Interactive comment on "Personal exposure to $PM_{2.5}$ emitted from typical anthropogenic sources in Southern West Africa (SWA): Chemical characteristics and associated health risks" by Hongmei Xu et al.

Anonymous Referee #2

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We would like to thank the reviewer for all the suggestions and comments. Below we have responded to each comment in point to point format.

Megacities in Africa are pollution hotspots, for which very little data have been published. Personal exposure in such environments have also received virtually no research attention. Therefore, the authors are commended for this work.

General comments: I regard the language in this paper as poor. In many instances, this prevents proper knowledge transfer to the reader and can cause misunderstanding/interpretation of the text. Additionally, it makes the paper difficult/cumbersome to read. It is not the job of the reviewer/editor to do language and/or text editing. In my opinion, this paper should not have been published in ACPD before the language and text was acceptable. Therefore, I recommend that final review of this paper should only be considered once the language/text is improved. In the current form, too many uncertainties exist in the paper, because of the poor language.

Response: We truly altered this issue. The revised manuscript will be proofread by a native speaker before re-submission.

I am also not 100% convinced that the content of this paper fits into the scope of ACP. According to the journal, ACP ". . . is an international scientific journal dedicated to the publication and public discussion of high-quality studies investigating the Earth's atmosphere and the underlying chemical and physical processes." My uncertainty arises from the fact that this paper focused more on personal exposure and not on ". . . underlying chemical and physical processes." Would the paper not fit better into a journal specifically considering atmospheric exposure and/or health impacts? I leave the decision to the editor. This comment should not be considered as negative in any way and it is also not a reflection of the science presented.

Response: We understood the reviewer's concern and respected the decision from the editor. We would like to explain that our work is absolutely related to the $PM_{2.5}$ chemical composition, emission sources and variability. This topic should be within the scope of ACP. Moreover, there are some related works on air quality conducted in sWA (same work package on Air pollution and Health) published in the ACP/AMT DACCIWA special issue, forming a coherent whole.

Specific comments: The authors must please not use the name “South West Africa” as they did in line 105, but rather keep to the term “southern West Africa”, as it the rest of the paper. “South West Africa” was the name for modern-day Namibia from 1915 to 1990. I would even go so far as to recommend that the term “southern West Africa” that is abbreviated at “sWA” (“southern” in lower case) be consistently used, instead of “Southern West Africa” that is abbreviated at “SWA” (“Southern” in upper case), to ensure that the reader does not confuse the area investigated with “South West Africa” that was abbreviated at “SWA”.

Response: We totally agreed. The abbreviation for southern West Africa (sWA) has been revised throughout the document.

The authors refer to “. . . garbage spontaneous combustion. . .” a couple of times. Is the garbage really combusting spontaneously, or are garbage dumps being set alight to reduce the volume of waste, to reduce pests (rats and mice) and prevent disease?

Response: Akouédo dump in Abidjan is a vast and old landfill. We used the term of “spontaneous combustion” as we observed several smoke plumes in the middle of the dump, far from the working area. Spontaneous combustion is a well-known phenomenon in such outdated landfill, but there was difficulty in counting its frequency of occurrence compared to control ignition. Landfill workers often burn waste when they collect trashes and recycle some of useful items. The “controlled burnt” occurs in the active part of the dump. Both processes have a high occurrence during daytime and in the dry and hot season. Here we referred waste burning in a more general manner rather than specifying spontaneous combustion.

The quality of the Google Earth images presented in Figure 1 and the photos presented in Figure 2 are not good and might deteriorate further in page setting during publication (e.g. if the images are printed even smaller). I encourage the authors to ensure the best possible quality for these images.

Response: We would definitely pay more attention to the pixel issues on the figures in the revised manuscript.

In its current form, the paper is long. If the authors and editor agree, I would suggest that Appendixes A, B, C and D, which present the questionnaires, rather be included as supplementary material, instead of appendixes. Appendixes are published as part of the paper, while supplementary material are published separately. Readers who want to assess the content of the questionnaires can download the supplementary material, instead of the paper becoming excessively long.

Response: We agreed with the reviewer’s comment. We will move the current

Appendix A-D to the supplementary material as supporting information.

I agree with Referee #1 that the authors cannot interpolate their results obtained from individuals with specific occupations and at specific locations to the wider southern West African region. All such statements should be revised.

