as follows: *"The high absorbing properties (SSA~0.81 at 550nm) and the presence of particles both in the accumulation and super-micron modes (i.e. section 3.2.) in background plumes are consistent with being a mixture of aged absorbing biomass burning and Atlantic marine aerosol."*
We have removed the sentence comparing our measurements with those of Zuidema et al. (2018), since it is discussed in more details in the next section.

P22 L207 : A variation of SAE could be due to a size shift if you know that chemical composition, mixing state and morphology are similar. In this case you don't show any chemical results but you have the SD. Why using SAE to highlight the size variation ?

As mentioned in section 2.3.1., the value of SAE depends primarily on the size of the particles, such that small values of SAE indicate larger aerosol particle, and large values of SAE indicate relatively smaller aerosol. This parameter is widely used in the community for studying the variability in aerosol size distribution because it is a sensitive parameter that needs limited corrections (only the truncation of the nephelometer). Using the measured aerosol size distribution would bring a higher degree of uncertainty due to the need to correct measurements with the aerosol refractive index.

L 611-613 Could you be more precise with the reference you cited ? Where were this studies perform and what were the particle diameter at the formation and after few hours ?

We have completed the sentence as follows: "*with previous observations of size distribution in aged North American biomass burning plumes*". The range of particle diameter reported in these studies was already provided section 3.2.

P23 L622 : Ân by the variability in composition of biomass burning aerosol Âz Please add a ref that prove that k is only linked with the aerosol chemical composition. Could you also show the spectral dependency of k ?

k is not only linked with the aerosol chemical composition, it also depends on the size distribution of the particle species. However, given that SSA was previously found to be independent of the aerosol size distribution (Figure 8a), Figure 8b suggests that SSA variability was strongly influenced by the variability in composition of biomass burning aerosol. The paragraph has been rewritten as follows: "*In contrast, Figure 8b shows that there was a consistent decrease in SSA with increasing k, although there is some variability between the results from different plumes. The observed variability of SSA is reflected in a large variability of k, which is estimated to span the large range 0.048–0.080 at 550 nm. k depends both on the aerosol chemical composition and size distribution (Mita and Isono, 1980). Given that SSA was found to be independent of the aerosol size distribution (Figure 8a), Figure 8b suggests that SSA variability was strongly influenced by the variability in composition of biomass burning aerosol, implying a high contribution for light-absorbing particles.*"

P25 L699 : You need to prove using chemical composition data that BB were not affected by the mixing with anthropogenic emissions (at least for the lowest SLR).

This has been intensively discussed in section 4.1 by looking at the vertical variability of SSA, SAE and NOx concentration. It was also discussed in section 3.2 with the analysis of aerosol size distribution in BB plumes.

P26 L717 : You never show the background SSA spectral dependency.

It is already shown Fig. 5 (a) in grey.

P26 L 721 : Â The large proportion of aerosols emitted from cities that resided in the ultrafine mode particles" Are you talking about African Cities or any city in general ? Do you have a reference? From your observations, you always have the background aerosol mixed with the city plume, right ? So you cannot tell what is coming from cities and what is coming from the background, right ? Or you made a difference between both SD ?

The interpretation of the ultrafine mode has been already intensively discussed in section 3.2. As stated in the text, this mode is only observed in plumes originating from the polluted cities of Lomé, Accra and Abidjan.

In the conclusion, we have clarified the origin of the anthropogenic aerosols: *"The large proportion of aerosols emitted from the cities of Lomé, Accra and Abidjan that resided in the ultrafine mode particles have limited impact on already elevated amounts of accumulation mode particles having a maximal absorption efficiency".*

The appendixes need to be called in the main text.
Appendixes were already called in the text (P5 for Appendix 1 and P7 for Appendix 2).

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Overview of aerosol optical properties over southern West Africa from DACCIWA aircraft measurements

~~Light absorption properties of aerosols over Southern West Africa~~

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Abstract. Southern West Africa (SWA) is an African pollution hotspot but a relatively poorly sampled region of the world. We present an overview of *in-situ* aerosol optical measurements collected over SWA in June and July 2016 as part as the DACCIWA (Dynamics-Aerosol-Chemistry-Clouds Interactions in West Africa) airborne campaign. The aircraft sampled a wide range of air masses, including anthropogenic pollution plumes emitted from the coastal cities, long-range transported biomass burning plumes from Central and Southern Africa and dust plumes from the Sahara and Sahel region, as well as mixtures of these plumes. The specific objective of this work is to characterize the regional variability of the vertical distribution of aerosol particles and their spectral optical properties (single scattering albedo: *SSA*, asymmetry parameter, extinction mass efficiency, scattering Ångström exponent and absorption Ångström exponent: *AAE)*. First findings indicate that aerosol optical properties in the planetary boundary layer were dominated by a widespread and persistent biomass burning loading from the Southern Hemisphere. Despite a strong increase of aerosol number concentration in air masses downwind of urban conglomerations, spectral *SSA* were comparable to the background and showed signatures of

35   the absorption characteristics of biomass burning aerosols. In the free troposphere, moderately to
36   strongly absorbing aerosol layers, dominated by either dust or biomass burning particles, occurred
37   occasionally. In aerosol layers dominated by mineral dust particles, *SSA* varied from 0.81 to 0.92 at
38   550 nm depending on the variable proportion of anthropogenic pollution particles externally mixed
39   with the dust. For the layers dominated by biomass burning particles, aerosol particles were
40   significantly more light absorbing than those previously measured in other areas (e.g. Amazonia,
41   North America) with *SSA* ranging from 0.71 to 0.77 at 550 nm. The variability of *SSA* was mainly
42   controlled by variations in aerosol composition rather than in aerosol size distribution.
43   Correspondingly, values of *AAE* ranged from 0.9 to 1.1, suggesting that lens-coated black carbon
44   particles were the dominant absorber in the visible range for these biomass burning aerosols.
45   Comparison with literature shows a consistent picture of increasing absorption enhancement of
46   biomass burning aerosol from emission to remote location and underscores that the evolution of
47   *SSA* occurred a long time after emission.

48   The results presented here build a fundamental basis of knowledge about the aerosol optical
49   properties observed over SWA during the monsoon season and can be used in climate modelling
50   studies and satellite retrievals. In particular and regarding the very high absorbing properties of
51   biomass burning aerosols over SWA, our findings suggest that considering the effect of internal
52   mixing on absorption properties of black carbon particles in climate models should help better
53   assessing the direct and semi-direct radiative effects of biomass burning particles.

54

1. Introduction

56   Atmospheric aerosols play a crucial role in the climate system by altering the radiation budget
57   through scattering and absorption of solar radiation and by modifying cloud properties and
58   lifetime. Yet considerable uncertainties remain about the contribution of both natural and
59   anthropogenic aerosol to the overall radiative effect *(Boucher et al., 2013)*. Large uncertainties are
60   related to the complex and variable properties of aerosol particles that depend on the aerosol
61   source and nature as well as on spatial and temporal variations. During transport in the
62   atmosphere, aerosol particles may undergo physical and chemical aging processes altering the
63   composition and size distribution and henceforth the optical properties and radiative effects. The
64   capability of reproducing this variability in climate models represents a real challenge *(Myhre et*
65   *al., 2013; Stier et al., 2013; Mann et al., 2014).* Therefore, intensive experimental observations in
66   both aerosol source and remote areas are of paramount importance for constraining and evaluating
67   climate models.

68

69 Key parameters from a climate perspective are the aerosol vertical distribution and respective
70 spectral optical properties. Radiative transfer codes commonly incorporated in climate models and
71 in satellite data retrieval algorithms use single scattering albedo (*SSA)*, extinction mass efficiency
72 (*MEE)* and asymmetry factor (*g*) as input parameters. These parameters depend on the aerosol size
73 distribution, the real and imaginary parts of the refractive index (*m-ik*), and the wavelength of
74 incident light, λ. The knowledge of the vertical distribution of these fundamental parameters is
75 crucial to accurately estimate the direct and semi-direct radiative effects of aerosols as well as the
76 vertical structure of atmospheric heating rates resulting from absorption by particles. Above
77 information is also required to retrieve aerosol properties (aerosol optical depth, size distribution)
78 from remote sensing data.
79
80 Southern West Africa (SWA) is one of the most climate-vulnerable region in the world , where the
81 surface temperature is expected to increase by ~3°K at the end of the century (2071-2100) in the
82 Coupled Model Intercomparison Project Phase 5 (CMIP5) *(Roehrig et al., 2013)*. It is
83 characterized by a fast-growing population, industrialization and urbanization *(Liousse et al.,*
84 *2014)*. This is particularly the case along the Guinea Coast where several already large cities are
85 experiencing rapid growth *(Knippertz et al., 2015a)*. Despite these dramatic changes, poor
86 regulation strategies of traffic, industrial and domestic emissions lead to a marked increase of
87 anthropogenic aerosol loading from multiple sources including road traffic, industrial activities,
88 waste burning, ship plumes, domestic fires, power plants, etc. Tangible evidence for regional
89 transport of anthropogenic pollutants associated with urban emissions has altered air pollution
90 from a local issue to a regional issue and beyond *(Deetz et al., 2018; Deroubaix et al. 2019)*. This
91 is particularly the case during summer when land-sea breeze systems can develop and promote the
92 transport of pollutants inland, away from the urbanized coastal strip of SWA *(Flamant et al.,*
93 *2018a)*. In addition to this anthropogenic regional pollution, SWA is impacted by a significant
94 import of aerosols from remote sources. Biomass burning mainly from vegetation fires in Central
95 Africa are advected to SWA in the marine boundary layer and aloft *(Mari et al., 2008; Menut et al.*
96 *2018; Haslett et al., 2019)*. The nearby Sahara desert and the Sahel are large sources of natural
97 wind-blown mineral dust aerosol throughout the year with a peak in springtime *(Marticorena and*
98 *Bergametti, 1996)*. Biomass burning, dust and anthropogenic pollution aerosols can be mixed
99 along their transport pathways *(Flamant et al., 2018a; Deroubaix et al. 2019)*, resulting in
100 complex interactions between physical and chemical processes and even meteorological
101 feedbacks.
102

