ACP-973-2017

"Mass concentration, optical depth and carbon composition of particulate matter in the major Southern West Africa cities of Cotonou (Benin) and Abidjan (Côte d'Ivoire)."

By Djossou et al.

Dear Editor,

We are grateful to the reviewers for their positive reviews and for their help in improving the quality of this paper. We have answered all the comments and we have revised the paper in light of them. The structure of the paper has been changed, resulting in different figure and section numbers. Please find below our response to each reviewer's comment and the changes we have done in the manuscript following their suggestions.

Yours sincerely.

Answers to reviewers

Reviewer #1

Black : review
Blue : answer
Red : modification of the paper

This work reports on in situ measurements of two locations in west Africa that include daily integrated filter measurements of PM and a daily measurement of AOD. This region is very much understudied, and these are an important set of measurements. However, there are a number of significant limitations to this work, addressed below, that need to be evaluated and considered for further consideration by this reviewer. These include concerns with extensive editorial revisions, a generally superficial analyses of a novel dataset, and a need for additional methodological information for this data to be interpretable. Though the authors should be commended for collecting a reasonable extensive primary dataset, the interpretation and utility of this data, as presented, is quite limited. It is my view that this would should be rejected, as the needed revisions are probably too extensive to warrant publication in ACP.

Major comments:

In a number of cases, the authors focus on the importance of dust contribution to haze in the region, which is a reasonable interpretation given proximity to Sahara, and the presence of regular Harmattan. Yet, the focus on chemical analysis is on EC and OC, typically a trivial fraction of crustal materials. While the available of EC and OC data can be useful for assessment of biomass burning, anthropogenic combustion, and the like, it does not make sense to use them to explain crustal affects on PM

Giving a comprehensive chemical closure of PM2.5 over the 2 years isn't the scope of the paper. The aim of the paper is to focus on the carbonaceous content of particulate matter in sub-Saharan major cities. We don't use the EC/OC to explain crustal effect on PM2.5. Throughout the paper, EC and OC are used to characterize the combustion sources. However, when giving an interpretation to the timeseries it has to be mentioned that mineral dust has a significant impact on PM2.5.

Onlines 90-92, the authors seem to conclude that PM anthropogenic emissions from primarily from '2 wheeled vehicles' whereas 'cars and buses' dominate emission in Abidjan. Is there any evidence to support this statement?

Traffic emissions are dominated by '2 wheeled vehicles' in Cotonou and 'cars and buses' in Abidjan. See Assamoi and Liousse (2010) and Fanou et al. (2006). This has been corrected in the text.

The Dynamic Aerosol-Cloud-Chemistry Interaction in West Africa (DACCIWA) research program has been started in 2014 (Knippertz et al. 2015). One of the aims of the program is to characterize the health impact of atmospheric pollution on SWA populations. An enhanced observation period (EOP) for assessing PM2.5 mass concentration and particulate carbon species has been conducted from February 2015 to March 2017 in two coastal cities representative of SWA conurbations: Abidjan in Côte d'Ivoire and Cotonou in Bénin. Three sites have been chosen in Abidjan with a focus on specific sources: domestic fires due to smoking activities, traffic at a crossroad and waste burning at the landfill. The traffic in Abidjan is dominated by car and buses while in Cotonou it is dominated by 2-

wheels vehicles (Ayi Fanou et al., 2006; Assamoi and Liousse, 2019; Mama et al., 2013). For this reason, an additional traffic site has been selected in Cotonou.

Please describe the sampling approach you took to collect PM2.5.

We have improved the description of the sampling approach. See also remark by reviewer #2. The following text has been added in the Method section.

The particles were collected on 47mm diameter on a weekly basis. Two types of filters are used depending of analysis performed. PTFE filters were used for gravimetric measurements while quartz fiber filters were used for carbonaceous aerosol analysis. The use of different types of filter requires the setup of two filtration lines operating in parallel. The sampling system uses 2 mini Partisol sampling impactors for PM2.5 working at a flow of 5L/min. Both lines are stored in a same box standing outside at ambient temperature and humidity. Figure 2 provide a quicklook of the inside of the sampling box, which is equipped for each line with a KNF pump with a flow rate of 9 l/min (N89 KNE-K version 220v), a Cole Palmer ball flow meter with a micrometric valve (flow range adjustable from 0 to 10 l/ min, accuracy 5%), a GALLUS type G4 gas meter (accuracy of 0.01 m3) giving the total volume of air sampled during the week, tubing, and a NILU online filter holder (see also Ouafo et al., 2017).