Response: Thank you for this point. We have made the necessary changes within the text.

In general, there is little comparison of the results presented in this paper to results obtained elsewhere. I appreciate that very little, if any, personal exposure data have been presented for African cities. However, even if the results presented are compared to ambient/indoor air quality results obtained in the rest of Africa (or Asia, or some other developing settings, if African reference cannot be found), the reader will be able to easier contextualize the exposure concentrations reported here. For instance, indoor air quality in semi- and informal settlements (low-income households) in South Africa (Kapwata et al., Atmosphere 2018, 9(4), 124; https://doi.org/10.3390/atmos9040124) could be compared to "Night" personal exposure of individual in this study. Also, characterization of the plume of fire grilling of meat in an African context (Venter et al., S. Afr. J. Chem., 2015, 68, 181–194; DOI: http://dx.doi.org/10.17159/0379-4350/2015/v68a25) could possibly be compared to the exposure of woman by Domestic Fires (DF) ("grilling meat or roasting peanuts") in this study. Such comparisons will help the reader to contextualize the results presented – currently only comparing the different groups with one another does not enable the reader to contextualize the results. There might be many more references, such as the afore-mentioned, these two are just examples that I found with a quick online search.

Response: We understood this point of view. The citations on our work are related to either ambient concentrations or source characterization. The suggested references should not be applicable. Venter et al. (2015) presented a study on charcoal combustion with a specific type of barbecue, totally different from the conditions at DF (wood in barrel). Kapwata et al. (2018) showed the work on PM_4 (dissimilar with our $PM_{2.5}$) and no chemical composition was provided.

In lines 370 and 384 of our original manuscript, we firstly compared the average PE $PM_{2.5}$ levels to the weekly ambient $PM_{2.5}$ concentrations obtained in the same area and similar sampling period, and also compared the daytime PE $PM_{2.5}$ mass concentrations with the daytime ambient $PM_{2.5}$ in the same area and exactly the same sampling dates (Djossou et al., 2018). We also used the results of the PAHs exposure measured in Cotonou in a previous study (Fanou et al., 2006). We provide additional references of previous works on personal exposure to $PM_{2.5}$ in household in Tanzania (Titcombe and Simcik, 2015) and for students in Ghana (Arku et al., 2014). And we compared the results with our data in this study as follows:

"The 5-h $PM_{2.5}$ average personal exposure concentration was 1574 μg m^{-3} (±287, n = 3) for open wood fires in households in the Njombe district of Tanzania (Titcombe and Simcik, 2015), and was comparable to the highest 12-h exposure level to $PM_{2.5}$ for women at DF site in this study (1164.7 μg m^{-3}, daytime in wet season, July 5th), and was 4.7 times of the daily average PE $PM_{2.5}$ concentration in dry and wet seasons (331.7±190.7 μg m^{-3})."

"In the study of Titcombe and Simcik (2015), we also found that the 5-h average total PAH personal exposure concentration was 5040 ng m^{-3} (±909, n = 3) for open wood fires in households in the Njombe district of Tanzania, which was much higher (~65 times) than the women exposure PAHs at DF site in the current research. The highest 12-h exposure PAHs for women at DF site in this study was 469.7 ng m^{-3} (daytime in wet season, July 6th), approximately one-tenth of the PAHs concentration from open wood fires in Tanzania mentioned above. The large PE PAH concentrations difference between these two studies may be influenced by many factors such as wood type, combustion state, stove structure and sampling time."

"Student (10-17 years old) $PM_{2.5}$ exposures ranged from less than 10 μg m^{-3} to more than 150 μg m^{-3} (mean 56 μg m^{-3}) in four neighborhoods in Accra, Ghana (Arku et al., 2014), much lower than that for students at WB site (356.9±71.9 μg m^{-3}). It can be seen that the high exposure of students in this study is likely to be related to the waste burning emissions, while there was no obvious strong $PM_{2.5}$ emission source in the study of Arku et al. (2014)."

We have also added the comparisons as mentioned in the revised manuscript.

Reference:

Arku, R. E., Dionisio, K. L., Hughes, A. F., Vallarino, J., Spengler, J. D., Castro, M. C., Agyei-Mensah, S., and Ezzati, M.: Personal particulate matter exposures and locations of students in four neighborhoods in Accra, Ghana. J. Expo. Sci. Environ. Epidemiol., 1–10, 2014.

