# Physico-chemical characterization of urban aerosols from specific combustion sources in West Africa at Abidjan in Côte d'Ivoire and Cotonou in Benin in the frame of the DACCIWA program

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#### 31 Abstract

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Urban air pollution in West Africa has yet to be well characterized. In the frame of 32 DACCIWA (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) program, 33 intensive measurement campaigns were performed in Abidjan (Côte d'Ivoire) and Cotonou 34 (Benin), in dry (January 2016 and 2017) and wet (July 2015 and 2016) seasons, at different 35 sites chosen to be representative of African urban combustion sources (i.e. domestic fires 36 (ADF), traffic (AT) and waste burning (AWB) sources in Abidjan and traffic source in Cotonou 37 (CT)). Both the size distribution of particulate matter (PM) and their chemical composition 38 including elemental carbon (EC), organic carbon (OC), water-soluble organic carbon (WSOC), 39 water-soluble inorganic ions (WSI) and trace metals were examined. Results show very high 40 PM concentrations at all sites and a well-marked seasonality as well as a strong spatial variation. 41 42 The average PM<sub>2.5</sub> mass concentrations during the wet season are 517.3, 104.1, 90.3 and 69.1 ug.m<sup>-3</sup> at the ADF, CT, AT and AWB sites, respectively. In the dry season, PM<sub>2.5</sub> concentrations 43 decrease to 375.7 µg.m<sup>-3</sup> at the ADF site, while they increase to 269.7, 141.3 and 175.3 µg.m<sup>-3</sup> 44 at the CT, AT and AWB sites, respectively. The annual PM2.5 levels at almost all sites are 45 significantly higher than the WHO guideline level of 10 µg.m<sup>-3</sup>. As for PM mass, (EC) and 46 (OC) concentrations are also maximum at ADF site, accounting up to 69% of the total PM mass. 47 Such high content is mainly linked to wood burning for domestic cooking and commercial food 48 smoking activities. Dust contributions are dominant at CT (57-80%), AT (20-70%) and AWB 49 (30-69%) sites and specially in the coarse and fine particle modes at CT and in the coarse 50 fraction at AT site, which may be explained by the impact of long-range desert-dust transport 51 and re-suspended particles from the roads, in addition to anthropogenic sources. The 52 contributions of WSI to the total PM mass, mainly driven by chloride, nitrate and calcium in 53 the fine and/or large particles are highly variable according to the sites but remain less than 54 30%. Values are generally 1-3 times higher in the wet season than the dry season. This is due 55 to anthropogenic emissions but also to nitrate formation by reaction processes and natural 56 57 emissions. The concentrations of trace element well reflect the trends of dust at the traffic and AWB sites, with a predominance of Al, Na, Ca, Fe and K, keys markers of crustal dust. This 58 59 study constitutes an original database that characterizes specific African combustion sources.

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Keywords: atmospheric pollution, chemical composition, physicochemical characterization,
Particulate matter, traffic, waste burning, domestic fires.

#### 63 **1.** Introduction

The impact of anthropogenic pollution on environment and health has been demonstrated by 64 numerous studies in Europe and North America, which have contributed to the implementation 65 of emission reduction policies. By contrast, air pollution in Africa is far from being well 66 characterized, although it is suspected to be responsible for negative health outcomes (WHO, 67 2014). This is a major problem since Africa is an intense emitter of pollution from 68 69 anthropogenic sources that includes domestic fires, vehicular traffic, waste burning as well as 70 growing oil and mining industries. It has also one of the fastest growing urban populations in the world, especially in West and East Africa (UN, 2019). As a consequence, it has been shown 71 that massive urbanization and rapid economic growth could be responsible for tripling 72 73 anthropogenic emissions in Africa between 2000 and 2030 (Liousse et al., 2014). Moreover, it is important to recall the impact of biomass burning and dust sources in the African atmospheric 74 75 composition, especially occurring during the dry season. All of this results in a major 76 degradation of urban air quality and an impact on the health of exposed populations. Only a few 77 studies on this subject have been conducted in West Africa (Val et al., 2013; Dieme et al., 2012; Kouassi et al., 2009) despite the high atmospheric pollutant concentrations already measured 78 79 to be of the same order as in Asian megacities and well above WHO international standards (WHO, 2014). 80

West Africa is then an "unique laboratory" to study urban pollution. Previous studies conducted 81 82 under the framework of the AMMA (Analyses Multidisciplinaires de la Mousson Africaine) 83 and POLCA (POLlution des Capitales Africaines) programs, have revealed very high average particulate mass concentrations in Cotonou (Benin), Bamako (Mali), Dakar (Senegal) and 84 Yaoundé (Cameroun) during the dry season (Doumbia et al., 2012; Val et al., 2013), suggesting 85 86 that the population may be affected by negative health outcomes. For example, during the dray season in Bamako (Mali) and Dakar (Senegal), Val et al. (2013) showed that the inflammatory 87 impact of combustion aerosol depends on the type of emission sources and determined the 88 predominant role of particulate organic matter. This is consistent with global findings showing 89 that fine and ultrafine aerosol fractions, as well as their content in trace metals and organic 90 compounds, induce biological effects due to their ability to reach the distal lung (Cassee et al., 91 92 2013). Such reasons highlight the need to better understand the size-speciation of aerosol chemical composition for the main West African anthropogenic sources during the different 93 94 seasons. Within this context, the DACCIWA (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) program, dedicated a specific work package to "Air Pollution and 95

Health" dealing with pollutant characterization related to health issues through toxicologicalstudies and epidemiological studies.

98 Campaigns have been organized from December 2014 to March 2017 in Abidjan and Cotonou. The strategy was to measure aerosol chemical composition in different sites, representative of 99 100 the main prevailing urban sources in West Africa following Liousse et al. (2014) and Keita et 101 al. (2018). Two typical traffic-sampling sites were chosen, one in Abidjan (Côte d'Ivoire) and 102 another one in Cotonou (Benin), to take into account differences in terms of fleets, type of fuel used and quality of roads. Indeed, in Cotonou, the majority of population uses two-wheel 103 vehicles susing gasoline fuel or gasoline and oil fuel, whereas in Abidjan, the vehicle fleet is 104 dominated by four-wheel engines using diesel fuel. Measurements were also performed at 105 domestic fire and waste burning sites, both located in Abidjan. 106

107 For a period of two years, PM<sub>2.5</sub> mass and carbonaceous aerosol were weekly measured and results are discussed in *Djossou et al. (2018)*. In this paper, we focus on the results from the 108 109 intensive campaigns. We present measurements obtained at each site during the wet and dry seasons of the studied periods: (i) PM size distribution and mass concentrations and (ii) PM 110 111 chemical composition including carbonaceous aerosol, water-soluble organic carbon, watersoluble inorganic ions, dust and trace elements in different size fractions. Experimental method 112 113 including description of sites, types of measurements and analyses, meteorological conditions, 114 will be presented in the section 2, whereas results and discussion are discussed in the sections 3 and 4 of the paper, respectively. 115

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#### 117 **2. Experimental method**

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# 2.1. Description of sites

Measurement campaigns have been performed in wet season (July 20-26, 2015 and July 4-13, 2016) and dry seasons (January 7-15, 2016 and January 5-14, 2017) at three sites in Abidjan (Côte d'Ivoire), representative of different sources, i.e. ADF for Abidjan Domestic Fires, AWB for Abidjan Waste Burning and AT for Abidjan Traffic (Figure 1), as well as one traffic site in Cotonou (Benin) (Figure 2).

As shown in Figure 3 which presents pictures of the different sampling sites, the ADF site (5°
19' 44 "N, 4° 06' 21" W) is situated on a platform, 5 m above ground level, in Yopougon

Bracody district near a market (Figure 1). This geographical area is highly populated with 126 various small commercial activities such as a fish and meat-smoking by women. There are also 127 many formal and informal settlements, which mainly use wood and charcoal as a source of fuel 128 129 for private and professional combustion activities. Other sources of concern contributing to the mix of pollutant emissions in the area include transportation-related emissions, biomass 130 burning, garbage bins or small landfills and various other fugitive sources. The AT site (5° 21' 131 14" N, 4° 01' 04" W) is located in Adjamé, on the roof of « 220 pharmacie logement building », 132 about 7 m above ground level and roughly 10 m away from the main road. This site, close to 133 134 the Adjamé market and to a bus station, is highly affected by traffic (Gbaka, bus, taxi, woroworo, private cars...). The AWB site (5° 21' 12" N, 3° 56' 16" W) is located at Akouédo in the 135 district of Cocody, on the roof of « Talafiguié », a building 15 m above ground level. This site 136 close to the big municipal landfill of Abidjan operational since 1965 and now covering an area 137 138 of 153 ha, is submitted to frequent waste burning pollution. The Cotonou Traffic (CT) (6° 22' 19" N, 2° 26' 5" E) site is located in Cotonou, on the «Sogema» building roof, about 7 m above 139 140 ground level. This site is close to the Dantokpa market and also to the biggest crossroad of Cotonou (intersection of 4 main roads). This site is highly influenced by intense traffic 141 142 activities. As previously mentioned, such a site is interesting because the vehicle fleet and fuels are different in Cotonou compared to Abidjan: (1) there are many two-wheel vehicles in 143 Cotonou whereas a few only in Abidjan; (2) in Cotonou, gasoline is of poor quality due to the 144 illegal fuel transport from Nigeria and (3) the roads are in worse conditions in Cotonou than in 145 Abidjan. 146

147 **2.2. Measurements** 

148 During each intensive campaign and on each site, two 3-hour samples collected with 149 cascade impactors operating in parallel are obtained for three consecutive days (i.e. six size-150 resolved samples per site during each campaign), to allow size-speciated characterization of the aerosol chemical composition. Note that the choice of the 3-hour periods is linked to the period 151 of maximum pollution for each site as shown by preliminary studies (e.g. morning at ADF site 152 (7-10am), afternoon at AT site (4-7pm, morning at CT site (7-10am)). There is no specific 153 period at AWB site since the activities are roughly the same during the day. The first impactor 154 with 4 stages (PM>2.5; PM 2.5-1; PM 1-0.2; PM<0.2) includes 4 quartz fiber filters (QMA, Whatman) 155 for mass and carbonaceous aerosol (EC, OC and WSOC analysis). The second impactor with 3 156 stages (PM>2.5; PM2.5-1; PM1-0.1) is equipped with three Teflon filters (Zefluor, Pall 157 Corporation®), dedicated for water-soluble ions species and trace elements. Due to operational 158

problems in july 2016, this second 3-stage cascade impactor is replaced by another 3-stage cascade impactor with different size cuts ( $PM_{>10}$ ,  $PM_{10-2.5}$ ,  $PM_{2.5-1}$ ). For consistency, results will be presented as an ultrafine (UF), fine (F) and coarse (C) classification. The two first stages ( $PM_{>2.5}$  and  $PM_{2.5-1}$ ) being considered as the coarse particulate fraction, the  $PM_{1-0.1}$  or  $PM_{1-0.2}$ stage, the fine particulate fraction and the  $PM_{<0.2}$  stage, the ultrafine fraction.

All the filters are prepared and analyzed at the Laboratoire d'Aerologie in Toulouse under different protocols described in the following paragraphs. Note that the quartz filters are prefired before sampling.

- 167 **2.3. Analyses**
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# 2.3.1. Gravimetric analyses

Aerosol mass concentrations are obtained using a high-precision balance (SARTORIUS MC21S), placed under a controlled temperature and humidity atmosphere (Person and Tymen, 2005). Before weighing, the filters are kept 24 hours in the weighing room at an ambient relative humidity of  $30 \pm 15\%$ . The filters are weighed before and after sampling. Result of a gravimetric measurement consists of the average of 2 to 4 weighing whose differences do not exceed 5µg. The standard error on a gravimetric measurement is therefore less than 10 µg, typically representing less than 5% of the particles mass.

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# 2.3.2. Carbonaceous aerosols

177 Carbonaceous aerosol is determined with thermal analysis with a two-step method178 adapted from Cachier et al. (1989). Two aliquots of the same filter are separately analyzed.

One portion is directly analyzed for its total carbon content (TC). The other portion is first submitted to a pre-combustion step (2 h at 340°C under pure oxygen) in order to eliminate Organic Carbon (OC), and then analyzed for its Elemental Carbon (EC) content. Organic carbon (OC) concentrations are calculated as the differences between TC and EC. Note that the aerosol carbon content is quantified by a non-dispersive infrared (NDIR) detector with G4 ICARUS instrument with a detection limit of the order of 2  $\mu$ gC.cm<sup>-2</sup>. Uncertainty is in the order of 5% for TC, while being in the range of 5-20%, for EC and OC.

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#### 2.3.3 Water Soluble Organic Carbon analysis

187 WSOC measurements are performed using a total organic carbon analyzer (Sievers M9).
188 A detailed description of this technique is reported in Favez et al. (2008). Briefly, the full

oxidation of total organic carbon into CO<sub>2</sub> is obtained by coupling chemical oxidation (with 189 ammonium persulphate) and UV light. CO<sub>2</sub> is then quantified by conductivity. Analyses are 190 conducted on 20 ml of solution extracts. For UF samples, solutions to be analyzed are obtained 191 using a total filter surface of 3cm<sup>2</sup> (6x0.5 cm<sup>2</sup> punches symmetrically taken out of each QMA 192 filter), whereas, for C and F sizes, due to the geometry of the spots at the surface of the filters, 193 samples are divided into equivalent parts (1/2 or 1/4 of 47 mm filters, rest of the filters being 194 used for carbonaceous analysis). The extraction protocol consists of 16h soaking under soft 195 shaking in an Erlen-Meyer containing 20mL of ultra-pure water. Prior to WSOC analysis, water 196 197 extracts are filtered through Teflon (PTFE) filters (0.2µm pore size diameter) in order to remove any suspended particle. Measurement uncertainty, given by the manufacturer, is of the order of 198 7%. The overall calculated blank value is of the order of  $2.27 \pm 0.33 \,\mu gC.cm^{-2}$ , which represents 199 16.4± 8.5% of the mean WSOC content. For each sample, duplicate analyses show a good 200 201 reproducibility.

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#### 2.3.4. Water-soluble ionic species

Water-soluble ionic species (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>) are 203 204 analyzed using ion chromatograph (IC), following the analytical protocol described in Adon et 205 al. (2010). Briefly, the aerosol water-soluble fraction is first extracted from half-sampled Teflon filter (the other part being used for trace element analysis), with a 10-min sonication in plastic 206 vials including 6 ml or 10 ml of purified water with a controlled resistivity of  $18.2M\Omega$ . Then 207 these vials are subjected to ionic chromatograph analysis or stored at +4°C if not analyzed 208 immediately. Cations are analyzed with Dionex DX-100 and anions with Dionex DX-500 with 209 a detection limit of 1 to 6 ppb depending on ionic species. Uncertainties in the range of 1-50% 210 is found depending on ionic species. 211

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#### 2.3.5. Trace elements

The protocol to measure trace element concentrations is developed and performed at the 213 Laboratory of Environmental Geosciences of Toulouse. Half-sampled Teflon filters (the other 214 part being used for water-soluble ionic species, see below) are mineralized by acid digestion 215 216 with a 10 ml concentrated HNO<sub>3</sub> and 0.5 ml HF solution (Lamaison, 2006) using a closed vessel microwave accelerated reaction system (MARS 5, CEM Corporation) at high pressure (700 psi) 217 (Celo et al. 2010). The digestion is realized in 3 steps: a rise in temperature at 130°C in 3min 218 and holding for 1 min, then, a second rise at 160°C in 1 min and holding for 30 seconds and 219 finally a third rise to 180°C in 1 min and holding for 3min. After a 12 h cooling period, the 220

solutions are evaporated at 80°C, and concentrated in 7 ml of 2% concentrated HNO<sub>3</sub> solution, before analysis by ICP-MS which are performed with a 7500 ce Agilent Technologies instrument equipped with a collision cell, and using In and Re as internal standards. The detection limit is less than 10 ppt. For all the samples, the final blank values and detection limit on filters are taken into account for final concentrations calculations. 13 trace metals are considered in this work: Al, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Ba, La, Th, Pb and Cd.

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# 2.3.6. Dust calculation

Many methods can be used to quantify dust concentrations. We have selected three methods (Sciare et al. 2005, Guinot et al. 2007, Terzi et al. 2010) to underline the uncertainties linked to dust estimates.

- 231 (1) Sciare et
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(1) Sciare et al. (2005) method consists of using soluble calcium data obtained with Ionic Chromatography (IC), to estimate the dust concentrations following the relationship: dust = 10.96\*nss-Ca<sup>2+</sup>, where nss-Ca<sup>2+</sup> (=1.02\*Ca<sup>2+</sup>-0.038\*Na<sup>+</sup>) refers to non-sea-salt calcium concentration.

