Anonymous Referee #1

General Comments

This study analyzes aerosol properties and PM2.5 concentrations using sunphotometers (handheld and automatic-AERONET) and samples at selected sites (coastal and inland) in the south west Africa. This region is very interesting for aerosol studies due to seasonally changing meteorological conditions i.e, Harmattan north dry winds during winter and monsoon humid south winds in summer. This reverse atmospheric circulation, along with the influence of dust events and forest/agricultural fires result in contrasting aerosol types and properties. From this point of view, the current work may have its own importance, taking also into consideration the very good correlation between satellite AODs and PM2.5 concentrations over selected sites that allows the PM2.5 estimates at a long-term period and examination of the trends. All the above issues, along with a rough classification of aerosol types are examined in this paper. However, the analysis, discussion, linkage of the present results with previous ones over the region and physical explanations of the aerosol properties related them with seasonality, meteorology and sources are missing or are rather poorly discussed throughout the manuscript. The analysis and discussions for each figure seem rather brief and do not emphasize on important issues of aerosol properties, mixing processes and connection of AOD with surface PM2.5 concentrations. In the following, I have specific comments for authors in a way to improve the scientific quality of the paper. In synopsis, detailed discussion of the results, enrichment of the literature and new analysis in some issues are necessary.

Dear Reviewer, Thank you for your time. We greatly appreciate your review and the references you have provided. The paper has been significantly modified and you will find below point-to-point answers to your review.

First, some general comments and information about the revised version for both reviewers.

- Firstly, we want to make clear about the region. We focus on the northern coastal part of the gulf of Guinea. Western Africa has a marked latitudinal gradient in ecosystems that largely impacts the emission and deposition of particules and trace gases (Adon et al., 2010). We define South Western Africa as delimited by the shore of gulf of Guinea and 9°N in agreement with previous authors (Kniffka et al., 2019). Most the previous studies on AOD observations were performed north of our region of interest, i.e. in the Sahel. The observations we report here are unprecedented in this area of the world so the comparison with previous studies is very limited.
- Reviewer #1 has raised a very important point on the misuse of MODIS AE over land to classify aerosols from AOD observations. So the methodology used for analysing the sun photometer data can't be directly transposed to MODIS record. Nonetheless similar conclusions can be drawn from the seasonal analysis.
- The methodology to estimate PM2.5 from AOD is based on a simple relationship of proportionality between both quantities that depends on the season and the aerosol type. The description of the method has been improved. Accurate estimate of the impact of the aerosol type on PM2.5/AOD ratio is limited by (i) the weekly frequency in the ground sample while aerosol type can change daily, (ii) the classification of aerosol type using Ångström exponent in a geographical area where mixing of aerosol type is high and pure characteristics are seldom observed. Notheless, the effect of the dust layer uplift on surface concentrations can be observed during the

intermediate period when the monsoon onset occurs.

1) The methodology for the PM2.5 estimations from space is not described in detail. Authors should provide the analysis or even the scatter plots between MODIS-AODs and ground-based PM2.5 concentrations for each site and present the linear regressions and the converted factors.

The methodology for PM2.5 estimation from AOD relies on a simple conversion factor that depends on the season and possibly on the Ångström exponent when this parameter is available. We provide equation 1 to explain the conversion. Conversion factors are presented in Figure 8 and retrieved PM2.5 are compared to observations for both sites in Figure 9. We discuss the error associated to the MODIS-derived PM2.5 in the text.

2) The accuracy of the handheld sun-photometer measurements of the spectral AOD is very important, not only for the validation of the MODIS AODs and the PM estimations,but also for the extraction of intensive aerosol properties like the Angstrom exponent...So, these retrievals can be assured as of high accuracy. Authors may check it by applying a 2nd-order polynomial fit to the sun-photometer wavelengths (Eck et al., 2001;JGR). Since the only three wavelengths will give an excellent (R2=1) polynomial fit,the two constant terms A2, A1 may be used and compared with the Angstrom exponent at same wavelength band and in case of very low errors the A2-A1 should equal to Angstrom exponent (Schuster et al., 2006; JGR). You may see this application in Sharma et al. (2014, Aer.Air.Qual.Res), Tiwari et al. (2018, Env.Sci.Poll.Res.) and references therein. With such sensitivity test, you assure the accuracy of the manual sunphotometer retrievals, which may have perturbed due to invisible clouds or even due to not exact matching of the sun disc in the instrument's FOV.

Thank you for the suggestion and the references. You suggest to check the AE value with regards to possible cloud contamination or error in collimation. A caveat should, however, be noted in your proposed method. We can't use a 2nd-order regression with only 3 points. The coefficients of the polynom are directly given by solving the linear equation system using the coordinates of the points. In the references you have given, the authors always use more wavelengths, 7 wavelenghts for Eck et al (2001) and 5 for Tiwari et al. (2018) and for a larger spectral range than in our study.

As AE is derived from the AOD-wavelenght log-log plot, it is subject to large error. The equation given by Hamonou et al. (JGR, 1999) shows that the absolute uncertainty in AE is sensitive to the spectral range and the relative uncertainty in AOD. Higher spectral range gives lower uncertainty on AE. However, as mentioned by Tiwari et al (2018), the departure from linearity leads to different AEs when considering different spectral ranges. In this paper, we use the same limited spectral range for the three different instrument. We estimate the AOD at 540 nm from AOD at 465 nm and AE then compare it to the measured AOD at 540 nm and should be within the measurement uncertainty, ± 0.02. Any point showing an inconsistency in the wavelength dependence is filtered out. We have clarified this point in the text as well as added references to Sharma et al (2014) and Kaskaoutis and Kambezidis et al. (2008) on L97.

3) Why did you use so long period (1 week) for filter samples? Usually, they are taken

on daily basis... Is this a technical reason or just to smooth the correlations with MODIS AODs, in order to avoid a larger daily variation? It is recommended to write something and discuss it more.

For logistic reasons it was not possible to run 4 sites during 2.5 years on a daily basis. We use gravimetric measurements, not automatic microbalance. A weekly sampling period seems reasonable to catch the main temporal pattern of atmospheric aerosols over long period of time (Ouafo-Leumbe et al., 2017). However the comparison of our weekly samplings with daily sun photometer observations is not trivial, specifically when looking at intensive parameters like Ångström exponent. This has been clarified in the text L152.

4) A critical point that has to be discussed regarding the measurement time series is the availability of sun photometer observations throughout the year and if there is an extent period (months, or season) with data missing. These large gaps may modify the AOD seasonality.

All the sun photometer observations are already presented in Figure 2 and Figure 3 as daily values. As you can notice, there are some gaps in the observations as already mentioned in the text Line 142. We have discuss the seasonaly of sun photometer time series for the sites having a significant number of observations throughout the year. For the seasonal cycle presented in Figure 10, we have used the MODIS observations and no significant gap has been noticed. We define a new larger area (see text L336) that has a least one observations per day over the MODIS record.

5) The present one constitutes a rather crude aerosol classification, as also recognizedby the authors. It is based on three AE groups, while there are not thresholds at all for the AOD. Alternatively, it is recommended to use the classic AOD vs AE scatterplot for all the examined sites to identify major aerosol types, where the dominance of dust and urban pollution will be defined, especially if such a plot is applied for the dry and wet seasons. That plot would constitute a much better representation of the aerosol types and is highly recommended. There are numerous studies over the globe (some are cited in the manuscript) dealing with such analysis. Finally, the aerosol classification based on AOD vs AE scatter plot is a first rough classification able to discriminate between major aerosol types i.e. biomass burning, desert dust, sea salt,but not between absorbing urban aerosols from various combustion sources, i.e. such a classification is incapable of determining absorption aerosol properties (see Giles etal., 2012, JGR; Cazorla et al., 2013, ACP). F urthermore, at the end of the results, this classification is expanded on long-term periods using the MODIS AE values, which are not accurate enough for such aerosol classification studies and the results and trends may be subject of biases. All these should be clearly discussed in the manuscript.

Thank you for your remark. Correct. The AOD-versus-AE scatter plot is a well-known methodology to get a classification of the aerosol types, also the classification remains crude as well. Adding a threshold on AOD values is interesting to make a speciation between clean marine air (low AOD and low AE) and dust (high AOD and low AE), while for the separation between biomass burning events (high AOD and high AE) and urban pollution events (high AOD and high AE) is rather difficult since, as you say in your

comment, this classification with AOD can't be used to determine aerosol absorption. In our dataset, the category 'clean marine air' is statistically unsignificant because of mixing with polluted air and continental aerosols. Moreover, there is a risk of over interpretation of the AE results for weak AOD because of uncertainty in AE depends on the AOD level (eg. Hamonou et al. 1999). However, we have followed your remark and we present the AOD vs AE plots for the differents sites and seasons. We also define a threshold on AOD to better identified dust event compared to mixing cases.

You comment on the MODIS AE has rised an important point. We have further investigated the use of MODIS AE (DB, OCEAN and Land algorithm) and finally end to the same conclusions as yours. At the coastal sites, both AE from the DT and OCEAN algorithm tends to reproduce the seasonal variability that is observed by the sun photometer. However there is no correlation between MODIS AE and sun photometer AE and MODIS AE can't be used to classify the daily observations.

Please refer to L250 and the new figure 6

6) Personally, I do not remember any other study that examined the PM2.5/AOD ratio and also there are no references for such studies here. However, I really doubt about the importance of this ratio, since it is strongly affected by the seasonally changing AOD and PM2.5 values. So, a similar ratio may define very contrasting aerosol regimes of low or high aerosol loading. Furthermore, I do not see a standard variation depending on season or aerosol type, which may indicate specific characteristics of aerosols in a certain period. For example, in periods with high dust activity, I would expect a lower AOD ratio, since dust is mainly transported above. Also, I would expect higher values for the "urban-like" types, since urban pollution mainly confines within the boundary layer and not in the vertical, so the PM2.5 is usually increasing with higher rates than AOD. However, all these may be highly variable from site to site and here, the averaged values from all the sites mask the results. Furthermore, authors fail to discuss in detail the physical meaning of their results and/or the importance of them. In case they want to maintain this analysis, they should be more detailed in the discussion of the main results and what a low or high ratio value represents. In the current version, this analysis and discussions are not considered important for the paper.

As mentionned in the introduction, there is an abundant litterature on AOD to PM conversion and a large diversity in the proposed methods albeit no consensus. The PM2.5/AOD is basically the amount of PM2.5 that is expected per unit of AOD. It was first promoted by van Donkelaar et al. (2010) as a conversion factor (Zheng et al., 2017; Yang et al., 2019). You are right, a similar ratio may define very constrasted aerosol regimes, including different aerosol types or vertical profiles. However we know that the most important phenomum that impact the AOD to PM2.5 relationship in this area is the transport of mineral dust from the Sahel and the shift of the dusty layer toward upper altitude during the pre-monsoon period. So we have analyzed the PM2.5/AOD ratio as a function of the season and aerosol type (Figure 8).

Standard errors on PM2.5/AOD ratios are already reported in Figure 6. We are sorry to say that your analysis on the PM2.5/AOD ratio is not correct. During winter dry period the dust is advected near the ground by the Harmattan wind so the PM2.5/AOD ratio is high (about 50 $\mu g/m^3$ per unit of AOD).

During the transition period (April-June), the dust layer is uplifted by the monsoon flow. Although the AOD is high, the PM2.5 remains low, leading to a PM2.5 ratio for dusty days of about half the one for the winter period. This phenomemum is similar for the

whole area and we pooled the dataset to increase statistical significance of the results. However, as suggested in your review, we provide the comparison between observed and derived PM2.5 for each site separatly.

Text has been changed on L280. Figure 9 has been changed to show both sites.

Specific Comments

Line 29: Add "in Chad" after depression.

Done.

Line 35: You may also see the recent global study about aerosol hot spot regions of dust, polluted-dust and smoke by Mehta et al. 2018. Mehta, M., Singh, N., Anshumali,2018. Global trends of columnar and vertically distributed properties of aerosols with emphasis on dust, polluted dust and smoke - inferences from 10-year long CALIOP observations. Rem. Sens. Environ., 208, 120-132.

Thank you for providing usefull references. This reference has been added Line 35.

Line 36: You may refer some of the important studies that linked the ARF with the monsoon circulation and precipitation redistribution in the Arabian Sea and India, since such studies are rather rare or even absent in the south west-Africa.

Thank you for the suggestion. Actually our sentence was awkward.

Please see new paragraph L36 in the revised document.

Lines 36-39: These sentences can be merged.

Done.

Line 44: Especially for the issue of aerosol-type classification, authors may also see the global study by Hamill et al. (2016; Atmos. Envron.), which also covers the study region.

Thank you for the reference. We have added Hamill et al. (2016) to the list of references Line 44.

Lines 53-54: Another study (Sinha et al., 2015; Intern. J. Rem. Sens.) relates this AOD-PM regression with vertical profile of aerosols and meteorological parameters (RH, Theta, wind) that strongly affect the PM vs. AOD correlation.

This new reference has been added Line 63.

Line 63: Add "and" after time series.

Done.

Lines 91-92: Revise this sentence.

Done.

Lines 105-107: It is not clear if these are your results or other ones from previous validation studies... Please, clarify it. Also, a revision is needed in this sentence.

CALITOO handheld sunphotometer were calibrated using Langley plot at high altitude Izana observatory. The calibration was check using coincident AERONET observations before and after field experiment We have modified the text as follows.

CALITOO observations were compared to coincident AERONET observations before and after the field experiment. The total uncertainty in AOD is estimated to ±0.02 for all the wavelengths. Post-field measurements indicates a change of about 1% per year in the calibration.

Line 143: "The overall range of AOD is (0.07, 3.8)." This does not make sense. In what station do you refer here? Also, the parenthesis confuses the reader that something else is missing here.

The overall range means considering all the stations together. Brackets indicates (minimum value, maximum value). As it appears not clear for the reader, we have changed this sentence. See L170.

Lines 148-149: The rather significant variability in the mean AODs at Lamto site due to different periods of the measurements and number of observations necessitates discussing the availability of measurements throughout the year and if a specific season dominates (in number of measurements) against others...

The timeseries at Lamto are presented on Fig. 2. No season dominates for HCC or CALITOO although the observations corresponds to different years. AERONET dataset is rather limited (see table 1, revised as there were an inconsistency in the ending date of the measurements). We have recall the observation period for Lamto in the main text L176.

Lines 156-157: Add ", respectively" at the end of the sentence.

Done.

Line 160: Revise as "... R=0.82, being R=0.90 between ..."

Done.

Lines 167-169: These characteristics should be discussed in view of aerosol sources,transport routes and dominant aerosol types in each season. Only presentation of the results without any explanation about their physical meaning is rather awkward.
Lines 170-172: Also, these results should be further discussed about aerosol types,sources and physical meaning. For example, why AE values are, on average, higher at the coastal stations despite the relative higher influence from sea-salt aerosols that are known to have low AEs? Furthermore, all sites throughout the year present rather low AEs, well below 1.0, except of some few cases, which has to be further commented by authors.

This section describes the observations while the physical meaning is discussed later in the text. However we have followed your recommandations and give the explanation of the physical meaning in this paragraph.Coastal sites are located in large conurbations so the AE remains higher than at the inland sites. AE values are further discussed in the classification section. See L199 and L205.

Line 178: Add "values" after RMSE.

Done.

Line 192-195: Some further discussions are needed here, regarding the suitability of using MODIS-derived AE values. According to my knowledge and several previous studies, MODIS-AE over land is highly biased since both Deep Blue and Dark Target algorithms used standard models and mixtures of them for the determination of the AE, which is not a measured but a computed parameter. So, the uncertainty increases significantly and this is also shown from the frequency distribution of the AE values from MODIS and sunphotometers. It would be nice, authors to present via graph, or even to give some values of comparison between MODIS-AE and sunphotometer AE in order to reveal the magnitude of the bias.

Correct. See your preceding comment new Figure 6 and corresponding text.

Line 197: AE does not depend on the aerosol optical properties. It is an intensive aerosol optical property by itself.

Correct but following the definition given by Ångström, AE depends on the aerosol extinction coefficients, which is an aerosol optical properties. Sentence has been revised as follows.

AE is an intensive aerosol optical parameter and depends on the spectral aerosol extinction coefficient (Nakajima et al., 1996; Eck et al., 1999; Holben et al., 2001). AE is influenced by the aerosol size distribution and is commonly used to identify aerosol types (Léon et al., 1999; Kaskaoutis et al., 2009; Perrone et al., 2005).

Line 209: Correct as “enables”

Done.

Line 210: Correct as “category”.

Done.

Lines 215-216: This sentence needs revision in English grammar, syntax and typos errors.

Done.