The concentrations of PM2.5 particles and carbonaceous aerosols (OC and EC) have been measured on a weekly time step by the ambient air pumping technique over a two-year period (February 2015 to March 2017) for the 4 sites cited above. The air is sampled for 15-min every hour, leading to a total volume of sampled air of about 12.6 m_3 by week. Due to power failure, some weeks were not sampled. The samples are stored in packs before sampling and then individually in Petri dishes and covered with aluminium fold once the sample has been collected. They are then return to Toulouse, France for analysis at the Laboratoire d'Aérologie.

Where appropriate size selection devices (cyclones, impactors) used?

Yes. We used a mini Partisol PM2.5 sampling impactor. The description of the sampling system can be found in Ouafo et al. (2017). See your previous remark.

Given the semicontinuous monitoring scheme, what was used to assess flow rate to ensure accurate size selection and total volume sampled?

Sampling lines are equipped with a Cole Palmer ball flow meter with a micrometric valve (flow range adjustable from 0 to 10l/min and a Gallus G4 gas meter measuring the total volume of air sampled. The flow rate was checked each time the filters were changed by the operator. The sampling frequency (weekly) was set up accordingly to avoid pressure loss due to filter clogging.

At the AWB site near Abidjan, the samples were collected at 12 meters (line 140). Is this accurate? How can we be sure ground-based emissions from the waste transfer industry were captured by a sampler located so high off the ground? And how frequently was this burning conducted?

The plume rising from the waste burning site is clearly above 12 m and the building is located on the other side the road in front of the dump. However we can't claim that we are ideally located to sample the emission from the waste burning site, mostly because of local winds that blows the waste burning plume on and off. This is clearly stated in the text. We don't have exact information on the burnt frequency in the dump. Spontaneous ignition leading to smoldering conditions occurs during the dry season. Dump workers use to burn waste for collecting material.

In the methods section (line 171), the authors report EC, TC, and OC detection limits that includes an uncertainty. It is unusual to report uncertainty with imputed detection limits, which are typically defined as three times the standard deviation of a blank, or some other standardized approach. In either case, more information towards how these MDLs were calculated is necessary.

Correct. Thank you for your comment. This statement was confusing. We now report the detection limit of the DRI analyzer based on the instrument blank, which are 0.4, 0.1, 0.3 µg/cm2 for TC, EC and OC. The accuracy estimated from our measurements is 5% for TC and 10 % for EC and OC.

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In Lines 197-200, the authors seems to concluded that AOD is different between the two locations based chiefly on population size differences between the two. This statement needs to be supported with evidence from either the literature, or the data that was collected.

We agree that this statement is awkward. Indeed, the relationship between AOD and the size of the city or the number of inhabitants is not clearly expected. This sentence has been suppressed from the text. We now rely only on the data presented in the paper.

More information is needed on sampler placement at each location (pictures in the figures, though aesthetically interesting, are not particularly helpful). Further, presuming these samplers were stationary, it would seem to make sense if the authors would conduct some sector analyses where known wind speed/direction is used to censor data to show how concentrations change when the sampling is likely downwind of the source, or when it is not. The authors have lumped all data together, and any useful signal is likely to be lost to averaging or noise.

Following your recommendation and the ones given by reviewer #2, we now provide a map reporting the position of the sampling sites (see new Figure 2). We also now provide the time series of wind components. However it has to be recalled that we sampled the air 15min/hour on a weekly basis. Moreover, the sampling was performed in the vicinity of the sources at least for AT, CT and ADF (eg. traffic cross-road or smoking place) so the site are always affected by the source. As stated in the text, each station was not equipped with a met. station and we use the NOAA ISD synoptic data available at the airport of each city.