(2) Guinot et al. (2007) method is based on a chemical closure where fine and coarse 235 236 particle aerosols are separated in 4 components (EC, POM, WSI and dust). EC, WSI, and total aerosol mass are directly experimentally determined (see below paragraphs). 237 238 POM concentrations are obtained from OC concentrations experimentally determined and k, the OC/POM conversion factor. Dust concentrations are obtained from measured 239  $Ca^{2+}$  concentrations and f, the abundance of calcium in dust. The k and f values are 240 obtained from a linear regression (L) between the reconstructed and the weighed aerosol 241 mass concentrations. Briefly, first step consists of focusing on the aerosol coarse 242 fraction. k is fixed to 1.8 and as a result of (L) just mentioned, f is obtained to be in the 243 range of 0.012 to 0.15 depending on our sites. Second step deals with the aerosol fine 244 245 fraction. The f values just obtained for the aerosol coarse fraction are applied to the fine fraction and k ratios are estimated using (L) to be in the range of 1.2 to 2.1. Note that at 246 all of our sites, the correlation between  $Ca^{2+}$  and the missing mass between the weighed 247 and the reconstructed aerosol mass is sufficiently good  $(r^2=0.9)$  to support the 248 consistency of this simple approach for the evaluation of dust. Also, f and k values are 249 included in the range of values provided in the literature (He et al. 2001; Sun et al. 2004; 250 Guinot et al. 2007). However, it is important to mention that the range of f and k 251 coefficients are large which is due to the source mixing observed in this study. 252

(3) In Terzi et al. (2010) method, dust is obtained with the following relationship: dust = 1.89[Al] + 1.21[K] + 1.95[Ca] + 1.66[Mg] + 1.7 [Ti] + 2.14[Si] + 1.42[Fe]. In our study, all these elements are determined except Silica (Si). Consequently, we used mean Si values obtained from different relationships available in the literature (SiO<sub>2</sub> =  $3*Al_2O_3$ for Alastuey et al., 2005, Si = 4.0\*Al for Zhang et al., 2003 and Si = 2.03\*Al for Chiapello et al., 1997).

259 The results of dust concentrations estimated from the three methodologies above described are 260 summarized in the Table 1 for wet season (WS) 2016 and dry season (DS) 2017. Indeed, Ca, 261 Al, and Fe concentrations measured by ICP-MS are only available in WS2016 and DS2017 due to experimental problems, whereas  $Ca^{2+}$  concentrations measured by IC are available for all 262 campaigns. As shown in table 1, the dust obtained from Ca<sup>2+</sup> measured by IC (Sciare et al., 263 2005) and by the Guinot et al. (2007) method is lower than that obtained from trace elements 264 265 (Terzi et al., 2010) for DS2017 whereas in the same order of magnitude in WS2016. Such results are in agreement with methodological aspects. Indeed, Al, Fe, Ca ... obtained by ICP-266 MS include both soluble and insoluble particles whereas Ca<sup>2+</sup> measured by IC only include 267 soluble particles. During the dry season, comparison of Ca measured by ICP-MS (not shown 268 here) is higher than that of the IC, by a factor of 1.7, 1.8, 2.2 and 1.1, at the ADF, AWB, AT, 269 and CT sites respectively. By contrast, this factor is low and constant (1.3) in the wet season 270 for all the sites. In our study, due to the lack of trace element data for WS2015 and DS2016, 271 dust estimations are performed from Guinot et al. (2007) method. This choice globally implies 272 an underestimate of dust concentrations by a factor of 1.5 to 3.5 in DS2017 as shown in Table 273 274 1.

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#### 6 2.3.7. Aerosol chemical closure methodology

As previously mentioned and detailed, aerosol chemical closure is performed followingthe Guinot et al. (2007) methodology.

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# 2.4. Meteorological conditions

In Figure 4, meteorological data (surface temperature, wind directions and speed) issued from the NOAA Integrated Surface database (ISD; see https:// www.ncdc.noaa.gov/isd) and the ASECNA (Agence pour la Sécurité de la Navigation Aérienne en Afrique et à Madagascar) are presented for the South-West Africa region including Abidjan and Cotonou. As expected, this

area is under the influence of the Convergence Zone of two air masses of a different nature, i.e. 285 Harmattan (hot and dry continental trade winds) from the north and Monsoon (humid maritime 286 trade winds) from the south (Figure 4). Ground contact between these two air masses constitutes 287 the intertropical front (ITF) of which the fluctuations during the year determine the seasons in 288 the Gulf of Guinea (Tapsoba, 1997). During the dry season (from November to March), 289 temperatures are relatively high with maximum around 30°C on the coast. The humidity is low, 290 since the prevailing Harmattan wind blows from the desert, usually bringing dust (Figure 4, 291 lower line). The period from June to September, especially in July is the wet season period 292 293 when daytime temperatures are slightly lower, with maximum around 26/28°C on the coast 294 (Figure 4, upper line). At this season, the humidity level is high across the region. On the coast, 295 rains may occur from March to November.

During our campaigns (not shown here), temperatures are roughly the same at Abidjan and 296 297 Cotonou, reaching 28°C and 26°C in the dry and wet seasons, respectively. Gentle to moderate wind speeds are observed during the measurement campaigns at the two cities, with average 298 values of 15-20 and 15-22 km.h<sup>-1</sup> at Abidian and Cotonou, respectively. There is no 299 precipitation at CT site during the studied periods. In Abidjan on the contrary, low rains occur 300 301 both in wet and dry periods with cumulative precipitation higher in DS2017 (7mm), than in 302 WS2016 (4.7mm) and WS2015 (2mm). There is no rain in DS2016 (https://www.historique-303 meteo.net/afrique/).

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# 2.5. Backward trajectories

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) modelling 305 306 system (Air resources laboratory, Draxler and Rolph, 2012) is used for the trajectory analysis. 307 HYSPLIT model is run to compute 120 h back trajectories ending at Abidjan and Cotonou at 308 50 m a.g.l. (Figure 5). Global Data Assimilation System reanalysis database is used as 309 meteorological input, with a  $0.25 \times 0.25$  degrees horizontal resolution. Results presented in Figure 5, confirm that air masses mainly come from the north with a few from the south-west 310 in dry season (January), whereas from the south-west and the south in wet season (July). 311 Therefore, in January, Abidjan and Cotonou are mainly impacted by polluted air masses from 312 surrounding areas and northern countries with possible dust and west African biomass burning 313 influences, whereas in July, the impact of oceanic sources possibly polluted by long-range south 314 African biomass burning aerosols may be observed. 315

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# 317 **3. Results**

#### **3.1.** Aerosol size distribution and mass concentration

In Figure 6, the relative mass distribution of PM for Coarse (C), Fine (F) and Ultra-Fine 319 320 (UF) particle sizes in percentages are presented with bulk mass concentration averages indicated in the black boxes for each site and for each campaign. As it may be seen, bulk 321 concentrations vary widely from site to site and from campaign to campaign. During the wet 322 season, the average total concentrations range from 82 to 676 µg.m<sup>-3</sup> in 2015 and 56 to 358 323 ug.m<sup>-3</sup> in 2016, with the maximum at the Abidjan Domestic Fire (ADF) site. While during the 324 dry season, values range from 168 to 269  $\mu$ g.m<sup>-3</sup> in 2016 and from 114 to 559  $\mu$ g.m<sup>-3</sup> in 2017, 325 with maximum concentration obtained at the Cotonou Traffic (CT) and ADF sites. In terms of 326 size distribution, concentration peaks may be observed for all aerosol size-fractions which are 327 found to exhibit different seasonal patterns. UF particles (<0.2 µm) represent the highest 328 contributor to the bulk mass at the ADF site, by up to 60 % (335.3 µg.m<sup>-3</sup>) in DS2017. F particles 329 (1-0.2 µm) are the second most important contributor and both combined particle sizes account 330 for more than 85 % of the total mass at the ADF site. In this site, ultra-fine and fine fractions 331 are also found to be maximum during WS2015 and WS2016 by up to 90 and 83%, respectively. 332 Let us note that C particle contribution in bulk is relatively higher in the traffic and waste 333 burning sites than in ADF site (40%) whereas F and UF particle contributions are on the order 334 335 of 60%.

In terms of PM2.5, the results of this work are presented in Figure 7. The mass concentration of PM2.5 averaged over DS2016 and DS2017 are  $154 \pm 74$ ,  $144 \pm 42$ ,  $134 \pm 7$ and  $211 \pm 51 \mu$ g.m<sup>-3</sup> at the ADF, AWB, AT and CT sites, respectively and  $338 \pm 24$ ,  $45 \pm 3$ ,  $52 \pm 4$  and  $70 \pm 1 \mu$ g.m<sup>3</sup> over the wet seasons (2015-2016). The increase in PM2.5 is of the order of 54% at ADF from dry to wet season, whereas a sharp reduction (more than 60%) is obtained at AWB, AT, and CT sites.

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# 3.2. Carbonaceous aerosol

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#### **3.2.1. EC and OC concentrations**

In Figure 8, EC relative mass contributions are presented for each size, site and campaign: wet season 2015 (WS2015), wet season 2016 (WS2016), dry season 2016 (DS2016) and dry season 2017 (DS2017). Mean EC bulk mass concentrations are added in the black boxes for each size and for each campaign. The most striking feature is that the ADF site concentrations are higher than at the other sites in WS2016 and in DS2017, whereas of the same order of CT site concentrations in the other seasons. Mean concentration at the CT site ( $16\mu g.m^{-3}$ ) is slightly higher than at the AT site ( $10\mu g.m^{-3}$ ), whereas the lowest concentrations are found at the AWB site. Results of the EC size distribution are very consistent among the different sites (Figure 8). Whatever the site and the season, higher EC concentrations are found in C (42%) and UF (43%) particles compared to F particles.

Same data are presented for OC concentrations in Figure 9. It may be underlined that ADF OC values are always higher than in the other sites by a factor ranging from 6 to 30, for all seasons and particle sizes, with highest and lowest values respectively in DS2017 and DS 2016. In terms of size distribution, maximum OC concentrations at the ADF site may be found in UF (53%), then F (29%) and finally C (18%) particles. The same distribution is observed for the traffic sites in DS2016, however, for the other campaigns, OC size distribution looks like the EC ones with higher concentrations in UF and C particles than in F particles.

As shown in Figure 10, the highest OC/EC ratios are always obtained at the ADF site 361 with a value as high as 25 for F particles in WS2016 whereas the lowest values are found in 362 DS2017. This is the same feature for the other sites with ratios lower than 2 in DS2017. OC/EC 363 364 ratios in AWB site are higher than in the traffic sites. Note that values at AT site are higher than CT values in the wet season whereas lower in the dry season. Finally, it is interesting to 365 underline that linear correlations between EC and OC are obtained in the ultrafine and fine 366 modes in all campaigns, particularly in DS2017 ( $r^2 = 0.8, 0.8, 0.9$  and 0.9) at the ADF, AWB, 367 AT and CT sites, respectively. This suggests that different studied sources can be assessed as 368 significant sources of both EC and OC. 369

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#### 3.2.2. Water-Soluble Organic Carbon

Concentrations of WSOC and WSOC/OC ratios are presented in Table 2 for each size 372 (UF, F, C and PM2.5) and campaign. As seen, WSOC are always higher at the ADF site than 373 in other sites, at least by a factor of 12. Maximum values are obtained in WS2016 with an 374 average of 16.47, 17.08 and 79.68 µgC.m<sup>-3</sup> for coarse, fine and ultra-fine fractions, respectively, 375 followed by WS2015 and DS2017. WSOC concentrations are the lowest in DS2016, with an 376 average of 4.14, 6.95 and 21.89 µg.m<sup>-3</sup> for coarse, fine and ultrafine fractions, respectively. In 377 378 terms of seasonality, there is not a clear trend in WSOC values at the AWB and AT sites, whereas at the CT and ADF sites, WSOC values are found to be respectively higher and lower 379

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in dry seasons compared to wet seasons. It is also interesting to note that WSOC are maximum
in UF sizes in the AT, ADF and AWB sites. At the CT site, the highest values are found in the
coarse particulate fractions, except in DS2016.

As expected, WSOC is strongly correlated with OC (r=0.7 at ADF, 0.8 at AT, 0.5 at 383 AWB and 0.7 at CT), whereas correlations with EC are weaker, especially at the AWB and CT 384 sites with values ranging from 0.1 to 0.4, respectively. Finally, when looking at WSOC/OC 385 ratios (Table 2), maximum values are obtained at the ADF site with PM2.5 ratios as high as 386 43%, followed by the AT and AWB sites with 32%. The lowest value (23%) is found at the CT 387 388 site. Also, Table 2 shows that there is no clear seasonality in WSOC/OC values, excepted at ADF where maximum values occur during the wet season. Note as for WSOC, that ratios are 389 maximum in UF and F fractions for all sites except at the CT site where the ratio for coarse 390 391 fraction is the highest.

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# 3.3. Water-soluble ionic species

393 Figure 11 shows the relative contribution of the major ions to the total concentration (also given) of the ions in the different particle modes (C and F) at the ADF, AWB, AT and CT 394 395 sites for the different measurement campaigns. Let us recall here that only C and F fractions may be documented due to the our experimental protocol. Total concentrations present 396 397 maximum values in ADF and CT sites. Values in AWB and AT sites are of the same order of magnitude and lower by a factor of 2 than in ADF and CT sites. The contribution of different 398 ions show significant variations from site to site. The dominant ionic species at the ADF site 399 over all campaigns is chloride (Cl<sup>-</sup>), with a 26% contribution, followed by nitrate (NO<sub>3</sub><sup>-</sup>) (16%), 400 calcium (Ca<sup>2+</sup>) (13%) and potassium (K<sup>2+</sup>) (12%). Sulfate (SO<sub>4</sub><sup>2-</sup>), ammonium (NH<sub>4</sub><sup>+</sup>), sodium 401  $(Na^+)$  and to a lesser extent magnesium  $(Mg^{2+})$  contributions are lower, ranging from 4 to 7% 402 403 of the total ion species. The lowest contribution is for organic acids with their total value lower than 5%. NO<sub>3</sub><sup>-</sup> is the major ionic component at the AWB and AT sites, representing 24% and 404 29% of the total water soluble inorganic concentration, respectively. The second major 405 contributor in AWB and AT is SO<sub>4</sub><sup>2-</sup>, accounting for 21% and 17% of the ion mass, respectively 406 followed by Ca<sup>2+</sup> (12% and 15%) and Cl<sup>-</sup> (15% and 13%). In CT, Ca<sup>2+</sup> is predominant with a 407 relative abundance of 24%, followed by  $NO_3^-$  (23%),  $SO_4^{2-}$  (19%) and  $Cl^-$  (13%).  $Na^+$ ,  $NH_4^+$ 408 and K<sup>+</sup> contributions are lower and in the same order of magnitude in AT, AWB and CT sites, 409 ranging from 4 to 9% of the total ion species. Note that organic ion contributions at AT, CT and 410 AWB is of the same order than in ADF, with lower values at CT. It is interesting to underline 411 in the Figure 11, that  $NO_3^-$  contribution is always higher in the coarse than in the fine size. 412

413 Conversely,  $K^+$  is always higher in the fine than in the coarse size. In CT,  $Ca^{2+}$  in the fine 414 fraction is as high as in the coarse fraction whereas in AT, AWB and ADF,  $Ca^{2+}$  coarse fraction 415 is predominant. Fine particle contribution may be noticed for Cl<sup>-</sup> in ADF whereas in the other 416 sites, Cl<sup>-</sup> is most likely dominated by coarse particles. Finally, SO<sub>4</sub><sup>2-</sup> is mainly found in the fine 417 mode at the AT, AWB and CT sites, but in the coarse mode in ADF site.

In terms of seasonal variations, it may be shown in Figure 11 that higher Cl<sup>-</sup> values are found 418 in wet seasons than in the dry seasons everywhere, except in ADF site where there is no marked 419 difference between seasons. For example, the mean relative total percentages of Cl<sup>-</sup> at the CT 420 site are 38 and 24% in the WS2015 and WS2016, respectively, while these percentages decrease 421 significantly to 18 and 13% in the DS2016 and DS2017, respectively. The Cl<sup>-</sup>/Na<sup>+</sup> ratios are 422 about 1.5 everywhere in both seasons, in agreement with the typical sea water ratio (1-1.2) 423 (Hara et al., 2004), except at the ADF site where these ratios increase to 4 and 5 in wet and dry 424 season, respectively and at the AWB site in the dry season (2).  $K^+$  and  $Ca^{2+}$  are always higher 425 in dry season than in wet season except for  $Ca^{2+}$  in ADF where values are of the same order. 426 Finally, the same trend is observed for  $NO_3^-$  and  $SO_4^{2-}$  with higher values in dry than in wet 427 seasons at AWB and CT sites whereas values at ADF and AT sites are of the same order of 428 429 magnitude for the two seasons.

430

#### **3.4. Trace element concentrations**

Table 3 shows the mean values of the major trace elements in bulk aerosol at the 431 different studied sites in WS2016 and DS2017, with their corresponding relative abundances in 432 the total aerosol mass into brackets. Let us recall that data are not available in WS2015 and 433 DS2016. The concentrations of trace elements span a wide range, from 0.2 to 25.2 µg.m<sup>-3</sup>. 434 Among the measured elements, Al, K, Na and Ca are the most abundant, followed by Fe and 435 Mg. In DS2017, Al and Na concentrations are higher in AWB than in the other sites. The 436 minimum value for these species is found in ADF site. Values in traffic sites are of the same 437 order of magnitude and higher than in ADF site. Maximum of Ca and K values may be found 438 in CT and ADF site respectively. It is interesting to note that Al, K, Na concentrations are higher 439 440 in the dry season than in the wet season. Such feature is less clear for Ca, whose seasonal variability is less marked except in AWB and AT sites. In terms of Mg, maximum values are 441 observed in ADF site and of the same order of magnitude whatever the season. Fe abundance 442 is higher in AWB and CT sites than in ADF and AT sites and higher in DS2017 than in WS2016 443 everywhere. The other metals (Ti, P, Zr, Zn, Cr, Mn, Pb and Ni) represent less than 0.5% and 444

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2% of the total mass in WS2016 and DS2017, respectively, at all sites, with Cr, Mn, Pb and Ni
exhibiting less seasonal variability compared to the rest of the metal elements.