103   In West Africa, most of the aerosol–radiation interaction studies focused on optical properties of

104   dust and biomass burning aerosols in remote regions far from major sources of anthropogenic

105   pollution aerosol. They include ground-based and airborne field campaigns such as DABEX (Dust

106   and Biomass Experiment, *Haywood et al., 2008)*, AMMA (Analysis Multidisciplinary of African

107   Monsoon, *Lebel et al., 2010)*, DODO (Dust Outflow and Deposition to the Ocean, *McConnell et*

108   *al., 2008)*, SAMUM-1 and SAMUM-2 (Saharan Mineral Dust Experiment, *Heintzenberg, 2009;*

109   *Ansmann et al., 2011)* and AER-D (AERosol Properties – Dust, *Ryder al. 2018)*. These projects

110   concluded that the influence of both mineral dust and biomass burning aerosols on the radiation

111   budget is significant over West Africa, implying that meteorological forecast and regional/global

112   climate models should include their different radiative effects for accurate forecasts and climate

113   simulations. Over the Sahel region, *Solmon et al. (2008)* have highlighted the high sensitivity of

114   mineral dust optical properties to precipitation changes at a climatic scale. However, the optical

115   properties of aerosols particles in the complex chemical environment of SWA are barely studied.

116   This is partly due to the historically low level of industrial developments of the region. Motivated

117   by the quickly growing cities along the Guinea Coast, the study of transport, mixing, and feedback

118   processes of aerosol particles is therefore very important for better quantification of aerosol

119   radiative impact at the regional scale and improvement of climate and numerical weather

120   prediction models.

121

122   In this context, the DACCIWA (Dynamics-Aerosol-Chemistry-Clouds Interactions in West Africa,

123   *Knippertz et al., 2015b*) campaign, designed to characterize both natural and anthropogenic

124   emissions over SWA, provides important and unique observations of aerosols in a region much

125   more affected by anthropogenic emissions than previously thought. A comprehensive field

126   campaign took place in June–July 2016 including extensive ground-based *(Kalthoff et al., 2018)*

127   and airborne measurements *(Flamant et al., 2018b)*. In this study, we present an overview of *in-*

128   *situ* airborne measurements of the vertical distribution of aerosol particles and their spectral optical

129   properties acquired with the ATR-42 French research aircraft over the Guinea Coast.

130

131   Section 2 presents the flight patterns, instrumentation and data analysis. Section 3 provides an

132   overview of the aerosol microphysical and optical properties. The impact of aging and mixing

133   processes on aerosol optical properties is discussed in section 4 before conclusions are presented in

134   section 5.

135

136 **2. Methodology**

137 **2.1. ATR-42 measurements overview**

138 This analysis focuses on flight missions conducted by the ATR-42 aircraft of SAFIRE (Service des
139 Avions Français Instrumentés pour la Recherche en Environnement - the French aircraft service
140 for environmental research) over the Gulf of Guinea and inland. A full description of flight
141 patterns during DACCIWA is given in *Flamant et al. (2018b)*. Here we present results from 15
142 flights focused on the characterization of anthropogenic pollution, dust and biomass burning
143 plumes. The flight tracks are shown in Figure 1 and a summary of flight information is provided in
144 Appendix 1. The sampling strategy generally consisted of two parts: first, vertical soundings were
145 performed from 60 m up to 8 km above mean sea level (amsl) to observe and identify interesting
146 aerosol layers. Subsequently, the identified aerosol layers were probed with the *in-situ* instruments
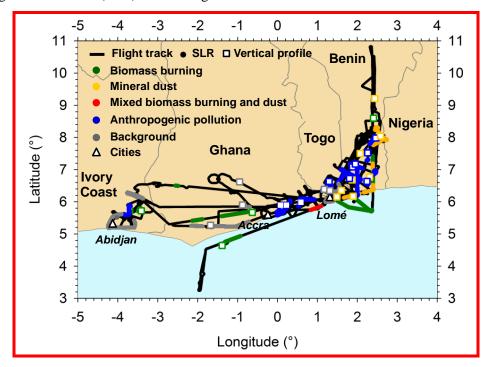147 by straight levelled runs (SLR) at fixed flight altitudes.

148
149 **Figure 1. Tracks of the 15 flights analyzed in this study. The colors indicate aircraft flight**
150 **sampling layers dominated by biomass burning (green), mineral dust (orange), mixed dust-**
151 **biomass burning (red), anthropogenic pollution (blue) and background particles (grey) from**
152 **both vertical profiles (squares) and straight and level runs (SLRs; dots).**
153

154 The ATR-42 aircraft was equipped with a wide variety of instrumentation performing gas and
155 aerosol measurements. The measured meteorological parameters include temperature, dew point
156 temperature, pressure, turbulence, relative humidity, as well as wind speed and direction. Gas
157 phase species were sampled through a rear facing ¼ inch Teflon tube. Carbon monoxide (CO) was

measured using ultra-violet and infrared analysers (PICARRO). The nitric oxide (NO) and nitrogen dioxide (NO_2) measurements were performed using an ozone chemiluminescence instrument (Thermo Environmental Instrument TEi42C with a Blue Light Converter for the NO2 conversion). On-board aerosol instruments sampled ambient air via stainless steel tubing through the Community Aerosol Inlet (CAI). This is an isokinetic and isoaxial inlet with a 50 % sampling efficiency for particles with a diameter of 5 μm *(Denjean et al., 2016).*

The total number concentration of particles larger than 10 nm (*Ntot*) was measured by a condensation particle counter (CPC model MARIE built by University of Mainz; *Mertes, Schröder, and Wiedensohler, 1995; Russell et al., 1996; Wiedensohler et al., 1997*). The aerosol size distribution was measured using an ultra-high sensitivity aerosol spectrometer (UHSAS, DMT), a custom-built scanning mobility sizer spectrometer (SMPS) and an optical particle counter (OPC, GRIMM model 1.109). Instrument calibration was performed with PSL nanospheres and oil particles size-selected by a differential mobility analyser (DMA) for diameters from 90 nm to 20 μm. The SMPS data acquisition system failed after two-third of the campaign and could not be repaired. We found the UHSAS to show false counts in the diameters below 100 nm. Therefore, these channels were disregarded in the data analysis.

The particle extinction coefficient (σ_{ext}) at the wavelength of 530 nm was measured with a cavity attenuated phase shift particle light extinction monitor (CAPS-PMex, Aerodyne Research). The particle scattering coefficients (σ_{scat}) at 450, 550 and 635 nm were measured using a TSI 3-wavelength nephelometer and corrected for angular truncator error in the data inversion procedure using a Mie code (i.e. Section 2.3.1 and Appendix 2). The absorption coefficients (σ_{abs}) at 467, 520 and 660 nm were measured by a Radiance Research Particle Soot Absorption Photometer (PSAP). The PSAP measures changes of filter attenuation due to the collection of aerosol deposited on the filter, which were corrected for the scattering artifacts according to the *Virkkula (2010)* method. Prior to the campaign, the CAPS was evaluated against the combination of the nephelometer and the PSAP. An instrument intercomparison was performed with purely scattering ammonium sulfate particles and with strongly absorbing black carbon particles (BC). Both types of aerosol were generated by nebulizing a solution of the respective substances and size-selected using a DMA. For instrument intercomparison purposes, σ_{ext} from the combination of nephelometer and PSAP was adjusted to that for 530 nm by using the scattering and absorption Ångstrom exponent (*SAE* and *AAE*, respectively). The instrument evaluation showed an excellent accuracy of the CAPS measurements by comparison to the nephelometer and PSAP combination.

2.2. Ancillary products

In order to determine the history of air masses prior aircraft sampling, backward trajectories and satellite images were used. The trajectories were computed using the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) and the National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS) data with 0.5° horizontal resolution for sequences and times of interest. We compared the backward trajectory heights with information of fire burning times (e.g. MODIS Burnt Area Product) and dust release periods (e.g. Meteosat Second Generation (MSG) dust RGB composite images) to assess the aerosol source regions of the investigated air masses. The air masses represented by the trajectory are assumed to obtain their aerosol loading from source regions when the trajectory passes over regions with significant dust activation and/or fire activity at an altitude close to the surface. Trajectory calculations with slightly modified initial conditions with respect to the arrival time, location and altitude were performed to check the reliability of the location of source regions. Uncertainties in this approach, caused by unresolved vertical mixing processes, and by general uncertainties of the trajectory calculations are estimated to be in the range of 15–20 % of the trajectory distance *(Stohl et al., 2002)*.