We keep the pictures because we think they help the reader to have a better idea of the sampling environment.

The description of the sites has been modified as follows:

The CT site in Cotonou is located in Dantokpa area, one of the biggest market in western Africa. The instrument is located on a balcony at 4 m height above a major crossroad. As shown in Figure 3a the traffic at the crossroad is largely dominated by 2-wheel vehicles including "zemidjian" moto-taxi.

In Abidjan, we set up the instrumentation in 3 sites representative of traffic (hereinafter called AT), waste burning at landfill (AL) and domestic fires (ADF) emissions. ADF is located in the market of Yopougon-Lubafrique (5° 19.746' N; 4° 6.353' W) in a large courtyard with about 25 fireplaces (Figure 3-d). Fireplaces are active from 6 a.m to 3 p.m. Highest activity is between 6 and 10 a.m. The smoke is due to smoking meat and fish or grilling of peanuts. The instrumentation is setup on a 3-m height tower. The fuel used is essentially hevea wood stored outside in the vicinity of the fireplaces. AT is located at Adjamé bus station (5° 21.252' N; 4 ° 1.095' W) and more precisely on the roof of the pharmacy "220 logements" . Adjamé bus station is one of the major traffic areas for small buses called "baka" in Abidjan and so this area is largely influenced by 4-wheel vehicles emissions ((Fig. 3.b). The traffic is dominated by old vehicles (4 wheels) using diesel. AL is near the public landfill of Akouédo (5 ° 21.215' N; 3° 56.277' W) on the flagstone roof a 3 story building at about 12 m above ground. The public dump receives the totality of waste produced in the district of Abidjan since 50 years, currently more than 1.000.000 tons of waste by year (Adjiri et al, 2015;) It negatively affects the environment and the living environment of the populations of Abidjan in general (UNEP, 2015). Dump workers burn waste mainly during the dry season. Spontaneous ignition also occurs in the landfill during the dry season. Fig. 3.d shows a combustion plume rising from the dump.

Your remark about lumping the data is unclear to us. We now present the data in a different way to better highlight the specific behaviour of each site. see Figure 5 below.

Fig. 3. Pictures showing the immediate environment of the stations (a) 2-wheels traffic in Cotonou (CT station), (b) small buses traffic in Abidjan (AT station), (c) waste burning at the Abidjan landfill (AL station) (d) smoking activities in Abidjan-Yopougon (ADF station).
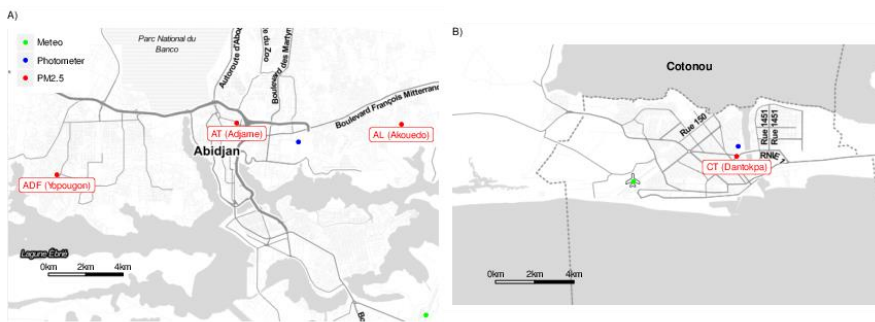
Figure 2: Maps of the cities of (A) Abidjan and (B) Cotonou reporting the location of the (red) PM2.5 sampling sites, (blue) photometer measurements and (green) meteorological station.

Figure 5: Time series of the (left) PM2.5 and (right) carbonaceous weekly concentrations at the 4 sites from February 2015 to March 2017. Shaded areas show the different seasons (see text): the long rainy seasons W1 and W2; the short dry seasons D1' and D2'; the short rainy seasons W1' and W2'; and the long dry seasons D1 and D2.

Did the authors make any assessment (modelling or otherwise) of high altitude aerosols which would affect AOD, but not ground-level PM?