To assess the relative contribution of crustal and non-crustal origin of elemental aerosol loadings, source enrichment factor (EF) of a trace element X have been first calculated with the following formula using both literature data of the typical elemental composition of the upper continental crust (Mason and Moore, 1982; Taylor, 1964), measured elemental composition from this study and Al as a reference element :

$$EF_{X} = \frac{\frac{[X]_{atm}}{[Al]_{atm}}}{[X]_{soil}/[Al]_{soil}}$$

Where [X]<sub>atm</sub> and [Al]<sub>atm</sub> are the concentrations of the chemical element X and Al in the 453 atmosphere, respectively, and [X]<sub>soil</sub> and [Al] soil are the typical concentrations of the element 454 X and Al in the earth's crust, respectively. Al is frequently used as a reference element assuming 455 that its anthropogenic sources in the atmosphere are negligible (Gao et al., 2002; Cao et al., 456 457 2005; Xu et al., 2012). In all sampling sites, EF values typically lower than 5 are obtained for several trace elements (Be, Sc, Ti, V, Fe, Ga, Sr, Nb, Rh, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, 458 459 Dy, Ho, Er, Tm, Yb, Lu, Ta, Th and U). This suggests a natural origin of these species (Freitas et al., 2007; Gao et al., 2002). The most enriched elements (EF>100) are Sb, Sn, Zn, Se, Te, 460 Cd, Pb, Bi and Mo at nearly all of the sites, indicating significant anthropogenic origin (Wang 461 et al., 2006). These elements are mainly emitted into the atmosphere through fossil fuel 462 463 combustion, traffic emission, wear of brake lining materials and industrial processes (Watson and Chow, 2001; Samara and al., 2003). Secondly, source contributions have been estimated 464 from these EF values following the method described by Arditsoglou and Samara (2005). Note 465 that this study refers to ratios for a limited list of sources, perhaps not including the African 466 source specificities. As a result, it may be seen that 30% of trace element concentrations is of 467 468 anthropogenic origin at ADF site whereas about 17 % at the others sites.

469 **3.5. Dust** 

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Figure 12 shows dust concentrations calculated from Guinot et al. (2007) methodology (see paragraph 2.3.6) for C and F particle sizes at the different sites for each season. Note that as for WSI and trace element and due to our sampling procedure, there are values for fine and coarse particles for all seasons excepted for WS2016 with values for coarse particles only.

474 During the wet season, coarse dust concentrations range from 5 to 25  $\mu$ g.m<sup>-3</sup> in 2015 and 9 to 475 37  $\mu$ g.m<sup>-3</sup> in 2016, with higher values at the CT and ADF sites in 2015 and at AT, CT and ADF in 2016. In WS2015, fine dust concentrations range from 12 to 49 with maximum values at ADF and CT sites also. During the dry season, values range from 38 to 156  $\mu$ g.m<sup>-3</sup> in 2016 and from 41 to 116  $\mu$ g.m<sup>-3</sup> in 2017, with maximum concentrations obtained at the CT site, followed by AWB site. When considering mean values of the dry seasons, total dust at CT is 2.4 times the values found at AT, 1.6 times at AWB and 3.4 times at ADF. Seasonal comparison shows that total dust concentration is higher in the dry seasons than in WS2015 by a factor of 3 in AT, 2.6 in CT and 4 in AWB, but of the same order of magnitude in ADF site.

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#### 3.6. Aerosol chemical closure

484 The aerosol chemical closure obtained using the Guinot et al. (2007) method (see below) 485 at the different sites for each season is presented in Figure 13. Results show clear intra- and inter-annual variations at all of the sites, as well as significant differences among the sites. In 486 total, dust accounts for 39 to 75% of the bulk PM mass at both traffic sites, with no clear 487 seasonal cycle and higher contributions in Cotonou (Figures 13c and 13d). These percentages 488 vary from 32 to 64% at the AWB site, and from 18 to 35% at the ADF site, with percentages 489 1.8 times higher in the dry season than in the wet season in AWB and no clear seasonal 490 491 difference in ADF (Figures 13a and 13b). Carbonaceous aerosol, the sum of EC and POM, show large contributions at the ADF site (from 49 to 69% of the total PM mass), with relatively 492 similar proportions in each season (Figure 13a). The absence of a clear seasonal pattern is also 493 observed in CT whereas carbonaceous aerosol is slightly higher in WS than in DS in AWB (23 494 and 16% respectively) and AT (37 and 21% respectively) (Figures 13b-d). Carbonaceous 495 aerosol contribution accounts for about 11-49% of the total mass at both traffic sites with higher 496 values in AT (mean of 30%) than in CT (13%). The ion percentages in PM fractions present 497 the same pattern at AT, CT and AWB sites with higher values in wet than in dry seasons. In 498 499 these sites, we may notice that mass concentration in coarse particles is larger in the wet season whereas of the same order of magnitude than the one in fine particles in the dry season. In ADF, 500 501 no marked difference may be found between the seasons and the sizes (Figure 13).

502

# 503 **4. Discussion**

A discussion of the results site by site (Abidjan domestic fire site, traffic sites both together and waste burning site) will be first proposed. We will scrutinize (1) the proximity between the sites and the sources; (2) the source specificity with more or less incomplete combustion (e.g. wood combustion and two-wheel vehicle emission factors are higher than gasoline emission factors (Keita et al., 2018); (3) the relative influence of other local sources or transported sources to
the studied sites such as dust and biomass burning; (4) the occurrence of continental air masses;
(5) the variation of the boundary layer height (as reported by Colette et al., 2007); and (6) the
meteorological parameters (e.g. temperature, relative humidity and wet deposition) to explain

- the differences of pollutant concentrations and their seasonal and inter-annual variabilities.
- 513 In a second part, we will present comparison of our values with other DACCIWA values and
- also with literature values for other intensive campaigns in Africa.
- 515

# 516 **4.1. Abidjan Domestic Fires (ADF)**

As shown in the above paragraphs, maximum values are obtained at the ADF site, for aerosol mass, EC, OC, WSOC, water-soluble ionic species (e.g.  $Cl^-$ ,  $NO_3^-$ ,  $Ca^{2+}$  and  $K^+$ ) and some trace elements such as Mg and K (whereas Al, Na and Fe are lower than in the other sites). Also, aerosol PM2.5 values are well above the annual and daily WHO guidelines of 25 and 10 µg.m<sup>-</sup> <sup>3</sup> respectively, whatever the season.

- 522 Such pattern is due to the proximity of the ADF site to the studied combustion source: in that area, the use of wood combustion is very active due to commercial activities of women drying 523 524 fish and meat and domestic cooking. This is also confirmed by the high relative importance of 525 total carbon in aerosol mass whatever the size (49 to 69%) and by values of source enrichment factor. Indeed, at least 30% of trace element concentrations are of anthropogenic origin at ADF 526 site. In addition, wood combustion is well known to be highly polluting due to incomplete 527 combustion: this is shown here by the measurements of very high OC/EC ratios at ADF, on the 528 order of the one measured at the source level by Keita et al. (2018). This is also shown by 529 WSOC relative importance which is expected for wood burning following Yu et al. (2018), 530 Tang et al. (2016), Feng (2006) and Saxena and Hildemann (1996) and by the strong correlation 531 of WSOC with biomass burning K<sup>+</sup> tracer. 532
- Chloride is most likely associated with sea salt origin (normal chloride concentrations represent 533 at least 55% of marine aerosols following Goldberg (1963)) or secondary aerosol production 534 535 (Li et al., 2016). Since chloride relative concentration at the ADF site remains lower than the that of sea-sal aerosols, the secondary production source would be the better explanation for 536 high chloride concentrations observed at ADF. The size distributions of Cl<sup>-</sup>, K<sup>+</sup>, NH<sub>4</sub><sup>+</sup> and SO<sub>4</sub><sup>2-</sup> 537 support this conclusion : the predominance of these elements in fine particle mode at the ADF 538 site would be associated with anthropogenic emissions, particularly biomass combustion and 539 domestic fires, or with secondary inorganic aerosols origin. This is confirmed by Cl<sup>-</sup>/Na<sup>+</sup> ratio 540 values as shown earlier. Contrarily,  $Ca^{2+}$  and  $NO_{3}^{-}$  contributions to the total ions at the ADF 541

site peak mainly in the large particle fraction and may be attributed to quasi natural origin, primarily to dust emissions and nitrate formation by reaction processes, respectively. In addition, Na<sup>+</sup> and Mg<sup>2+</sup> display similar size distributions at the ADF site, with the major contribution in the coarse particle fraction, suggesting the common sea salt origin of these two elements (Belis et al., 2013).

As we have shown above, the lower proportion of metal elements at the ADF site (6.5% of the 547 bulk concentration) can be explained by the less dominant influence of re-suspended dust 548 particles compared to traffic sources. Elements such as Cr, Mn, Pb and Ni have less seasonal 549 550 variability than other metallic elements. These small proportions of these non-crust elements suggest a low contribution of elements emitted mainly by anthropogenic activities such as 551 industrial processes (Viana et al., 2007 and 2008; Minguillón et al., 2014). Finally, the Zn/Cd 552 553 ratio has been also examined. A value of 29 close to ratio reported for gasoline vehicle (27, Qin 554 et al., 1997) is obtained for the ADF site, indicating that this site is also impacted by traffic sources. 555

- 556 High values of WSOC/OC ratios are expected to be harmful to health (Ramgolam et al., 2009, Val et al., 2013). This effect is being enhanced by the particulate size measured at this site (Kim 557 558 et al., 2003; Wilson et al., 2002). Indeed, the relative mass distribution of PM and OC particle sizes shows a major contribution of particles less than 1 µm (as high as 85% of PM). This could 559 be due to the fact that carbonaceous aerosols are formed near emission sources and are mainly 560 of submicron size (Boucher, 2012). Nevertherless, note that EC also presents large coarse 561 particle contribution. This could be due to the importance of wood burning at this site with less 562 563 efficient combustion and large particle emissions (Watson et al., 2011).
- In terms of seasonality, higher concentrations of aerosol mass, OC, WSOC, EC and total water 564 soluble ionic species (SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>) are observed in WS2015 and WS2016 than in 565 DS2016. This may be explained by a more incomplete combustion in the wet seasons than in 566 567 DS2016 due to the use of moist wood for cooking and smoking fish, which leads to large amount of smoke and higher particulate emission factor values. Note that DS2017 values are 568 569 as important as the ones of wet seasons, which will be explained later in the text. With regard to WSOC, their variabilities may be also linked to meteorological factors, such as solar radiation 570 571 (Tang et al., 2016; Favez et al., 2008) and relative humidity (Liang et al., 2016). At ADF site, temperatures are roughly similar in both seasons. However, RH variability may play a role since 572 it is higher in wet season than in dry season. Finally, our results indicate no clear seasonal cycle 573 for Cl<sup>-</sup>, which confirms its anthropogenic origin, as previously shown. 574

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#### 576 **4.2. Traffic sites (Abidjan traffic and Cotonou Traffic sites)**

Let us recall first that the two traffic sites have been chosen since they are representative of the traffic diversity in West Africa. At CT site, both personal cars, taxis and an important twowheel fleet may be found whereas at AT site, there are buses, taxis and personal cars. Also, the distance between the site and the traffic sources is the same for the two traffic sites, slightly larger than the distance between the site and the wood burning sources at ADF site.

In these two sites, concentrations are high with PM2.5 values well above the WHO guidelines. 582 Average aerosol mass, EC, OC, dust and water soluble ionic concentrations (with NO3<sup>-</sup> and 583 Ca<sup>2+</sup> maximum at AT and CT sites respectively) are higher at CT than at AT site by a factor of 584 1.5 to 2. Note that this poor air quality found in Cotonou has been reported by Cachon et al. 585 586 (2014). The higher values found in Cotonou could be due to more intense traffic in Cotonou 587 than in Abidjan. Also in Cotonou, this traffic is associated with the lack of public transportation 588 and the use of highly polluted mopeds (aged over 15 years) (Gounougbe, 1999; Avogbe et al., 2011), despite the effort in the last 10 years to restrict their use. Several studies such as MMEH 589 590 (2002) have shown that more than 94,000 mopeds and 350,000 second-hand vehicles are in circulation in Cotonou. Other factors contributing to the local pollution include outdoor 591 592 restaurants using charcoal and motorcycle garages, which are more present around the Cotonou 593 traffic site compared to Abidjan site. It also includes anthropogenic dust. Indeed, at Cotonou, the lack of road infrastructure favours the resuspention of dust particles. Finally, other sources 594 may potentially influence aerosol seasonal composition in these two sites, including marine 595 aerosols, transported dust and biomass burning particles as well as anthropogenic aerosols from 596 597 the surrounding countries (Figure 5). Note also that source enrichment factor values show that about 17% of trace element concentrations are of anthropogenic origin at both traffic sites and 598 that the relative importance of total carbon in mass is higher at AT than at CT sites. 599

600 Aerosol mass, composition and size depend on the season and the two traffic sites are differently 601 affected. The EC and OC concentrations measured in both traffic sites and averaged per season are higher in dry than in wet season. Such variations may be explained by several factors: 602 603 particulate wet deposition occurring during the wet season, reduction of traffic flow due to school vacations and meteorological influence. Higher EC and OC concentrations are obtained 604 605 at CT than at AT sites in dry seasons whereas no statistical difference may been found between 606 the two sites in wet seasons. Such a result is mainly explained by figures 5. In wet seasons, similar backtrajectory pattern may be observed for both sites whereas in dry seasons, CT traffic 607 site only would be influenced by Nigerian anthropogenic sources. 608

In terms of WSOC concentrations, concentrations at the AT site are on average higher than 609 those recorded at the CT site in the wet season, but lower in dry season. The presence of dust 610 can produce semi-volatile organic gas scavenging and therefore WSOC and OC enhancement. 611 Such a phenomenon can explain the highest WSOC concentrations observed in dry season at 612 the CT site where dust concentrations are highest (see dust paragraph). Moreover, this can also 613 explain why the maximum WSOC are in coarse particles at CT, while at AT maximum values 614 are in ultra-fine particles.

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- Total WSI concentrations are larger at AT site in the wet than in the dry season with higher 616 617 values in coarse particles. At CT site, total WSI concentrations in fine particles are higher in the dry than in the wet season whereas same values are obtained in coarse particles for both 618 seasons. Note that CT values are generally higher than AT values with a more important 619 contribution of fine particles in the dry season. These WSI variations can be explained by the 620 relative importance of  $Ca^{2+}$ ,  $SO_4^{2-}$  and  $NO_3^{-}$  in both sites. 621
- First, Ca<sup>2+</sup> contribution to total WSI is higher in CT site than in AT site with no clear seasonal 622 623 variation at CT site and higher values in dry season than in wet season at AT site. Also at CT, fine and coarse  $Ca^{2+}$  particles are in the same range, whereas coarse  $Ca^{2+}$  particles are 624 625 predominant at AT site. Such feature may be explained by the impact of dust sources including long-range dust transport at Abidjan and a combination of long-range dust transport and road 626 resuspension at Cotonou. 627
- Second, the relative contribution of  $SO_4^{2^-}$ ,  $NH_4^+$  and  $NO_3^-$  as a percentage of total WSI in the 628 different particle modes is reduced in the wet season. During the wet season, the clean winds 629 surrounding the ocean before reaching the measurement sites could contribute to lower the 630 proportion of these species, in addition to the scavenging processes during the rainy days. 631 Unlike the wet season, a relatively good correlation with  $r^2$  of 0.87 (SO<sub>4</sub><sup>2-</sup> versus NH<sub>4</sub><sup>+</sup>), 0.73 632  $(NO_3^- \text{ versus } NH_4^+)$  and 0.87  $(SO_4^{2^-} \text{ versus } NO_3^-)$  has been found in coarse particles, indicating 633 similar sources for these three species during the dry season. In order to try to identify these 634 sources, the ratio of  $SO_4^{2-}/Ca^{2+}$  and  $NO_3^{-}/Ca^{2+}$  has been determined. The average  $SO_4^{2-}/Ca^{2+}$ 635 and  $NO_3/Ca^{2+}$  ratios in combined coarse particles (1.07 and 2.58 during the wet season and 0.33 636 and 1.60 during the dry season) are higher than the corresponding ratios for typical soil (0.026 637 and 0.003, respectively). On the other hand, the  $SO_4^{2-}/Ca^{2+}$  ratio increases in the fine particles 638 (5.07 during the wet season and 2.53 during the dry season), while that of  $NO_3^{-}/Ca^{2+}$  remains 639 almost constant (2.86 during the wet season and 1.65 during the dry season). This implies that 640 the atmosphere at AT and CT sites is enriched by  $SO_4^{2-}$  formed as anthropogenic secondary 641 particles, possibly from sulfur containing pollution sources (Seinfield and Pandis, 1998), 642

particularly in fine particle mode, and by NO3<sup>-</sup> mostly coming from nitrogen containing sources 643 in all particle sizes. The higher contributions of these elements during the dry season could 644 result from a combination of several factors: 1) an atmosphere loaded with dust favoring 645 heterogeneous chemistry to obtain secondary aerosol and the rise of biomass burning emissions; 646 2) the increase of photochemical activity and higher concentrations of hydroxyl radicals in the 647 dry season, which can oxidize SO<sub>2</sub> from combustion (Arndt et al., 1997) to SO<sub>4<sup>2-</sup></sub> (Li et al., 648 2014); and 3) the wind transport of anthropogenic secondary particles from the industrial zone 649 located upstream from our sites. Finally, the proportion of Cl<sup>-</sup> relative to the total mass of ions 650 651 is highest for coarse particles at both traffic sites especially during the wet season, suggesting that Cl<sup>-</sup> at AT and CT sites if from natural origin and probably from sea salt emissions. 652

653 If we focus now on dust during the two wet seasons, concentrations are higher in 2016 than in 2015 at CT and AT sites for coarse particles (no data of fine particles are available in WS2016). 654 655 This is consistent with observed aerosol optical depth (AOD) values at CT, which increased by a factor of 2 between 2015 and 2016. No AOD value is given by Léon et al. (2019) at Abidjan 656 657 in WS2015 to allow such comparison in Abidjan. Moreover, during the wet season, an Angström coefficient (AE) on the order of 1 has been found at CT site, indicating smaller 658 659 particles that could be due to road resuspension. It is interesting to note that during WS2016, AOD and AE are respectively higher and lower at Abidjan than at Cotonou. Again, this is 660 consistent with our dust concentrations at CT site. In Abidjan, we could assume that another 661 source of  $Ca^{2+}$ , which is not taken into account in our dust calculations, may explain our dust 662 concentration data. That may be the result of anthropogenic Ca<sup>2+</sup> emissions from residential 663 in 2016 than in 2015 more important as shown earlier 664 combustion, (http://naei.beis.gov.uk/overview/pollutants?pollutant\_id=84). 665

The relative contribution of dust generally peaks in the coarse mode and, to a lesser extent, in the fine mode, reflecting their natural origin. It is interesting to note that the dust contribution observed in this study for the year 2016 at the Abidjan site is in agreement with the results of Xu et al. (2019) which show a PM2.5 dust contribution of 35-50% compared to our values of 18-52%.