2.3. Data analysis

In the following, extensive aerosol parameters (concentrations, scattering, absorption and extinction coefficients) are converted to standard temperature and pressure (STP) using $T = 273$ K and $P = 1013.25$ hPa. The STP concentration data correspond to mixing ratios, which are independent of ambient pressure and temperature during the measurement. In the analysis, the data were averaged over sections of SLR with homogeneous aerosol conditions outside of clouds.

2.3.1. Derivation of aerosol microphysical and optical properties

Appendix 2 and Table 1 show the iterative procedure and the equations used to calculate the aerosol microphysical and optical parameters as briefly explained below.

The particle number concentration in the coarse mode (N_{coarse}) was calculated by integrating the OPC size distributions over the range 1 to 5 µm. The signal to noise ratio of the OPC for particles in this size range was higher than 3, which makes the instrument well suited to quantify variations in N_{coarse}. The number concentration of particles in the fine mode (N_{fine}) was obtained as the difference between total number concentration (N_{tot} particle diameter range above 5 nm) measured by the CPC and N_{coarse}.

226

For optical calculations, the 3λ-σ_{abs} from the PSAP were adjusted at the 3 wavelengths measured by the nephelometer using the *AAE* calculated from the 3λ measured σ_{abs}. Once σ_{scat} and σ_{abs} obtained at the same wavelength, an optical closure study estimated the complex refractive index based on optical and size data. Optical calculations were performed using Mie theory, implying a sphericity assumption, because it facilitates a quantitative comparison with past data, mostly using this simplification and because most climate models assume spherical properties. The retrieval algorithm consists of iteratively varying the real part of the complex refractive index *(m)* from 1.33 to 1.60 and the imaginary part of the complex refractive index *(k)* from 0.000 to 0.080 in steps of resolution of 0.001. *m* and *k* were fixed when the difference between calculated values of σ_{scat} and σ_{abs} and measurements was below 1%. Given that the size distribution measured by the UHSAS and the OPC depends on *n*, the optical-to-geometrical diameter conversion was recalculated at each iteration based on the assumed *n*. The resulting number size distributions from SMPS, UHSAS and OPC were parameterized by fitting four log-normal distributions and used as input values in the optical calculations. Once *n* and *k* were obtained at 3λ, we estimated the following optical parameters:

- *SAE* depends on the size of the particles. Generally, it is lower than 0 for aerosols dominated by coarse particles, such as dust aerosols, but it is higher than 0 for fine particles, such as anthropogenic pollution or biomass burning aerosol *(Seinfeld and Pandis, 2006; Schuster et al., 2006)*.

- *AAE* provides information about the chemical composition of atmospheric aerosols. BC absorbs radiation across the whole solar spectrum with the same efficiency, thus it is characterized by *AAE* values around 1. Conversely, mineral dust particles show strong light absorption in the blue to ultraviolet spectrum leading to *AAE* values up to 3 *(Kirchstetter et al., 2004; Petzold et al., 2009)*.

- *SSA* describes the relative importance of scattering and absorption for radiation. Thus, it indicates the potential of aerosols for cooling or warming the lower troposphere.

- *g* describes the probability of radiation to be scattered in a given direction. Values of *g* can range from −1 for entirely backscattered light to +1 for complete forward scattering light.

- *MEE* represents the total light extinction per unit mass concentration of aerosol. The estimates of *MEE* assume mass densities of 2.65 g cm^3 for dust aerosol, 1.35 g cm^3 for biomass burning aerosol, 1.7 g cm^3 for anthropogenic aerosol and 1.49 g cm^3 for background aerosol *(Hess et al., 1998; Haywood et al., 2003a)*.

8

Aerosol parameters	Symbol	λ (nm)	Method
Aerosol microphysical properties			
Total number concentration	N_{tot}	-	Measured by a CPC in the particle diameter range above 5 nm
Number concentration in the coarse mode	N_{coarse}	-	GRIMM size distributions integrated on the range 1 to 5 μm.
Number concentration in the fine mode	N_{fine}	-	Difference N_{tot} and N_{coarse}.
Number size distribution	dN/dlogDp	-	$dN/dlogDp = \Sigma_{i=1}^{4} (N_{tot,i}\, exp(-(logD_p - logD_{p,g,i})^2/(2\, log\, \sigma_i))/(\sqrt{2}\, log\, \sigma_i))$ with $N_{tot,i}$ the integrated number concentration, $D_{p,g,i}$ the geometric median diameter and σ_i geometric standard deviation for each mode i
Volume size distribution	dV/dlogDp	-	$dV/dlogDp = \Sigma_{i=1}^{4} (N_{tot,i}\, D_p^3\, \pi/6\, exp(-(logD_p - logD_{p,g,i})^2/(2\, log\, \sigma_i))/(\sqrt{2}\, log\, \sigma_i))$
Aerosol optical properties			
Scattering coefficient	σ_{scat}	450, 550, 635	Measured by the nephelometer and corrected for truncator error
Absorption coefficient	σ_{abs}	467, 520, 660	Measured by the PSAP and corrected for filter based artefacts
Extinction coefficient	σ_{ext}	530	Measured by the CAPS
Scattering Ångström exponent	SAE	450 to 700	Calculated from the nephelometer measurements : $SAE = -ln\,(\sigma_{scat}(450)/\sigma_{scat}(700))\,/\,ln\,(450/700)$
Absorption Ångström exponent	AAE	440 to 660	Calculated from the PSAP measurements : $AAE = -ln(\sigma_{abs}(467)/\sigma_{abs}(660))\,/\,ln\,(467/660)$
Complex refractive index	n	450, 550, 660	Inversion closure study using Mie theory (Fig. A2) $n(\lambda) = m(\lambda)-ik(\lambda)$
Single scattering albedo	SSA	450, 550, 660	Inversion closure study using Mie theory (Fig. A2) $SSA(\lambda) = \sigma_{scat}(\lambda)/\sigma_{ext}(\lambda)$
Mass extinction efficiency	*MEE*	450, 550, 660	Inversion closure study using Mie theory (Fig. A2) $MEE(\lambda) = \sigma_{ext}(\lambda)/C_m$ with C_m the aerosol mass concentration
Asymmetry parameter	g	450, 550, 660	Inversion closure study using Mie theory (Fig. A2) $g(\lambda) = 1/2 \int_0^{\pi} cos(\Theta)sin(\Theta)P(\Theta,\lambda)d(\Theta)$ with $P(\Theta,\lambda)$ the scattering phase function and Θ the scattering angle.

259   **Table 1. Aerosol microphysical and optical properties derived in this work**

260

2.3.2. Classification of aerosols plumes

Data were screened in order to isolate plumes dominated by anthropogenic pollution from urban emissions, biomass burning and mineral dust particles, resulting in a total number of 19, 12 and 8 genuine plume interceptions, respectively, across the 15 flights. As shown in Figure 2, identification of the plumes was based on a combination of CO and NOx (sum of NO and NO_2) concentrations, as well as *AAE* and *SAE* that have been shown to be good parameters for classifying aerosol types *(Kirchstetter et al., 2004; Petzold et al., 2009)*. The classification was then compared with results from the back trajectory analysis (Figure 3) and satellite images described in section 2.2.
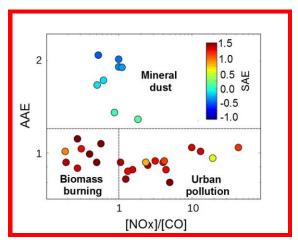

Figure 2. Absorption Ångström Exponent (AAE) as a function of the ratio NOx to CO. The markers are colored according to the Scattering Ångström Exponent (SAE). Classification of mineral dust, biomass burning and urban pollution particles has been added to the figure.

The guidelines for classification are as follows:

- *Anthropogenic pollution*: *SAE* was beyond threshold 0, indicating a large number fraction of small particles in urban plumes, and CO and NOx concentrations 2 times higher than the background concentrations. During the DACCIWA campaign background CO and NOx values were around 180 ppb and 0.28 ppb, respectively. The trajectories show large differences in the flow patterns and source regions with urban plumes originating from the polluted cities of Lomé, Accra and Abidjan. The aircraft sampling over land mostly followed the north-eastward direction (Figure 3d).

- *Biomass burning*: The criteria are the same as for urban pollution plumes except that trajectories track theses plumes back to active fire hotspots as observed by MODIS and the ratio NOx to CO was set below 1. CO and NOx are byproducts of combustion sources but CO is preserved longer along the plume when compared with NOx, which makes the ratio NOx to CO a good indicator for distinguishing fresh anthropogenic pollution plumes from biomass burning plumes transported

288 over long distances *(Wang et al., 2002; Silva et al., 2017)*. During this time of the year, most of the

289 forest and grassland fires were located in Central and Southern Africa (Figure 3a).

290 - *Mineral dust*: *AAE* higher than 1 indicates a large mass fraction of mineral dust and a *SAE* below

291 0 indicate a high effective particle diameter. The source region of the dust loaded air masses was

292 located in the Saharan desert and in the Sahel (Figure 3b).

293 - *Dust and biomass burning mixing:* Combining remote sensing observations and model

294 simulations, *Flamant et al. (2018a)* identified a biomass burning plume mixed with mineral dust.