The answer is no. Assessing the apportionment between local produced particulate pollution and aloft one would require a dedicated study. It is the purpose of another paper dealing with assessment of PM2.5 ground-level concentration from satellite AOD.

The percentage of OC and EC to PM2.5 contribution (Lines 299) are unusually high. For example, at the ADF site, 11% of PM was from EC, and 51% was from OC. Inorder to be more meaningful, OC really should be converted to OM (typically 1.5*OC = OM, though it does vary). If one makes this assumption, then more than ~90% of PM2.5 is explained by OM and EC, leaving very little for crustal, metal, or ionic contribution to PM2.5.

ADF is located in the heart of a smoking place so having a major contribution of carbonaceous species in PM2.5 is not surprising.
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In Lines 311-359, the authors make comparisons to other sites found around the world (Paris, Milan, Morocco, Agra, Querol, etc). What is the point of this comparison? How does this support your findings?

The purpose of the discussion section is to put our results in a more general context. Air quality in urban centers is obviously not a issue specific to SWA. However this is the first time that such a field experiment has been conducted there. Our findings are consistent with satellite estimation (Van Donkelaar et al., 2010) and in the range of other similar studies worldwide. Following

recommendation from reviewer #2, we have organized this discussion in a different way so we explain better explain why we report measurements from other studies. Please refer to the main text.

The conclusions in this work are largely unfocused, and are overly generic. The authors, quite unexpectedly, include new arguments (Line 391) to suggest that 'humidification of wood fuel' is driving the relationship at one location. There is only a limited analysis of AOD measurements, which could be an important and useful lower-cost measurement of PM25 in this locations. But as presented, one only sees a time series of AOD. It would be far more useful to include back trajectory analyses or receptor models to support your data.

We have improved and shorten the conclusion as a bullet list to better highlight our findings. The statement regarding 'humidification of wood fuel' has already been presented in main text at line 253. But you are right it has to be remove from the conclusion. We have clarified this point. The analysis of PM2.5 /AOD has been improved by adding new figures as recommended by reviewer #2. However the analysis of PM2.5 and AOD at the regional scale along with satellite data is the subject of another paper. We don't provide a full receptor model analysis but now better discussed possible regional scale transport based on wind time series.

This study reports new and unique observations of weekly PM2.5 mass concentration and particulate carbon species in the vicinity of major sources of combustion aerosols, ie. traffic, burning of waste at landfill, and smoking activities in coastal cities of SWA. Traffic emissions were investigated in 2 different environments, one is dominated by 2-wheel vehicles (for Cotonou) and the other one by 4-wheels (Abidjan). Additionally, the AOD was also measured for the first time in Abidjan and Cotonou on a daily basis. The period of observations spans from February 2015 to March 2017. Our findings can be summarized as follows:

- The mean PM2.5 concentration for the urban sites is about 30 $\mu g.m^{-3}$ is coherent with previous studies for sub-Saharan western Africa and is 3 times higher than the concentrations recommended by the World Health Organization.
- We observe large similarities in the seasonal cycle of PM2.5 and AOD between both urban sites with an overall increase in concentration in AOD and PM2.5 during the major dry season. During

this period AOD and PM2.5 are well correlated suggesting that most of the particles are located in the lower part of the atmosphere.

- The spikes in PM2.5 weekly time series can be associated to the contribution of dust transport or biomass burning activities as reveals by the analysis of the EC/OC ratio, Angstrom exponent and MODIS burnt area. Those spikes are observed during the major dry season, while the minor dry season shows low PM2.5 concentrations possibly due to no biomass burning and the enhancement of the atmospheric dispersion in relation with the increase in the wind intensity.

- The mean OC/EC ratio is on average 4 in Cotonou and 2 in Abidjan, clearly indicating the larger contribution emission by the 2-wheel motorcycles in Cotonou compared to Abidjan, mostly dominated by diesel Vehicle Park.

- The observations of domestic fire emission at the open air smoking courtyard in Abidjan shows an average weekly PM2.5 concentration of 145 $\mu g.m^{-3}$, indicating that open air smoking activities could be a large contributor to air pollution. We observe there a seasonal cycle different from the one of the urban sites.