Lines 217-218: For aerosol type classification in west Africa, authors may see the results by Hamill et al. (2016; Atm.Env.) about dominant aerosol types at several sites of the region.

Thank you for the reference. Actually Hamill et al (2016) is out of the region. So no comparison can directly apply. However we have used this reference for dust source activity north of our domain. We now cite this paper L271.

Line 229: Correct as “ratios”.

Done.

Line 235: Correct as “remain”.

Done.

Lines 235-236: Which are the moderate PM2.5 and significant AOD values reported here? There is not a clear view of the levels of these values, which should be mentioned here.

We now give the corresponding values. Values are also reported in Figure 9.

Lines 236-238: This is only true for the "dust-like" aerosols and not for the other types. So, this is not a main findings of the analysis. We don’t get your point here.

Lines 240-241: There is no analytic description of the satellite-derived PM2.5 concentrations. The methodology of these retrievals should be included in the manuscript. This analysis, based on weekly averages, is rather rough and not analytic about the association between dominant aerosol and PM2.5 levels. The scatter plot of AOD vs. AE may also

include as third variable (color coded), the PM2.5 concentrations, so the reader would be able to see what type is associated with highest PM2.5. Statistics of such analysis may be also included in a Table. Authors may also provide the seasonality of the aerosol types and correlations between AOD and PM2.5 for each type separately.

The text associated to Figure 7 (now Figure 9) has been improved. Several points were unclear. The color code on Figure 9 directly give which type is associated to PM2.5 levels. In the first version we have mixed MODIS observations and sun photometer observations to increase the number of comparison points (specifically for Abidjan where we have less sun photometer observations). However as in this revised version we don't rely on MODIS AE, we only use sun photometer observations in Figure 9.

Lines 247-251: The current excellent results regarding the PM2.5 estimates from satellite measurements should be compared with other studies over the globe, several of them cited in the Introduction section. In general, there is a lack of any comparison or even discussion of the present results with previous studies in the region. This constitutes a main drawback in the scientific presentation of the present study.

We agree on this point however it is not a main drawback. Not all the authors report the same metrics and there is no scientific agreement on which algoritm should be used to derive PM2.5. A general discussion on RMSE for the different type of algoritm is out of the scope of this paper. Moreover there is no similar study in the area targeted in our paper, so the comparison remains limited. Nonetheless we propose to mention L336 that the retrieved RMSE are within the range of previous studies and we cite Sinha et al. (2015) and Ma et al. (2015).

Line 254: It is not clear, since there is no discussion in the method, if authors used a unique PM2.5/AOD conversion fraction for the whole data or seasonal-dependent conversion factors, based on AOD vs PM2.5 scatter plots in each season. In other places over the globe, this seasonal dependence has been shown to be very important for the accuracy of the retrievals.

Correct. Now it is explicitly written that we use the seasonaly adjusted ratios presented in Figure 9.

Lines 255-257: The critical here is to provide information about the PM2.5 levels above $25\mu gm^{-3}$, which is the threshold for pollution established by EU. Authors should provide analysis about the seasonality of the PM2.5 exceedances and to associate them with the dusty period and the monsoon circulation.

The EU target of $25\mu gm^{-3}$ is for the annual mean. We report the years being below this threshold. We also report daily exceedances above $35\mu gm^{-3}$.

Line 257: Correct as "Maximum is...".

Done.

Line 261: Explanation is needed for the much higher PM2.5 values in Cotonou, which also reflect higher AODs than those in Abidjan. Also, correct as "Abidjan".

The data used for figure 8 as well as the figure have been revised. We still observed a large difference in April (23% higher in Cotonou) that is clearly attributed to a change in the contribution of coarse dust(+30% in Cotonou) while the contribution of other types remains the same. So our hypothesis is a higher contribution of dust during the dry and intermediate period over Cotonou. Further validation of this hypothesis will need both numerical modelling of the dust transport and in situ chemical speciation.

See text L365 and new figure 10.

Line 263: "PM2.5 concentrations are then multiplied by a factor of about 3 in 4 months.". This sentence is rather confusing without a clear meaning. It should be rephrased.

The sentence described the increase in PM2.5 concentrations between September and December. Now rephrased

see revised text L371

Figure 9: Why did you mix the observations from both sites? I think that this smooths the variability and possibly the trends. At any case, the trends should be examined separately for these sites. Also, authors are based on MODIS-AE values for the determination of the urban-like aerosol type. As discussed above, the MODIS-derived AEs are significantly biased and this should be mentioned in the text. However, in annual basis, the errors and biases are significantly reduced and trends may be examined but with caution. At any case, a full discussion about the biases occurred in such approaches should be given.

In the first version we have mixed the observations from both sites to increase the statistical significance of the results. We now introduce a new wider geographical area called SWA that includes both cities but provides more observations. Figure 9 have been changed to show (i) the monthly mean MODIS-derived PM2.5 (ii) the trend in the short dry period. MODIS AE is not used any more in the long-term analysis (see your preceeding remark). We report the 95% confidence interval for the trend indicating that the trend remains uncertain. As PM2.5 is proportional to AOD, any bias occuring in AOD, including calibration drift, will affect the PM2.5 concentrations.

see revised text L385 and new reference to Sayer et al. 2019

Line 265: Correct as "represents"Lines 269 and 271. This is Figure 9.

Done.

Line 271: For such an explanation you may see the differences in MODIS AODs (likely significant lower values in 2018) or in the frequency distribution of the urban-like type. However, the annual PM2.5 and the trends in the two sites should be analyzed separately, so the current figures should be changed.

Yes. AODs for 2018 (and 2019 added now) are lower than the previous years. We observed a 20% decrease in AOD annual average between 2017 and 2019. As stated earlier, the change in the frequency of urban-like can't be addressed from MODIS. However we now plot the monthly mean PM2.5 (SWA area in Figure 9) that best highlights the decrease in the recent years what ever is the season. The differences in monthly means between Abdijan and Cotonou and the SWA are not significant (see also the monthly annual cycle in Figure 10) for the trend analysis and not displayed on Figure 11.

see revised text L376 and Figure 11.

Lines 272-274: These results should be re-evaluated separately for the two sites. Also, here it is not referred that this increasing tendency corresponds to the urban-like aerosols. Also, there is no discussion about increasing trends in PM and anthropogenic pollution in the urban areas of the region, or even any comparison with previous works.

Due to the large uncertainty in the trend analysis, discussing the difference between the two sites is not relevant. We choose to present the trend for a broader area that encompassed both cities have at least one observations per day so any bias due to sampling is avoided. There no such previous work over the area so comparison remains very limited and moreover regional scale emission scenario for this particular area are scarce. As stated in the conclusion, the trend is consistent with existing emission scenarii.

Please refer to the conclusion section.

Conclusions section should be revised in view of the new results and discussions in the text. Conclusions and abstract have been revised accordingly.

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Anonymous Referee #2

This paper combines satellite (MODIS) and ground-based (AERONET and hand-held)AOD with PM2.5 data to (1) evaluate MODIS AOD and (2) investigate the relationshipbetween MODIS AOD and PM in southern West Africa. Empirical relationships between AOD and PM are created (based on season and an aerosol type proxy) and used to estimate trends in ground-level PM in the region. The topic is in scope for the journal and the subject matter is important. The hand-held and PM data were presented by the authors in a previous paper in ACP (Djousse etal., 2018); this work is a natural extension of that (using the data collected and morein concert with MODIS) and contains sufficient novel material. The quality of language is quite good, and the analysis, while fairly simple in parts, is explained well and uses statistics fairly appropriately (for example the authors acknowledge the skew in AOD distributions and treat this appropriately, while many authors do not). There are some bits that are a bit unclear, I have a few thoughts on the data use, and I found some typographical issues. I recommend major revisions, mainly due to more detail needed in the AOD/PM ratio part. I would like to review the revised version.
Comments are as follows:

Dear Reviewer,
Thank you for your time. We greatly appreciate your review and the references you have provided. The paper has been significantly modified and you will find below point-to-point answers to your review.

First, some general comments and information about the revised version for both reviewers.

- Firstly, we want to make clear about the region. We focus on the northern coastal part of the gulf of Guinea. Western Africa has a marked latitudinal gradient in ecosystems that largely impacts the emission and deposition of particules and trace gases (Adon et al., 2010). We define South Western Africa as delimited by the shore of gulf of Guinea and 9°N in agreement with previous authors (Kniffka et al., 2019). Most the previous studies on AOD observations were performed north of our region of interest, i.e. in the Sahel. The observations we report here are unprecedented in this area of the world so the comparison with previous studies is very limited.
- Reviewer #1 has raised a very important point on the misuse of MODIS AE over land to classify aerosols from AOD observations. So the methodology used for analysing the sun photometer data can't be directly transposed to MODIS record. Nonetheless similar conclusions can be drawn from the seasonal analysis.
- The methodology to estimate PM2.5 from AOD is based on a simple relationship of proportionality between both quantities that depends on the season and the aerosol type. The description of the method has been improved. Accurate estimate of

the impact of the aerosol type on PM2.5/AOD ratio is limited by (i) the weekly frequency in the ground sample while aerosol type can change daily, (ii) the classification of aerosol type using Ångström exponent in a geographical area where mixing of aerosol type is high and pure characteristics are seldom observed. Notheless, the effect of the dust layer uplift on surface concentrations can be observed during the intermediate period when the monsoon onset occurs.

1. As a general point about the MODIS retrievals: the authors use daily level 3 (L3) products (1 degree) from Aqua as the basis for both the comparison with Sun photometers and the PM prediction. For validation against Sun photometers it is more usual to use level 2 (L2) data with an averaging radius around 25 km to decrease discrepancies arising from real spatial and temporal variability. The authors might acknowledge that here. It is probably ok to use the L3 data if the goal is to make regional-scale PM analyses. But the purpose here (and the data collected) seems focused on the two cities. So I would suggest the authors might get L2 data and perform the same sunphotometer comparison to see whether the same patterns hold. I suspect they might but without seeing the data we don't know. I acknowledge that this might not be feasible dependent on the computational resource available to the authors. I suppose doing both is a good way to test whether there is significant variability below the L3 scales.

We agree with the reviewer that validation exercises are usually applied to the MODIS level 2 pixels rather than gridded products. However our objective is not to valide the retrieval algorithms but to address the ability of MODIS data to reflect aerosol changes in a specific area. Doing the validation of both L2 and L3 products will lead to further investigations in the statistical representativity of L3 versus L2 that is not the purpose of the paper.

Please refer to L125 in the revised version.

2. The authors are also using (from their statements) an outdated version of the MODIS product: Collection 6 rather than 6.1. This was released several years ago now (in late 2017: `https://atmosphere-imager.gsfc.nasa.gov/documentation/collection-61`) so it is unfortunate to see Collection 6 still being used. Collection 6.1 has some algorithm updates as well as calibration updates which might affect the results. It would be preferable to repeat the analysis with the latest data version.

You have rised an important point. Our archive was not uptodate and actually a mixed between Version 6.0 and 6.1. This problem has been fixed although with there is no large impact on the results.

3. It would also be interesting to add a second satellite data set for an additional point of comparison. One option would be MODIS Terra, as the earlier overpass time might mean different sampling due to cloud cover changes. Alternatively the authors might consider a different sensor or algorithm. There are many available during the 2014-2017 study

period, but for the longer-term PM trend analysis the options are fewer. MISR has a narrower swath so there will be many fewer matchups with the Sun photometers, but its retrieval of aerosol properties has some more flexibility so it might perform better. OMI also has a nice smoke/dust aerosol type identification which might be useful here since part of the analysis involves relating AOD to PM based on aerosol type. So that could be a good addition. Another alternative is using a reanalysis product (e.g. MERRA2) which might also have surface concentration estimates. I am not saying this should be a requirement for publication, just something for the authors to consider.

Thank you for your suggestion. We agree that a mixed of different satellite products along with reanalysis like MERRA2 could provide interesting information on AOD-PM relationship in our area of interest. However there are not so much options for long-term analysis as you have noticed. We have selected the MODIS record as it is comprehensive and well referenced. CALIPSO would be also an interesting alternative that requires further investigations. We have followed Wei et al. (2019) recommandations of using AQUA products.

Please refer to L127 of the revised document.

4. Line 6: "Angstrom" should be written as "Ångström" here and throughout the paper. The paper is not always consistent. Line 110 and Table 1 have the ö but not the Å,for example, and Figure 3 has neither. This is not needed on line 119 though because there the authors are referring directly to the variable name.

We now use the correct writing of Ångström in the text, bibliography, and in the figure captions. AE is used for figure labels.

5. Line 16: I think "S=" can be removed here.

Done.

6. Figure 1: again, not essential, but rather than have a greyscale map the authors might consider using e.g. a population map upon which to show the site locations.

Figure 1 has been updated with the geographical location and population of cities having more than 1,000 inhab.

7. Line 124: I would add parentheses around the EE expression as the interpretation of the +/- is ambiguous as written. I believe the correct representation is +/-(0.05+0.15xAOD) and not (+/- 0.05)+0.15xAOD.

Done.

8: Line 137: this should be IQR not IRQ.

Done.

9: Lines 144-145: the sentence says that the highest AOD was 3.8, but then says it was 3.7. This should be checked and corrected.

Correct value is 3.76. The text has been modified.

See revised text L170

10. Lines 152-153: the offset in AE between two measurement types could well be related to calibration; the authors may wish to mention the study by Wagner and Silva (2008) on this topic: https://acp.copernicus.org/articles/8/481/2008/

Thank you for the reference. Very interesting. Rather than related to calibration, the difference might be due to the difference in the AOD statistical distribution. Indeed the observations were not acquired during the same period and the AOD distributions are not the same. We have added this possible explanation in the text with the corresponding reference. Moreover, we have better discussed the comparison on coincident observations between AERONET and CALITOO in Lamto. Despite a short sample period, the agreement is excellent (see added figure). Note there is an error in the sampling period in Table 1 but Figure 2 and 3 shows the overlap period.

See revised text L181.

11. Line 156: AERONET collects data from dawn to dusk, while the hand-held instruments say they were used twice a day. Are these daily averages from all points or from the same times as the hand-held instruments?

AERONET observations are daily average. Now mentionned in the text L84.

12 Figure 5: my assumption is that the AE shown from MODIS Deep Blue and ocean algorithm here are for all points of the domain, and not only for the grid cells wherethere are sun photometer data. Is that correct? If so, some differences might also be expected due to real spatial variability in the AE at locations without sun photometer data. This should be mentioned.

The comparison of MODIS AE values have been revised to better show the difference at both the coastal and inland sites. We used only the measurements corresponding to the grid cell where the sun photometers are located.

Please refer to revised Figure 6 and text L215.

13. Table 3: I am not sure I fully understand this as it took a few readings. It seems that the numbers not in brackets are the percentage of days from each category, using only the AE to split them. Then, the numbers in columns are the same, except considering only those days where the AOD was above the third quartile for that location, i.e.days where AOD was particularly high. So the table is contrasting the optical "type" ofaerosols between sites, and also between the data set as a whole and those particularhigh-AOD days of concern for air quality purposes. Is that correct? I wonder if thenumbers in parentheses should be given their own 3 columns with own subheader inthe table. This is because it is the tendency as a reader to look at the number and the one next to it, and in this case they're not directly related, as the relevant comparison is between columns.

We agree that the information provided in the Table 3 were not easy to read. We have simplified this table and now provide only the percentage of daily observations for each category and each site. Please note that the 'Dust' category has been updated by using a threshold on AOD.

See new table 3.

14. Figure 6, and associated discussion: this should be fleshed out more. First, is this figure showing mean and standard deviation of the ratio? This should be stated. However, another concern is whether mean and standard deviation are the right metrics to show; I suggest median and IQR could again be more appropriate. The authors do not show the raw data so there's no way to know whether there are e.g. outliers which are throwing off the mean ratios here. I suggest adding plots of the data (i.e. weekly AOD and PM for different seasons and type classifications). In theory, given constant meteorology and composition, it is true that the ratio between AOD and PM should be a constant (the mass extinction efficiency and a factor based on height). However in practice factors such as changing composition, variations in aerosol vertical structure (e.g. whether or not the aerosols are mostly in the boundary layer), and moisture (as AOD is dependent on ambient RH while PM is not) will be important. See for example Sayer et al (2016) for the dependence of the ratio on some of these factors for smoke in Thailand: https://doi.org/10.4209/aaqr.2015.08.0500 The situation is a bit different here because the present study is weekly filter measurements (not continuous) but the general point remains. The authors need to show the data to provide justification that adopting a direct ratio approach, and reporting the mean and standard deviation (or changing to median and IQR), is an appropriate empirical parametrization here. It is not possible to judge from the material presented in the paper. As this figure is key to most of the rest of the paper, changes to this part of the analysis could affect the later results and discussion as well.