295 This agrees well with the measured *AAE* of 1.2 and *SAE* of 0.3 observed in this layer. *Menut et al.*

296 *(2018)* have shown that one of the transport pathways of biomass burning aerosols from Central

297 Africa was associated with northward advection towards Chad and then westward displacement

298 linked to the African Easterly Jet. The plume originated from a broad active biomass burning area

299 including Gabon, the Republic of Congo and the Democratic Republic of Congo and passed over

300 areas with strong dust emissions further north within 1–3 days before being sampled by the aircraft

301 (Figure 3c).

302 - *Background:* We refer to background conditions as an atmospheric state in the boundary layer

303 without the detectable influence of mineral dust, biomass burning or local anthropogenic sources.

304 Most back trajectories originated from the marine atmosphere and coastal areas south of the

305 sampling area.

Longitude

(a) (b) (c) (d)

Latitude

Biomass burning
Mixed biomass burning and dust
Mineral dust
Anthropogenic pollution

Figure 3. Backward trajectories for the analyzed aerosol layers. Trajectories date back 10
days for (a), 5 days for (b) and (c), and 1 day for (d).

308

309 ## 3.1. Aerosol vertical distribution

310 Figure 4 shows a statistical analysis of N_{fine}, N_{coarse} and σ_{ext} derived from the *in-situ* measurements

311 of vertical profiles. The aerosol vertical structure is strongly related to the meteorological structure

312 of the atmosphere (see *Knippertz et al., 2017* for an overview of the DACCIWA field campaign*).*

313 Therefore wind vector and potential temperature profiles acquired with the aircraft have been

314 added to Figure 4 as a function of the dominating aerosol composition, introduced in Figure 1. The

315 data from individual vertical profiles were merged into 200 m vertical bins from the surface to 6

316 km amsl. The profiles were calculated using only individual profiles obtained outside of clouds.

317
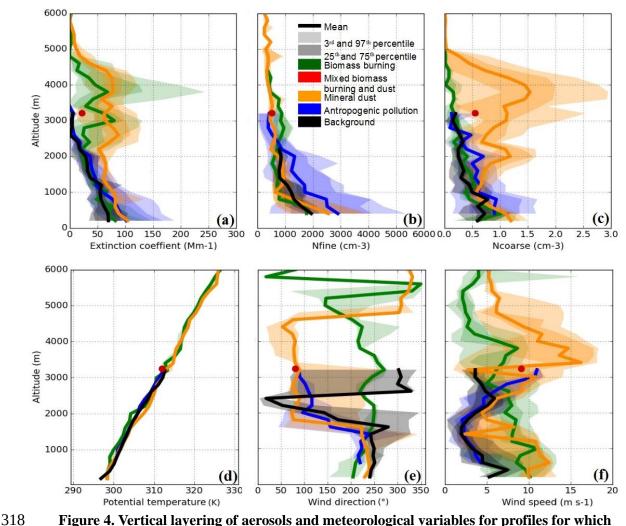

318 **Figure 4. Vertical layering of aerosols and meteorological variables for profiles for which**
319 **aerosols dominated by biomass burning (green), dust (orange), mixed dust-biomass burning**
320 **(red), anthropogenic pollution (blue) and background particles (black) were detected. The**
321 **panels show profiles of (a) the extinction coefficient at 530 nm, (b) the particle number**

327

328   The observed wind profiles highlight the presence of several distinct layers in the lower

329   troposphere. For cases related to dust, urban pollution and background condition, we clearly

330   observe the monsoon layer up to 1.5 km amsl which is characterized by weak to moderate wind

331   speeds (2 to 10 m s^{-1}, the later corresponding to dust cases) and a flow from the southwest (220-

332   250°). In all three air mass regimes, the monsoon layer is topped by a 500 to 700 m deep layer

333   characterised by a sharp wind direction change (from south-westerly to easterly). Weak wind

334   speeds (less than 5 m s^{-1}) are observed in urban pollution and background conditions, while higher

335   wind speeds are observed in the dust cases. Above 2.5 km amsl, the wind speed increases in the

336   urban pollution and dust cases and the wind remains easterly, indicating the presence of the

337   African easterly jet with its core typically farther north over the Sahel (Figure 8 in *Knippertz et al.,*

338   *2017* for the latitudinal variations of the African easterly jet during the DACCIWA field phase).

339   The maximum easterlies are observed in the dust cases slightly below 3.5 km amsl (> 15 m s^{-1}).

340   For the background cases, the wind above the shear layer shifts to north-westerly and remains

341   weak (~i.e. 5 m s^{-1}). Overall, the wind profile associated with the biomass burning cases is quite

342   different from the other three cases, with a flow essentially from the south-southwest below 5 km

343   amsl and higher wind speeds in the lower 2 km amsl than above, and a secondary maximum of 7

344   m s^{-1} at 4 km amsl.

345

346   The vertical distribution of aerosol particles was very inhomogeneous, both across separate

347   research flights and between individual plumes encountered during different periods of the same

348   flight. Measurements of aerosols within this analysis cover a broad geographic region, as shown in

349   Figure 1, which may explain some of the variability. SWA is subject to numerous anthropogenic

350   emission sources (e.g. road traffic, heavy industries, open agriculture fires, etc.) coupled to

351   biogenic emissions from the ocean and forests. These resulting large emissions are reflected in the

352   high variability of σ_{ext}, N_{fine} and N_{coarse} in the lower troposphere over SWA. Below 2.5 km amsl, σ_{ext}

353   showed a large heterogeneity with values ranging from 35 to 188 Mm^{-1} between the 3rd and 97th

354   percentile and a median value of 55 Mm^{-1}. The variability of σ_{ext} values was slightly enhanced near

355   the surface and was correlated to N_{fine} and N_{coarse} which ranged from 443 to 5250 cm^{-3} and from

356   0.15 to 1.6 cm^{-3}, respectively. Maximum surface σ_{ext} was recorded in the anthropogenic pollution

357   plume of Accra where high N_{fine} was sampled. The aerosol vertical profile is strongly modified

358   during biomass burning and dust events. The dust plume extends from 2 to 5 km amsl, and is

359   associated with transport from the dust sources in Chad and Sudan (see Figure 3) with the midlevel

360   easterly flow. The biomass burning plume extends from 1.5 to 5 km amsl and is associated with

361   transport from the southwest in a layer of enhanced wind speed just below 4 km amsl as discussed

362   above. Both layers showed enhanced σ_{ext} with median values of 68 Mm^{-1} (p$_{03}$ = 12 Mm^{-1}; p$_{97}$ = 243

363   Mm^{-1}) in biomass burning plumes and 78 Mm^{-1} (p$_{03}$ = 45 Mm^{-1}; p$_{97}$ = 109 Mm^{-1}) in dust plumes.

364   As expected, the extinction profile was strongly correlated to N_{fine} for biomass burning layers and

365   N_{coarse} for dust layers.

366

367   A prominent feature in the vertical profiles is the presence of fine particles up to 2.5 km amsl

368   outside of biomass burning or dust events. $\sigma_{ext,}$ N_{fine} and N_{coarse} continuously decrease with altitude,

369   most likely due to vertical mixing of local emissions from the surface to higher levels. Therefore,

370   the regional transport of locally emitted aerosols was not limited to the surface but occurred also at

371   higher altitude. Recently, numerical tracer experiments performed for the DACCIWA airborne

372   campaign period have demonstrated that a combination of land–sea surface temperature gradients,

373   orography-forced circulation and the diurnal cycle of the wind along the coastline favor the

374   vertical dispersion of pollutants above the boundary layer during daytime *(Deroubaix et al., 2019;*

375   *Flamant et al., 2018a)*. Because of these complex atmospheric dynamics, aerosol layers transiting

376   over the Gulf of Guinea in the free troposphere could be contaminated by background or urban

377   pollution aerosols from the major coastal cities.

378

379   **3.2. Aerosol size distribution**

380   Figure 5a shows the range of variability of the number and volume size distributions measured

381   during DACCIWA. These are extracted from the SLRs identified in Fig.1. Figure 5b shows the

382   same composite distribution normalized by CO concentration in order to account for differences in
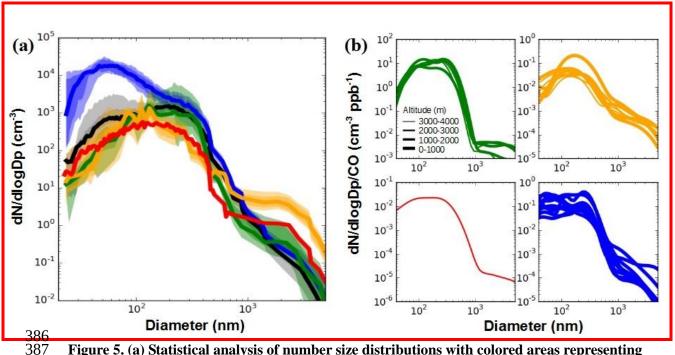
383   the amount of emissions from combustion sources.

384
385

386
387 **Figure 5. (a) Statistical analysis of number size distributions with colored areas representing**
388 **the 3th, 25th, 75th and 97th percentiles of the data and (b) number size distributions normalized**
389 **to CO for plumes dominated by biomass burning (green), dust (orange), mixed dust-biomass**
390 **burning (red), anthropogenic pollution (blue) and background particles (black). In panel b,**
391 **the line thickness is scaled by the altitude of the aerosol plume.**
392

393 Considerable variability in the number concentration of the size distributions, up to approximately

394 2 orders of magnitude, was observed for a large fraction of the measured size range. The size

395 distributions varied both for different aerosol types and for a given aerosol class. This reflects the

396 relative wide range of different conditions that were observed over the region, both in terms of

397 sources, aerosol loading, and lifetimes of ~~the~~ plumes.