671

#### 672 **4.3. Abidjan Waste Burning site**

Concentrations measured at AWB site are slightly lower than values found in the other sites.
This can be explained by the larger distance of the site to the main studied source (here waste
burning source) than in the other sites. However, PM2.5 values are also higher than WHO
guidelines. +

Aerosol mass, EC and OC concentrations are higher in dry than in wet season, which suggests 677 678 less waste burning activities during the wet season or impacts of other local anthropogenic sources or long-range biomass burning sources. Highest values are found in DS2017 with the 679 lowest OC/EC ratio, as at AT site. OC/EC ratio is highly variable at AWB (1-10) which 680 confirms that AWB site may be impacted by different types of sources as well as by secondary 681 aerosol organic formation which can be detected for OC/EC higher than 2 (Turpin et al. 1990; 682 Hildermann et al. 1991; Chow et al. 1996). Note that OC/EC typical for waste burning source 683 684 is of the order of 8 (Keita et al., 2018).

- 685 It is also observed that at the AWB site, PM mass concentrations are mainly distributed in C 686 mode (30-44%) over the entire period of study, excepted during the WS2015, and to a lesser 687 extend in F mode (21-44%). EC and OC being mainly distributed in C and UF modes. Watersoluble fraction of organic carbon is important (32%) and on the order of the one found at AT 688 689 site. Same for WSI concentrations and WSI composition. At AWB, WSI values are globally slightly higher in wet than in dry season. However, it is interesting to underline that  $Ca^{2+}$  is 690 691 much higher in dry season than in wet season, especially in DS2017. This is in agreement with 692 dust concentrations and trace element concentrations, which have been found to be maximum 693 at AWB, reaching 35.8% of the total PM mass in the dry season. These maximum percentages 694 are due to the large contribution of both Al and Na crustal elements which account for about 26%. Also note that Cu/Sb of 0.08 in DS2017, which indicate an influence of re-suspended 695 particles. A Zn/Cd value of 56 is obtained for the AWB site which is in close agreement with 696 values reported for oil burning (Watson et al., 2001, Samara et al., 2003). That could indicate 697 that oil might be one of the waste burning materials. 698
- 699 Our result suggests that AWB aerosol mass is influenced by a mix of sources, including fuel 700 combustion and mineral salt from sources around the measurement site, associated to long-701 range source impact of dust and biomass burning which will be further discussed in the next 702 paragraph.
- 703

#### 704 4.4. Interannual variability of aerosols in Abidjan and Cotonou

EC and OC concentrations are generally higher in DS2017 than in DS2016 for all the sites. This is not due to the meteorological condition, which is similar in both years. This is also not due to biomass burning impacts. Indeed, when looking at MODIS burnt areas for our period of study (http://www.aeris-data.fr/redirect/MODIS-MCD64A1), burnt areas of west African savannas are higher in 2016 than in 2017. Therefore, carbonaceous aerosol concentrations should be higher in 2016. Then, this could be due to a counter effect between biomass burning emission

strength and air mass transport efficiency. As a result, biomass burning impact could not explain 711 712 the difference in EC and OC during the dry season between 2016 and 2017. Rather, this is due to the variability of local sources. In DS2016 in Abidjan, there was a general strike of civil 713 714 servants of the State with important consequences on urban activities. Lower activities were observed (lower fish smoking emissions, lower traffic ...) in DS2016 compared to DS2017, thus 715 explaining the lower EC and OC concentrations at Abidjan sites. In Cotonou, highest 716 carbonaceous aerosol values in DS2017 may be explained by backtrajectory patterns: Cotonou 717 would be impacted by air masses coming from the high polluted Lagos (Nigeria) area in that 718 719 period whereas from less polluted northern areas in DS2016. Such an assumption is validated by the AOD values at 550nm from MODIS satellite images (http://www.aeris-720 721 data.fr/redirect/MODIS-MCD64A1), which show very high particulate concentrations in the 722 Guinean Golf (Figure 14).

This figure also shows the AOD difference between Cotonou and Abidjan for DS2017, with higher values at Cotonou than in Abidjan for the campaign period, in agreement with our measurements of aerosol mass, EC, OC and dust. This is confirmed by the DACCIWA sunphotometer AOD and Angström coefficient (AE) measurements at Abidjan and Cotonou (Léon et al., 2019; Djossou et al., 2018). Indeed, in DS2017, during our period of measurements, mean AOD in Cotonou is of the order of 1.3 versus 0.9 in Abidjan for an AE of 0.6 for both sites, which clearly indicates the presence of coarse dust particles.

Finally, aerosol mass and dust concentrations have been seen to be higher in DS2016 than in DS2017 in Abidjan whereas values are on the same order of magnitude at Cotonou. Such high values at Abidjan in DS2016 can be explained by the back-trajectory pattern with air-masses all coming from northern dusty areas in DS2016 (Bodélé depression in Tchad, Prospero et al. (2002), Washington et al., (2003), Knippertz et al. (2011), Balarabe et al., (2016)) and/or from northern dusty countries (Mali, Niger) (Ozer, 2005), whereas in DS2017, contribution of southern marine clean air-masses may also be noted.

In the wet season, aerosol mass, EC and OC are higher in WS2015 than in WS2016. This may
be due to particulate wet deposition, more efficient in WS2016 which have been seen earlier to
be more rainy (4.7mm) than in WS2015 (2mm). Moreover, at AT site, dust concentrations are
higher for coarse particles in WS2016 than in WS2015. Such variations may be explained by
long-range dust sources and/or road dust resuspension processes. As no dust event has been
noticed, local source explanation seems to be more evident.
In AT, CT and AWB, OC/EC ratios are globally on the same order for WS2015, WS2016 and

DS2016, with values lower than for DS2017. This could be due to lower traffic activities linked

to the DS2016 strike and the wet season vacation periods. Indeed, much higher OC/EC ratios
measured in DS2017 are typical of those of diesel vehicles (Mmari et al., 2013; Keita et al.,
2018). Finally, it is interesting to note that OC/EC ratios measured in this study are in the range
of those previously reported for other megacities such as Agra in India with 6.7 (Pachauri et al.,
2013), Helsinki in Finland with 2.7 (Viidanoja, 2002), Cairo in Egypt with 2.9 (Favez, 2008),

- Paris in France with 3.5 (Favez, 2008), and Milan in Italy with 6.6 (Lonati et al., 2007).
- 751

# 752 **4.5. Comparison with literature data**

Firstly, the comparison between our data and other DACCIWA results including other time 753 sampling focuses on PM2.5 levels, since these particle sizes are relevant for health impact 754 755 studies (Xing et al., 2016). In addition to our values, Figure 7 presents data from Xu et al. (2019) using personal samplers collected in the same area and at the same dates in 2016 during 12h on 756 757 women at the ADF site, students at the AWB site and drivers at the CT site, and from Djossou et al. (2018) study based on one week exposed filters collected at the same areas and for the 758 same periods as this study. We note that PM2.5 directly measured on women are 2.3 and 0.9 759 times our values obtained at the ADF site in dry and wet seasons, respectively, and 3.4 and 4.9 760 times higher on students than at the AWB site, and 1.6 and 2.1 times higher on drivers than at 761 the CT site. Also, our values are on average 1.6, 3, 5 and 8 times higher than weekly-integrated 762 values of Djossou et al. (2018) including our 3 days of measurements at the AWB, ADF, AT 763 764 and CT sites respectively. As it may be seen, the lowest concentrations are observed in Djossou 765 et al. (2018), whereas the highest concentrations are recorded in Xu. et al. (2019). This is valid 766 for all sites, seasons and campaigns. Differences between our values and Djossou values may be explained by the sampling times of the two studies. Indeed, as recalled, Djossou 767 768 measurements are weekly integrated, taking into account diurnal activities during all the week, including week-end and nights which have expected lower PM2.5 concentrations. Our study 769 770 includes only maximum pollution conditions for each site. The highest differences occur for 771 the traffic sites. This may be clearly understood since diurnal and weekly variations of traffic 772 sources are the most variable. Comparison between our values and Xu et al. (2019) values is 773 also interesting. Indeed, it is at the ADF site that on-site and women PM2.5 concentrations are 774 the closest, which shows that this site is the most representative of the pollution exposure to women. The biggest differences are found at the AWB site. As already mentioned, distance 775 from the site to the waste burning source is more important than for other sites, which explains 776 why concentrations obtained on students who are leaving close to the sources are much higher 777 778 than on-site concentrations. At the Cotonou traffic site, measurements taken from people are

also higher than on site measurements. Such differences can be explained by additional 779 780 pollution exposure as people move around. Note that the sampling technique may also play a role in such a comparison. In terms of seasonal variation, our results are in agreement with long-781 term EC measurements obtained by Djossou et al. (2018) for the same sites and period. Finally, 782 Table 4 compares our mean PM2.5 results obtained from 3-hour sampling for three consecutive 783 days to literature data for different traffic sites in the world given at a daily scale. It is interesting 784 to note that our values are situated at the higher end of the range of PM2.5 data observed from 785 786 the other sites.

787 Secondly, Table 5 compares our OC and EC values to those obtained by Djossou et al. (2018) and Xu et al. (2019) as previously described for the same period and the same sites. Again, it is 788 789 interesting to note that Djossou's values are in general lower than ours. Indeed, for the wet and 790 dry seasons, our OC measurements are 4 and 1.4 times higher than Djoussou's at the AT site, 791 2.1 and 5.7 times higher at the CT site, and 2.5 and 2.5 times higher at the ADF site, respectively. As for PM2.5, this can be explained by the different sampling times between our 792 793 experiments that were performed at the peak of urban activities, while Djossou's dataset represents weekly integrated values. Differences at the ADF site are largely explained by the 794 795 temporal pattern of fish smoking activities which take place every day, only in the morning, as 796 such the associated pollution is not well represented in the weekly sampling. Finally, there are 797 less differences at the AWB site between both datasets. As above explained, there are no marked temporal variations of concentrations at AWB site. The predominant waste burning emissions 798 impacting our site can occur night and day, week-days and week-end since origin of such 799 burning can be either anthropogenic or from spontaneous combustion. It may be also recalled 800 that another reason of agreement between both datasets may come from the large distance 801 between the site and the local and regional sources. Comparisons made between our values and 802 those of Xu's personal data show that both OC and EC are of the same order at the ADF site, 803 804 whereas Xu values are higher than ours at the CT and AWB sites. This result is in agreement with what we found with PM2.5 concentrations as detailed above. Finally, Table 6 presents OC 805 806 and EC for the PM2.5 comparison between our values and other recent studies dealing with traffic sites in other regions of the world and with similar operational conditions. We find that 807 808 our values are situated in the middle of the range observed in these different studies. Briefly, as presented in Table 7, it is interesting to compare our WSOC concentrations to literature data 809 for different traffic sites of the world. We note that our values are on the same order as values 810 found in Asia and higher than those found in Europe. 811

- 812 Thirdly, the percentages of the total WSI to PM mass (15-20%) at the three Abidjan sites (ADF,
- AWB and AT) are in the same order of magnitude than the data from PM2.5 personal exposure
- samples collected at the same locations in 2016 by Xu et al. (2019). Our results also are very
- close to the ionic contribution of 9% of the PM10 mass found at the urban curbside site in Dar
- es Salaam in Tanzania during the wet season 2005 by Mkoma (2008).
- 817

## 818 **5. Conclusion**

- This paper presents the mass and the size-speciated chemical composition of particulate matter (PM) obtained during the dry and wet seasons in 2015, 2016 and 2017. During each campaign, 3-hour sampling at the peak period of pollution for three consecutive days was performed at three sites in Abidjan, representative of domestic fire (ADF), waste burning (AWB) and traffic (AT) sources, and at one traffic site in Cotonou (CT).
- It is important to underline that our results and their temporal variations are very sensitive to (1) the source activities whose pollution levels are highly linked to socio-economic status of each city; (2) the impact of imported pollution (sea-salt, biomass burning, dust, anthropogenic emissions from neighboring countries), according to air mass origins; and (3) the particle wet deposition.
- The comparison between our results and other DACCIWA measurements underlines the importance of the distance of the chosen site to the sources. At the source level (such as ADF), pollution results at the site are in agreement with exposure of people living at this site. However, at the other sites, comparison is more difficult since the sites are under the influence of a mix of transported sources. That shows the key importance of exposure studies to estimate air quality and health impacts. That shows also the need of long-term studies to really understand role of imported sources in urban air quality.
- The main striking feature is that PM2.5 values are well above the annual and daily WHO 836 guidelines of 25 and 10 µg.m<sup>-3</sup>, respectively, whatever the site and the season. Also, measured 837 concentrations from this study are situated in the middle to the high part of the range of 838 839 worldwide urban aerosol concentrations given at a daily scale. In addition, we have stressed the importance of ultra-fine and fine particles in the studied aerosol and of species such as 840 841 particulate organic matter and water soluble organic carbon, which are well known to be particularly harmful. This is again a warning signal for pollution levels in African capitals if 842 843 nothing is done to reduce emissions in the future.
- 844 Our study constitutes an original database to characterize urban air pollution from 845 specific African combustion sources. The next step will be to cross such an exhaustive aerosol

- 846 chemical characterization to biological data in order to evaluate the impact of aerosol size and
- 847 chemical composition on aerosol inflammatory properties.
- 848

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# 855 Author Contributions

J.A. and C.L. conceived and designed the study. J.A., C.L. and E.T.D. contributed to the
literature search, data analysis/interpretation and manuscript writing. J.A., C.L., A.B. and
E.T.D. contributed to manuscript revision. J.A., C.L., J.F.L, H.C, V.Y., A.A, C.G, C.Z, E.C and
S.K. carried out the particulate samples collection and chemical experiments, analyzed the
experimental data.

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# 862 Additional Information

Figure S1 and Appendix A-D accompany this manuscript can be found inSupplementaryInformation.

865

### 866 Competing financial interests

- 867 The authors declare no competing financial interests
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Figure 2: Map of the city of Cotonou reporting the geographical location of DACCIWA urbansampling site.

- Figure 3: Pictures of the different sampling sites: (a) Traffic in Cotonou (Benin, CT station),
  (b) Waste burning in Abidjan (Côte d'Ivoire, AWB station), (c), Domestic fire, showing
  smoking activity in Yopougon, Abidjan (Côte d'Ivoire, ADF station), (d) " woro-woro
  and Gbaka " traffic in Abidjan (Côte d'Ivoire, AT station).
- Figure 4: Wind, pressure and temperature diagram at Abidjan and Cotonou during the differentcampaigns.
- Figure 5: Back trajectories arriving at Abidjan (a) and Cotonou (b) for each season (WS2015,
  WS2016, DS2016 and DS2017).

Figure 6: Aerosol Mass concentrations at the different study sites for each campaign and for the
different sizes (C in black, Fine in light Grey, Ultra-fine in grey). Bulk aerosol mass is
indicated in boxes.

- Figure 7: Comparison of PM2.5 mass concentrations in μg.m<sup>-3</sup> at the four sites with those
  obtained by Djossou et al. (2018) and Xu et al. (2019) for the same sites and periods.
  Data for the following weeks were then selected in Djossou et al. (2018): 20-27/07/2015
  for WS15, 4-11/01/2016 for DS16, 4-11/07/2016 for WS16 and 9-16/01/2017 for DS17.
- Figure 8: EC relative concentrations in each size classes (C in black, Fine in light grey, Ultrafine in grey) at the different study sites for each campaign. Bulk EC concentration for
  each site is indicated in boxes.
- Figure 9: OC relative concentrations in each size classes (C in black, Fine in light grey, Ultrafine in grey) at the different study sites for each campaign. Bulk OC concentration for
  each site is indicated in boxes.

Figure 10: OC/ EC ratio for the different campaigns and sites for each aerosol size (C in black,
Fine in light grey, Ultra-fine in grey). Each box shows the median and the first and the
third quartiles.

- Figure 11: Water-soluble ionic species speciation for each site, each campaign and each aerosolsize.
- Figure 12: Dust concentrations at the different study sites for each campaign and for thedifferent sizes (C in black, Fine in light grey, Ultra-fine in grey).

	U		1 0
1285	ea	each aerosol size.	
1286	Figure 14	4: MODIS Aerosol optical depth regional distribution over West Africa. D	ata are for
1287	20	2017, focusing on our campaign date at Abidjan (a-c 01/11-12-left part) and	d Cotonou
1288	(t	b-d 01/6-7, right part).	
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ABIDJAN DOMESTIC FIRES

(ADF)

Abidjan cityMeasurement sites

0 75 150 km 



- 1299 Figure1



- **Figure 2**



# **Figure 3**





1344 Figure 5-a



- 1346 Figure 5-b











- 1362 Figure 8

- . . . .



- 1369 Figure 9







1380 Figure 11



1384 Figure 12

WSI POM EC Dust n.d.