We agree with your remark that the actual PM2.5/AOD ratio is subject to a large variability due to the modification of meteorological conditions or changes in aerosol chemical composition. We have added your proposed reference (Sayer et al., 2016) in the introduc-

tion section where we discuss previous studies. So far there is no scientific agreement on a universal method to convert aerosol optical parameters into surface concentrations however it is generally admitted that AOD and PM2.5 are correlated. Conversely, for steady surface concentrations and columnar AODs, a poor correlation between both quantities doesn't mean that a PM2.5/AOD ratio is not appropriate. Errors on the PM2.5/AOD ratio can then be estimated from the standard deviations of both quantities. The time series of coincident weekly observations already reported in Figure 9 (now for each site) provide posterior justification that the PM2.5 is proportional to AOD by period of time and that the aerosol type may also has some influence on the proportionality. So the coefficient are estimated by season and by aerosol type. The standard error reported on Figure 6 reflects that any change in AOD is not associated to an exact proportional change in PM2.5 over time. However and as you mentionned, there is an additionnal difficulty due to the different sampling period (weekly PM2.5 versus daily AOD). We handle this difficulty by estimating the relative influence of each aerosol type during a week.

Please see additional information in the method section L298 and Figure 9.

15. Figure 7 caption: "weakly" should be "weekly".

Done.

16. Figure 9 and associated discussion: given the high seasonality in AOD (and PM),as well as the potential for uneven satellite sampling through the year (due to e.g. cloud cover variations), I am not convinced that it makes sense to examine only annual trends in PM. I suggest adding seasonal analyses as well. This will provide more insight as to any changes. The authors might also add a plot showing e.g. the number of days with data in the average Jan, Feb etc from MODIS, so we can see whether sampling variations exist. A second point about trends is that rather than talk only about p values, it would be useful to mention the uncertainty estimate on the trend as well. Statistical significance and importance are not the same thing. For example, a statistically significant result might have a small magnitude which is not important for practical purposes. And a result that is not significant might be because either the estimated magnitude is small and the uncertainty is also fairly small (i.e. we can be confident there is not a large trend), or because we have a large uncertainty so can't tell whether an effect is large or small (i.e. we can't be confident about the magnitude and/or sign of any trend). Reporting best estimate of trend and p value without the uncertainty estimate means we cannot directly tell which of these is the case here.

Correct. The trend analysis has been totaly revised. Following your recommandation, we now provide the monthly mean concentrations and the seasonaly adjusted trend. We have reduced the potential impact of uneven sampling by selecting a broader area along the shore line. This larger area has at least one observation per day. Yes we totally agree with your remark however the uncertainty on the Thiel-Sen's slope were already reported in original text L273. The monotonic increasing trend is significant however the exact magnitude remains highly uncertain.

17. Data availability: the authors give Giovanni as the MODIS data source. This is mostly a visualization portal and may not have the latest/official data versions. I just checked there and they list collection 6.1 which makes it more surprising that the authors used the older collection 6. Note the main NASA search tool is `https://earthdata.nasa.gov/` and the actual MODIS data portal for this product is LAADS, `https://ladsweb.modaps.eosdis.nasa.gov/`.

Correct. Actually we did a first attempt of using MODIS L3 products from the Giovanni portal that also provides an extraction tool. Then we used the a mirror archive but the archive wasn't uptodate. Now we use the data from the LAADS portal `https://ladsweb.modaps.eosdis.nasa.gov/`.

Thank you for your warning on the MODIS Version. We have changed the data availability section.

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PM2.5 surface concentrations in southern West African urban areas based on sun photometer and satellite observations

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Abstract. Southern West Africa (sWA) is influenced by large amounts of aerosol particles of both anthropogenic and natural origins. Anthropogenic aerosol emissions are expected to increase in the future due to the economical growth of African megacities. In this paper, we investigate the aerosol optical depth (AOD) in the coastal area of the Gulf of Guinea using sun photometer and MODIS satellite observations. ~~We use a~~ A network of lightweight handheld sun ~~photometer measuring the solar irradiance at 465, 540 and 619 nm operated manually every day~~ photometers have been deployed in sWA from December 2014 to April 2017 at 5 different locations in Côte d'Ivoire and Bénin. The handheld sun photometer measures the solar irradiance at 465, 540 and 619 nm and is operated manually once per day. Handheld sun photometer observations are complemented by available AERONET sun photometer observations and MODIS level 3 time series between 2003 and ~~2018.~~ 2019. MODIS daily level 3 AOD agrees well with sun photometer observations in ~~Abdidjan~~ Abidjan and Cotonou (correlation coefficient R=0.89 and RMSE=0.19). A classification based on the ~~Angstrom Exponent~~ sun photometer AOD and Ångström exponent (AE) is used to separate the influence of coarse mineral dust and urban-like aerosols. The AOD seasonal pattern is similar for all the sites and is clearly influenced by the mineral dust advection from December to May. Sun photometer AODs are analyzed in coincidence with surface PM2.5 concentrations to infer trends in the particulate pollution levels over conurbation of Abidjan (Côte d'Ivoire) and Cotonou (Bénin). PM2.5 to AOD conversion factors are evaluated as a function of the season and the aerosol type identified in the AE classification. Highest PM2.5 concentrations (up to 300 $\mu g/m^3$) are associated to the advection of mineral dust in the heart of the dry season (December-February). ~~From December to March the median concentration above Abidjan and Cotonou is around 40~~ Annual means are around 30 $\mu g/m^3$ ~~, while it is around 20~~ and 80% of days in the winter dry season have a value above 35 $\mu g/m^3$ ~~during the rest of the year. Considering only the days during which the AOD belongs to the~~ while concentrations remains below 16 $\mu g/m^3$ from May to September. No obvious trend is observed in the 2003-2019 MODIS-derived PM2.5 time series. However the short dry period (August-September) when urban-like ~~aerosol category, we observe a significant trend S=0.32~~ aerosols dominate, is associated to a monotonic trend between 0.04 and 0.43 $\mu g/m^3/year$ in the PM2.5 concentrations over the period 2003-2017. ~~This trend leads to an increase of 5 $\pm$ 3 $\mu g/m^3$ over 15 years and~~ The monotonic trend remains uncertain but is coherent with the expected increase in combustion aerosol emissions in sWA.

1 Introduction

The increasing trend in the anthropogenic emissions in Africa (Liousse et al., 2014) gives rise to the question of the impact of human activities on air quality, the monsoon system and the regional climate. The Gulf of Guinea and adjacent countries, hereinafter called southern West Africa (sWA), is influenced by large amounts of aerosol particles of both anthropogenic and natural origins advected from the African continent. The season cycle in sWA is driven by the monsoon system (Knippertz et al., 2015) with the alternation of a major winter (November to March) dry season and a summer (June-July) rainy season. The Inter Tropical Front (Lélé and Lamb, 2010) is at its southernmost position during the winter dry season enabling the northeasterly Harmattan wind to carry a dust-laden dry air southward (Adetunji et al., 1979). The major conurbations of sWA are then downwind the mineral dust emission of the Bodélé depression in Chad, the predominant dust emission source of West Africa (Todd et al., 2007; Washington, 2005; Koren et al., 2006; Schepanski et al., 2009). Carbonaceous aerosols that are emitted by open biomass burning (Liousse et al., 2010) are also advected southward to the main coastal cities of sWA during the dry period. The summer wet season corresponds to the continental intrusion of the southwesterly monsoon winds carrying moist air and precipitation. During this period, biomass burning emissions in central Africa can be advected to sWA by easterly wind and thus can impact the local air quality of coastal conurbations (Menut et al., 2018).

sWA is a hot spot of atmospheric aerosol concentrations as revealed by satellite-derived aerosol optical depth ~~(Kaufman et al., 2002).~~ (Kaufman et al., 2002; Mehta et al., 2018). Atmospheric aerosols can alter the development of monsoons by weakening the land-ocean thermal contrast, and the thermodynamic stability and the convective potential of the lower atmosphere (Li et al., 2016). Precipitation reduction in the West African monsoon region (Janicot, 1992) has been attributed to high aerosol concentrations near the Gulf of Guinea (Huang et al., 2009). Yoon et al. (2016) have pointed out the role of carbonaceous aerosols on rainfall reduction in the West African monsoon region. Aerosol effects on regional climate falls into two categories (Boucher et al., 2013). Direct effects refer to the influence of aerosol scattering and absorption on the atmospheric radiative balance. Indirect and semi-direct effects refer to the impact of aerosol on cloud properties with subsequent effects on the radiative balance. The aerosol optical depth (AOD) is ~~a key parameter~~ one of the key parameters for assessing the aerosol direct radiative impact ~~(Boucher et al., 2013), which is of primary importance for the regional climate and the monsoon system (Janicot, 1992).~~ (Liou, 2002).

AOD is the primary aerosol optical parameter derived from satellite remote sensing (Kaufman et al., 1997). AOD is related to the reduction in the atmospheric transmission due to aerosol particles in suspension in the atmosphere. AOD can be measured directly from the ground by using a sun photometer (Volz, 1959; Prospero et al., 1979; Tanré et al., 1988; Nakajima et al., 1996). The Aerosol Robotic Network (Holben et al., 1998, 2001) is one of the most important federated network of ground-based automatic sun photometers providing continuous AOD measurements in many places of the world. Western Africa benefits from a good geographical coverage of AERONET sun photometers in the Sahel transect. The stations are located in remote places dedicated to the monitoring of Saharan dust or biomass burning aerosols optical properties and atmospheric transport

(Tanré et al., 1988; Redelsperger et al., 2006; Mallet et al., 2008; Léon et al., 2009). However sun photometer observations in the large conurbations surrounding the gulf of Guinea remain scarce. AOD observations in the coastal part of sWA will thus provide additional ground-truths for satellite validation.

Long-term satellite-derived AOD can also make up for the lack of in situ particulate matter (PM) surface observations. As air quality in sWA conurbations is still poorly covered by operational observational networks, satellite-derived PM may have a significant added-value for air quality monitoring. There is an abundant literature on linking columnar satellite AOD to PM (Kacenelenbogen et al., 2006; Hoff and Christopher, 2009; Ma et al., 2015; van Donkelaar et al., 2016). The relationship between instantaneous AOD and PM measurements is not straightforward and several regression models have been tested, either linear (Kacenelenbogen et al., 2006), multi-linear (Gupta and Christopher, 2009) or non-linear (Gupta and Christopher, 2009; Yahi et al., 2013; Kamarul Zaman et al., 2017). The conversion model from AOD to PM depends on the aerosol physical properties (aerosol type), hygroscopicity and the atmospheric dynamics including boundary layer mixing. In particular, the variability of the planeraty boundary layer depth can act as a controlling factor to the ratio between surface PM and columnar AOD (Boyouk et al., 2010; Sayer et al., 2016). Vertical profil of aerosols and meteorological parameters affects the correlation between PM and AOD (Sinha et al., 2015). Additional local analysis of the PM-to-AOD relationship based on in situ observations will strengthen the systematic retrieval of PM for satellite remote sensing.

In a companion paper (Djossou et al., 2018), the AOD measurements obtained downtown major cities of Abidjan (Côte d'Ivoire) and Cotonou (Bénin) were presented along with the surface observations of the PM2.5 mass concentration and carbonaceous aerosol composition. A tentative analysis of the relationship between AOD and PM2.5 was made and show the potential of AOD to infer PM2.5 concentration in both conurbations. In this paper, we report additional AOD measurements over sWA using lightweight handheld and automatic sun photometer with the purpose of validating the MODIS-derived AOD at the regional scale and investigating further the use of AOD for local pollution assessment. Section 2 presents the data sets and the methods. Section 3 presents the sun photometer time series and the validation of the ~~validation to the~~ satellite AODs. The relationship between AOD and PM2.5 is investigated in section 4. The last section presents the interannual trends in PM2.5 derived from the MODIS observations.

2 Data and Method

All the observations were acquired in a geographical box ranging from approximately 4° N to 9° N and 6° W to 5° E (Figure 1). SWA has a marked latitudinal gradient in ecosystems that largely impacts the emission and deposition of particules and trace gases (Adon et al., 2010). We define SWA as delimited by the shore of gulf of Guinea and 9°N in agreement with previous authors (Kniffka et al., 2019). The domain is bounded at its southern part by the gulf of Guinea and at its northern part by the sudanian savanna and desertic areas of Sahel and encompasses guinea savanna and forest ecosystems. Major conurbations are located on the shore of the gulf of Guinea: Abidjan (Côte d'Ivoire), Accra (Ghana), Lomé (Togo), Cotonou (Bénin) and Lagos (Nigeria). We have collected observations at 3 coastal locations, namely Abidjan, Cotonou and Koforidua, and 4 inland locations, namely Savè, Lamto, Ilorin and Comoé, (see Table 1 for geographical coordinates). The sites labelled Abidjan and

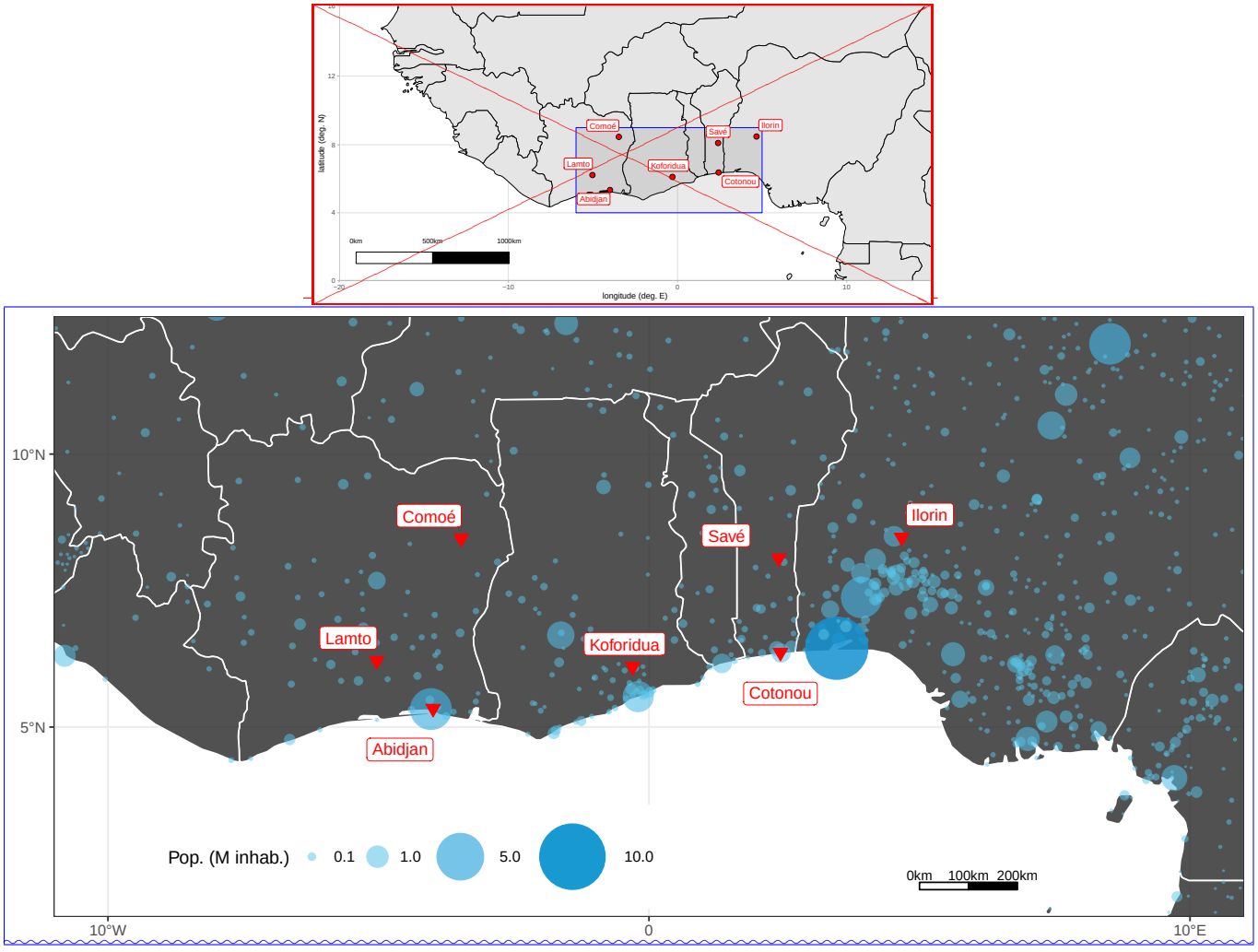

Figure 1. ~~Geographical~~ Map of south Western Africa (SWA) indicating the (red triangles) geographical locations of the sun photometer sites ~~in south Western Africa~~and (blue circles with legend) the location and population of urban areas having more than 1,000 inhabitants (source https://public.opendatasoft.com/).