398

399 In particular for ultrafine particles with diameters below 100 nm, large differences are observed,

400 with an increase as large as a factor of 50 in urban plumes, which reflects concentration increase

401 from freshly formed particles. Interestingly elevated number concentrations of these small-

402 diameter particles were also observed in some dust layers. Comparing the particle size distribution

403 of the different dust plumes sampled during the field campaign, a variation as large as a factor of

404 20 in the number concentration of ultrafine particles is found (i.e. Figure 4). Their contribution

405 decreased with height as reflected by higher small particle number recorded in dust plumes below

406 2.5 km amsl (Figure 4b and 5b). As the composite urban size distributions showed a relatively

407 similar ultrafine mode centered at 50 nm, dust layers have most likely significant contributions

408 from anthropogenic pollution aerosol freshly emitted in SWA. The ultrafine mode was not

15

409 observed in biomass burning size distributions, even though dust and biomass burning plumes
410 were sampled in the same altitude range. We interpret this observation with dust plumes
411 transported below 2.5 km amsl that were sampled over the region of Savè (8°01'N, 2°29'E; Benin)
412 near the identified urban air mass transported northeastwards from Lomé and/or Accra and which
413 may have collected significant fresh pollution on their way, whereas biomass burning plumes
414 collected at the same altitude and sampled over Ivory Coast south of the Abidjan pollution plumes
415 may not have been affected by significant direct pollution (Figure 1).

416

417 The accumulation mode was dominated by two modes centered at $D_{p,g}$ ~ 100 and 230 nm
418 depending on the aerosol plume. The particle size distributions for biomass burning plumes were
419 generally dominated by an accumulation mode centered at $D_{p,g}$ ~ 230 nm. Despite the relative wide
420 range of sources and lifetimes of the biomass burning plumes sampled throughout the campaign
421 (Figure 3), the $D_{p,g}$ in the accumulation mode showed little variation ($D_{p,g}$ from 210 to 270 nm)
422 between the plumes. Similarly, previous field studies found accumulation mode mean diameters
423 from 175 to 300 nm for aged biomass burning plumes, regardless of their age, transport time and
424 source location *(Capes et al., 2008; Janhäll et al., 2010; Weinzierl et al., 2011; Sakamoto et al.,*
425 *2015; Carrico et al. 2016)*. The coagulation rate can be very high in biomass burning plumes and
426 can shape the size distribution over a few hours (*Sakamoto et al., 2016*). It is worth noting that in
427 the biomass burning and dust size distributions there is a persistent particle accumulation mode
428 centered at ~100 nm that exceeds the amount of particles centered at 230 nm in some layers. This
429 small mode is unlikely to be related to long-range transport of biomass burning and Saharan dust
430 emissions, as it would be expected that particles in this size range would grow to larger particles
431 through coagulation relatively quickly. As similar concentrated accumulation modes of particles
432 have been observed in background plumes, it suggests the entrainment of background air from the
433 boundary layer in dust and biomass burning plumes. This is supported by remote sensing
434 observations on 2 and 5 July 2016 *(Flamant et al., 2018a; Deroubaix et al., 2019)*.

435

436 The number concentration of large super-micron particles was strongly enhanced in the mineral
437 dust layers. The peak number concentration displayed a broad shape at $D_{p,g}$ ~ 1.8 µm, which is
438 comparable to literature values of other long-range transported dust aerosols *(Weinzierl et al.,*
439 *2011; Ryder et al., 2013; Denjean et al., 2016; Liu et al., 2018)*. The super-micron mode of the
440 dust plume is expected to be impacted by the mixing with other particles in case of an internal
441 mixing, which should somewhat increase the particle size. The relatively homogeneous $D_{p,g}$ in the
442 coarse mode of dust reported here ($D_{p,g}$ from 1.7 to 2.0 µm) suggests low internal mixing with

443 other atmospheric species. Besides, the volume size distribution in urban plumes showed
444 significant presence (~ 65% of the total aerosol volume) of large particles with diameters of ~ 1.5
445 – 2 μm, which were also observed in background conditions. We measured AAE in the range 0.7-
446 1 in anthropogenic pollution plumes (Figure 2), which suggests negligible contribution of mineral
447 dust in these plumes. This coarse mode has most likely significant contributions from sea salt
448 particles, as plumes arriving from the cities were transported at low altitude over the ocean (Fig.
449 3).

450

3.3. Aerosol optical properties

452 *SSA* is one of the most relevant intensive optical properties because it describes the relative
453 strength of the aerosol scattering and absorption capacity and is a key input parameter in climate
454 models *(Solmon et al., 2008)*. Figure 6 shows the spectral *SSA* for the different SLRs considered in
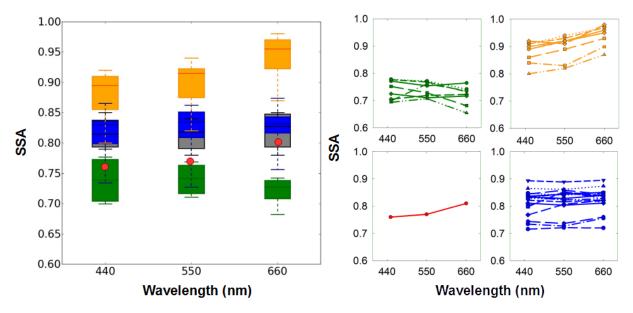455 this study.

456

Figure 6. (a) Statistical analysis of single scattering albedo at 450, 550 and 660 nm for plumes dominated by biomass burning (green), dust (orange), mixed dust-biomass burning (red), anthropogenic pollution (blue) and background particles (black). The boxes enclose the 25th and 75th percentiles, the whiskers represent the 5th and 95th percentiles and the horizontal bar represents the median. (b) Spectral SSA for the different individual plumes considered in this study. The mixed dust-biomass burning plume is represented by a dot because it is derived from measurements during only one SLR.

464

465 The highest absorption (lowest *SSA*) at all three wavelengths was observed for biomass burning
466 aerosols. *SSA* values ranged from 0.69–0.78 at 440 nm, 0.71–0.77 at 550 nm and 0.65–0.76 at 660

nm. This is on the low side of the range of values (0.73–0.93 at 550 nm) reported over West Africa during DABEX for biomass burning plumes mixed with variable proportion of mineral dust *(Johnson et al., 2008)*. No clear tendency was found for the spectral dependence of *SSA*, which in some of the cases decreased with wavelength and in others were very similar to each other at all three wavelengths. ~~The strongest spectral dependence of SSA was observed for biomass burning plumes with the lowest absorption (highest SSA) at 440 nm. Laboratory experiments have shown that strongly absorbing biomass burning particles tend to have a weak wavelength dependent absorption, while weakly light-absorbing particles tend to have a strong wavelength dependent absorption *(McMeeking et al., 2014; Zhai et al., 2017)*, which is consistent with results in this study.~~

SSA values of anthropogenic pollution aerosols were generally intermediate in magnitude with median values of 0.81 at 440 nm, 0.82 at 550 nm and 0.82 at 660 nm. Our data show that the value of *SSA* varied significantly for the different plumes. Some pollution aerosols absorb almost as strongly as biomass burning aerosols with *SSA(550nm)* values as low as 0.72, whereas the highest *SSA(550nm)* value observed was 0.86. In addition, the absorption properties of urban aerosol varied greatly between the sampled plumes for smoke of apparent same geographic origin. For example, we measured *SSA(550nm)* values from 0.72 to 0.82 in the Accra pollution outflow. The variability in SSA values may be due to the possible contribution of emissions from different cities to the sampled pollution plumes (*Deroubaix et al., 2019)*, thus having different combustion sources and chemical ages. Past in-situ measurements of aerosol optical properties over SWA cities appear, unfortunately, to be absent from literature. However, the flat spectral dependence of SSA appears to be anomalous for anthropogenic pollution aerosols, as SSA ~~is~~ has been shown to ~~xpected~~ decrease with increasing wavelength for a range of different urban pollution plumes over European, American and Asian cities (*Dubovick et al., 2002; Di Biagio et al., 2016; Shin et al., 2019).* ~~As during DACCIWA SSAs of anthropogenic pollution aerosols reached similar values to those of background aerosols, it suggests a large contribution of the latter to the aerosol optical properties of the mixture.~~

The magnitude of *SSA* increased at the three wavelengths when dust events occurred. ~~It is important to note that the measurements exclude a significant portion of the coarse-mode aerosol due to poor inlet passing efficiency of larger aerosol particles (a 50% size cut around 5 μm), which may result in an overestimate of SSA. Despite this limitation, our measurements are comparable to one another and to previous *in-situ* measurements by taking into account the sampling inlet.~~ Large

variations in *SSA* were obtained with values ranging from 0.76–0.92 at 440 nm, 0.81–0.94 at 550 nm and 0.81–0.97 at 660 nm. The measurement of *SSA* is highly dependent on the extent to which the coarse mode is measured behind the aerosol sampling inlet. *Denjean et al. (2016)* found that the absolute error associated with *SSA, g and MEE* of dust aerosols due to the CAI inlet is in the range covered by the measurement uncertainties. However, different aerosol inlet systems were used during previous field campaigns, which makes comparison of our results with previous measurements difficult. Overall, compared with the literature for transported dust, lower values were obtained in the present study for few cases. For example, *Chen et al. (2011)* reported SSA(550 nm) values of 0.97±0.02 during NAMMA (a part of AMMA operated by NASA) using an inlet with a comparable sampling efficiency. The lower values from DACCIWA reflect inherently more absorbing aerosols in some dust plumes. In contrast to fire plumes, the SSA of dust aerosol showed a clear increasing trend with wavelength. This behavior is likely due to the domination of large particles in dust aerosol, which is in agreement to similar patterns observed in dust source regions (*Dubovik et al., 2002)*. Moreover, an increase of SSA is observed with wavelength for mixed dust-smoke aerosol, suggesting that the aerosol particles were predominantly from dust, albeit mixed with a significant loading of biomass burning.