1390



- 1393 Figure 14

1403 List of table

- 1405 Table 1: Comparison of dust concentrations obtained from different methodologies
- 1406 Table 2: WSOC concentrations ( $\mu$ g.m<sup>-3</sup>) and WSOC/OC ratios (%) for each site, each campaign and
- 1407 each aerosol size
- 1408 Table 3: Trace element concentrations for bulk aerosol for each site and for DS2017 and WS2016.
- Table 4: Comparison of PM2.5 concentrations with literature data. Only literature data given at a dailyscale have been selected.
- Table 5: PM2.5-EC and PM2.5-OC comparison with Djossou et al. (2018) and Xu et al. (2019) values.
  Units are μgC.m<sup>-3</sup>
- 1413 Table 6: EC and OC comparison with literature values. Only literature data given at a daily scale have1414 been selected.
- Table 7: Comparison of WSOC concentrations with literature data. Only literature data given at a dailyscale have been selected.

Dry 2017		Sciare et	Guinot et al.	Terzi et al.
		al. (2005)	(2007)	(2010)
ADF	С	18.5	11.2	86.9
	F	9.3	29.7	22.2
	bulk	27.7	40.9	109.1
AWB	С	12.3	48.5	126.4
	F	5.2	39.7	106.4
	bulk	17.6	88.2	232.8
AT	С	16.4	24.8	98.5
	F	4.3	13.4	34.2
	bulk	20.7	38.2	132.7
СТ	С	37.9	70.0	98.4
	F	23.4	45.6	55.8
	bulk	61.3	115.6	154.2

#### 1440 Table 1: Comparison of dust concentrations obtained from different methodologies in $\mu$ g.m<sup>-3</sup>

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Wet 2016		Sciare et Guinot et al.,		Terzi et
		al., 2005	2007	al.(2010)
ADF	С	35.34	21.5	27.9
AWB	С	13.46	8.6	21.1
AT	С	19.65	37.5	21.4
СТ	С	42.98	26.2	52.5
	Bulk	42.98	26.2	52.5

#### 1451 each aerosol size

Site		Site Abidjan Waste Burning		Abidjan Domestic Fire	
Period	Size	WSOC	WSOC/OC	WSOC	WSOC/OC
Wet season	Coarse	1.3	24,6	8.2	32.5
2015	Fine	0.7	19.9	12.8	22.7
	Ultra fine	4.1	43.6	51.3	72.5
	PM2.5	5.5	33.7	69.5	47.2
Dry season	Coarse	0.4	12.3	4.4	18.8
2016	Fine	0.9	46.9	7.0	20.4
	Ultra fine	1.5	38.4	21.9	61.5
	PM2.5	2.7	32.7	31.0	32.0
Wet season	Coarse	1.3	42.5	16.5	44.3
2016	Fine	0.8	26.3	17.1	33.0
	Ultra fine	2.0	41.2	79.7	84.5
	PM2.5	3.5	37.1	106.0	52.0
Dry season	Coarse	1.9	32.9	12.1	36.0
2017	Fine	1.4	38.4	19.9	35.0
	Ultra fine	1.6	11.5	38.6	19.0
	PM2.5	4.0	30.0	65.8	29.0

<sup>1452</sup> 

- 1453
- 1454 Table 2 (suite): WSOC concentrations (µg.m<sup>-3</sup>) and WSOC/OC ratios (%) for each site, each campaign
- 1455 and each aerosol size

Si	ite	Abidja	an Traffic	Cotone	ou Traffic
Period	Size	WSOC	WSOC/OC	WSOC	WSOC/OC
Wet season	Coarse	2.4	39.6	1.1	23.3
2015	Fine	1.3	46.7	0.5	22.1
	Ultra fine	4.7	29.0	0.4	12.7
	PM2.5	6.9	34.0	2.2	18.0
Dry season	Coarse	1.4	43.0	2.3	64.1
2016	Fine	1.9	59.0	0.6	10.5
	Ultra fine	4.9	62.0	6.3	42.9
	PM2.5	7.5	49.4	8.0	29.0
Wet season	Coarse	1.1	23.1	1.2	34.7
2016	Fine	0.5	16.8	0.5	32.2
	Ultra fine	1.4	34.8	0.9	23.0
	PM2.5	2.4	26.0	1.9	28.0
Dry season	Coarse	0.9	24.0	3.5	37.8
2017	Fine	0.3	24.3	2.4	39.6
	Ultra fine	1.8	14.8	1.9	10.4
	PM2.5	2.6	16.0	6.0	18.2

#### Bulk ng.m<sup>-3</sup> (%) DRY 2017 WET 2016 ADF AWB AT СТ ADF AWB СТ AT Al 10050.8 (1.8) 25186.1 (13.7) 14015.8 (12.26) 15480.4 (5.7) 1370.5 (0.4) 1990.1 (3.5) 2191.4 (2.4) 4010.5 (4.4) Κ 8634.3 (1.5) 6093.7 (3.3) 3677.7 (3.22) 5068.9 (1.9) 1105.0 (0.3) 472.0 (0.8) 275.9 (0.3) 1076.0 (1.2) 2070.6 (0.6) Na 6847.8 (1.2) 23430.5 (12.8) 15372.1 (13.44) 11529.3 (4.3) 3735.4 (6.6) 2861.5 (3.1) 5310.2 (5.8) Ca 4321.2 (0.8) 2923.7 (1.6) 4117.6 (3.60) 6233.5 (2.3) 4124.7 (1.1) 447.5 (0.8) 374.7 (0.4) 4954.02 (5.4 Mg 1940.6 (0.3) 384.0 (0.2) 410.3 (0.36) 823.2 (0.3) 1524.7 (0.4) 294.9 (0.5) 283.5 (0.3) 619.2 (0.7) 3807.9 (2.1) 709.3 (1.3) 987.3 (1.1) Fe 1709.9 (0.3) 1628.1 (1.42) 3406.8 (1.3) 1314.0 (0.4) 1549.4 (1.7) Р 1521.9 (0.3) 696.0 (0.4) 147.8 (0.13) 207.4 (0.1) 605.4 (0.2) 13.2 8.6 81.4 (0.1) Ti 457.9 (0.17) 170.8 (0.05) 75.7 (0.13) 96.8 (0.11) 488.9 (0.1) 2270.3 (1.2) 282.8 (0.25) 154.7 (0.17) Zn 189.7 (0.03) 80.9 (0.04) 57.9 (0.05) 149.4 (0.06) 60.3 (0.02) 1.9 41.1 (0.04) 36.2 (0.04) Zr 217.9 (0.19) 145.3 (0.05) 22.4 (0.04) 36.7 (0.04) 31.2 (0.03) 172.1 (0.03) 390.3 (0.21) Pb 87.1 (0.02) 11.0 (0.01) 4.8 11.5 8.3 2.1 2.3 9.3 (0.01) 37.4 (0.01) 0.77 0.09 0.0006 Sn 79.7 (0.01) 38.4 (0.02) 21.6 (0.02) 9.ç (0.01) Mn 74.2 (0.01) 35.2 (0.02) 33.7 (0.03) 160.6 (0.06) 48.9 (0.01) 12.01 (0.02) 9.1 (0.01) 41.41 (0.05) Rb 52.4 (0.01) 8.7 5.9 (0.01) 8.5 4.47 0.71 0.85 1.9 Sb 59.9 (0.01) 201.2 (0.11) 123.6 (0.11) 149.04 (0.06) 24.4 (0.01) 0 0.0006 2.9 Ba 37.3 (0.01) 53.3 (0.03) 47.4 (0.04) 65.8 (0.02) 18.5 8.02 (0.01) 9.9 (0.01) 32.0 (0.03) Ni 36.5 (0.01) 34.5 (0.02) 27.9 (0.02) 50.2 (0.02) 18.00) 33.1 (0.06) 9.7 (0.01) 14.9 (0.02) Cr 29.4 (0.01) 53.8 (0.03) 35.8 (0.03) 28.6 (0.01) 41.9 (0.01) 47.7 (0.08) 24.3 (0.03) 29.7 (0.03) 21.2 (0.02) 28.1 (0.01) 34.02 (0.01) 17.02 0 0.19 8.1 (0.01) Sr 15.5 (0.01) Cu 24.0 12.3 (0.01) 3.6 9.6 3.99 0.26 0.87 2.8) 0.22 Sr 12.6 17.1 8.9 (0.01) 7.8 (0.01) 0.23 Li 7.3 15.5 (0.01) 7.39 0.36 0.32 0.75 Cd 6.1 0.83 1.18 0.05 0.02 0.17 1.6 1.012.4 (0.01) v 5.5 5.1 10.62 2.14 1.84 2.0 3.35 Mo 5.5 8.0 4.9 3.19 4.56 6.84 (0.01) 2.04 3.2 Cs 5.4 0.9 1.2 0.94 0.11 0.12 0.01 0.17 0.97 Hf 4.5 10.8 (0.01) 6.8 (0.01) 4.63 0 0.67 1.03 4.2 0 0 (0) 0.05 0.60 4.5 1.22 As 3.1 Li 4.0 9.8 5.9 (0.01) 5.82 0.27 0.37 0.16 0.93 0.13 35.67 (0.01) 0.49 0.33 Co 3.8 1.1 2.1 0.86 3.7 6.0 (0.01) 9.85 1.06 0.50 0.42 2.03 Ce 6.8 0.92 2.9 4.78 0.54 0.24 0.25 La 1.8 3.5 0.98 0.5 0.63 Nb 1.5 2.6) 1.4 2.48 0.46 0.05 0.40 1.5 2.5 2.4 4.15 0(0)Nd 0.02 Sc 0.69 1.4 1.1) 1.31 0,00 0.03 0.13 0.28 0.003 Be 0.19 0.2 0.37 Ga 0.61 1.15 0.8 0.98 0.2 0.11 0.12 0.11 Ge 0.42 1.02 0.8 0.68 0.01 0.07 0.02 0.91 0.02 0.59 0.20 0.18 Se Rh 0.02 0.02 0.00002 0.002 0.002 0.0002 0.05 Te 0.06 0.08 0.08 0.02 0.02 0.02 0,00 Pr 0.40 0.74 0.7 1.13 0.06 0.01 0.004 0.15 0.27 0.46 0.45 0.76 0.01 0,00 0.07 Sm 0.05 0.08 0.01 0.003 0.0 Eu 0.08 0.15 0.05 Gd 0.31 0.57 0.54 0.86 0.02 0.001 0.15 0.04 0.07 0.07 0.005 0.02 Tb 0.10 0.24 0.42 0.40 0.57 0.001 0.05 Dy Но 0.05 0.09 0.09 0.12 0.01 0.008 0.01 0.03

Er

0.16

0.31

0.28

0.35

0.02

0.03

0.02

0.09

#### 1456 Table 3: Trace element concentrations for bulk aerosol for each site and for DS2017 and WS2016.

Tm	0.02	0.05	0.05	0.05	0.002	0.003	0.001	0.01
Yb	0.18	0.38	0.32	0.34	0.01	0.02	0.03	0.09
Lu	0.03	0.06	0.08	0.06	0.003	0.005	0.005	0.02
Та	0.07	0.14	0.09	0.16	0.06	0.02	0.02	0.03
W	0.80	1.63	0.69	0.54	0.26	0.41	0.4	0.3
Tl	0.22	0.01	0.03	0.06		0.009		
Bi	0.32	0.26	0.02	0.08	0.06	-	-	0.08
Th	0.41	0.88	0.79	1.29	0.15	0.09	0.09	0.24
U	0.22	0.43	0.49	0.51	0.03	0.03	0.02	0.09
Total	36459.9	65817.6	40312.2	44159.2	12562.9	7874.7	7227.2	18001.1
Mass(µg/m <sup>3</sup> )	558.8	183.6	114.4	270.0	374.7	56.3	91.6	91.9

 Location	PM2.5	Reference
	(µg.m <sup>-5</sup> )	
Abidjan, Cote d Ivoire	142	This work
Cotonou, Benin	154	I his work
Beijing, China	81.4	Ale et al., 2019
Christchurch, New Zealand	9.2	Tunno et al., 2019
Pune, India	$98 \pm 28$	Pipal et al., 2019
Delhi, India	123	Guttikunda and Calori, 2013
Lahore, Pakistan	91	Colbeck et al., 2011
Ahvaz, Iran	69	Shahsavani et al., 2012
Hong Chong, Hong Kong	$54.7\pm25.6$	Cheng et al., 2015
Lecce, Italia	16	Cesari et al., 2016
Libreville, Gabon	35.8	Note at $a1 - 2010$
Port Gentille, Gabon	60.9	Ng0 et al., 2019
Kenitra, Morocco	51.3	Zghaid et al., 2009
Bilecik, Turkey	247	Gaga et al., 2018
Algiers, Algeria	34.8	Bouhila et al., 2015
Shobra, Egypt	216	Lowenthal et al.,2015

Table 4: Comparison of PM2.5 concentrations with literature data. Only literature data given at a dailyscale have been selected.

Table 5: PM2.5-EC and PM2.5-OC comparison with Djossou et al. (2018) and Xu et al. (2019) values.
Units are μgC.m<sup>-3</sup>.

Location	Period	PM2.5 OC	PM2.5 EC	References
	July 2015	$22.6\pm3.4$	$4.3\pm0.2$	
	January 2016	$15.2 \pm 5.3$	$7.0 \pm 2.6$	This Work
	July 2016	$9.3 \pm 1.3$	$2.2\pm0.1$	THIS WORK
	January 2017	$16.1 \pm 1.7$	$18.9 \pm 1.4$	
Traffic Abidian	July 2015	$3.3 \pm 0.2$	$2.3\pm0.2$	
Cote d'Ivoire	January 2016	$7.7 \pm 0.0$	$3.9\pm0.0$	Diosson et al. 2018
	July 2016	$7.6\pm0.2$	$4.9\pm0.0$	Dj08500 et al. 2018
-	January 2017	$19.1\pm6.2$	$13.9\pm5.5$	
	July 2015	$13.1 \pm 1.2$	$3.5 \pm 0.7$	
	January 2016	$27.8 \pm 11.3$	$10.9\pm2.6$	This Work
	July 2016	$6.7\pm1.9$	$2.0\pm0.5$	THIS WORK
	January 2017	$33.1\pm4.6$	$27.3\pm0.9$	
	July 2015	$4.2\pm0.7$	$1.5\pm0.1$	
Traffic Cotonou.	January 2016	$3.0\pm0.3$	$1.5 \pm 0.2$	51 1 6640
Benin	July 2016	$6.7 \pm 0.2$	$1.6 \pm 0.1$	Djossou et al. 2018
	January 2017	$14.5 \pm 0.8$	$4.4 \pm 0.7$	
	,			
	January 2016	$49.5 \pm 12.5$	$13.6\pm3.6$	Yu et al. 2010
	July 2016	$37.0\pm3.5$	$9.3\pm0.8$	Au et al. 2019
-	July 2015	$147.2\pm14.5$	$16.1 \pm 1.6$	
	January 2016	$56.5\pm51.5$	$7.4 \pm 3.1$	
	July 2016	$172.3 \pm 39.0$	$17.9 \pm 4.8$	This Work
Domestic fire	January 2017	$283.9\pm34.9$	$37.9\pm4.3$	
Cote d'Ivoire	July 2015	$80.5 \pm 1.1$	$32.2 \pm 1.6$	
	January 2016	$76.3 \pm 13.7$	$11.4 \pm 0.2$	
	July 2016	$68.4 \pm 16.5$	$17.4 \pm 2.1$	Djossou et al. 2018
	January 2017	$66.4 \pm 7.5$	21.1 ± 6.6	
	1 2017	72.4.24.6	10.5 . 7.2	
	January 2016	$72.4 \pm 24.6$	$19.5 \pm 7.3$	Xu et al. 2019
-	July 2016	189.3 ± 197.8	$11.5 \pm 10.8$	
	July 2015	$14.8 \pm 1.1$	$4.4 \pm 0.1$	
	January 2016	$7.7 \pm 1.3$	$3.0 \pm 0.3$	This Work
	July 2016	$10.0\pm2.4$	$1.5 \pm 0.3$	THIS WORK
	January 2017	$21.9\pm4.2$	$19.2\pm2.4$	
Waste Burning Abidian	July 2015	$3.7 \pm 2.2$	$4.3 \pm 0.3$	
Cote d'Ivoire	January 2016	$13.9\pm9.0$	$3.6 \pm 1.8$	
	July 2016	$9.8 \pm 4.4$	$2.8\pm0.9$	Djossou et al. 2018
	January 2017	$22.4\pm7.8$	$8.7\pm3.0$	
	January 2016 July 2016	$85 \pm 57.4$ $65.2 \pm 65.2$	$15 \pm 4.7$ $12.3 \pm 11.4$	Xu et al. 2019

- Table 6: EC and OC comparison with literature values. Only literature data given at a daily scale havebeen selected.

Location	OC (µg.m <sup>-3</sup> )	BC (µg.m <sup>-3</sup> )	Reference
Abidjan (Côte d'Ivoire)	16	8.1	This study
Cotonou (Benin)	20.2	11	This study
Bilecik (Turkey)	49.6-62.8	38.8-58.1	Gaga et al., 2018
Pune (India)	30	5	Pipal et al., 2019
Shanghai (China)	4.9-13.1	1.9-5	Ding et al., 2017
Lahore (Pakistan)	85.7-152	13.8-21	Stone et al., 2010
Agra (India)	25.4-70	3.3-9.5	Satsangi et al., 2012, Pipal et al., 2014
Delhi (India)	34.1-50	5.3-10.6	Bisht et al., 2015a, Pipal et al., 2014
Ahmedabad (India)	18.3	3	Rengarajan et al., 2011
Yokohama (Japan)	4	2	Khan et al., 2010
Beijing (China)	2.9-28.2	1.2-16.3	Guinot et al., 2007

- 1534 Table 7: Comparison of WSOC concentrations with literature data. Only literature data given at a daily
- 1535 scale have been selected.