Cotonou are respectively located downtown the city of Abidjan (≈ 4.4 million inh.) and the conurbation of Cotonou (≈ 1,7 million inh. including satellite cities). Savè site is located in the medium-sized city of Savè (≈ 90,000 inh.). Lamto is a rural remote site located 200 km north of Abidjan. Comoé site is located near the village of Nassian at the southern edge of La Comoé natural reserve. Ilorin site is located at the Department of Physics on the campus of the University of Ilorin (≈ 800,000 inh.) in Nigeria. Koforidua site is located at the main campus of All Nations University College about 5 km from Koforidua City (≈ 120,000 inh.), 50 km north of Accra, Ghana.

2.1 Sun photometers

Table 1 summarizes the location, type of instrument and observation periods. We have used different types of sun photometers, automatic and handhelds. The automatic CIMEL sun photometer is the reference instrument used in the AERONET network (Holben et al., 1998) for measuring the AOD and retrieve columnar aerosol optical properties and size distribution. We have used the level 2 quality assured ~~data~~ daily averages processed with the version 3 of the aerosol optical depth algorithm (Giles et al., 2019). We used the data for Ghana (station named Koforidua_ANUC located at 6° 6' N, 0° 6' W), Nigeria (station named Ilorin located at 8° 29' N, 4° 40' E) and Côte d'Ivoire (station named Lamto located at 6° 13' N, 5° 2' W). The geophysical station of Lamto was early equipped in 1997-1998 then the automatic sun photometer was restored back in 2017.

Handheld sun photometer is a well-known scientific instrumentation for measuring atmospheric transmission (Porter et al., 2001; Volz, 1959, 1974). The first type of handheld photometer we used is the one manufactured by CIMEL, hereinafter called HHC. HHC was operated during 2 years between April 2006 and March 2008 at Lamto geophysical station. The operating wavelengths are 440, 670 and 870 nm. The second handheld sun photometer is a new lightweight instrument manufactured by TENUM (http://www.calitoo.fr) and named CALITOO (Djossou et al., 2018). CALITOO operating wavelengths ~~for the CALITOO~~ are 465 nm, 540 nm and 619 nm. The sun photometer measures the Sun irradiance at the 3 wavelengths so no additional check on the AOD curvature (Kaskaoutis and Kambezidis, 2008; Sharma et al., 2014) can be applied however the spectral consistency between the AODs (observed at 540 nm and computed using the Ångström exponent) is checked. The atmospheric optical depth is then retrieved following the Beer-Lambert law knowing the calibration constant for each instrument and the relative air mass. The AOD is then retrieved after subtracting the Rayleigh and trace gases optical depth.

For the HHC, observations were acquired twice a day at around 9:00 and 15:00 UTC. For the CALITOO sun photometer, the observations were acquired at around 13:00 LT. The operators were asked to make measurements only when the sun was not obscured by clouds and have proceed with a sequence of 5 measurements within about 15 minutes. The presence of sub-visible cirrus or broken clouds within the field of view induces spurious variation in the atmospheric transmission (Smirnov et al., 2000) that can be easily detected by looking at the standard deviation of the 15-minute series of AOD measurements. An arbitrary threshold of 0.2 on the standard deviation has been selected to remove the cloud-contaminated observations. The diurnal variability range is expected to be less than 10% for our site conditions (Smirnov, 2002). The sun photometer observations are reported as daily averages.

The total uncertainty in AOD for the AERONET instruments is ± 0.01 for $\lambda > 440nm$ and ± 0.02 for shorter wavelengths (Holben et al., 1998). CALITOO sun photometers were calibrated prior to the site deployment using the Langley-plot method (Soufflet et al., 1992; Schmid and Wehrli, 1995) at the Izaña high-altitude observatory (Basart et al., 2009). ~~A direct comparison with a AERONET instrument indicates that the~~ CALITOO observations were compared to coincident AERONET observations before and after the field experiment. The total uncertainty in AOD ~~for CALITOO is~~ is estimated to ± 0.02 for all the wavelengths. ~~The post-field calibration was done using a reference AERONET instrument and~~ Post-field measurements indicates a change of about 1% per year in the calibration.

AOD measurements are all reported at 550 nm because this wavelength is a reference for visibility calculation (Boers et al., 2015) and satellite mission (e.g. Remer et al., 2008). The ~~Angström~~ Ångström exponent (AE) (Ångström, 1961) is computed between wavelengths 465 and 619 nm for the CALITOO and 440 and 670 nm for the HHC, and between 440 and 675 nm for the AERONET.

2.2 Satellite data

The Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol products (Remer et al., 2005, 2008) have been widely used by the scientific community for assessing the impact of aerosols on global climate (Boucher et al., 2013) or air quality (van Donkelaar et al., 2016). The MODIS AOD is also used in operational data assimilation for weather forecast (Benedetti et al., 2009; Lynch et al., 2016). ~~We have used the daily MODIS AOD at 550 nm from~~ The MODIS Level 2 product has a spatial resolution of 10 km $\times$ 10 km at Nadir but increasing to 20 km $\times$ 40 km at the edges of the swath. The MODIS Level 3 is a regular gridded product having a spatial resolution of $1^\circ \times 1^\circ$. Most of the time, the validation exercise of MODIS derived aerosol parameters consists in a comparison between sun photometers observations and MODIS L2 pixels co-located in space and time (Remer et al., 2020). A box of 5 $\times$ 5 MODIS pixels and a time slot of $\pm$ 30 min the satellite overpass is admitted to be a good compromise, however the window-size dependence is small and this compromise is more dictated by statistics rather than physics (Ichoku et al., 2002).

Gridded daily or monthly mean MODIS level 3 AODs have also been demonstrated to fit the AERONET retrievals (Ruiz-Arias et al., 201 As the objective of the paper is to address the ability of MODIS data to reflect aerosol changes in a specific area rather than the validation of retrieval algorithm, hereinafter we rely on the gridded level 3 MODIS product of AQUA satellite (namely MYD08_D3) from 2003 to ~~2018. The spatial resolution of the MODIS product is 1$\times$ 1. We~~ 2019. MODIS AQUA has been selected following the recommendations of Wei et al. (2019) for long-term trend analysis. This product provides several values for the AOD depending on the underlying surface and the algorithm used. For a sake of consistency between the different sites, we use the product named AOD_550_Dark_Target_Deep_Blue_Combined_Mean ~~and Deep_Blue_Angstrom_Exponent_Land_Mean~~ from the version ~~6~~ 6.1 (Levy et al., 2013) of the MODIS processing algorithm, which is a combination of the "Dark target" (Levy et al., 2010) and "Deep blue" (Sayer et al., 2013) methods. For the coastal sites, both AOD over land (namely Aerosol _Optical_Depth_Land_Mean or Deep_Blue_Aerosol_Optical_Depth_550_Land_Mean) and over ocean (Aerosol_Optical_Depth _Average_Ocean_Mean) are also provided. We use the "Deep Blue" AE (namely Deep_Blue_Angstrom_Exponent_Land _Mean) and compute an Ångström exponent from the spectral AODs over land and ocean, respectively. For the purpose of satellite validation, the satellite AOD and AE of the nearest cell to the photometer location are extracted. We have adopted the evaluation metrics proposed by Sayer et al. (2014) including the linear correlation coefficient, the median bias, the root mean square error, the mean absolute percentage error, and the fraction of data falling within the MODIS expected error (EE) given by ~~$EE = \pm 0.05 + 0.15 \times AOD$~~ $EE = \pm(0.05 + 0.15 \times AOD)$.

2.3 Surface concentration observations

From February 2015 to March 2017, Abidjan and Cotonou were equipped with PM2.5 monitoring stations (Djossou et al., 2018). Particles were collected on 47 mm diameter filters (quartz and PTFE filter types) at a flow rate of 5 L/min. Samplers were equipped with a PM2.5 mini Partisol impactor. PTFE filter were weighted before and after the sampling with a microbalance Sartorius MC21S.

The total volume of filtered air is measured by a GALLUS-type G4 gas meter. Mass concentrations of PM2.5 ~~mass concentrations~~ are estimated from the mass load of particles on the filters and the total volume of air~~sampled measured by a GALLUS-type G4 gas meter. Exposure~~ . The exposure duration of the ~~filter~~ filters is one week. ~~We have used the PM2.5 weekly observations collected at the urban site named "traffic" in Cotonou and the mean value at the two urban sites named "traffic" and "landfill" in Abidjan. In section 4, the AOD and AE are weekly averaged for the sake of comparison between PM2.5 and sun photometer observations.~~ A period of one week is sufficient to capture the main temporal pattern of atmospheric aerosols over a long period of time (Ouafo-Leumbe et al., 2017) and has been selected as a trade-off between logistics and observations.

3 Sun photometer results

3.1 Daily statistics

A total of 2323 handheld sun photometer observations (including data collected during the 2006 campaign) have been acquired. Starting and ending dates are reported in table 1 along with the number of observations, median and interquartile range (~~IRQ~~IQR) of AOD and AE distributions. We select the AERONET data until the end of the CALITOO observation period, i.e ~~October~~ March 2017 for a total number of 1248 daily observations. There is an excellent time coverage for the stations of Lamto and Cotonou by the CALITOO observations. Observations were performed for 66% of the time in Cotonou, and 68% in Lamto. As a comparison, this rate is 68% for the automatic sun photometer in Koforidua, indicating that handheld measurements can be as representative as the automatic ones. This rate drops to 24% in Abidjan, 39% in Savè due to operating issues leading to gaps in the time series.

~~The overall range of AOD is (~~Considering all the stations, the AOD ranges between a minimum of 0.07 ~~,3.8).~~ and a maximum of 3.76. The highest AOD acquired by the CALITOO instrument is ~~3.5~~ 3.50 in Cotonou in March 2015 and the highest AOD recorded by the AERONET sun photometer is ~~3.7~~ 3.76 in Ilorin in Dec. 2016. The median AOD ranges between a minimum of 0.47 at Lamto and maximum of 0.66 at Comoé. Considering all the daily measurements for all the sites, the median AOD is 0.52, IQR=(0.33, 0.82). AOD observations at Cotonou and Abidjan are rather similar, with a median AOD=0.55 (0.38, 075) and 0.58 (0.35, 0.86), respectively. The observations with the automatic sun photometer in Koforidua shows AOD in the same range as the 2 aforementioned stations, with a median AOD=0.49 (0.33, 0.84). The observations performed at Lamto with the 3 kinds of sun photometers show similar AOD: 0.47 (0.3, 0.72), 0.55 (0.35, 0.80) and 0.56 (0.38, 0.85) for CALITOO, HHC and AERONET respectively. The difference in the statistics for the 3 instruments at Lamto is due to different sampling periods

(see Table 1) although the coincident observations between CALITOO and AERONET in 2017 shows an excellent agreement (see below).

The median AE is between a minimum of 0.33 (0.13, 0.55) at Comé and a maximum of 0.78 (0.56, 1.09) at Koforidua. Observations performed with CALITOO at Lamto shows a slightly lower range of AE values than the ones performed with the HHC~~and AERONET. The bias toward lower AE is about -0.1. AEs in Cotonou AE~~. The difference between CALITOO median AE and HCC median AE is -0.09. However it should be noted that the statistical distribution of AOD values has an impact on the corresponding AE distribution (Wagner et al., 2008). AOD observed at Lamto during the HHC period (2006-2008) being higher than the ones observed during the CALITOO period (2014-2016), it is not expected to have the same AE values. Nonetheless, the difference is within the expected accuary.

Coincident AERONET and CALITOO observations (N=~~0.58 (0.32, 0.89) are lower than in Abidjan AE~~ 31) were acquired between January and March 2017 at Lamto (see Figure 3 and Table 1). Figure 2 shows the scatter plot for the corresponding daily AOD and AE There is an excellent agreement for both AOD (regression coefficient R=~~0.73 (0.44, 0.97), possibly reflecting a larger influence of mineral dust.~~ 0.93) and AE (R=0.87) between the two instruments.

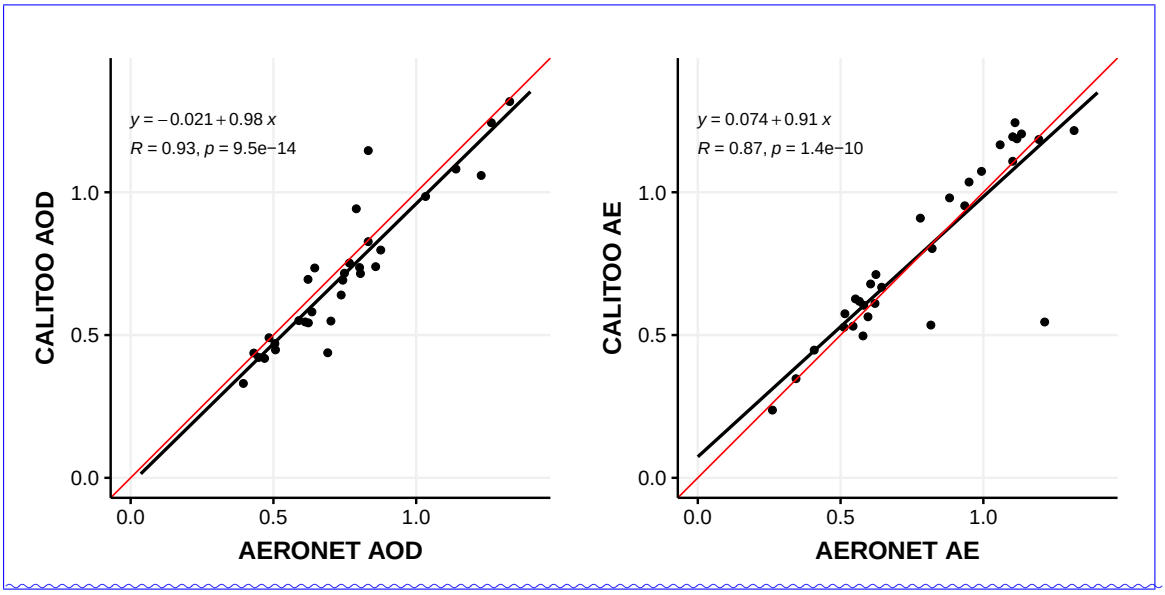

Figure 2. Scatter plot of AOD and AE observed at Lamto by CALITOO and AERONET instrument between January and March 2017 (number of points = 31).

Table 1. Summary of observations period, number of days of observations (N) per instrument and location. Median and interquartile range for aerosol optical depth (AOD) and ~~Angström~~ Ångström exponent (AE).

Site	Type	Latitude	Longitude	period	N	AOD median (IQR)	AE median (IQR)
Lamto	HHC	6° 13' N	5° 2' W	Mar. 2006- Mar. 2008	524	0.55 (0.35, 0.80)	0.68 (0.42, 0.96)
Abidjan	CALITOO	5° 20' N	3° 59' W	Feb. 2015- Apr. 2017	190	0.55 (0.38, 0.75)	0.73 (0.44, 0.97)
Lamto	CALITOO	6° 13' N	5° 2' W	Nov. 2014- ~~Jun. 2016~~ Mar. 2017	499	0.47 (0.30, 0.72)	0.59 (0.35, 0.86)
Save	CALITOO	8° 01' N	2° 28' E	Sep. 2015- Oct. 2017	411	0.61 (0.42, 0.86)	0.49 (0.26, 0.73)
Comoe	CALITOO	8° 27' N	3° 28' W	Jan. 2016- Feb. 2017	82	0.66 (0.43, 0.95)	0.33 (0.13, 0.55)
Cotonou	CALITOO	6° 22' N	2° 26' E	Nov. 2014- Jun. 2016	615	0.58 (0.35, 0.86)	0.58 (0.32, 0.89)
Lamto	AERONET	6° 13' N	5° 2' W	Jan. 2017- ~~Oct~~Mar. 2017	35	0.74 (0.59, 0.83)	0.82 (0.58, 1.08)
Ilorin	AERONET	8° 29' N	4° 40' E	Jan. 2014- ~~Oct~~Mar. 2017	472	0.52 (0.30, 0.89)	0.63 (0.39, 1.00)
Koforidua	AERONET	6° 6' N	0° 6' W	Dec. 2015- ~~Oct~~Mar. 2017	264	0.54 (0.32, 0.92)	0.78 (0.56, 1.09)

3.2 Time series

The daily AODs and AEs for each site and each instrument between 2015 and 2017 (between 2006 and 2008 for HHC) are presented in Figure 3 and 4~~.~~ , respectively. A similar seasonal pattern is observed in the different time series. There is an increase in AOD during the main dry season (December to March) and a decrease during the rainy season (April-July). The 2-week smoothing average reveals a high degree of correlation between time series. The correlation coefficient between Cotonou and Lamto time series is R=0.82~~and~~ , being R=0.90 between Cotonou and Koforidua. During the short overlap period in March 2017, the CALITOO and AERONET instruments shows similar AOD values at Lamto station. The Comoé time series is the weakest one with only 82 data points. The 2006-2008 HHC Lamto time series has the same pattern as the one recorded by CALITOO in 2015-2017 and showing two maxima in the dry season, one in December and another one in January-February.