		SSA(450)	SSA(550)	SSA(660)	MEE(450)	MEE(550)	MEE(660)	g(450)	g(550)	g(660)	SAE
Mineral dust	median	0.88	0.90	0.93	0.74	0.68	0.66	0.74	0.72	0.69	-0.35
	3th	0.82	0.82	0.86	0.38	0.38	0.39	0.69	0.67	0.65	-0.56
	25th	0.85	0.87	0.90	0.43	0.43	0.43	0.73	0.72	0.67	-0.48
	75th	0.91	0.93	0.96	0.94	0.85	0.85	0.75	0.74	0.72	-0.25
	97th	0.92	0.95	0.97	1.57	1.37	1.21	0.78	0.76	0.72	-0.12
Biomass burning	median	0.74	0.76	0.72	1.91	1.62	1.34	0.69	0.68	0.61	1.07
	3th	0.70	0.72	0.66	0.94	1.45	1.22	0.64	0.65	0.59	0.59
	25th	0.70	0.76	0.71	1.67	1.48	1.27	0.69	0.65	0.60	0.83
	75th	0.77	0.77	0.74	1.86	1.65	1.55	0.72	0.68	0.62	1.15
	97th	0.78	0.77	0.76	2.38	1.92	1.58	0.73	0.68	0.63	1.64
Mixed dust-Biomass burning	median	0.76	0.77	0.81	1.58	1.40	1.30	0.73	0.66	0.64	0.38
	3th	-	-	-	-	-	-	-	-	-	-
	25th	-	-	-	-	-	-	-	-	-	-
	75th	-	-	-	-	-	-	-	-	-	-
	97th	-	-	-	-	-	-	-	-	-	-
Anthropogenic Pollution	median	0.83	0.84	0.85	2.60	2.49	1.90	0.60	0.61	0.62	0.75
	3th	0.78	0.79	0.81	0.70	1.24	0.54	0.60	0.59	0.54	0.30
	25th	0.80	0.82	0.83	2.14	2.25	1.53	0.62	0.60	0.56	0.65
	75th	0.84	0.86	0.85	3.51	2.96	2.53	0.69	0.62	0.67	0.89
	97th	0.87	0.88	0.90	3.70	4.83	2.74	0.73	0.64	0.70	0.94

518
519   **Table 2. Single scattering albedo, extinction mass efficiency, asymmetry parameter, single**
520   **scattering albedo and scattering Ångstrom exponent for the dominant aerosol classification.**
521
522 As shown in Table 2, the observed variability of *SSA* reflects a large variability for *MEE* at 550nm,

523 which spans a wide range from 0.38 to 1.37 m^2 g^{-1}, 1.45 to 1.92 m^2 g^{-1} and 1.24 to 4.83 m^2 g^{-1} for

524 dust, biomass burning for anthropogenic polluted aerosols, respectively. MEE is heavily

525 influenced by the mass concentrations in the accumulation mode where the aerosol is optically

526 more efficient in extinguishing radiation. We found MEE to be positively correlated with SAE (not

527 shown), which was expected because of the dependence of MEE on particle size. In contrast, the

528 values of *g* appear to differ only little between the sampled plumes for a given aerosol class. We

529 found *g* in the range of 0.67–0.76 for dust, 0.65–0.68 for biomass burning and 0.59–0.64 for

530 anthropogenic polluted aerosols at 550 nm. *g* values in dust plumes were high, which is expected

531 due to the presence of coarse particles contributing to forward scattering.

532

533 This analysis includes sampled aerosols originating from different source regions and having

534 undergone different aging and mixing processes, which could explain some of the variability. The

impact of these factors on the magnitude and spectral dependence of optical parameters will be investigated in the following section.

4. Discussion

4.1. Contribution of local anthropogenic pollution on aerosol absorption properties

Figure 7 shows the vertical distribution of *SSA, SAE* and *NOx* mixing ratio for the dominant aerosol classification. We exclusively consider measurements acquired during SLRs, since only during these phases the whole set of aerosol optical properties were measured. In dust plumes, if we exclude the case of mixing with biomass burning aerosol, SSAs were fairly constant above 2.5 km amsl with values ranging between 0.90 and 0.93 at 550 nm, in agreement with values reported over dust source regions *(Schladitz et al., 2009; Formenti et al., 2011; Ryder et al., 2013, 2018)*. Despite the range of sources identified during DACCIWA, dust absorption properties do not seem to be clearly linked to particle origin or time of transport. Aerosols were more absorbing within the low-altitude dust plumes with SSA values dropping to 0.81. *SAE* values exhibited simultaneously a sharp increase close to zero below 2.5 km amsl. This is consistent with a higher concentration of fine particles, though the value of *SAE* was still much lower than for pollution or background aerosol (i.e. where it is typically > 0.2), which means that scattering was still dominated by larger particles. ~~The decrease in NOx with height further indicates the concurrent influence by emissions from pollution sources in the low-altitude dust plumes.~~ Based on these observations, the strong variation in the light-absorption properties of dust-dominated aerosol over SWA could be attributed to the degree of mixing into the vertical column with either freshly emitted aerosols from urban/industrial sources or long-range transported biomass burning aerosol.

One of the critical factors in the calculation of aerosol direct and semi-direct radiative effects is the mixing state of the aerosols, which can significantly affect absorbing properties. There were no direct observational constraints available on this property during the DACCIWA airborne campaign. However, we investigated the probable aerosol mixing state by calculating composite SSA from the aerosol size distribution. On the basis of Figure 5, dust size distribution showed only minor discrepancies in the mean and standard deviation of the coarse mode but significant differences in the balance between fine and coarse modes, which suggests low internal mixing of dust with other atmospheric species. The size distributions of mixed dust-pollution have been deconvoluted by weighting the size distributions of mineral dust and anthropogenic pollution aerosol averaged over the respective flights. This assumes that dust was externally mixed with the anthropogenic pollution particles and assumes a homogeneous size distribution for the dust and

569   anthropogenic pollution aerosol throughout a flight. σ_{scat} and σ_{abs} were then calculated using Mie

570   theory from each composite size distributions and the corresponding k and m. The refractive

571   indices at 550 nm were assumed to be 1.52-0.002i and 1.60-0.040i for dust and anthropogenic

572   pollution particles, respectively, which are the mean values deduced from the data inversion

573   procedure (i.e. section 2.3.1) throughout the campaign. The resulting σ_{scat} and σ_{abs} were used to

574   calculate a composite *SSA*. A similar calculation was performed for the mixed dust-biomass

575   burning case. Figure 7 shows a good agreement with the observations of *SSA*, implying that

576   external mixing appears to be a reasonable assumption to compute aerosol direct and semi-direct

577   radiative effects in these dust layers for modeling applications. This is consistent with the filter

578   analysis performed during AMMA and SAMUM-2, which did not reveal any evidence of internal

579   mixing in both mixed dust-biomass burning and dust-anthropogenic pollution layers *(Chou et al.,*

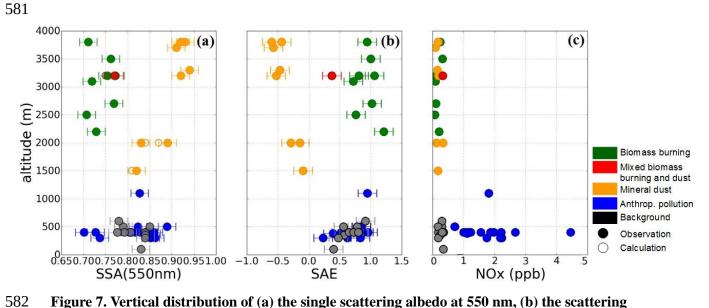580   *2008; Lieke et al., 2011; Petzold et al., 2011).*

581

582 **Figure 7. Vertical distribution of (a) the single scattering albedo at 550 nm, (b) the scattering**
583 **Ångstrom exponent and (c) NOx mixing ratio for the dominant aerosol classification. In**
584 **panel (a), full circles represent *SSA* measurements and empty circles represent composite**
585 ***SSA* calculated by deconvoluting size distribution measurements in mixed dust layers and**
586 **assuming an external mixing state.**

587

588 Figure 7 indicates markedly different processes affecting optical properties of biomass burning

589 aerosols. *SSA*, *SAE* and NOx of biomass burning plumes did not significantly vary with height

590 from 2.2 to 3.8 km amsl. Moreover, the size distribution of biomass burning aerosols for the

591 observed cases did not show significant contribution of ultrafine particles (Figure 5). These

592 observations seem to indicate that the absorption properties of biomass burning plumes were not

593 affected by direct pollution emissions, probably due to the remote location of the sampled biomass