Location	WSOC (µg.m <sup>·3</sup> )	Reference
Abidjan, Côte d'Ivoire	2-8	This work
Cotonou, Benin	2-8	This work
Beijing, China	9-27	Yu et al., 2018
Beijing, China	4-6	Xiang et al., 2017
Beijing, China	8-12	Tang et al., 2016
Beijing, China	7	Du et al., 2014
Beijing, China	6-8	Feng et al., 2006
Shanghai, China	2-7	Feng et al., 2006, Huang et al., 2012
Guangzhou, Hong Kong	2	Huang et al., 2012
Guangzhou, Hong Kong	5-10	Feng et al., 2006
Gwangju, Korea	2-3.5	Park et al., 2015
Tokyo, Japan	3-23	Sempere and Kawamura, 1994
Cairo, Egypt	3	Favez et al., 2008
Amsterdam, Netherland	1-2	Feng et al., 2007
Barcelone, Spain	1-2	Viana et al., 2007 and 2008
Brindisi, Italy	1.5	Genga et al., 2017
Saint Jean de Maurienne, France	1-5	Sullivan et al., 2004, Jaffrezo et al., 2005a

# Physico-chemical characterization of urban aerosols from specific combustion sources in West Africa at Abidjan in Côte d'Ivoire and Cotonou in Benin in the frame of the DACCIWA program

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#### 31 Abstract

32

DACCIWA (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) program, 33 intensive measurement campaigns were performed in Abidjan (Côte d'Ivoire) and Cotonou 34 (Benin), in dry (January 2016 and 2017) and wet (July 2015 and 2016) seasons, at different 35 sites chosen to be representative of African urban combustion sources (i.e. domestic fires 36 (ADF), traffic (AT) and waste burning (AWB) sources in Abidjan and traffic source in Cotonou 37 (CT)). Both the size distribution of particulate matter (PM) and their chemical composition 38 including elemental carbon (EC), organic carbon (OC), water-soluble organic carbon (WSOC), 39 water-soluble inorganic ions (WSI) and trace metals were examined. Results show very high 40 PM concentrations at all sites and a well-marked seasonality as well as a strong spatial variation. 41 42 The average PM<sub>2.5</sub> mass concentrations during the wet season are 517.3, 104.1, 90.3 and 69.1 ug.m<sup>-3</sup> at the ADF, CT, AT and AWB sites, respectively. In the dry season, PM<sub>2.5</sub> concentrations 43 decrease to 375.7 µg.m<sup>-3</sup> at the ADF site, while they increase to 269.7, 141.3 and 175.3 µg.m<sup>-3</sup> 44 at the CT, AT and AWB sites, respectively. The annual PM2.5 levels at almost all sites are 45 significantly higher than the WHO guideline level of 10 µg.m<sup>-3</sup>. As for PM mass, (EC) and 46 (OC) concentrations are also maximum at ADF site, accounting up to 69% of the total PM mass. 47 Such high content is mainly linked to wood burning for domestic cooking and commercial food 48 smoking activities. Dust contributions are dominant at CT (57-80%), AT (20-70%) and AWB 49 (30-69%) sites and specially in the coarse and fine particle modes at CT and in the coarse 50 fraction at AT site, which may be explained by the impact of long-range desert-dust transport 51 and re-suspended particles from the roads, in addition to anthropogenic sources. The 52 contributions of WSI to the total PM mass, mainly driven by chloride, nitrate and calcium in 53 the fine and/or large particles are highly variable according to the sites but remain less than 54 30%. Values are generally 1-3 times higher in the wet season than the dry season. This is due 55 to anthropogenic emissions but also to nitrate formation by reaction processes and natural 56 57 emissions. The concentrations of trace element well reflect the trends of dust at the traffic and AWB sites, with a predominance of Al, Na, Ca, Fe and K, keys markers of crustal dust. This 58 59 study constitutes an original database that characterizes specific African combustion sources.

60

61 **Keywords**: atmospheric pollution, chemical composition, physicochemical characterization,

62 Particulate matter, traffic, waste burning, domestic fires.

#### 63 **1.** Introduction

The impact of anthropogenic pollution on environment and health has been demonstrated by 64 numerous studies in Europe and North America, which have contributed to the implementation 65 of emission reduction policies. By contrast, air pollution in Africa is far from being well 66 characterized, although it is suspected to be responsible for negative health outcomes (WHO, 67 2014). This is a major problem since Africa is an intense emitter of pollution from 68 69 anthropogenic sources that includes domestic fires, vehicular traffic, waste burning as well as 70 growing oil and mining industries. It has also one of the fastest growing urban populations in the world, especially in West and East Africa (UN, 2019). As a consequence, it has been shown 71 that massive urbanization and rapid economic growth could be responsible for tripling 72 73 anthropogenic emissions in Africa between 2000 and 2030 (Liousse et al., 2014). Moreover, it is important to recall the impact of biomass burning and dust sources in the African atmospheric 74 75 composition, especially occurring during the dry season. All of this results in a major 76 degradation of urban air quality and an impact on the health of exposed populations. Only a few 77 studies on this subject have been conducted in West Africa (Val et al., 2013; Dieme et al., 2012; Kouassi et al., 2009) despite the high atmospheric pollutant concentrations already measured 78 79 to be of the same order as in Asian megacities and well above WHO international standards (WHO, 2014). 80

West Africa is then an "unique laboratory" to study urban pollution. Previous studies conducted 81 under the framework of the AMMA (Analyses Multidisciplinaires de la Mousson Africaine) 82 83 and POLCA (POLlution des Capitales Africaines) programs, have revealed very high average particulate mass concentrations in Cotonou (Benin), Bamako (Mali), Dakar (Senegal) and 84 Yaoundé (Cameroun) during the dry season (Doumbia et al., 2012; Val et al., 2013), suggesting 85 86 that the population may be affected by negative health outcomes. For example, during the dray season in Bamako (Mali) and Dakar (Senegal), Val et al. (2013) showed that the inflammatory 87 impact of combustion aerosol depends on the type of emission sources and determined the 88 predominant role of particulate organic matter. This is consistent with global findings showing 89 that fine and ultrafine aerosol fractions, as well as their content in trace metals and organic 90 compounds, induce biological effects due to their ability to reach the distal lung (Cassee et al., 91 92 2013). Such reasons highlight the need to better understand the size-speciation of aerosol chemical composition for the main West African anthropogenic sources during the different 93 94 seasons. Within this context, the DACCIWA (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) program, dedicated a specific work package to "Air Pollution and 95

Health" dealing with pollutant characterization related to health issues through toxicologicalstudies and epidemiological studies.

98 Campaigns have been organized from December 2014 to March 2017 in Abidjan and Cotonou. The strategy was to measure aerosol chemical composition in different sites, representative of 99 100 the main prevailing urban sources in West Africa following Liousse et al. (2014) and Keita et 101 al. (2018). Two typical traffic-sampling sites were chosen, one in Abidjan (Côte d'Ivoire) and 102 another one in Cotonou (Benin), to take into account differences in terms of fleets, type of fuel used and quality of roads. Indeed, in Cotonou, the majority of population uses two-wheel 103 vehicles susing gasoline fuel or gasoline and oil fuel, whereas in Abidjan, the vehicle fleet is 104 dominated by four-wheel engines using diesel fuel. Measurements were also performed at 105 domestic fire and waste burning sites, both located in Abidjan. 106

107 For a period of two years, PM<sub>2.5</sub> mass and carbonaceous aerosol were weekly measured and results are discussed in *Djossou et al. (2018)*. In this paper, we focus on the results from the 108 109 intensive campaigns. We present measurements obtained at each site during the wet and dry seasons of the studied periods: (i) PM size distribution and mass concentrations and (ii) PM 110 111 chemical composition including carbonaceous aerosol, water-soluble organic carbon, watersoluble inorganic ions, dust and trace elements in different size fractions. Experimental method 112 113 including description of sites, types of measurements and analyses, meteorological conditions, 114 will be presented in the section 2, whereas results and discussion are discussed in the sections 3 and 4 of the paper, respectively. 115

116

#### 117 **2. Experimental method**

#### 118

#### 2.1. Description of sites

Measurement campaigns have been performed in wet season (July 20-26, 2015 and July 4-13, 2016) and dry seasons (January 7-15, 2016 and January 5-14, 2017) at three sites in Abidjan (Côte d'Ivoire), representative of different sources, i.e. ADF for Abidjan Domestic Fires, AWB for Abidjan Waste Burning and AT for Abidjan Traffic (Figure 1), as well as one traffic site in Cotonou (Benin) (Figure 2).

As shown in Figure 3 which presents pictures of the different sampling sites, the ADF site (5°
19' 44 "N, 4° 06' 21" W) is situated on a platform, 5 m above ground level, in Yopougon

Bracody district near a market (Figure 1). This geographical area is highly populated with 126 various small commercial activities such as a fish and meat-smoking by women. There are also 127 many formal and informal settlements, which mainly use wood and charcoal as a source of fuel 128 129 for private and professional combustion activities. Other sources of concern contributing to the mix of pollutant emissions in the area include transportation-related emissions, biomass 130 burning, garbage bins or small landfills and various other fugitive sources. The AT site (5° 21' 131 14" N, 4° 01' 04" W) is located in Adjamé, on the roof of « 220 pharmacie logement building », 132 about 7 m above ground level and roughly 10 m away from the main road. This site, close to 133 134 the Adjamé market and to a bus station, is highly affected by traffic (Gbaka, bus, taxi, woroworo, private cars...). The AWB site (5° 21' 12" N, 3° 56' 16" W) is located at Akouédo in the 135 district of Cocody, on the roof of « Talafiguié », a building 15 m above ground level. This site 136 close to the big municipal landfill of Abidjan operational since 1965 and now covering an area 137 of 153 ha, is submitted to frequent waste burning pollution. The Cotonou Traffic (CT) (6° 22' 138 19" N, 2° 26' 5" E) site is located in Cotonou, on the «Sogema» building roof, about 7 m above 139 140 ground level. This site is close to the Dantokpa market and also to the biggest crossroad of Cotonou (intersection of 4 main roads). This site is highly influenced by intense traffic 141 142 activities. As previously mentioned, such a site is interesting because the vehicle fleet and fuels are different in Cotonou compared to Abidjan: (1) there are many two-wheel vehicles in 143 Cotonou whereas a few only in Abidjan; (2) in Cotonou, gasoline is of poor quality due to the 144 illegal fuel transport from Nigeria and (3) the roads are in worse conditions in Cotonou than in 145 Abidjan. 146

147 **2.2. Measurements** 

148 During each intensive campaign and on each site, two 3-hour samples collected with 149 cascade impactors operating in parallel are obtained for three consecutive days (i.e. six size-150 resolved samples per site during each campaign), to allow size-speciated characterization of the aerosol chemical composition. Note that the choice of the 3-hour periods is linked to the period 151 of maximum pollution for each site as shown by preliminary studies (e.g. morning at ADF site 152 (7-10am), afternoon at AT site (4-7pm, morning at CT site (7-10am)). There is no specific 153 period at AWB site since the activities are roughly the same during the day. The first impactor 154 with 4 stages (PM>2.5; PM 2.5-1; PM 1-0.2; PM<0.2) includes 4 quartz fiber filters (QMA, Whatman) 155 for mass and carbonaceous aerosol (EC, OC and WSOC analysis). The second impactor with 3 156 stages (PM>2.5; PM2.5-1; PM1-0.1) is equipped with three Teflon filters (Zefluor, Pall 157 Corporation®), dedicated for water-soluble ions species and trace elements. Due to operational 158

problems in july 2016, this second 3-stage cascade impactor is replaced by another 3-stage cascade impactor with different size cuts ( $PM_{>10}$ ,  $PM_{10-2.5}$ ,  $PM_{2.5-1}$ ). For consistency, results will be presented as an ultrafine (UF), fine (F) and coarse (C) classification. The two first stages ( $PM_{>2.5}$  and  $PM_{2.5-1}$ ) being considered as the coarse particulate fraction, the  $PM_{1-0.1}$  or  $PM_{1-0.2}$ stage, the fine particulate fraction and the  $PM_{<0.2}$  stage, the ultrafine fraction.

All the filters are prepared and analyzed at the Laboratoire d'Aerologie in Toulouse under different protocols described in the following paragraphs. Note that the quartz filters are prefired before sampling.

- 167 **2.3. Analyses**
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#### 2.3.1. Gravimetric analyses

Aerosol mass concentrations are obtained using a high-precision balance (SARTORIUS MC21S), placed under a controlled temperature and humidity atmosphere (Person and Tymen, 2005). Before weighing, the filters are kept 24 hours in the weighing room at an ambient relative humidity of  $30 \pm 15\%$ . The filters are weighed before and after sampling. Result of a gravimetric measurement consists of the average of 2 to 4 weighing whose differences do not exceed 5µg. The standard error on a gravimetric measurement is therefore less than 10 µg, typically representing less than 5% of the particles mass.

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#### 2.3.2. Carbonaceous aerosols

177 Carbonaceous aerosol is determined with thermal analysis with a two-step method
178 adapted from Cachier et al. (1989). Two aliquots of the same filter are separately analyzed.

One portion is directly analyzed for its total carbon content (TC). The other portion is first submitted to a pre-combustion step (2 h at 340°C under pure oxygen) in order to eliminate Organic Carbon (OC), and then analyzed for its Elemental Carbon (EC) content. Organic carbon (OC) concentrations are calculated as the differences between TC and EC. Note that the aerosol carbon content is quantified by a non-dispersive infrared (NDIR) detector with G4 ICARUS instrument with a detection limit of the order of 2  $\mu$ gC.cm<sup>-2</sup>. Uncertainty is in the order of 5% for TC, while being in the range of 5-20%, for EC and OC.

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#### 2.3.3 Water Soluble Organic Carbon analysis

187 WSOC measurements are performed using a total organic carbon analyzer (Sievers M9).
188 A detailed description of this technique is reported in Favez et al. (2008). Briefly, the full

oxidation of total organic carbon into CO<sub>2</sub> is obtained by coupling chemical oxidation (with 189 ammonium persulphate) and UV light. CO<sub>2</sub> is then quantified by conductivity. Analyses are 190 conducted on 20 ml of solution extracts. For UF samples, solutions to be analyzed are obtained 191 using a total filter surface of 3cm<sup>2</sup> (6x0.5 cm<sup>2</sup> punches symmetrically taken out of each QMA 192 filter), whereas, for C and F sizes, due to the geometry of the spots at the surface of the filters, 193 samples are divided into equivalent parts (1/2 or 1/4 of 47 mm filters, rest of the filters being 194 used for carbonaceous analysis). The extraction protocol consists of 16h soaking under soft 195 shaking in an Erlen-Meyer containing 20mL of ultra-pure water. Prior to WSOC analysis, water 196 197 extracts are filtered through Teflon (PTFE) filters (0.2µm pore size diameter) in order to remove any suspended particle. Measurement uncertainty, given by the manufacturer, is of the order of 198 7%. The overall calculated blank value is of the order of  $2.27 \pm 0.33 \,\mu gC.cm^{-2}$ , which represents 199 16.4± 8.5% of the mean WSOC content. For each sample, duplicate analyses show a good 200 201 reproducibility.

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#### 2.3.4. Water-soluble ionic species

Water-soluble ionic species (Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>) are 203 204 analyzed using ion chromatograph (IC), following the analytical protocol described in Adon et 205 al. (2010). Briefly, the aerosol water-soluble fraction is first extracted from half-sampled Teflon filter (the other part being used for trace element analysis), with a 10-min sonication in plastic 206 vials including 6 ml or 10 ml of purified water with a controlled resistivity of  $18.2M\Omega$ . Then 207 these vials are subjected to ionic chromatograph analysis or stored at +4°C if not analyzed 208 immediately. Cations are analyzed with Dionex DX-100 and anions with Dionex DX-500 with 209 a detection limit of 1 to 6 ppb depending on ionic species. Uncertainties in the range of 1-50% 210 is found depending on ionic species. 211

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#### 2.3.5. Trace elements

The protocol to measure trace element concentrations is developed and performed at the 213 Laboratory of Environmental Geosciences of Toulouse. Half-sampled Teflon filters (the other 214 part being used for water-soluble ionic species, see below) are mineralized by acid digestion 215 216 with a 10 ml concentrated HNO<sub>3</sub> and 0.5 ml HF solution (Lamaison, 2006) using a closed vessel microwave accelerated reaction system (MARS 5, CEM Corporation) at high pressure (700 psi) 217 (Celo et al. 2010). The digestion is realized in 3 steps: a rise in temperature at 130°C in 3min 218 and holding for 1 min, then, a second rise at 160°C in 1 min and holding for 30 seconds and 219 finally a third rise to 180°C in 1 min and holding for 3min. After a 12 h cooling period, the 220

solutions are evaporated at 80°C, and concentrated in 7 ml of 2% concentrated HNO<sub>3</sub> solution, before analysis by ICP-MS which are performed with a 7500 ce Agilent Technologies instrument equipped with a collision cell, and using In and Re as internal standards. The detection limit is less than 10 ppt. For all the samples, the final blank values and detection limit on filters are taken into account for final concentrations calculations. 13 trace metals are considered in this work: Al, Ti, Cr, Mn, Fe, Ni, Cu, Zn, Ba, La, Th, Pb and Cd.

227

#### 2.3.6. Dust calculation

Many methods can be used to quantify dust concentrations. We have selected three methods (Sciare et al. 2005, Guinot et al. 2007, Terzi et al. 2010) to underline the uncertainties linked to dust estimates.