This 2 year-long field campaign focused on combustion aerosol sources in the emergent cities of coastal SWA provides a first and unique data set for a better understanding of the impact of such pollutants on health and environment in this part of the world.

Minor comments: Some of the information in the introduction (e.g. lines 60-65) is too elementary.

Since this paper deals with carbonaceous aerosols, we believe it is important to remind some basic definition about OC et EC and would like to keep this small paragraph in the introduction.

Missing references for recent campaigns listed in lines 77-78 and for the IMPROVE protocol used (line 161)

Line 77-78 have rewritten (see remark by reviewer #2) and we have added references for the IMPROVE protocol.

Carbonaceous aerosols in West Africa are known to result from biomass burning, traffic and domestic fire emissions and to a lesser extent from other combustion sources such as industries, power plants and flaring (Doumbia et al., 2012; Liousse et al., 1996, 2014; and Liousse and Galy-Lacaux, 2010). According to Liousse et al., (1996, 2010), the emissions from biomass burning have an importance

on the atmospheric composition in rural area of West Africa. Atmospheric aerosol in West African rural areas and typical inter-tropical ecosystems has been the subject of research programs such as DECAFE (Cachier et al., 1995 ; Lacaux et al., 1995), IDAF (Galy-Lacaux et al., 2009), EXPRESSO (Delmas et al. 1999) (1996), DECAFE program allowed to characterize the chemical composition of aerosols during savanna fires in West Africa (Côte d'Ivoire). However, very little information exists on aerosols in West African cities.

In cities of sub-Saharan Africa such as Cotonou (Bénin), the emissions from 2-wheeled vehicles running on mixtures of smuggled gasoline from Nigeria and motor oil are a large source of pollutants (Liousse et al., 2014). The atmosphere in the city of Cotonou is characterized by thick opaque fumes, especially at the level of major arteries and intersections (Mama et al., 2013). This phenomenon is linked to the use of two-wheeled motorcycles, especially motorcycle taxi-moto called "Zemidjan", whose number in 2005 was estimated at 96.095 according to the statistics of Cotonou City Council (Ayi Fanou et al., 2006; Mama et al., 2013). In Cotonou motorbike taxis account for 90% of the cases with intoxication symptoms 1,5 time higher than in the non-drivers of these motorbikes. The symptoms recorded on statement are intoxication disorders such as conjunctival hyperemia (18%) among which 12% of lacrimation, respiratory disorders (23%) (Fourn and Fayomi, 2006). According to Fourn and Fayomi, (2006) and Mama et al. (2013), motorcycle taxi emissions are the highest at the Dantokpa area.

The results obtained by Doumbia et al., (2012), confirm that in West and Sub-Saharan African big cities there is a "hot spot" of emissions, particularly due to the emissions from vehicle engines, and domestic fires. Nevertheless, pollution in urban African areas and their related health impacts have been poorly studied. Therefore, observations for fine particle (PM2.5) and particulate carbon species are needed for African cities.

(...)

Carbonaceous aerosols organic carbon (OC), elemental carbon (EC) and total carbon (TC, calculated by the sum of OC and EC) were measured on a 0.55 cm² punch from each quartz filter by thermo/optical reflectance following the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol (Chiappini et al., 2013; Ouafo-Leumbe et al., 2017).

> **Commenté [D2]:** Ligne 77-78 RC2 citation

Were the filters (line 120) used actually Teflon filters (which are trade names of a specific brand) or more generic PFTE filters?

The filters are generic PTFE. The sentence has been changed.

PTFE filters were used for gravimetric measurements while quartz fiber filters were used for carbonaceous aerosol analysis.

Unfortunately, the paper needs to be deeply edited by a native-english speaker. There are many instances of typos, unfinished sentences, grammatical errors, and improper punctuation; these are

probably too numerous to completely list here. Identified errors include lines: 30 31 45 47 87 106 108 142 200 201 223 310 317 318 368 372 384 388.

Thank you for your time and review. We did our best to correct the typos and improve the writing.