The seasonal pattern for AEs (~~right side of~~ Figure 4) shows an opposite cycle with lower values in the dry season and higher values during the rainy season. AE seasonal cycle is clearly affected by the winter dry period with dust laden air masses that decrease the AE values. The median AE value during the first half of the year (all site except Comoé) is 0.36 (0.23, 0.62) and 0.69 (0.43, 1.00) during the second half of the year.

AOD in the last quartile (AOD above 0.82) are mostly (72%) observed during the month of December, January and February and are associated with a median AE of 0.44 (0.24, 0.64). While low AODs (first quartile, i.e. below 0.33) are associated to a median AE of 0.89 (0.61, 1.12) and observed between August and October (51% of the observations).

The difference in AOD between the inland and the coastal sites is less than 0.05, with differences up to 0.1 between April and June, the AOD at the coastal stations being higher than inland. AE are higher at the coastal stations than at the inland stations by 0.15 on average, reflecting the influence of urban air pollution at the coastal stations.

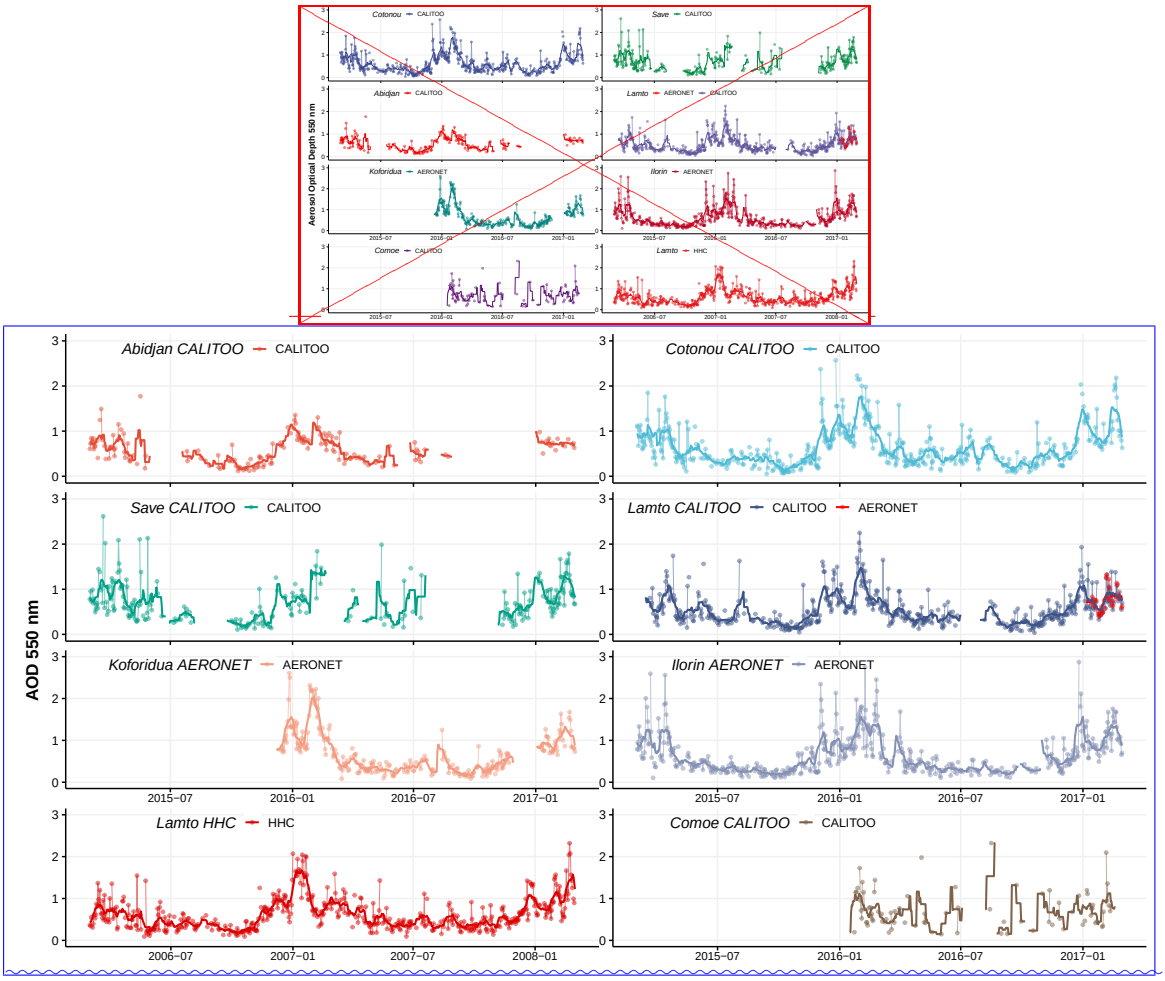

Figure 3. Daily Aerosol Optical Depth at 550 nm. The name of the site and the type of instrument used is given in the legend of each plot. Solid line is a 2-week smoothing average.

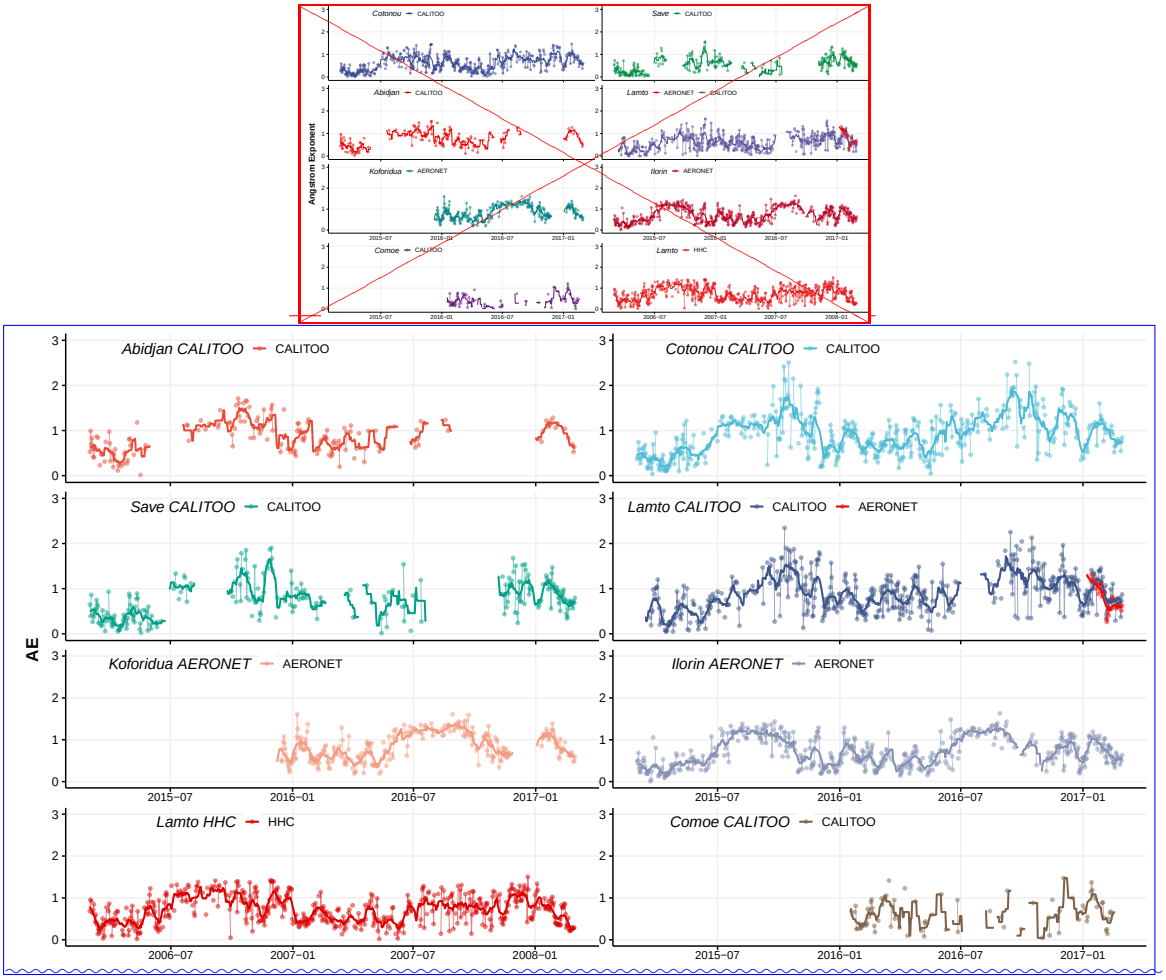

Figure 4. Same as Figure 3 for the visible ~~Angstrom Exponent~~Ångström exponent.

3.3 Comparison with MODIS aerosol products

Table **??** gives the statistics of the regressions for each site and per instrument presented in Figure 5. We have then adopted a log-log representation on the scatter plots presented in Figure 5 as the AOD distribution has a significant right skewness (O'Neill et al., 2000). Figure 5 presents also the MODIS expected error (EE, blue lines). Whatever the site is, there is a significant correlation between the MODIS and sun photometer observations. The Pearson correlation coefficient R ranges between 0.75 (Comoé) and 0.94 (Koforidua). For the CALITOO observations, R is between 0.75 and 0.90 (Cotonou). The lowest RMSE values are found for the measurements operated using the CALITOO at the coastal sites of Abidjan and Cotonou. The MAPE is on average 30%. The sites in Cotonou and Abdijan are not biased and present a fraction of data falling within the MODIS EE above 60%. All the inland sites are biased and it results in a rather low fraction of data falling within the MODIS expected error.

Table 2. Results of the MODIS and sun photometer AOD comparison by site location and type of instrument indicating the (N) number of data, (R) Pearson correlation coefficient, (RMSE) root mean square error, bias, (MAPE) mean absolute percentage error and (fEE) fraction falling within the MODIS expected error.

site	~~type~~ instru	N	R	RMSE	Bias	MAPE ~~(%)~~	fEE ~~(%)~~
~~Savé~~ Cotonou	CALITOO	~~295~~ 401	~~0.80~~ 0.88	~~0.31~~ 0.22	~~-0.16~~ -0.02	~~33~~ 24	~~35~~ 64
~~Abidjan~~ Save	CALITOO	~~142~~ 254	~~0.86~~ 0.79	~~0.14~~ 0.31	~~0.00~~ -0.19	~~21~~ 35	~~70~~ 35
~~Cotonou~~ Abidjan	CALITOO	~~458~~ 118	~~0.90~~ 0.86	~~0.21~~ 0.14	~~0.01~~ -0.02	~~29~~ 18	~~61~~ 76
Lamto	CALITOO	~~235~~ 185	~~0.89~~ 0.86	~~0.22~~ 0.26	~~-0.13~~ -0.15	~~28~~ 29	50
~~Comoé~~ Comoe	CALITOO	~~53~~ 47	~~0.75~~ 0.76	~~0.36~~ 0.37	~~-0.21~~ -0.22	~~29~~ 32	~~50~~ 44
Ilorin	AERONET	~~327~~ 264	~~0.93~~ 0.91	~~0.27~~ 0.32	~~-0.15~~ -0.19	~~30~~ 33	~~40~~ 39
Koforidua	AERONET	~~177~~ 144	~~0.94~~ 0.93	~~0.27~~ 0.30	~~-0.18~~ -0.20	~~27~~ 26	~~50~~ 46
Lamto	AERONET	~~21~~ 17	~~0.86~~ 0.87	~~0.32~~ 0.34	~~-0.29~~ -0.32	~~47~~ 50	~~9~~ 0
Lamto	HHC	~~219~~ 181	~~0.84~~ 0.83	~~0.34~~ 0.39	~~-0.24~~ -0.29	~~33~~ 37	~~34~~ 25

The bias has a seasonal behavior being highest during the dry season between December and March. An underestimation of the MODIS AOD is then observed at maximum in January with an absolute bias of -0.33 (39% in relative) at the inland sites. Sayer et al. (2014) have already pointed out the possible differences in the "Dark Target" and "Deep blue" algorithms. It appears from the Figure 6 in Sayer et al. (2014) that the dry to humid savanna transition zone in sWA is an area where large differences exist in both retrieval techniques during the dry season. Those differences can explain that the "Merge" product used in this study has a large bias during the dry season in the northern part for the inland sites. So the North-South AOD gradient in this area remains difficult to assess based on satellite products.

For all the sites considered in this study and whatever the sun photometer is, the correlation between MODIS AE and sun photometer AE is non significant. This finding is coherent with the results of Antuña-Marrero et al. (2018) and Sayer et al. (2013). The histogram of the Sun photometer and MODIS-derived AE are presented in Figure 6. The default values AE=1.5 and AE=1.8 in the MODIS AE Deep Blue product have been removed (Sayer et al., 2013). The MODIS median AE is bi-

ased by 0.32 and the distribution of AE values doesn't follow a normal distribution, whereas the distribution of AE values for the sun photometers does. ~~For all the sites considered in this study and whatever the sun photometer is, the correlation between MODIS and sun photometer AE is non significant. This finding is coherent with~~ At the coastal sites, the ~~results of Antuña-Marrero et al. (2018) and Sayer et al. (2013)~~MODIS AE Ocean algorithm reproduces well the left side of the histogram (lowest AE) while it significantly underestimates AE above 0.8. MODIS AE Land algorithm at 0.5 indicates that a dust-like aerosol model has been selected (Levy et al., 2010) however there is no systematic association with low sun photometer AE. However it could be possible to adapt the interval bounds (lower and upper limits of AE values) for each category, the statistical distribution of MODIS AE values doesn't fit the sun photometer ones and could lead to misclassification of daily observations.

4 Aerosol type and relationship with surface concentrations

~~AE~~

4.1 Aerosol type

AE is an intensive aerosol optical parameter and depends on the ~~aerosol~~ spectral aerosol extinction coefficient (Nakajima et al., 1996; Eck et AE is influenced by the aerosol size distribution and ~~aerosol optical properties (Nakajima et al., 1996; Eck et al., 1999; Holben et al., 2001)~~ is commonly used to identify aerosol types (Léon et al., 1999; Kaskaoutis et al., 2009; Perrone et al., 2005). Aerosol types having a dominant fraction of their size distribution in the coarse mode, like dust and sea-salt particles, are associated with a lower value of AE than aerosol types having a size distribution dominated by the accumulation mode, like secondary and combustion aerosols. The concurrent changes in AOD and AE help to distinguish generic aerosol types in sun photometer time series (Toledano et al., 2007; Verma, 2015). Mineral dust tends to increase atmospheric AOD and decrease AE (Hamonou et al., 1999) while biomass burning events tends to increase both AE and AOD (Eck et al., 2003). ~~The~~

The AOD versus AE scatter plot can be used to cluster the observations by aerosol broad categories corresponding to a main source, like coarse mineral dust of biomass burning aerosols. The thresholding in AOD and AE for aerosol type identification varies from one site to another and also depends on the distance from aerosol sources upwind the site (Verma, 2015; Benkhalifa et al., 2017). In particular, the classification based on AOD vs. AE values is incapable of determining aerosol absorption properties (Giles et al., 2012; Cazorla et al., 2013). Figure 7 presents the scatter plots of AODs (log scale) versus AEs for each site and splitted by seasons. We have considered 4 seasons corresponding to the long dry season (Dec.-Mar), the long wet season (Apr.-Jun.) and the short dry (Aug.-Sep.) and short wet season (Oct.-Nov.). For the sites with the most comprehensive data set over the different seasons (Lamto, Cotonou, Koforidua, Ilorin) the AOD vs. AE plots show a similar pattern with decreasing AODs almost linearly as AEs increases. The lowest AEs are observed during the long dry season and are associated to largest AODs indicating the presence of coarse mineral dust. The presence of dust can be also observed in the wet long season. During the short dry season, all the sites excepted Comoé show larger AEs and lower AODs than for the other seasons.

It can be noticed from Figure 7 that there is no clear definition of AOD and AE thresholds for each aerosol categories and the scatter plots of Figure 7 reflects the high mixing of different aerosol types. The absolute error on AE is a function of the relative error on AODs and depends on the spectral range investigated (Hamonou et al., 1999; Wagner et al., 2008). Typical error on AE is $\pm$ 0.3 for an AOD of 0.2 and there is a risk of over interpretating AE variations.