- 231 (1) Sciare e
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(1) Sciare et al. (2005) method consists of using soluble calcium data obtained with Ionic Chromatography (IC), to estimate the dust concentrations following the relationship: dust = 10.96\*nss-Ca<sup>2+</sup>, where nss-Ca<sup>2+</sup> (=1.02\*Ca<sup>2+</sup>-0.038\*Na<sup>+</sup>) refers to non-sea-salt calcium concentration.

(2) Guinot et al. (2007) method is based on a chemical closure where fine and coarse 235 236 particle aerosols are separated in 4 components (EC, POM, WSI and dust). EC, WSI, and total aerosol mass are directly experimentally determined (see below paragraphs). 237 238 POM concentrations are obtained from OC concentrations experimentally determined and k, the OC/POM conversion factor. Dust concentrations are obtained from measured 239  $Ca^{2+}$  concentrations and f, the abundance of calcium in dust. The k and f values are 240 obtained from a linear regression (L) between the reconstructed and the weighed aerosol 241 mass concentrations. Briefly, first step consists of focusing on the aerosol coarse 242 fraction. k is fixed to 1.8 and as a result of (L) just mentioned, f is obtained to be in the 243 range of 0.012 to 0.15 depending on our sites. Second step deals with the aerosol fine 244 245 fraction. The f values just obtained for the aerosol coarse fraction are applied to the fine fraction and k ratios are estimated using (L) to be in the range of 1.2 to 2.1. Note that at 246 all of our sites, the correlation between  $Ca^{2+}$  and the missing mass between the weighed 247 and the reconstructed aerosol mass is sufficiently good  $(r^2=0.9)$  to support the 248 consistency of this simple approach for the evaluation of dust. Also, f and k values are 249 included in the range of values provided in the literature (He et al. 2001; Sun et al. 2004; 250 Guinot et al. 2007). However, it is important to mention that the range of f and k 251 coefficients are large which is due to the source mixing observed in this study. 252

(3) In Terzi et al. (2010) method, dust is obtained with the following relationship: dust = 1.89[Al] + 1.21[K] + 1.95[Ca] +1.66[Mg] + 1.7 [Ti] +2.14[Si] + 1.42[Fe]. In our study, all these elements are determined except Silica (Si). Consequently, we used mean Si values obtained from different relationships available in the literature (SiO<sub>2</sub> =  $3*Al_2O_3$ for Alastuey et al., 2005, Si = 4.0\*Al for Zhang et al., 2003 and Si = 2.03\*Al for Chiapello et al., 1997).

259 The results of dust concentrations estimated from the three methodologies above described are 260 summarized in the Table 1 for wet season (WS) 2016 and dry season (DS) 2017. Indeed, Ca, 261 Al, and Fe concentrations measured by ICP-MS are only available in WS2016 and DS2017 due to experimental problems, whereas  $Ca^{2+}$  concentrations measured by IC are available for all 262 campaigns. As shown in table 1, the dust obtained from Ca<sup>2+</sup> measured by IC (Sciare et al., 263 2005) and by the Guinot et al. (2007) method is lower than that obtained from trace elements 264 265 (Terzi et al., 2010) for DS2017 whereas in the same order of magnitude in WS2016. Such results are in agreement with methodological aspects. Indeed, Al, Fe, Ca ... obtained by ICP-266 MS include both soluble and insoluble particles whereas Ca<sup>2+</sup> measured by IC only include 267 soluble particles. During the dry season, comparison of Ca measured by ICP-MS (not shown 268 here) is higher than that of the IC, by a factor of 1.7, 1.8, 2.2 and 1.1, at the ADF, AWB, AT, 269 and CT sites respectively. By contrast, this factor is low and constant (1.3) in the wet season 270 for all the sites. In our study, due to the lack of trace element data for WS2015 and DS2016, 271 dust estimations are performed from Guinot et al. (2007) method. This choice globally implies 272 an underestimate of dust concentrations by a factor of 1.5 to 3.5 in DS2017 as shown in Table 273 274 1.

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#### 6 2.3.7. Aerosol chemical closure methodology

As previously mentioned and detailed, aerosol chemical closure is performed following
the Guinot et al. (2007) methodology.

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#### 2.4. Meteorological conditions

In Figure 4, meteorological data (surface temperature, wind directions and speed) issued from the NOAA Integrated Surface database (ISD; see https://www.ncdc.noaa.gov/isd) and the ASECNA (Agence pour la Sécurité de la Navigation Aérienne en Afrique et à Madagascar) are presented for the South-West Africa region including Abidjan and Cotonou. As expected, this

area is under the influence of the Convergence Zone of two air masses of a different nature, i.e. 285 Harmattan (hot and dry continental trade winds) from the north and Monsoon (humid maritime 286 trade winds) from the south (Figure 4). Ground contact between these two air masses constitutes 287 the intertropical front (ITF) of which the fluctuations during the year determine the seasons in 288 the Gulf of Guinea (Tapsoba, 1997). During the dry season (from November to March), 289 temperatures are relatively high with maximum around 30°C on the coast. The humidity is low, 290 since the prevailing Harmattan wind blows from the desert, usually bringing dust (Figure 4, 291 lower line). The period from June to September, especially in July is the wet season period 292 293 when daytime temperatures are slightly lower, with maximum around 26/28°C on the coast 294 (Figure 4, upper line). At this season, the humidity level is high across the region. On the coast, 295 rains may occur from March to November.

During our campaigns (not shown here), temperatures are roughly the same at Abidjan and 296 297 Cotonou, reaching 28°C and 26°C in the dry and wet seasons, respectively. Gentle to moderate wind speeds are observed during the measurement campaigns at the two cities, with average 298 values of 15-20 and 15-22 km.h<sup>-1</sup> at Abidian and Cotonou, respectively. There is no 299 precipitation at CT site during the studied periods. In Abidjan on the contrary, low rains occur 300 301 both in wet and dry periods with cumulative precipitation higher in DS2017 (7mm), than in 302 WS2016 (4.7mm) and WS2015 (2mm). There is no rain in DS2016 (https://www.historique-303 meteo.net/afrique/).

#### 304

#### 2.5. Backward trajectories

The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) modelling 305 306 system (Air resources laboratory, Draxler and Rolph, 2012) is used for the trajectory analysis. 307 HYSPLIT model is run to compute 120 h back trajectories ending at Abidjan and Cotonou at 308 50 m a.g.l. (Figure 5). Global Data Assimilation System reanalysis database is used as 309 meteorological input, with a  $0.25 \times 0.25$  degrees horizontal resolution. Results presented in Figure 5, confirm that air masses mainly come from the north with a few from the south-west 310 in dry season (January), whereas from the south-west and the south in wet season (July). 311 Therefore, in January, Abidjan and Cotonou are mainly impacted by polluted air masses from 312 surrounding areas and northern countries with possible dust and west African biomass burning 313 influences, whereas in July, the impact of oceanic sources possibly polluted by long-range south 314 African biomass burning aerosols may be observed. 315

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#### 317 **3. Results**

#### **3.1.** Aerosol size distribution and mass concentration

In Figure 6, the relative mass distribution of PM for Coarse (C), Fine (F) and Ultra-Fine 319 320 (UF) particle sizes in percentages are presented with bulk mass concentration averages indicated in the black boxes for each site and for each campaign. As it may be seen, bulk 321 concentrations vary widely from site to site and from campaign to campaign. During the wet 322 season, the average total concentrations range from 82 to 676 µg.m<sup>-3</sup> in 2015 and 56 to 358 323 ug.m<sup>-3</sup> in 2016, with the maximum at the Abidjan Domestic Fire (ADF) site. While during the 324 dry season, values range from 168 to 269  $\mu$ g.m<sup>-3</sup> in 2016 and from 114 to 559  $\mu$ g.m<sup>-3</sup> in 2017, 325 with maximum concentration obtained at the Cotonou Traffic (CT) and ADF sites. In terms of 326 size distribution, concentration peaks may be observed for all aerosol size-fractions which are 327 found to exhibit different seasonal patterns. UF particles (<0.2 µm) represent the highest 328 contributor to the bulk mass at the ADF site, by up to 60 % (335.3 µg.m<sup>-3</sup>) in DS2017. F particles 329 (1-0.2 µm) are the second most important contributor and both combined particle sizes account 330 for more than 85 % of the total mass at the ADF site. In this site, ultra-fine and fine fractions 331 are also found to be maximum during WS2015 and WS2016 by up to 90 and 83%, respectively. 332 Let us note that C particle contribution in bulk is relatively higher in the traffic and waste 333 burning sites than in ADF site (40%) whereas F and UF particle contributions are on the order 334 335 of 60%.

In terms of PM2.5, the results of this work are presented in Figure 7. The mass concentration of PM2.5 averaged over DS2016 and DS2017 are  $154 \pm 74$ ,  $144 \pm 42$ ,  $134 \pm 7$ and  $211 \pm 51 \mu$ g.m<sup>-3</sup> at the ADF, AWB, AT and CT sites, respectively and  $338 \pm 24$ ,  $45 \pm 3$ ,  $52 \pm 4$  and  $70 \pm 1 \mu$ g.m<sup>3</sup> over the wet seasons (2015-2016). The increase in PM2.5 is of the order of 54% at ADF from dry to wet season, whereas a sharp reduction (more than 60%) is obtained at AWB, AT, and CT sites.

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#### 3.2. Carbonaceous aerosol

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#### **3.2.1. EC and OC concentrations**

In Figure 8, EC relative mass contributions are presented for each size, site and campaign: wet season 2015 (WS2015), wet season 2016 (WS2016), dry season 2016 (DS2016) and dry season 2017 (DS2017). Mean EC bulk mass concentrations are added in the black boxes for each size and for each campaign. The most striking feature is that the ADF site concentrations are higher than at the other sites in WS2016 and in DS2017, whereas of the same
order of CT site concentrations in the other seasons. Mean concentration at the CT site ( $16\mu g.m^{-3}$ ) is slightly higher than at the AT site ( $10\mu g.m^{-3}$ ), whereas the lowest concentrations are found at the AWB site. Results of the EC size distribution are very consistent among the different sites (Figure 8). Whatever the site and the season, higher EC concentrations are found in C (42%) and UF (43%) particles compared to F particles.

Same data are presented for OC concentrations in Figure 9. It may be underlined that ADF OC values are always higher than in the other sites by a factor ranging from 6 to 30, for all seasons and particle sizes, with highest and lowest values respectively in DS2017 and DS 2016. In terms of size distribution, maximum OC concentrations at the ADF site may be found in UF (53%), then F (29%) and finally C (18%) particles. The same distribution is observed for the traffic sites in DS2016, however, for the other campaigns, OC size distribution looks like the EC ones with higher concentrations in UF and C particles than in F particles.

As shown in Figure 10, the highest OC/EC ratios are always obtained at the ADF site 361 with a value as high as 25 for F particles in WS2016 whereas the lowest values are found in 362 DS2017. This is the same feature for the other sites with ratios lower than 2 in DS2017. OC/EC 363 364 ratios in AWB site are higher than in the traffic sites. Note that values at AT site are higher than CT values in the wet season whereas lower in the dry season. Finally, it is interesting to 365 366 underline that linear correlations between EC and OC are obtained in the ultrafine and fine modes in all campaigns, particularly in DS2017 ( $r^2 = 0.8, 0.8, 0.9$  and 0.9) at the ADF, AWB, 367 AT and CT sites, respectively. This suggests that different studied sources can be assessed as 368 significant sources of both EC and OC. 369

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#### 3.2.2. Water-Soluble Organic Carbon

Concentrations of WSOC and WSOC/OC ratios are presented in Table 2 for each size 372 (UF, F, C and PM2.5) and campaign. As seen, WSOC are always higher at the ADF site than 373 in other sites, at least by a factor of 12. Maximum values are obtained in WS2016 with an 374 average of 16.47, 17.08 and 79.68 µgC.m<sup>-3</sup> for coarse, fine and ultra-fine fractions, respectively, 375 followed by WS2015 and DS2017. WSOC concentrations are the lowest in DS2016, with an 376 average of 4.14, 6.95 and 21.89 µg.m<sup>-3</sup> for coarse, fine and ultrafine fractions, respectively. In 377 378 terms of seasonality, there is not a clear trend in WSOC values at the AWB and AT sites, whereas at the CT and ADF sites, WSOC values are found to be respectively higher and lower 379

in dry seasons compared to wet seasons. It is also interesting to note that WSOC are maximum
in UF sizes in the AT, ADF and AWB sites. At the CT site, the highest values are found in the
coarse particulate fractions, except in DS2016.

As expected, WSOC is strongly correlated with OC (r=0.7 at ADF, 0.8 at AT, 0.5 at 383 AWB and 0.7 at CT), whereas correlations with EC are weaker, especially at the AWB and CT 384 sites with values ranging from 0.1 to 0.4, respectively. Finally, when looking at WSOC/OC 385 ratios (Table 2), maximum values are obtained at the ADF site with PM2.5 ratios as high as 386 43%, followed by the AT and AWB sites with 32%. The lowest value (23%) is found at the CT 387 388 site. Also, Table 2 shows that there is no clear seasonality in WSOC/OC values, excepted at ADF where maximum values occur during the wet season. Note as for WSOC, that ratios are 389 maximum in UF and F fractions for all sites except at the CT site where the ratio for coarse 390 391 fraction is the highest.

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## 3.3. Water-soluble ionic species

393 Figure 11 shows the relative contribution of the major ions to the total concentration (also given) of the ions in the different particle modes (C and F) at the ADF, AWB, AT and CT 394 395 sites for the different measurement campaigns. Let us recall here that only C and F fractions may be documented due to the our experimental protocol. Total concentrations present 396 397 maximum values in ADF and CT sites. Values in AWB and AT sites are of the same order of magnitude and lower by a factor of 2 than in ADF and CT sites. The contribution of different 398 ions show significant variations from site to site. The dominant ionic species at the ADF site 399 over all campaigns is chloride (Cl<sup>-</sup>), with a 26% contribution, followed by nitrate (NO<sub>3</sub><sup>-</sup>) (16%), 400 calcium (Ca<sup>2+</sup>) (13%) and potassium (K<sup>2+</sup>) (12%). Sulfate (SO<sub>4</sub><sup>2-</sup>), ammonium (NH<sub>4</sub><sup>+</sup>), sodium 401  $(Na^+)$  and to a lesser extent magnesium  $(Mg^{2+})$  contributions are lower, ranging from 4 to 7% 402 403 of the total ion species. The lowest contribution is for organic acids with their total value lower than 5%. NO<sub>3</sub><sup>-</sup> is the major ionic component at the AWB and AT sites, representing 24% and 404 29% of the total water soluble inorganic concentration, respectively. The second major 405 contributor in AWB and AT is SO<sub>4</sub><sup>2-</sup>, accounting for 21% and 17% of the ion mass, respectively 406 followed by Ca<sup>2+</sup> (12% and 15%) and Cl<sup>-</sup> (15% and 13%). In CT, Ca<sup>2+</sup> is predominant with a 407 relative abundance of 24%, followed by  $NO_3^-$  (23%),  $SO_4^{2-}$  (19%) and  $Cl^-$  (13%).  $Na^+$ ,  $NH_4^+$ 408 and K<sup>+</sup> contributions are lower and in the same order of magnitude in AT, AWB and CT sites, 409 ranging from 4 to 9% of the total ion species. Note that organic ion contributions at AT, CT and 410 AWB is of the same order than in ADF, with lower values at CT. It is interesting to underline 411 in the Figure 11, that  $NO_3^-$  contribution is always higher in the coarse than in the fine size. 412

413 Conversely,  $K^+$  is always higher in the fine than in the coarse size. In CT,  $Ca^{2+}$  in the fine 414 fraction is as high as in the coarse fraction whereas in AT, AWB and ADF,  $Ca^{2+}$  coarse fraction 415 is predominant. Fine particle contribution may be noticed for Cl<sup>-</sup> in ADF whereas in the other 416 sites, Cl<sup>-</sup> is most likely dominated by coarse particles. Finally, SO<sub>4</sub><sup>2-</sup> is mainly found in the fine 417 mode at the AT, AWB and CT sites, but in the coarse mode in ADF site.

In terms of seasonal variations, it may be shown in Figure 11 that higher Cl<sup>-</sup> values are found 418 in wet seasons than in the dry seasons everywhere, except in ADF site where there is no marked 419 difference between seasons. For example, the mean relative total percentages of Cl<sup>-</sup> at the CT 420 site are 38 and 24% in the WS2015 and WS2016, respectively, while these percentages decrease 421 significantly to 18 and 13% in the DS2016 and DS2017, respectively. The Cl<sup>-</sup>/Na<sup>+</sup> ratios are 422 about 1.5 everywhere in both seasons, in agreement with the typical sea water ratio (1-1.2) 423 (Hara et al., 2004), except at the ADF site where these ratios increase to 4 and 5 in wet and dry 424 season, respectively and at the AWB site in the dry season (2).  $K^+$  and  $Ca^{2+}$  are always higher 425 in dry season than in wet season except for  $Ca^{2+}$  in ADF where values are of the same order. 426 Finally, the same trend is observed for  $NO_3^-$  and  $SO_4^{2-}$  with higher values in dry than in wet 427 seasons at AWB and CT sites whereas values at ADF and AT sites are of the same order of 428 429 magnitude for the two seasons.