Titcombe, M. E., and Simcik, M.: Personal and indoor exposure to $PM_{2.5}$ and polycyclic aromatic hydrocarbons in the southern highlands of Tanzania: a pilot-scale study. Environ. Monit. Assess., 180, 461–476, 2011.

Line 415. The author state that "The previous studies (Cao et al., 2008; Li et al., 2009; Tian et al., 2017) suggested that average OC/EC characterizes 1.1 as motor vehicle exhaust, 2.7 as coal combustion and 9.0 as biomass burning. The OC/EC in the present study points out that biomass burning emission was the main contributor to carbonaceous aerosols for women at DF, and the mixed emissions from biomass and coal burning, even or/and motor vehicle exhaust dominated the carbonaceous aerosol sources for students at WB and drivers at MT." However, the authors should clarify these statements, since OC/EC ratio will change in a plume with aging, with the formation of secondary OC and deposition of EC. Therefore, if the above OC/EC ratios

are used to characterize fresh emissions/plumes, it should be stated as such and not left to the reader to interpret.

Response: The statements have been revised as follows:

> "The previous studies (Cachier et al., 1989; Cao et al., 2005a; Cao et al., 2008; Li et al., 2009; Tian et al., 2017; Watson et al., 2001) suggested that average OC/EC characterizes 1.1 as motor vehicle exhaust, 2.7 as coal combustion and 9.0 as biomass burning for source samples (fresh emissions/plumes)".

The reason why we could compare the OC/EC results of the personal exposure data in this study with the above source samples is because that the participants were close or around to the typical anthropogenic sources in this study. In addition, $PM_{2.5}$ emitted from the pollution sources still maintained a relatively fresh state (less aging) without long-distance transport, and then was inhaled into human body. Therefore, the OC/EC ratio comparison results in this study could yield reliable conclusions as described in the texts (originally lines 418-421).

Reference:

Cachier, H., Bremond, M. P., and Buat-Menard, P.: Carbonaceous aerosols from different tropical biomass burning sources. Nature, 340, 371–373, 1989.

Cao, J. J., Wu, F., Chow, J. C., Lee, S. C., Li, Y., Chen, S. W., An, Z. S., Fung, K. K., Watson, J. G., Zhu, C. S., and Liu, S. X.: Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China. Atmos. Chem. Phys., 5, 3127–3137, 2005a.

Watson, J. G., Chow, J. C., and Houck, J. E.: $PM_{2.5}$ chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in northwestern Colorado during 1995. Chemosphere, 43, 1141–1151, 2001.

Line 247. Fe and the heave metals reported (V, Cr, Mn, Co, Ni, Cu, Zn, Sb, Ba and Pb) were analyzed with ED-XRF. How sure are the authors that some of the heavy metals were not part of the GM and are therefore partially double accounted for in the mass balance (Figure 5, line 493 onwards), i.e. accounted as heavy metal mass and also contributing to the mass of the GM?

Response: The abundance of Fe in the earth's crust is about 4% (discussed in next comment detailly), and the abundances of the other nine elements (i.e., V, Cr, Mn, Co, Ni, Cu, Zn, Sb, Ba and Pb) in the earth's crust ranged from 0.2×10^{-6} (Sb) to 660×10^{-6} (Mn), which are 2-5 orders of magnitude lower than Fe (Taylor and McLennan, 1985). From the above crustal content comparison, the authors had reason to believe that the overlap contributions to $PM_{2.5}$ from the other nine elements and the estimated crustal material content (by using Fe) can be neglected.

Line 511. Although the authors give a citation (i.e. "Taylor and McLennan, 1985") to support the use of Fe as a tracer for geological material (GM), how accurate is this

method? The authors state "Fe constitutes about 4.0% of the Earth's crust in dust of the earth's crust (Cao et al., 2005)". Are there any indications of Fe contents of local soils (and the variation on Fe contents) and how it differs from the global average value of 4%, which was used? Basically, I am asking how accurate the method is. Can the authors give any indication of accuracy? This is important, since ". . . it is found that GM contributed 35.8%±2.1%, 46.0%±3.7% and 42.4%±4.7% of PE $PM_{2.5}$ mass concentrations for women at DF, students at WB and drivers at MT, respectively."