In this paper we classify the daily observations according to the AE values using a simple statistical analysis and a threshold on AOD. The whole sun photometer dataset is divided into 3 quantiles. The first third corresponds to AE$\leq$0.45 and observations having an AOD $\geq$ 0.8 are labelled "coarse dust". ~~The~~ , while observations having an AOD<0.8 are labelled as "mixed". The threshold on AOD corresponds to the third quantile of AOD distribution and is used to better identify dust events. The last third corresponds to AE$\geq$0.80 and is labelled "urban-like". The data having 0.45<AE<0.80 falls into a "mixed" category~~.~~, being more populated than the two others. This rather crude classification ~~enable~~ enables to identify the main aerosol influence with a significant number of observations in each ~~categories~~category.

Table 3. Percentage of daily observations ~~for~~ in the aerosol categories at each site~~and by aerosol category considering data falling within the AOD interquartile range and above the last quartile (numbers in brackets)~~.

site	~~Urban-like~~ Coarse dust	Mixed	~~Coarse Dust~~ Urban-like
Abidjan	~~34.4 (25.6)~~3.7	~~37.7 (35.9)~~ 56.8	~~27.9 (38.5)~~39.5
Comoe	~~10.9 (3.8)~~ 22.0	~~41.3 (1.5)~~ 69.5	~~47.8 (84.6)~~8.5
Lamto	~~37.2 (11.7)~~8.6	~~35.7 (32.5)~~ 56.5	~~27.2 (55.8)~~34.9
Save	~~22.1 (13.0)~~12.2	~~32.8 (30.1)~~ 67.9	~~45.1 (56.9)~~20.0
Cotonou	~~34.3 (15.9)~~10.2	~~26.3 (35.2)~~ 57.6	~~39.4 (48.9)~~32.2
Ilorin	~~28.9 (12.8)~~12.7	~~39.3 (32.6)~~ 50.6	~~31.8 (54.6)~~36.7
Koforidua	~~46.5 (27.3)~~4.9	~~39.5 (55.7)~~ 47.3	~~14.0 (17.0)~~47.7

Table 3 presents the typology of the sites according to the aforementioned classification. ~~The classification is given for the observations falling within the AOD IQR (see table 1). We also report the percentage of observations having AOD in the last quartile of the AOD distribution to highlight the contribution of the different aerosol types to the aerosol events . As expected~~ Comoe is the ~~percentage of each catergory for a given site represents roughly one third of the data set~~most influenced by coarse dust aerosol (20%), followed by Ilorin (12.7%), Savè (12.2%), Cotonou (10.2%) and Lamto (8.6%). The southwestern sites, Abidjan (3.7%) and Koforidua (4.9%) are less influed by dust events than the eastern sites. As expected from Figure 7 all the site shows 'urban-like' category that also corresponds to low AODs. Two sites are less influenced by urban-like aerosols than the others, namely Save ~~and Comoe. For all the sites excepted Koforidua, most of the days showing a large AOD are associated with coarse dust events. Cotonou is also also more influenced bu Coarse Dust (39%) than Abidjan (28%). The northernmost sites are more affected by dust events and~~ (20%) and Comoe (8.5%). For the other sites, the 'urban-like' category ranges between 47.7% (Koforidua) to 32.2% (Cotonou).

As SWA lacks dedicated studies on aerosol characterization, there are few other data to compare with. Hamill et al. (2016) have proposed a sophisticated aerosol classification for Africa based on AERONET observations however none of the sites are

located in our area. Hamill et al. (2016) have classified Djougou (Northern Benin located at 9° 42' N) as a dust site that is seldom affected by biomass burning. Savè and Ilorin are located around 200 km south of Djougou and the influence of ~~coarse dust on the AOD decreases from North to South and East to West.~~ dust is still significant compared to the coastal sites. Comoé is located 600 km westward of Djougou and probably less influenced by the dust transport from the Bodélé area in Chad however measurements acquired at Comoé don't cover a full season and the exact frequency of dust or biomass burning events remains uncertain.

~~As reported by Djossou et al. (2018), the~~

4.2 Relationship to surface concentrations

The changeover between the monsoon and the harmattan ~~circulation leads to a drastic~~ results in a change in the ~~aerosol type and stratification~~vertical distribution of aerosol layers and in the type of aerosols Djossou et al. (2018). The harmattan flow carries continental aerosols in the lowest part of the atmosphere during the ~~long~~ dry winter season ~~. During this period, the~~ (December to March). During the dry winter season the days with high AOD are often associated with an increase in the PM2.5 surface concentration ~~. As a consequence, the~~ leading to a high correlation coefficient between AOD and PM2.5~~is the highest during the dry long season (Djossou et al., 2018). Considering the whole dataset of~~ .

The correlation coefficient between weekly mean AOD and PM2.5 ~~and AOD (weekly basis)~~ measured in Cotonou and Abidjan ~~, the correlation between PM2.5 and AOD is significant with a correlation coeff.~~ is R=0.75 ~~,~~ (N=105) ~~. The correlation~~ when considering the whole observation period. The correlation coefficient can reach R=0.96 (N=6) during specific aerosol events observed from December 2015 to January 2016 in the heart of the dry season. During other periods of the year, the correlation remains weak because the concentrations are less fluctuating than during the winter period.

~~The variation in the~~ PM2.5/AOD ratios are estimated using the daily AOD observations and the weekly PM2.5. The PM2.5/AOD is basically the amount of PM2.5 that is expected per unit of AOD. It was first promoted by van Donkelaar et al. (2010) as a conversion factor (Zheng et al., 2017; Yang et al., 2019). The PM2.5/AOD ratio ~~is estimed as a function of the season and the aerosol type. The data set is divided into 4 periods: the long dry season (Dec. -Mar), the long wet season(Apr.-Jun.) and the short dry (Aug. -Sep.) and short wet season(Oct. -Nov.) Weekly mean AODs are calculated by aerosol category and the~~ reflects how a change in the AOD affects the ground surface concentrations, however there is no evidence of a unique relationship between both quantities. The PM2.5/AOD ratio depends on the vertical stratification of the aerosol layers in the atmosphere due to mixing processes in the boundary layer or large scale advection (Sayer et al., 2016). The ratio depends also on the aerosol size distribution and chemical properties that are changing during the transport and the aging of the aerosols.

In the specific case of the coastal cities of the Gulf of Guinea, we are interested in evaluating how the change in aerosol type during the season, and in particular the seasonal advection of mineral dust from the desert area, may affect the PM2.5 surface concentrations. For this purpose, we estimate a PM2.5/AOD ratio per aerosol type and per season.

Each daily AOD observation is associated to an aerosol type (Coarse dust, mixed or urban-like) depending on the corresponding daily AE value. The daily AODs are associated to the corresponding PM2.5 observation using equation 1.

$$PM2.5_{weekly} = \frac{1}{n}\sum_{i=1}^{n}(\sum_{t=1}^{3}\beta_{t,s}\tau_{i,t,s}) \quad (1)$$

The corresponding PM2.5/AOD ~~ratii are determined from~~ coefficients, $\beta_{t,s}$ in equation 1 where t represents the aerosol type and s the season, are evaluated using a multilinear regression ~~analysis between the weekly PM2.5 and the AODs.~~ on the observations collected in Cotonou and Abidjan for each season independently. Cotonou and Abidjan samples are pooled together to increase the statistical significance and to retrieve average coefficients at the regional level. As the season are not equal in length and the number of observations differs in Abidjan and Cotonou, the number of samples differs, ranging between 71 samples during to long dry season to 24 samples during the dry short season. The significance of the regression and standard error on the coefficients depends on the number of samples. None of the weeks in the short dry period are affected by dusty days, so the coefficient for coarse dust is not retrieved for this period. During the short wet season, only 2 weeks over 26 have a dust contribution and the coefficient is not significant. The PM2.5/AOD ratios by aerosol category are presented in Figure 8 as a function of the season and with their respective uncertainties. The average PM2.5/AOD ratio ~~ranges between 11 $\pm$ 4 $\mu g/m^3/AOD$ in the short dry season for the Mixed category upto 78 $\pm$ 12 $\mu g/m^3/AOD$ in the short wet season for coarse dust~~without accounting for the aerosol category for a given season is also reported. The uncertainties corresponds to the standard error of the coefficients found by regression. The standard error depends on the ~~occurence~~ occurrence of an aerosol category ~~for~~ and its relative weight in a given season. For all the season the coefficients for each aerosol type are significant ($p<0.05$) except for the coarse dust category during the short dry (no data) and the short wet season. The resulting adjusted coefficient of determination for the regression is between 0.76 (long dry season) and 0.83 (wet short season).

The PM2.5/AOD ratio for ~~coarse dust decreases from 49~~ Coarse dust aerosols ranges between 54 $\pm$ ~~6~~ 8 $\mu g/m^3/AOD$ ~~to 29~~ in the dry long season to 20 $\pm$ ~~3~~ 4 $\mu g/m^3/AOD$ ~~between the long dry and long~~ in the long wet season. ~~This change in the ratio reflects the seasonal change in the altitude of coarse dust transport~~The seasonal changes in the PM2.5/AOD ratio for coarse dust reflects well the vertical shift of the dust layer between the dry and the wet season. During the wet (~~long~~Apr-Jul) season, the air masses are uplifted by the monsoon flow. PM2.5 concentrations ~~remains moderate~~ remain moderate (21 $\mu g/m^3$ in April) while AODs are still significant ~~due to~~ (0.57 on average in April) due to the aloft transport. ~~Conversely the~~ The impact of coarse dust on PM2.5 is higher during the dry season (higher ratio and high AODs) when the dusty air masses are advected close to the ground surface.

The PM2.5/AOD ratio ~~remains rather constant from the long dry to~~ for Mixed aerosols ranges between 53 $\pm$ 7 $\mu g/m^3/AOD$ in the dry long season to 27 $\pm$ 11 $\mu g/m^3/AOD$ in the short dry season~~, between 45~~. During the short dry season, only 30% of the weeks are are affected by mixed type aerosols, the remaining being classified as urban-like. The corresponding PM2.5/AOD ratio for mixed type aerosol is close to the one found for dust during the previous season (long wet) indicating the aloft dust transport can be still active but incorrectly classified to the mixed aerosol type due to a low intensity (small AOD).

The PM2.5/AOD ratio for Urban-like aerosols ranges between 96 ± ~~4~~ 15 $\mu g/m^3/AOD$ ~~and 47~~ in short wet season to 37 ± ~~16~~ 5 $\mu g/m^3/AOD$ ~~. This ratio increases in the short season and reaches 78 ± 12 $\mu g/m^3/AOD$. This later period shows the largest uncertainties on the estimation~~ in the long wet season. The PM2.5/AOD ratio retrieved in the long dry season for the urban-like category are affected by a larger uncertainty due to a limited impact of Urban-like aerosol during the long dry season compared to Coarse dust. There is a shift of the PM2.5/AOD ratio ~~due to moderate AODs leading to a less accurate regression.~~ toward higher value during the short wet season. The short wet season is a transition period during which the stagnation of air masses over land favors the accumulation of pollutants, and also combustion by-products emitted over Nigeria (Marais et al., 2014).

Figure 9 presents the weekly average AODs, satellite-derived and in situ PM2.5 for both Abidjan and Cotonou~~together~~. The label attributed to each week corresponds to the ~~dominant aerosol type during the week based on daily AOD~~ aerosol type having the largest mean AOD over the week. The period from March to May is dominated by the coarse dust type and there is a clear shift to urban-like type in June-July. A second period of coarse dust is observed in December (2015 and 2016) and is associated to a significant increase in both AOD and PM2.5. PM2.5 during the dusty period of December rise ~~close to~~ over 100 $\mu g/m^3$. Another sharp increase is observed in February and is associated to the mixed aerosol type. For both years, the two intense periods (December and February) are ~~separeted~~ separated by an interim period showing moderate PM2.5 and AOD and classified as urban-like aerosols.

On average, satellite-derived PM2.5 are ~~in an excellent agreement~~ agrees with the in situ PM2.5 observations. Indeed the mean difference between retrieved and observed PM2.5 during the 2015-2016 period is less than 1 $\mu g/m^3$ (3%). The MAE is ~~9.8~~ 14 $\mu g/m^3$ and the RMSE is ~~14.6~~ 21 $\mu g/m^3$. The RMSE found here is within the range of previous studies (Ma et al., 2015; Sinha et al., 2015) for other regions of the world and different algorithms. The very intense periods are underestimated, e.g. the mean difference between retrieved and observed PM2.5 is ~~-20~~ -51 $\mu g/m^3$ (~~-25~~-70%) in December ~~2015. However the~~ 2015 in Cotonou and nearly a factor of 2 lower in December 2016. The satellite-derived concentrations in January and in March are overestimated. ~~The smoothing effect of~~ Despite introducing a characterization of the aerosol type, there is still a clear smoothing effect on the weekly concentrations ~~can result from the ajdustment~~ that results from the adjustment of the regression coefficients on a seasonal basis. ~~Indeed the the mean difference between retrieved and observed~~ Using seasonally adjusted coefficients only rather than the seasonally and aerosol type adjusted coefficients has a limited impact on the comparison and decrease the RMSE by only 2 $\mu g/m^3$ on average. The biggest impact is during the long wet season (20% decrease in RMSE) when a lower PM2.5~~during the 2015-2016 period is only 3 $\mu g/m^3$ (6%)~~/AOD coefficient is selected for the identified dust cases.

5 Trend in the MODIS-derived PM2.5 time series

We have applied the PM2.5/AOD conversion factors to the daily MODIS AOD observations between 2003 and ~~2018.~~ 2019. As the MODIS AE can't be used to classified the daily aerosol observations, we have applied the mean seasonaly adjusted PM2.5/AOD ratios. The database of daily AOD observations consists in ~~3170~~ 2675 observations for the area of Abidjan and

~~3573 for the the~~ 3018 for the area of Cotonou~~. Daily mean~~ , corresponding to about 160 observations per year on average. To increase the number of observations per year, we introduce a new area named SWA located between 7° W and 5° E and between 6° N and 4° N. There is at least one MODIS L3 observation per day in this largest coastal area that encompasses both cities.

Mean retrieved PM2.5 is ~~31.4~~ 28.3 ± ~~19.6 and 34.5~~ 22.2, 30.5 ± 24 $\mu g/m^3$ in Abidjan and Cotonou, respectively. Almost all the years have a annual average above the EU target value of 25 $\mu g/m^3$, except 2003, 2013, 2014 and 2019. More than 90% of the daily observations are above 10 $\mu g/m^3$ for both cities. ~~Maximum are~~ A maximum is observed as high as 300 $\mu g/m^3$ during dust event in winter 2010 in Abidjan. During the long dry season 80% of the days have a value above 35 $\mu g/m^3$ while this number drops to 4% during the short dry season.

The MODIS-derived PM2.5 monthly mean ~~seasonal cycle is~~ annual cycle given in Figure 10 for both cities and SWA area reflects this large seasonal change in the concentrations. A first period is observed between December to March when concentrations are at highest. During this period, the overall ~~median~~ mean PM2.5 value is ~~43~~ 47 $\mu g/m^3$, concentrations in Cotonou being ~~20%~~ higher than in ~~Abdijan.~~ Abidjan (max 11% in January). We observe a large difference in April (18% higher in Cotonou) that is clearly attributed to a change in the contribution of coarse dust(+34% in Cotonou) while the contribution of other types remains the same. This higher contribution of dust during the dry period and even more during the intermediate period over Cotonou could be associated to the higher proximity of Cotonou to major dust sources (Bodélé depression) and preferential advection pathways.

A second period is observed between ~~April~~ May and September showing ~~median~~ mean PM2.5 ~~around 20~~ below 16 $\mu g/m^3$ for both cities and the whole area. The third period corresponds to a steady increase in PM2.5 between September to the December. PM2.5 ~~concentrations are then multiplied~~ mean concentration over SWA area is around 11 $\mu g/m^3$ in September and increases up to 37 $\mu g/m^3$ in December, corresponding to an increase by a factor of about 3 in 4 months. A similar increase is observed for Abidjan and Cotonou.

~~The percentage of daily observations falling in the coarse dust aerosol category is about 60% from January to April. The urban-like category represent 10% of the daily observations during the same period. The percentage of observations identified as coarse dust decreases from April to July down to 20% on average. From July to October, the urban-like category represents 50% of the observations.~~

~~There is no obvious trend in the MODIS-derived~~ The monthly mean PM2~~.5 annual time series~~.5 displayed on Figure 11A shows the strong seasonal variation with highest values in January or February every years. The trend on monthly means is retrieved after a seasonal decomposition using a procedure based on Loess (Cleveland et al., 1990). The trend does'nt have an obvious pattern however one can observe a pseudo-cycle of 4 to 5 years. We can notice a decrease in the mean concentrations after 2017. The drop in 2018 and 2019 is due to lower AODs for those two years. The annual mean AOD decreases by 20% between 2017 and 2019 however we didn't investigated further a possible explanation for the decrease. Mann-Kendall'~~s trend test on annual median values~~ seasonal trend test (Hirsch et al., 1982) applied on monthly means is not significant ~~(tau=-0.31, p-value=0.11). Figure 10 represents the annual boxplots of daily concentrations between 2003 and 2018. The annual boxplots considering only the days during which the AOD belongs to the~~ over the whole 2003-2019 period.