594 ~~burning plumes as discussed in section 3.2. The optical properties of aerosols are determined by~~
595 ~~either the aerosol chemical composition, the aerosol size distribution, or both. Changes in the size~~
596 ~~distribution of biomass burning aerosol due to coagulation and condensation have been shown to~~
597 ~~alter the *SSA*, as particles increase towards sizes for which scattering is more efficient *(Laing et al.,*~~
598 ~~*2016).* Variations in particle chemical composition, caused by source emissions and aging~~
599 ~~processes associated with gas to particle transformation and internal mixing, has been shown to~~
600 ~~change the *SSA (Abel et al., 2003; Petzold et al., 2011).*~~

601

602 In the boundary layer, the similar *SSA* and *SAE* in anthropogenic pollution and background plumes
603 suggests that background aerosol may be rather called background pollution originating from a
604 regional background source in the far field. Our analysis of the spectral dependence of *SSA*
605 showed no apparent signature of anthropogenic pollution aerosols (see section 3.3) despite a strong
606 increase of aerosol number concentrations in air masses crossing urban centers (see section 3.2).
607 This can be explained by two factors: First, the majority of accumulation mode particles were
608 present in the background, while the large proportion of aerosols emitted from cities resided in the
609 ultrafine mode particles that have less scattering efficiencies (Figure 5). Second, large amounts of
610 absorbing aerosols in the background can minimize the impact of further increase of absorbing
611 particles to the aerosol load. ~~We did not find any correlation between the values of *SSA* and their~~
612 ~~spectral dependence, which suggests that the variability in *SSA* cannot be attributed to different~~
613 ~~contributions of marine aerosol in pollution plumes.~~ The high CO values (~180 ppb) observed in
614 background conditions further indicates a strong contribution of combustion emissions at the
615 surface. Recent studies showed a large background of biomass burning transported from the
616 Southern Hemisphere in SWA that dominated the aerosol chemical composition in the boundary
617 layer *(Menut et al., 2018; Haslett et al., 2019).* The high absorbing properties (*SSA*~0.81 at
618 550nm) and the presence of particles both in the accumulation and super-micron modes (i.e.
619 section 3.2.) in background plumes are consistent with being a mixture of aged absorbing biomass
620 burning and Atlantic marine aerosol. ~~Moreover *SSA* of background aerosol was lower than~~
621 ~~previously reported over the Southern Atlantic (Ascension Island) outside the fire season in~~
622 ~~Central Africa *(Zuidema al., 2018),* which supports this conclusion.~~ These results highlight that
623 aerosol optical properties at the surface were dominated by the widespread biomass burning
624 particles at regional scale.

625

626 **4.2. Aging as a driver for absorption enhancement of biomass burning aerosol**

627	The optical properties of aerosols are determined by either the aerosol chemical composition, the

628	aerosol size distribution, or both. Changes in the size distribution of biomass burning aerosol due

629	to coagulation and condensation have been shown to alter the *SSA*, as particles increase towards

630	sizes for which scattering is more efficient *(Laing et al., 2016)*. Variations in particle chemical

631	composition, caused by source emissions and aging processes associated with gas-to-particle

632	transformation and internal mixing, has been shown to change the *SSA (Abel et al., 2003; Petzold*

633	*et al., 2011)*.

634

635	In order to determine the contributions from size distribution and chemical composition to the

636	variation of *SSA* in biomass burning plumes, *SSA* is presented as a function of *SAE* and *k* in Figure

637	7a and b, respectively. *k* was iteratively varied to reproduce the experimental scattering and

638	absorption coefficients, as described in section 2.3.1. It appears that the variation of the size

639	distribution (assessed via *SAE* in Figure 8a) had minimal impact in determining the variability of

640	*SSA*. Thus, the observations suggest that there was no effect of plume age on the size distribution,

641	consistent with previous observations of size distribution in aged North American biomass

642	burning plumes *(Sakamoto et al., 2015; Carrico et al., 2016; Laing et al., 2016)*. Using a

643	Lagrangian microphysical model, *Sakamoto et al. (2015)* have shown a rapid shift to larger sizes

644	for biomass burning plumes within the first hours of aging. Less drastic but similarly rapid growth

645	by coagulation was seen by *Capes et al. (2008)* in their box model. Given that the biomass burning

646	plumes sampled during DACCIWA had more than 5 days in age, the quick size-distribution

647	evolution within the early plume stages might explain the limited impact of the size distribution on
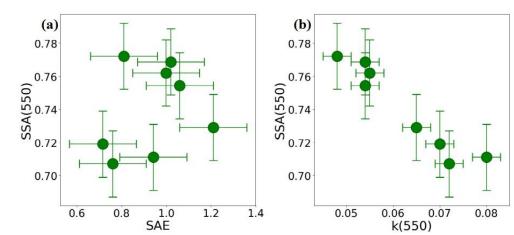
648	the *SSA*.

649

Figure 8. Contribution to single scattering albedo (a) from particle size (assessed via *SAE*) and (b) from composition (assessed via *k*) in biomass burning plumes.

In contrast, Figure 8b shows that there was a consistent decrease in *SSA* with increasing *k*, although there is some variability between the results from different plumes. The observed variability of *SSA* is reflected in a large variability of *k*, which is estimated to span the large range 0.048–0.080 at 550 nm. k depends both on the aerosol chemical composition and size distribution *(Mita and Isono, 1980)*. Given that *SSA* was found to be independent of the aerosol size distribution (Figure 8a), Figure 8b suggests that *SSA* variability was strongly influenced by the variability in composition of biomass burning aerosol, implying a high contribution for light-absorbing particles. ~~Although there is some variability between the results from different plumes, overall there was a consistent decrease in *SSA* with increasing *k*, implying a high contribution for light-absorbing particles. The observed variability of *SSA* is reflected in a large variability of *k*, which is estimated to span in the large range 0.048–0.080 at 550 nm.~~ No clear tendency was found for the wavelength dependence of *k*, which in some of the cases increases with wavelength and in others decreases (not shown). Correspondingly, values of *AAE* in biomass burning plumes ranged from 0.9 to 1.1 with a median value of 1.0. Theoretically, fine-mode aerosol with absorption determined exclusively by BC would have *AAE* equal to 1.0, since BC is expected to have a spectrally constant *k (Bond et al., 2013)*. Therefore, the low *SSA* values observed in biomass burning plumes over SWA and the small spectral variation of *k* both suggest that BC is the dominant absorber in the visible and near-IR wavelengths for these biomass burning aerosols.

Compared with past in-situ measurements of aged biomass burning aerosol, *SSA* values over SWA (0.71–0.77 at 550 nm) are at the lower end of those reported worldwide (0.73–0.99 at 550 nm) *(Maggi et al., 2003; Reid et al. 2005; Johnson et al., 2008; Corr et al. 2012; Laing et al. 2016).*

25

675   This can be attributed in part to the high flaming versus smoldering conditions of African smoke
676   producing more BC particles *(Andreae and Merlet, 2001; Reid et al., 2005)*, which inherently have
677   low *SSA* compared to other regions *(Dubovick et al*. 2002). However, *SSA* values over SWA are
678   significantly lower than the range reported near emission sources in sub-Saharan Africa and over
679   the southeast Atlantic, where values span over 0.84–0.90 at 550 nm *(Haywood et al., 2003b;*
680   *Pistone et al., 2019)*. Recent observations carried out on Ascension Island to the south-west of the
681   DACCIWA region showed that smoke transported from Central and South African fires can be
682   very light absorbing over the July-November burning season but *SSA* values were still higher
683   (0.80±0.02 at 530 nm; *Zuidema et al., 2018*) than those reported over SWA. A possible cause of
684   the lower *SSA* in SWA is that Ascension Island is much closer to the local sources and the aerosol
685   is therefore less aged.

686

687   Currently there are few field measurements of well-aged biomass burning emissions. Our
688   knowledge of biomass burning aerosol primarily comes from laboratory experiments and near-
689   field measurements taken within a few hours of a wildfire *(Abel et al., 2003; Yokelson et al., 2009;*
690   *Adler et al., 2011; Haywood et al., 2003b; Vakkari et al., 2014; Zhong and Jang, 2014; Forrister*
691   *et al., 2015; Laing et al., 2016; Zuidema et al., 2018)*. Exception made of the study by *Zuidema et*
692   *al. (2018)* over the southeast Atlantic, it is generally found that the aged biomass burning aerosol
693   particles are less absorbing than freshly emitted aerosols due to a combination of condensation of
694   secondary organic species and an additional increase in size by coagulation. This is in contrasts to
695   our results showing that *SSA* of biomass burning aerosols were significantly lower than directly
696   after emission and that the evolution of *SSA* occurred long time after emission.