Response: Fe is the most important metal and one of the major constituents of the lithosphere. Its average content of the Earth's crust is about 5%. The global abundance of Fe is around 4.5% (Kabata-Pendias and Mukherjee, 2007). The typical range of Fe contents in soils is between 0.1 and 10% and its distribution in soil is variable, which is controlled by several soil parameters (Kabata-Pendias and Mukherjee, 2007). The authoritative study from Taylor and McLennan (1985) of elemental content in soils showed that the global abundance of Fe is around 3.5%.

Because of the lack of elemental composition obtained in topsoil of Africa, the authors used 4.0% (global mean Fe content from the mentioned literatures) as a percentage of Fe in the topsoil of African study area for geological material estimation in this study. The value was widely used in other literatures (Cao et al., 2005b; Hao et al., 2007; Sun et al., 2014; Wu et al., 2012; Xu et al., 2016b). In our opinion, this method can roughly indicate the contribution of the geological material to atmospheric particle matters to a certain extent. Moreover, since the geological material of three sampling sites in this study were estimated using a consistent method, the relative results were comparable.

Reference:

Hao, Y. C., Guo, Z. G., Yang, Z. S., Fang, M., and Feng, J. L.: Seasonal variations and sources of various elements in the atmospheric aerosols in Qingdao, China, Atmos. Res., 85, 27-37, 2007.

Kabata-Pendias, A., and Mukherjee, A. B.: Trace Elements from Soil to Human. Springer-Verlag, Berlin Heidelberg, Germany, pp. 381-393, 2007.

Sun, Y. Y., Hu, X., Wu, J. C., Lian, H. Z., and Chen, Y. J.: Fractionation and health risks of atmospheric particle-bound As and heavy metals in summer and winter, Sci. Total Environ., 493, 487-494, 2014.

Wu, F., Zhang, D. Z., Cao, J. J., Xu, H. M., and An, Z. S.: Soil-derived sulfate in atmospheric dust particles at Taklimakan desert, Geophy. Res. Lett., 39, L24803, doi:10.1029/2012GL054406, 2012.

Line 529 "From Figure 5, evident diurnal distinguishes are observed in two major chemical compositions (OM and GM) in this study. We can see that GM exhibits the lower proportion at night (35.3%) than daytime (47.5%), indicating its close relationship with human activities." However, does meteorology not also play a role? In Line 483 the authors imply that precipitation is higher during night-time, i.e. ". . . spontaneous combustion of waste occurs frequently during the day, because of less precipitation and higher air temperature at daytime. . ."

Response: As our best knowledge, the main source of the geological material in $PM_{2.5}$ is from crust dust. In addition to the effects of crust erosion by water and wind, it is largely related to the physical activities of human, such as resuspension from individual activities, construction activities and etc. The second reason (meteorological factor) shown in original line 483 explained the day-night ratio of heavy metals in $PM_{2.5}$, that is, the absolute concentrations of heavy metals; while the content in original line 529 discussed the proportion of geological material in $PM_{2.5}$. The meteorological factors, including precipitation, indeed cause scouring action on particulate matters (i.e., wet deposition), but they have little effect on altering the proportion of geological material in $PM_{2.5}$. Therefore, lower geological material proportion in $PM_{2.5}$ at night in this study was mainly due to the resuspension of the geological material from the less individual activities.

Line 539 ". . . due to the influence from the damp wood burning at the working time." I could not find any place where the wood moisture content was reported. Therefore, this statement and previous, as well as subsequent deductions, based on this statement, are not fact based. However, I do agree with later statements (line 566) that the wood will be damper in the wet season, i.e. ". . . increase in humidity (moisture content) of the wood used for grilling meat in wet season. . .".

Response: In this study, we did not measure the moisture of the wood used for barbecue in women's work. Since the grilling fuel (wood) was placed in the open area (no shield or roof), the abundant rainfall in wet season inevitably led to moisture increase in the wood. With on-field observation, it frequently took a long time to ignite the wood, which emitted more plume from damp fuels in the wet season during the sampling period. In previous studies (Chen et al., 2010; Grandesso et al., 2011; Keita et al., 2018; Shen et al., 2012, 2013), the results showed that biomass fuel with high moisture often required additional energy to vaporize the water and hence resulted in low combustion efficiency and high pollutant emissions. The emission factor (EF) of OC increase with the fuel moisture content (Chen et al., 2010).