To further investigate a possible trend in the urban-like ~~aerosol category are also plotted on Figure 10. The drop in the concentration in 2018 is not clearly explained~~aerosol, we have selected the data acquired during the short dry period (August-September) during which no dust event have been detected in our sun photometer dataset. Over the period 2003-2017, we observe a ~~significant increasing trend~~ monotonic trend (Mann-Kendall's tau=0.48) in the annual ~~values (tau=0.50, p-value=0.01~~mean MODIS-derived PM2.5 over the SWA box (Figure 11B). The Thiel-Sen's slope over 2003-2017 is ~~0.32~~ 0.20 with an 95% confidence interval of ~~(0.11, 0.51). This weak relatively weak trend leads to an increase of about 5 $\pm$ 3~~ [0.04, 0.43] corresponding to a monotonic increase in PM2.5 of 3.0 $\mu g/m^3$ over 15 years~~(25% in relative).~~ . The large uncertainty in the observed trend during the short dry period is due to the low PM2.5 concentrations observed during this period (see Figure 10). As PM2.5 are directly linked to AOD, any bias occuring in AOD will affect the PM2.5 concentrations. Moreover the drift in the MODIS AQUA calibration expressed in AOD per decade is 0.01 (Sayer et al., 2019) and will lead to an increase of the same order of magnitude when considering the corresponding PM2.5/AOD conversion coefficient.

6 Conclusions

An increase in the anthropogenic emission of atmospheric pollutants is expected as a result of the massive urbanization of the Gulf of Guinea. The scarcity of ground-based observations in sWA is still a limiting factor for a comprehensive understanding of the short-time trend over growing African cities. Moreover, the large influence of natural aerosol emission in sWA produces a complex mixing of particles in the ~~cities atmosphere~~ urban atmosphere of sWA cities. In this paper, sun photometer and satellite observations have been used to characterize the magnitude and seasonal behaviour of the aerosol optical depth in sWA. We have set up a small network of lightweight handheld sun photometers, that provides an unprecedented data set on the AOD over sWA between 2015 and 2017. This data set was complemented by additional measurements from AERONET data and observations obtained during a previous campaign in 2006 in Côte d'Ivoire. The comparison of our observations with the MODIS Level 3 gridded satellite observations shows that the satellite AOD derived in the vicinity of the coastal conurbation are excellent, while there is a possible negative bias for the retrievals farther inland, that must be further investigated. Reversely the MODIS AE doesn't fit the sun photometer observations.

A basic classification using the AOD spectral dependency reveals the large impact of the advection of mineral dust on the AOD seasonal cycle. Dust impacts the cities of the northern part of the gulf of Guinea (namely Abidjan and Cotonou in the present study) from December to May and brings the largest AODs during the months of December and February.

Weekly surface PM2.5 in Abidjan and Cotonou and daily AOD observations were used to estimate a set of AOD to PM2.5 conversion coefficients that accounts for the aerosol ~~typing~~ category and the season. Despite a good agreement for most of the year, the retrieved PM2.5 underestimates the actual concentrations during the large aerosol events in the dry season. Reversely the PM2.5 are overestimated in early march as a consequence of the shift in altitude of the Harmattan wind. Nonetheless the seasonal variability of the PM2.5 concentrations is in a good agreement with the actual ones. ~~The~~

The seasonal PM2.5/AOD conversion coefficients are applied to the MODIS AOD time series from 2003 to ~~2018. A significant increase is observed during the 2003-2017 period in~~ 2019. It was not possible to adjust the PM2.5~~during the days~~

~~falling in the urban-like aerosol category~~/AOD ratio both seasonally and by aerosol type due to the lack of precision in the MODIS AE. No obvious trend is observed in the mean monthly concentrations however trend fluctuates with pseudo period of 4 to 5 years. A link to the 5-year cycle of rainfall in the Sahel (Brandt et al., 2019) could be involved as rainfall is one of main driver of dust emission (Prospero and Nees, 1977) and also as it controls the amount of biomass that can be burnt.

An increase in MODIS-derived PM2.5 is observed over the 2003-2017 period during the short dry period (August-September). The trend corresponds to an increase of ~~25~~20% over 15 years.

There are several mechanisms that can lead to the increase in the anthropogenic PM2.5 concentrations. Combustion sources are subject to an increase in sWA as well as for the rest of Africa, e.g. organic carbon emissions are multiplied by a factor between 1.5 and 3.0 over 2005-2030 ~~Liousse et al. (2014). However recent~~ (Liousse et al., 2014). The conurbations of the Gulf of Guinea are under the influence of gas flaring emissions in the Niger delta area (Ologunorisa, 2001). Recent studies show a decrease in gas flaring emissions in the Niger delta area (Deetz and Vogel, 2017; Doumbia et al., 2019) ~~, the conurbations of the Gulf of Guinea are directly under the influence of such emission the~~ but the impact of the year-to-year variability of such emissions on regional atmospheric concentrations has to be ~~investigated~~ further investigated. The increase found ~~in the urban-like PM2.5~~ during the short dry period corresponds to an average annual growth rate of ~~1.48% in agreement with~~ 1.1% being in the lower bound of the emission scenarii ~~. However~~ however there is no evidence that the observed trend ~~in our data is~~ is direclty linked to an increase in the ~~local city~~ urban emissions. The phenomena can also be linked to the possible advection of biomass burning byproducts from central Africa and crossing the gulf of Guinea resulting from the zonal transport (Menut et al., 2018; Flamant et al., 2018). ~~In addition to satellite data, unraveling the causes and consequences of the changes in aerosol~~

While sWA has received little attention regarding anthropogenic urban emissions, our study reports new observations and original analysis. Additional ground-truths and advanced satellite aerosol products or combination of products targeting on aerosol attribution are required to unravel the relative impact of anthropogenic versus natural aerosol emissions on atmospheric concentrations in this area of the world that is under a ~~significant anthropogenic pressurerequires sustaining long-term in situ observations of the aerosol concentrations and chemical composition~~growing anthropogenic pressure.

Data availability. Handheld sun photometer and PM2.5 data are available at http://baobab.sedoo.fr/. AERONET sun photometer data are available at https://aeronet.gsfc.nasa.gov/. MODIS aerosol data can be downloaded from https://ladsweb.modaps.eosdis.nasa.gov/.

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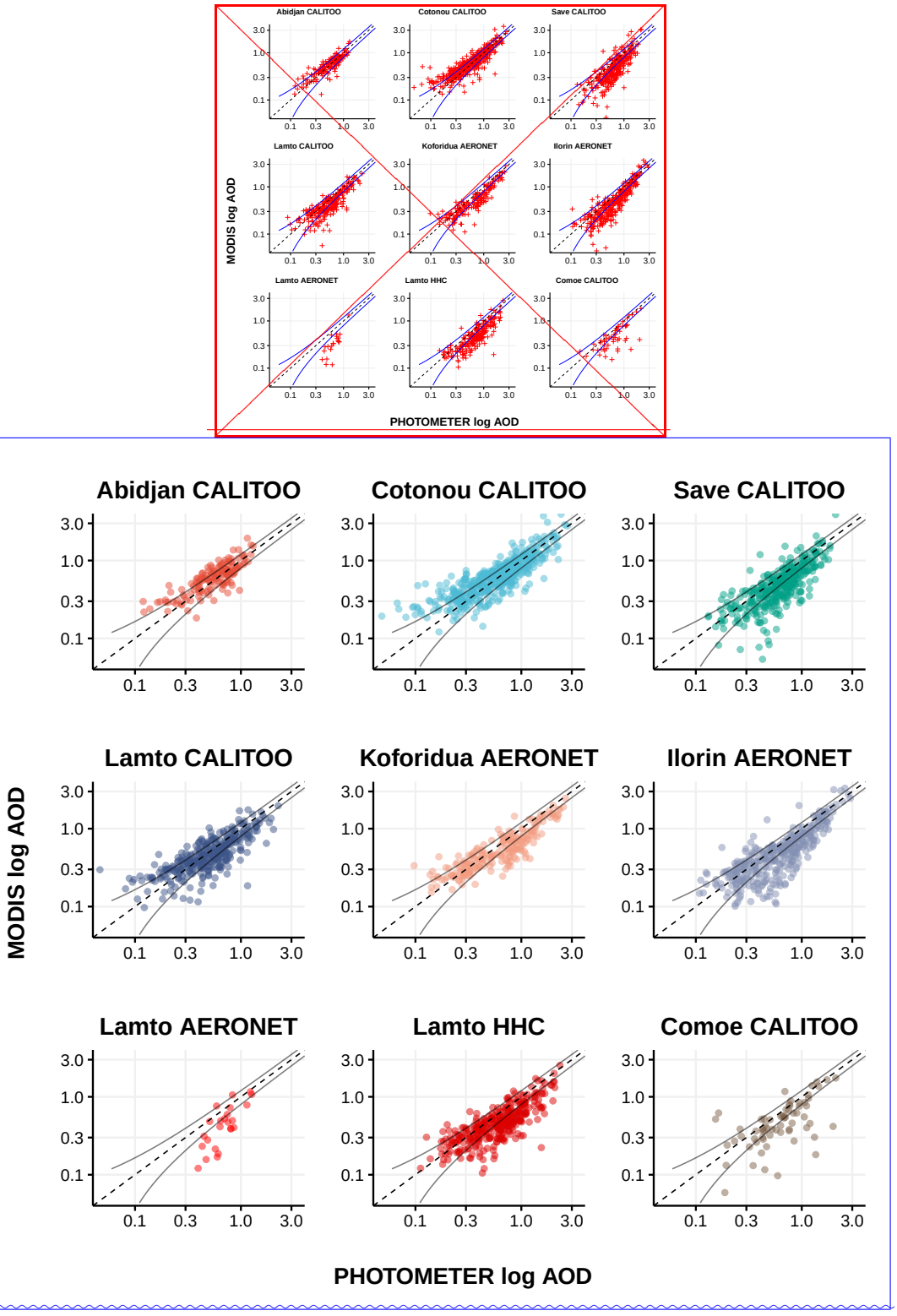

Figure 5. Scatterplots of MODIS versus sun photometer AOD for the 3 types of sun photometers (automatic AERONET, and handheld CALITOO and CIMEL) and different sites (Lamto, Comoé, Savè, Cotonou, Abidjan, Ilorin, Koforidua)

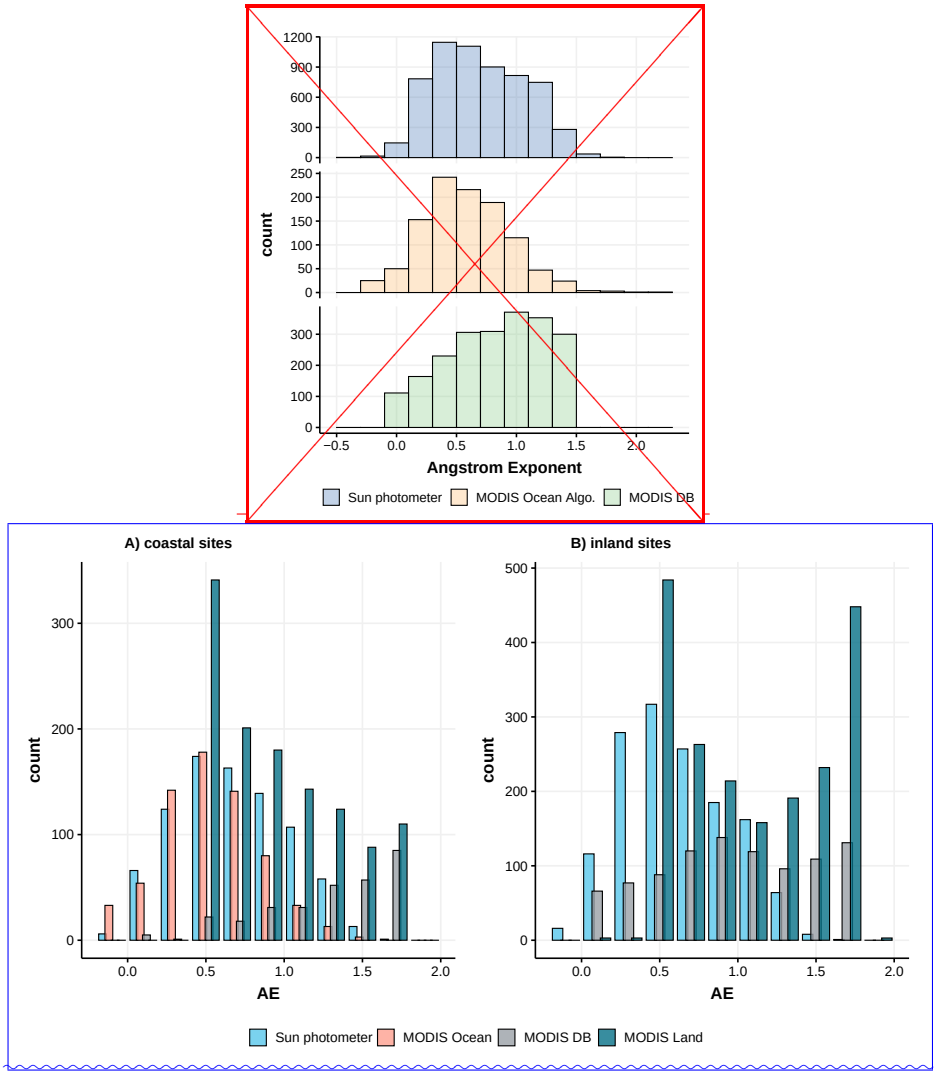

Figure 6. ~~Histogram~~ Comparative histograms of ~~Angstrom Exponents~~ Ångström exponents for ~~all~~ coincident sun photometer~~observations and~~ , MODIS ~~DB~~ Deep Blue, MODIS Land and Ocean algorithms, for (A) coastal sites and (B) inland sites.

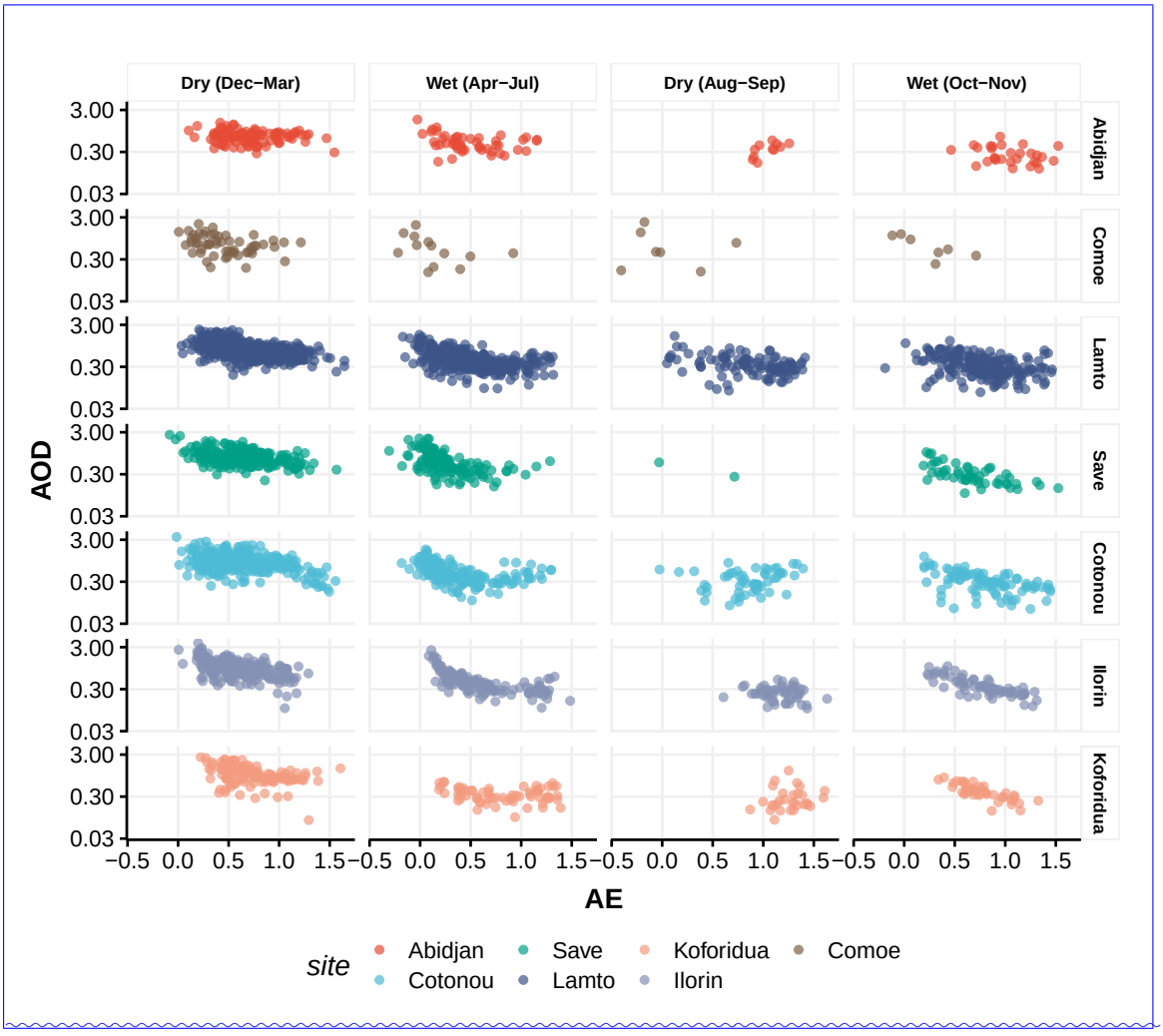

Figure 7. Scatter plots of sun photometer aerosol optical depth (AOD) versus Ångström exponent (AE) splitted by sites and by seasons.