697

698   There are three possible explanations for these results. First, one must consider sample bias. As
699   regional smoke ages, it can be enriched by smoke from other fires that can smolder for days
700   producing large quantities of non-absorbing particles, thereby increasing the mean *SSA (Reid et al.,*
701   *2005; Laing et al., 2016)*. However, during DACCIWA, biomass burning plumes were transported
702   over the Atlantic Ocean and were probably less influenced by multiple fire emissions. Second,
703   there is evidence that fresh BC particles become coated with sulfate and organic species as the
704   plume ages in a manner that enhances their light absorption *(Lack et al., 2012; Schwarz et al.,*
705   *2008)*. Finally, organic particles produced during the combustion phase can be lost during the
706   transport through photobleaching, volatilization and/or cloud-phase reactions *(Clarke et al., 2007;*
707   *Lewis et al., 2008; Forrister et al., 2015)*, which is consistent with the low *SSA* and *AAE* values we
708   observed. Assessing whether these aging processes impact the chemical components and

709 henceforth optical properties of transported biomass burning aerosol would need extensive
710 investigation of aerosol chemical composition that will be carried out in a subsequent paper.
711

712 **5. Conclusions**

713 This paper provides an overview of *in-situ* airborne measurements of vertically resolved aerosol
714 optical properties carried out over SWA during the DACCIWA field campaign in June-July 2016.
715 The peculiar dynamics of the region lead to a chemically complex situation, which enabled
716 sampling various air masses, including long-range transport of biomass burning from Central
717 Africa and dust from Sahelian and Saharan sources, local anthropogenic plumes from the major
718 coastal cities, and mixtures of these different plumes. This work fills a research gap by providing,
719 firstly, key climate relevant aerosol properties *(SSA, MEE, g, SAE, AAE)* and secondly,
720 observations of the impact of aging and mixing processes on aerosols optical properties.

721

722 The aerosol vertical structure was very variable and mostly influenced by the origin of air mass
723 trajectories. While aerosol extinction coefficients generally decreased with height, there were
724 distinct patterns of profiles during dust and biomass burning transport to SWA. When present,
725 enhanced values of extinction coefficient up to 240 Mm^{-1} were observed in the 2–5 km amsl range.
726 These elevated aerosol layers were dominated by either dust or biomass burning aerosols, which is
727 consistent with what would be expected on the basis of the atmospheric circulations during the
728 monsoon season *(McConnell et al., 2008; Knippertz et al., 2017)*. However, during one flight a
729 mixture of dust and biomass burning was found in a layer at around 3 km amsl, implying that there
730 may be substantial variability in the idealized picture. In the lower troposphere, the large
731 anthropogenic pollution plumes extended as far as hundreds of kilometers from the cities emission
732 sources and were not limited to the boundary layer but occurred also at higher levels up to 2.5 km
733 amsl, which is explained by vertical transport and mixing processes, partly triggered by the
734 orography of SWA *(Deroubaix et al., 2019; Flamant et al., 2018a)*. The analysis of the aerosol size
735 distributions, *SAE* and NOx suggests a strong mixing of dust with anthropogenic pollution
736 particles in dust layers transported below 2.5 km amsl, whereas biomass burning plumes that were
737 transported more northward were not affected by this mixing. Both transport pathways and vertical
738 structures of biomass burning and dust plumes over SWA appear to be the main factors affecting
739 the mixing of anthropogenic pollution with dust and biomass burning particles.

740

741 The aerosol light absorption in dust plumes was strongly enhanced as the result of this mixing. We
742 find a decrease of *SSA(550nm)* from 0.92 to 0.81 for dust affected by anthropogenic pollution

743   mixing compared to the situation in which the dust plumes moved at higher altitudes across SWA.

744   Comparison of the particle size distributions of the different dust plumes showed a large

745   contribution of externally mixed fine mode particles in mixed layers, while there was no evidence

746   for internal mixing of coarse particles. Concurrent optical calculations by deconvoluting size

747   distribution measurements in mixed layers and assuming an external mixing state allowed to

748   reproduce the observed *SSA*s. This implies that an external mixing would be a reasonable

749   assumption to compute aerosol direct and semi-direct radiative effects in mixed dust layers.

750

751   Despite a strong increase of aerosol number concentration in air masses crossing urban

752   conglomerations, the magnitude of the spectral *SSA*s was comparable to the background.

753   Enhancements of light absorption properties were seen in some pollution plumes, but were not

754   statistically significant. A persistent spectral signature of biomass burning aerosols in both

755   background and pollution plumes highlights that the aerosol optical properties in the boundary

756   layer were strongly affected by the ubiquitous biomass burning aerosols transported from Central

757   Africa *(Menut et al., 2018; Haslett et al., 2019).* The large proportion of aerosols emitted from the

758   cities of Lomé, Accra and Abidjan that resided in the ultrafine mode particles have limited impact

759   on already elevated amounts of accumulation mode particles having a maximal absorption

760   efficiency. As a result, in the boundary layer, the contribution from local city emissions to aerosol

761   optical properties were of secondary importance at regional scale compared with this large

762   absorbing aerosol mass. While local anthropogenic emissions are expected to rise as SWA is

763   currently experiencing major economic and population growth, there is increasing evidence that

764   climate change is increasing the frequency and distribution of fire events *(Joly et al., 2015).* In

765   terms of future climate scenarios and accompanying aerosol radiative forcing, whether the large

766   biomass burning events that occur during the monsoon season would limit the radiative impact of

767   increasing anthropogenic emissions, remains an open and important question.

768

769   The *SSA* values of biomass burning aerosols transported in the free troposphere were very low

770   (0.71–0.77 at 550 nm) and have only rarely been observed in the atmosphere. The variability in

771   *SSA* was mainly controlled by the variability in aerosol composition (assessed via k) rather than by

772   variations in the aerosol size distribution. Correspondingly, values of *AAE* ranged from 0.9 to 1.1,

773   suggesting that BC particles were the dominant absorber in the visible for these biomass burning

774   aerosols. In recent years the southern Atlantic Ocean, especially the area of the west coast of

775   Africa, became an increasing focus in the research community, through the ORACLES/LASIC

776   (ObseRvations of Aerosols above CLouds and their intEractionS/Layered Atlantic Smoke

777   Interactions with Clouds), AEROCLO-sA (AErosol RadiatiOn and CLOuds in Southern Africa –

778   AEROCLO-SA) and CLARIFY (Cloud and Aerosols Radiative Impact and Forcing) projects

779   *(Zuidema et al.; 2016; Zuidema et al.; 2018; Formenti et al.; 2019)*. Comparison with literature

780   showed a consistent picture of increasing absorption enhancement of biomass burning aerosol

781   from emission to remote locations. Further, the range of *SSA* values over SWA was slightly lower

782   than that reported on Ascension Island to the south-west of the DACCIWA region, which

783   underscores that the evolution of *SSA* occurred long time after emission. While the mechanism

784   responsible for this phenomenon warrants further study, our results support the growing body of

785   evidence that the optical parameters used in regional/global climate modeling studies, especially

786   absorption by biomass burning aerosols, have to be better constrained using these recent

787   observations to determine the direct and semi-direct radiative effects of smoke particles over this

788   region *(Mallet et al. 2019)*. In particular and regarding the very high absorbing properties of

789   smoke, specific attention should be dedicated to the semi-direct effect of biomass burning aerosols

790   at the regional scale and its relative contribution to the indirect radiative effect.

791

792   We believe the set of DACCIWA observations presented here is representative of the regional

793   mean and variability in aerosol optical properties that can be observed during the monsoon season

794   over SWA, as the main dynamical features were in line with climatology *(Knippertz et al., 2017)*.

795   This is why results from the present study will serve as input and constraints for climate modeling

796   to better understand the impact of aerosol particles on the radiative balance and cloud properties

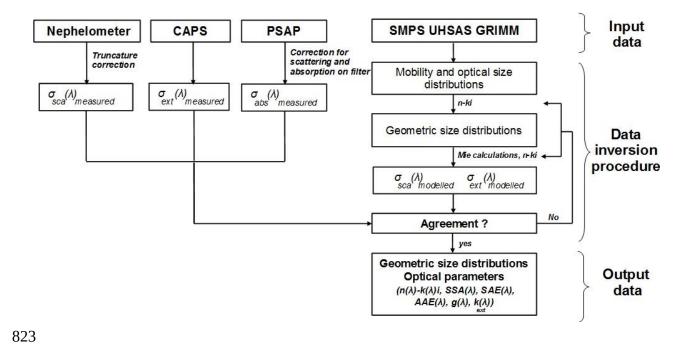797   over this region and also will substantially support remote sensing retrievals.

819 **Appendix 1. Summary of flight information. All flights were conducted during 2016.**

Flight number	Date	Take off time (UTC)	Landing time (UTC)	Events observed
F17	29 June	14:17	17:10	Export of pollution from Lomé
F18	30 June	12:52	16:29	Export of pollution from Lomé
F19	1 July	10:35	14:06	Export of pollution from Accra
F20	2 July	09:53	13:21	Export of pollution from Lomé Dust outbreak
F21	2 July	15:04	18:29	Export of pollution from Lomé Biomass burning outbreak Mixed dust-biomass burning outbreak
F22	3 July	09:54	13:29	Export of pollution from Lomé
F24	6 July	07:17	11:03	Export of pollution from Abidjan
F27	8 July	05:52	09:28	Export of pollution from Accra
F28	8 July	10:53	14:22	Dust outbreak
F29	10 July	10:31	14:11	Export of pollution from Lomé Dust outbreak
F30	11 July	07:19	11:01	Export of pollution from Abidjan Biomass burning outbreak
F31	11 July	13:48	16:42	Biomass burning outbreak
F32	12 July	13:56	17:20	Export of pollution from Accra
F33	13 July	12:40	16:11	Biomass burning outbreak
F35	15 July	09:32	13:00	Export of pollution from Lomé

820

821   **Appendix 2. Data inversion procedure to calculate the aerosol microphysical and optical**

822   **parameters.**

823

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