The authors have modified the original statement in lines 538-541 as follows:

> "There is an exception in the last case, i.e., OM proportion at daytime women PE $PM_{2.5}$ was much higher (50.8%) than nighttime (38.2%) in wet season, due to the influence from the damp wood burning at the working time. As we know, burning biomass fuel with high moisture often results in lower combustion efficiency, longer smoldering period and high air pollutant emissions (Grandesso et al., 2011; Shen et al., 2012, 2013). The emission factor of OC increases with the fuel moisture content (Chen et al., 2010; Keita et al., 2018). Therefore, burning the damp wood led to more OC emission than dry wood to women at DF in this study".

Reference:

Chen, L.-W. A., Verburg, P., Shackelford, A., Zhu, D., Susfalk, R., Chow, J. C., and Watson J. G.: Moisture effects on carbon and nitrogen emission from burning of wildland

biomass. Atmos. Chem. Phys., 10, 6617–6625, 2010.

Grandesso, E., Gullett, B., Touati, A., and Tabor, D.: Effect of moisture, charge size, and chlorine concentration on PCDD/F emissions from simulated open burning of forest biomass. Environ. Sci. Technol., 45, 3887–3894, 2011.

Shen, G., Wei, S., Wei, W., Zhang, Y., Min, Y., Wang, B., Wang, R., Li, W., Shen, H., Huang, Y., Yang, Y., Wang, W., Wang, X., Wang, X., and Tao, S.: Emission factors, size distributions, and emission inventories of carbonaceous particulate matter from residential wood combustion in rural China. Environ. Sci. Technol., 46, 4207−4214, 2012.

Line 584. "Students at WB: nighttime PE PAHs were higher in dry season and lower in wet season compared with daytime levels, with the average D/N ratios of 0.7 and 1.8, respectively. The higher concentrations of combustion markers-BbF and BeP were observed during the day, while the higher concentrations of gasoline vehicle emission markers-DahA and BghiP were found at night (Baek et al., 1991; Wang et al., 2006), which was related to the garbage truck for waste transportation from city to the landfill during night." I am not sure that the latter explanation can be so simple, i.e. only due to "garbage truck".

Response: We apologize for the misleading. Referring to the middle panel of Figure 6A, it shows that the distributions of PAHs in students' $PM_{2.5}$ personal exposure samples in the dry and wet seasons were basically the same, with the higher concentrations in the dry season. The authors discussed the diurnal variation of PAHs concentration in different seasons and the dominant PAH species in daytime and nighttime. Both PAH profiles had a similar feature of high combustion markers of BbF and BeP, and gasoline emission markers of DahA and BghiP. Besides, with the large error bars (standard deviations) of PAH concentrations shown in this Figure, we believe that the previous statements about the dominant PAHs species in the daytime and nighttime are not so supportive, and thus the statements have been deleted from the revised manuscript.

In addition, the statements in lines 586-591 have been revised as following:

> "Both of the PAH profiles in the day and night were with the features of higher combustion markers-BbF and BeP, and higher gasoline vehicle emission markers-DahA and BghiP (Baek et al., 1991; Wang et al., 2006)".

Line 706. Can the authors please explain the selection of species included in the "Noncancer risks", wherein only "four heavy metals (Mn, Ni, Zn and Pb)" were considered?

Response: The non-carcinogenic risks of heavy metals in $PM_{2.5}$ via inhalation were calculated according to the U.S. EPA health risk assessment model (USEPA, 2004, 2011). U.S. EPA health risk assessment model is the process to estimate the nature and

probability of adverse health effects in humans who may be exposed to chemicals in contaminated environmental media, now or in the future. The reason we only chose Mn, Ni, Zn and Pb to assess the health risks in personal exposure $PM_{2.5}$ samples is because these four metals (among all the chemicals we analyzed in this study) are included in this U.S. EPA health risk assessment model and they are assessed to be hazardous to human health in the previous studies (e.g., Hu et al., 2018; Kong et al., 2015; Sun et al., 2014; Xu et al., 2018).