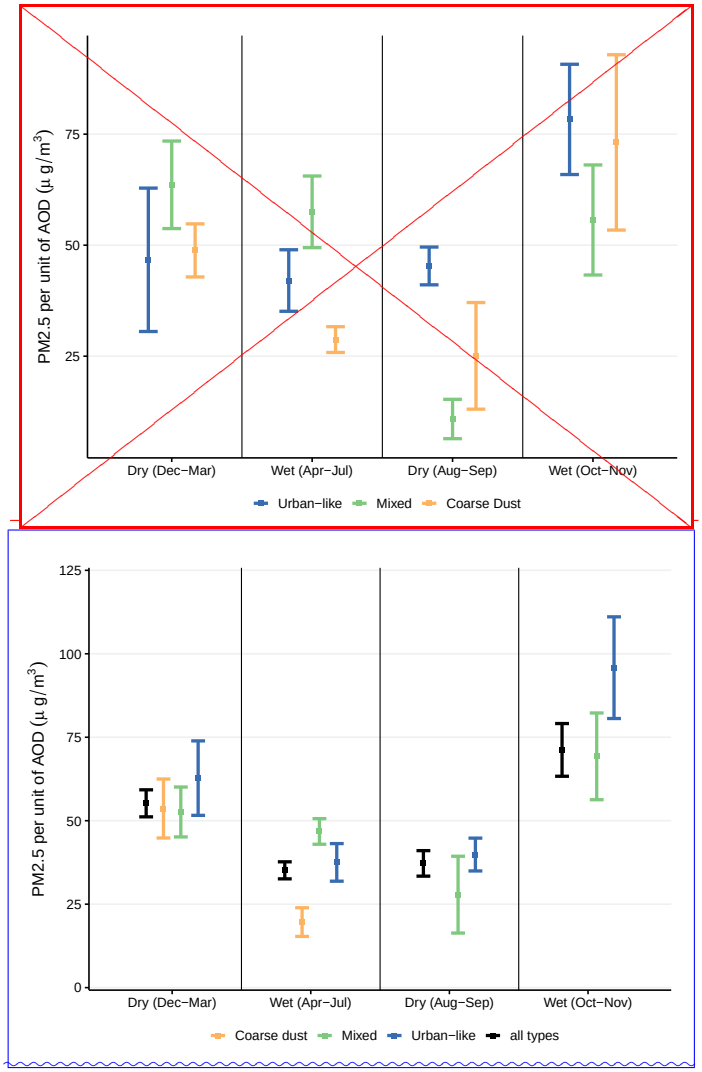

Figure 8. Ratio of PM2.5 to AOD for each of the aerosol types during the long dry (Dec.-Mar) and long wet (Apr.-Jun.) seasons and the short dry (Aug.-Sep.) and short wet (Oct.-Nov.) seasons. Data are collected in Abidjan and Cotonou cities from 2015 to 2017.

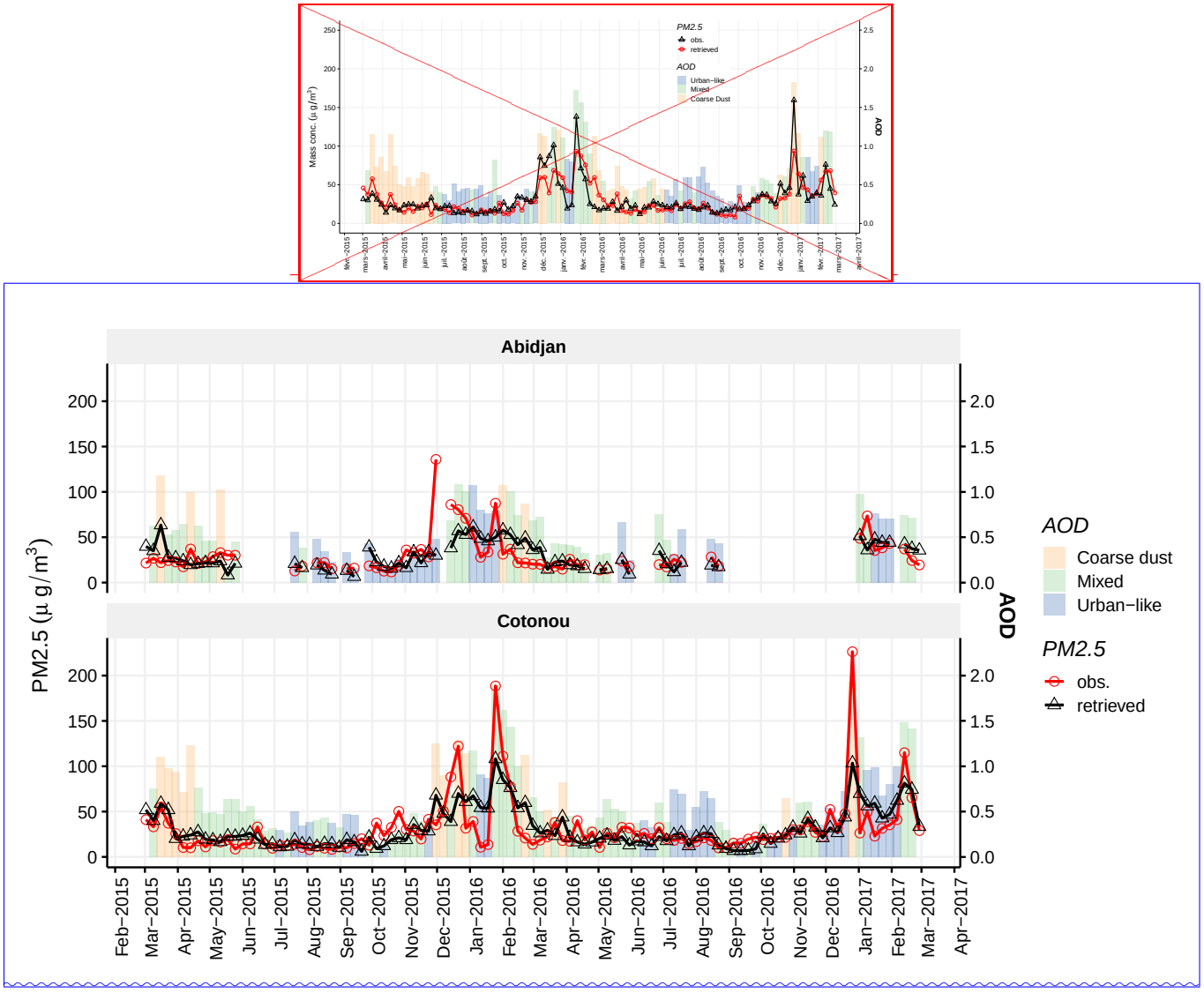

Figure 9. In situ and AOD-derived mean weekly PM2.5 from March 2015 to March 2017 in Abidjan and Cotonou. Vertical color bars give the ~~weakly~~ weekly AOD by aerosol category.

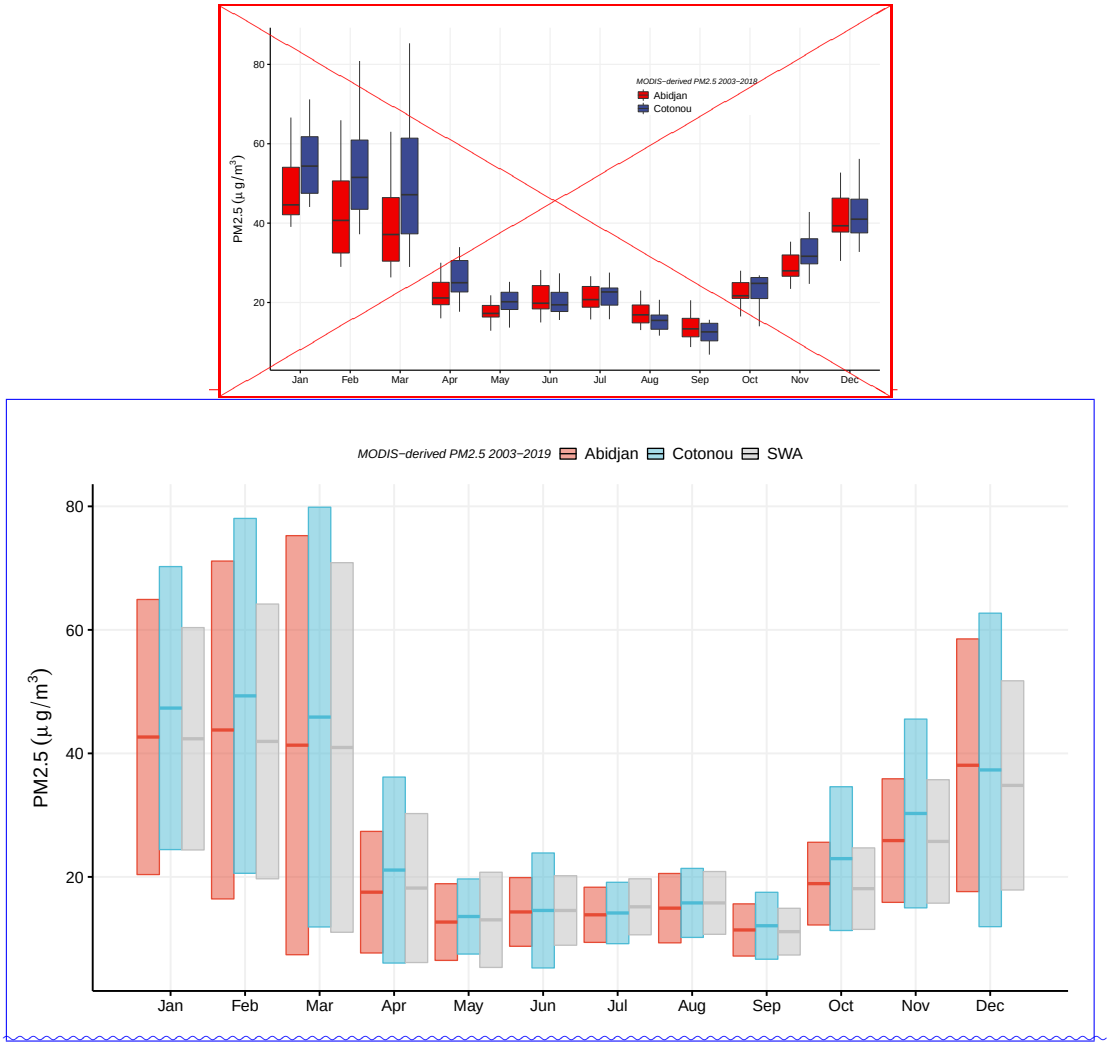

Figure 10. ~~Seasonal~~ Monthly mean annual cycle of ~~monthly average~~ MODIS-derived PM2.5 in Abidjan and Cotonou cities ~~from~~ and SWA area between 2003 ~~to 2018. Lower~~ and ~~upper hinges correspond to~~ 2019. Boxes represents the ~~first and third quartiles~~mean ± one standard deviation.~~Upper and lower whiskers extend to largest value (respectively lower) no further (respectively at most) 1.5*IQR.~~

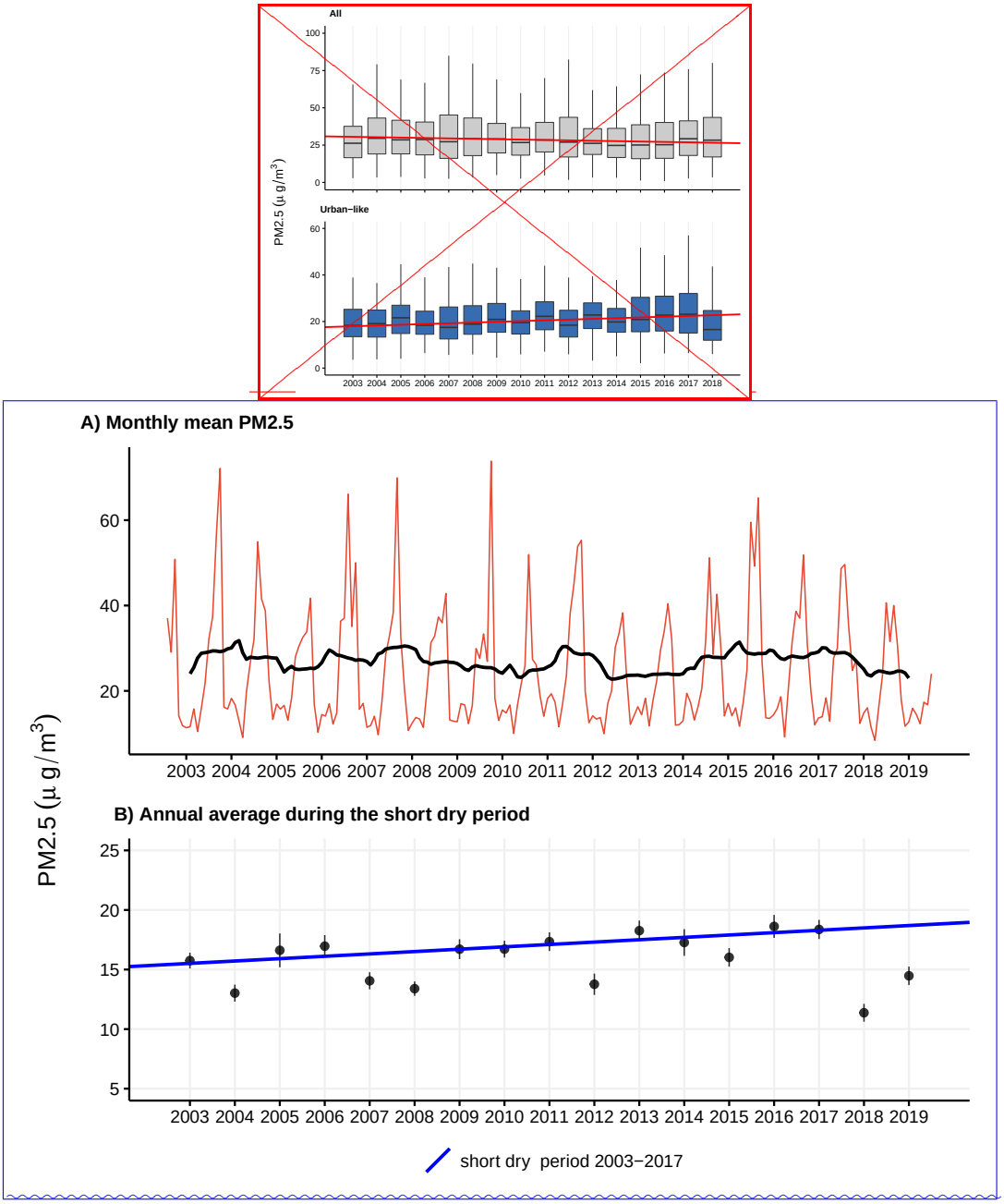

Figure 11. MODIS-derived PM2.5 ~~annual mean for Abidjan and Cotonou~~ from 2003 to ~~2018.~~ 2019. (~~top~~A) ~~all type of aerosols~~Monthly mean over SWA and seasonal adjusted trend, (~~bottom~~B) ~~urban-like type only~~annual average during the short dry period and monotic trend computed over 2003-2017.