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#### **3.4. Trace element concentrations**

Table 3 shows the mean values of the major trace elements in bulk aerosol at the 431 different studied sites in WS2016 and DS2017, with their corresponding relative abundances in 432 the total aerosol mass into brackets. Let us recall that data are not available in WS2015 and 433 DS2016. The concentrations of trace elements span a wide range, from 0.2 to 25.2 µg.m<sup>-3</sup>. 434 Among the measured elements, Al, K, Na and Ca are the most abundant, followed by Fe and 435 Mg. In DS2017, Al and Na concentrations are higher in AWB than in the other sites. The 436 minimum value for these species is found in ADF site. Values in traffic sites are of the same 437 order of magnitude and higher than in ADF site. Maximum of Ca and K values may be found 438 in CT and ADF site respectively. It is interesting to note that Al, K, Na concentrations are higher 439 440 in the dry season than in the wet season. Such feature is less clear for Ca, whose seasonal variability is less marked except in AWB and AT sites. In terms of Mg, maximum values are 441 observed in ADF site and of the same order of magnitude whatever the season. Fe abundance 442 is higher in AWB and CT sites than in ADF and AT sites and higher in DS2017 than in WS2016 443 everywhere. The other metals (Ti, P, Zr, Zn, Cr, Mn, Pb and Ni) represent less than 0.5% and 444

2% of the total mass in WS2016 and DS2017, respectively, at all sites, with Cr, Mn, Pb and Ni
exhibiting less seasonal variability compared to the rest of the metal elements.

To assess the relative contribution of crustal and non-crustal origin of elemental aerosol loadings, source enrichment factor (EF) of a trace element X have been first calculated with the following formula using both literature data of the typical elemental composition of the upper continental crust (Mason and Moore, 1982; Taylor, 1964), measured elemental composition from this study and Al as a reference element :

$$EF_{X} = \frac{\frac{[X]_{atm}}{[Al]_{atm}}}{[X]_{soil}/[Al]_{soil}}$$

453 Where [X]<sub>atm</sub> and [Al]<sub>atm</sub> are the concentrations of the chemical element X and Al in the atmosphere, respectively, and [X]<sub>soil</sub> and [Al] soil are the typical concentrations of the element 454 X and Al in the earth's crust, respectively. Al is frequently used as a reference element assuming 455 that its anthropogenic sources in the atmosphere are negligible (Gao et al., 2002; Cao et al., 456 457 2005; Xu et al., 2012). In all sampling sites, EF values typically lower than 5 are obtained for several trace elements (Be, Sc, Ti, V, Fe, Ga, Sr, Nb, Rh, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, 458 459 Dy, Ho, Er, Tm, Yb, Lu, Ta, Th and U). This suggests a natural origin of these species (Freitas et al., 2007; Gao et al., 2002). The most enriched elements (EF>100) are Sb, Sn, Zn, Se, Te, 460 Cd, Pb, Bi and Mo at nearly all of the sites, indicating significant anthropogenic origin (Wang 461 et al., 2006). These elements are mainly emitted into the atmosphere through fossil fuel 462 463 combustion, traffic emission, wear of brake lining materials and industrial processes (Watson and Chow, 2001; Samara and al., 2003). Secondly, source contributions have been estimated 464 from these EF values following the method described by Arditsoglou and Samara (2005). Note 465 that this study refers to ratios for a limited list of sources, perhaps not including the African 466 source specificities. As a result, it may be seen that 30% of trace element concentrations is of 467 468 anthropogenic origin at ADF site whereas about 17 % at the others sites.

469 **3.5. Dust** 

- Figure 12 shows dust concentrations calculated from Guinot et al. (2007) methodology (see paragraph 2.3.6) for C and F particle sizes at the different sites for each season. Note that as for WSI and trace element and due to our sampling procedure, there are values for fine and coarse particles for all seasons excepted for WS2016 with values for coarse particles only.
- 474 During the wet season, coarse dust concentrations range from 5 to 25  $\mu$ g.m<sup>-3</sup> in 2015 and 9 to 475 37  $\mu$ g.m<sup>-3</sup> in 2016, with higher values at the CT and ADF sites in 2015 and at AT, CT and ADF

in 2016. In WS2015, fine dust concentrations range from 12 to 49 with maximum values at ADF and CT sites also. During the dry season, values range from 38 to 156  $\mu$ g.m<sup>-3</sup> in 2016 and from 41 to 116  $\mu$ g.m<sup>-3</sup> in 2017, with maximum concentrations obtained at the CT site, followed by AWB site. When considering mean values of the dry seasons, total dust at CT is 2.4 times the values found at AT, 1.6 times at AWB and 3.4 times at ADF. Seasonal comparison shows that total dust concentration is higher in the dry seasons than in WS2015 by a factor of 3 in AT, 2.6 in CT and 4 in AWB, but of the same order of magnitude in ADF site.

483

#### 3.6. Aerosol chemical closure

484 The aerosol chemical closure obtained using the Guinot et al. (2007) method (see below) 485 at the different sites for each season is presented in Figure 13. Results show clear intra- and inter-annual variations at all of the sites, as well as significant differences among the sites. In 486 total, dust accounts for 39 to 75% of the bulk PM mass at both traffic sites, with no clear 487 seasonal cycle and higher contributions in Cotonou (Figures 13c and 13d). These percentages 488 vary from 32 to 64% at the AWB site, and from 18 to 35% at the ADF site, with percentages 489 1.8 times higher in the dry season than in the wet season in AWB and no clear seasonal 490 491 difference in ADF (Figures 13a and 13b). Carbonaceous aerosol, the sum of EC and POM, show large contributions at the ADF site (from 49 to 69% of the total PM mass), with relatively 492 similar proportions in each season (Figure 13a). The absence of a clear seasonal pattern is also 493 observed in CT whereas carbonaceous aerosol is slightly higher in WS than in DS in AWB (23 494 and 16% respectively) and AT (37 and 21% respectively) (Figures 13b-d). Carbonaceous 495 aerosol contribution accounts for about 11-49% of the total mass at both traffic sites with higher 496 values in AT (mean of 30%) than in CT (13%). The ion percentages in PM fractions present 497 the same pattern at AT, CT and AWB sites with higher values in wet than in dry seasons. In 498 499 these sites, we may notice that mass concentration in coarse particles is larger in the wet season whereas of the same order of magnitude than the one in fine particles in the dry season. In ADF, 500 501 no marked difference may be found between the seasons and the sizes (Figure 13).

502

# 503 4. Discussion

A discussion of the results site by site (Abidjan domestic fire site, traffic sites both together and waste burning site) will be first proposed. We will scrutinize (1) the proximity between the sites and the sources; (2) the source specificity with more or less incomplete combustion (e.g. wood combustion and two-wheel vehicle emission factors are higher than gasoline emission factors (Keita et al., 2018); (3) the relative influence of other local sources or transported sources to
the studied sites such as dust and biomass burning; (4) the occurrence of continental air masses;
(5) the variation of the boundary layer height (as reported by Colette et al., 2007); and (6) the
meteorological parameters (e.g. temperature, relative humidity and wet deposition) to explain

- the differences of pollutant concentrations and their seasonal and inter-annual variabilities.
- 513 In a second part, we will present comparison of our values with other DACCIWA values and
- also with literature values for other intensive campaigns in Africa.
- 515

# 516 **4.1. Abidjan Domestic Fires (ADF)**

As shown in the above paragraphs, maximum values are obtained at the ADF site, for aerosol mass, EC, OC, WSOC, water-soluble ionic species (e.g.  $Cl^-$ ,  $NO_3^-$ ,  $Ca^{2+}$  and  $K^+$ ) and some trace elements such as Mg and K (whereas Al, Na and Fe are lower than in the other sites). Also, aerosol PM2.5 values are well above the annual and daily WHO guidelines of 25 and 10 µg.m<sup>-</sup> <sup>3</sup> respectively, whatever the season.

- 522 Such pattern is due to the proximity of the ADF site to the studied combustion source: in that area, the use of wood combustion is very active due to commercial activities of women drying 523 524 fish and meat and domestic cooking. This is also confirmed by the high relative importance of 525 total carbon in aerosol mass whatever the size (49 to 69%) and by values of source enrichment factor. Indeed, at least 30% of trace element concentrations are of anthropogenic origin at ADF 526 site. In addition, wood combustion is well known to be highly polluting due to incomplete 527 combustion: this is shown here by the measurements of very high OC/EC ratios at ADF, on the 528 order of the one measured at the source level by Keita et al. (2018). This is also shown by 529 WSOC relative importance which is expected for wood burning following Yu et al. (2018), 530 Tang et al. (2016), Feng (2006) and Saxena and Hildemann (1996) and by the strong correlation 531 of WSOC with biomass burning K<sup>+</sup> tracer. 532
- 533 Chloride is most likely associated with sea salt origin (normal chloride concentrations represent
- at least 55% of marine aerosols following Goldberg (1963)) or secondary aerosol production
- 535 (Li et al., 2016). Since chloride relative concentration at the ADF site remains lower than the
- that of sea-sal aerosols, the secondary production source would be the better explanation for
- high chloride concentrations observed at ADF. The size distributions of  $Cl^{-}$ ,  $K^{+}$ ,  $NH_{4}^{+}$  and  $SO_{4}^{2-}$
- support this conclusion : the predominance of these elements in fine particle mode at the ADF
- site would be associated with anthropogenic emissions, particularly biomass combustion and
- domestic fires, or with secondary inorganic aerosols origin. This is confirmed by  $Cl^{-}/Na^{+}$  ratio
- values as shown earlier. Contrarily,  $Ca^{2+}$  and  $NO_3^{-}$  contributions to the total ions at the ADF

site peak mainly in the large particle fraction and may be attributed to quasi natural origin, primarily to dust emissions and nitrate formation by reaction processes, respectively. In addition,  $Na^+$  and  $Mg^{2+}$  display similar size distributions at the ADF site, with the major contribution in the coarse particle fraction, suggesting the common sea salt origin of these two elements (Belis et al., 2013).

As we have shown above, the lower proportion of metal elements at the ADF site (6.5% of the 547 bulk concentration) can be explained by the less dominant influence of re-suspended dust 548 particles compared to traffic sources. Elements such as Cr, Mn, Pb and Ni have less seasonal 549 550 variability than other metallic elements. These small proportions of these non-crust elements suggest a low contribution of elements emitted mainly by anthropogenic activities such as 551 industrial processes (Viana et al., 2007 and 2008; Minguillón et al., 2014). Finally, the Zn/Cd 552 553 ratio has been also examined. A value of 29 close to ratio reported for gasoline vehicle (27, Qin 554 et al., 1997) is obtained for the ADF site, indicating that this site is also impacted by traffic sources. 555

556 High values of WSOC/OC ratios are expected to be harmful to health (Ramgolam et al., 2009, Val et al., 2013). This effect is being enhanced by the particulate size measured at this site (Kim 557 558 et al., 2003; Wilson et al., 2002). Indeed, the relative mass distribution of PM and OC particle sizes shows a major contribution of particles less than 1 µm (as high as 85% of PM). This could 559 be due to the fact that carbonaceous aerosols are formed near emission sources and are mainly 560 of submicron size (Boucher, 2012). Nevertherless, note that EC also presents large coarse 561 particle contribution. This could be due to the importance of wood burning at this site with less 562 efficient combustion and large particle emissions (Watson et al., 2011). 563

In terms of seasonality, higher concentrations of aerosol mass, OC, WSOC, EC and total water 564 soluble ionic species (SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup>) are observed in WS2015 and WS2016 than in 565 DS2016. This may be explained by a more incomplete combustion in the wet seasons than in 566 567 DS2016 due to the use of moist wood for cooking and smoking fish, which leads to large amount of smoke and higher particulate emission factor values. Note that DS2017 values are 568 569 as important as the ones of wet seasons, which will be explained later in the text. With regard to WSOC, their variabilities may be also linked to meteorological factors, such as solar radiation 570 571 (Tang et al., 2016; Favez et al., 2008) and relative humidity (Liang et al., 2016). At ADF site, temperatures are roughly similar in both seasons. However, RH variability may play a role since 572 it is higher in wet season than in dry season. Finally, our results indicate no clear seasonal cycle 573 for Cl<sup>-</sup>, which confirms its anthropogenic origin, as previously shown. 574

#### 576 **4.2. Traffic sites (Abidjan traffic and Cotonou Traffic sites)**

Let us recall first that the two traffic sites have been chosen since they are representative of the traffic diversity in West Africa. At CT site, both personal cars, taxis and an important twowheel fleet may be found whereas at AT site, there are buses, taxis and personal cars. Also, the distance between the site and the traffic sources is the same for the two traffic sites, slightly larger than the distance between the site and the wood burning sources at ADF site.

In these two sites, concentrations are high with PM2.5 values well above the WHO guidelines. 582 Average aerosol mass, EC, OC, dust and water soluble ionic concentrations (with NO3<sup>-</sup> and 583 Ca<sup>2+</sup> maximum at AT and CT sites respectively) are higher at CT than at AT site by a factor of 584 1.5 to 2. Note that this poor air quality found in Cotonou has been reported by Cachon et al. 585 586 (2014). The higher values found in Cotonou could be due to more intense traffic in Cotonou 587 than in Abidjan. Also in Cotonou, this traffic is associated with the lack of public transportation 588 and the use of highly polluted mopeds (aged over 15 years) (Gounougbe, 1999; Avogbe et al., 2011), despite the effort in the last 10 years to restrict their use. Several studies such as MMEH 589 590 (2002) have shown that more than 94,000 mopeds and 350,000 second-hand vehicles are in circulation in Cotonou. Other factors contributing to the local pollution include outdoor 591 592 restaurants using charcoal and motorcycle garages, which are more present around the Cotonou 593 traffic site compared to Abidjan site. It also includes anthropogenic dust. Indeed, at Cotonou, the lack of road infrastructure favours the resuspention of dust particles. Finally, other sources 594 may potentially influence aerosol seasonal composition in these two sites, including marine 595 aerosols, transported dust and biomass burning particles as well as anthropogenic aerosols from 596 597 the surrounding countries (Figure 5). Note also that source enrichment factor values show that about 17% of trace element concentrations are of anthropogenic origin at both traffic sites and 598 that the relative importance of total carbon in mass is higher at AT than at CT sites. 599

600 Aerosol mass, composition and size depend on the season and the two traffic sites are differently 601 affected. The EC and OC concentrations measured in both traffic sites and averaged per season are higher in dry than in wet season. Such variations may be explained by several factors: 602 603 particulate wet deposition occurring during the wet season, reduction of traffic flow due to school vacations and meteorological influence. Higher EC and OC concentrations are obtained 604 605 at CT than at AT sites in dry seasons whereas no statistical difference may been found between 606 the two sites in wet seasons. Such a result is mainly explained by figures 5. In wet seasons, similar backtrajectory pattern may be observed for both sites whereas in dry seasons, CT traffic 607 site only would be influenced by Nigerian anthropogenic sources. 608

In terms of WSOC concentrations, concentrations at the AT site are on average higher than those recorded at the CT site in the wet season, but lower in dry season. The presence of dust can produce semi-volatile organic gas scavenging and therefore WSOC and OC enhancement. Such a phenomenon can explain the highest WSOC concentrations observed in dry season at the CT site where dust concentrations are highest (see dust paragraph). Moreover, this can also explain why the maximum WSOC are in coarse particles at CT, while at AT maximum values are in ultra-fine particles.

- Total WSI concentrations are larger at AT site in the wet than in the dry season with higher values in coarse particles. At CT site, total WSI concentrations in fine particles are higher in the dry than in the wet season whereas same values are obtained in coarse particles for both seasons. Note that CT values are generally higher than AT values with a more important contribution of fine particles in the dry season. These WSI variations can be explained by the relative importance of  $Ca^{2+}$ ,  $SO_4^{2-}$  and  $NO_3^{-}$  in both sites.
- First,  $Ca^{2+}$  contribution to total WSI is higher in CT site than in AT site with no clear seasonal variation at CT site and higher values in dry season than in wet season at AT site. Also at CT, fine and coarse  $Ca^{2+}$  particles are in the same range, whereas coarse  $Ca^{2+}$  particles are predominant at AT site. Such feature may be explained by the impact of dust sources including long-range dust transport at Abidjan and a combination of long-range dust transport and road resuspension at Cotonou.
- Second, the relative contribution of  $SO_4^{2^-}$ ,  $NH_4^+$  and  $NO_3^-$  as a percentage of total WSI in the 628 different particle modes is reduced in the wet season. During the wet season, the clean winds 629 surrounding the ocean before reaching the measurement sites could contribute to lower the 630 proportion of these species, in addition to the scavenging processes during the rainy days. 631 Unlike the wet season, a relatively good correlation with  $r^2$  of 0.87 (SO<sub>4</sub><sup>2-</sup> versus NH<sub>4</sub><sup>+</sup>), 0.73 632  $(NO_3^- \text{ versus } NH_4^+)$  and 0.87  $(SO_4^{2^-} \text{ versus } NO_3^-)$  has been found in coarse particles, indicating 633 similar sources for these three species during the dry season. In order to try to identify these 634 sources, the ratio of  $SO_4^{2-}/Ca^{2+}$  and  $NO_3^{-}/Ca^{2+}$  has been determined. The average  $SO_4^{2-}/Ca^{2+}$ 635 and  $NO_3/Ca^{2+}$  ratios in combined coarse particles (1.07 and 2.58 during the wet season and 0.33 636 and 1.60 during the dry season) are higher than the corresponding ratios for typical soil (0.026 637 and 0.003, respectively). On the other hand, the  $SO_4^{2-}/Ca^{2+}$  ratio increases in the fine particles 638 (5.07 during the wet season and 2.53 during the dry season), while that of  $NO_3^{-}/Ca^{2+}$  remains 639 almost constant (2.86 during the wet season and 1.65 during the dry season). This implies that 640 the atmosphere at AT and CT sites is enriched by  $SO_4^{2-}$  formed as anthropogenic secondary 641 particles, possibly from sulfur containing pollution sources (Seinfield and Pandis, 1998), 642

particularly in fine particle mode, and by NO3<sup>-</sup> mostly coming from nitrogen containing sources 643 in all particle sizes. The higher contributions of these elements during the dry season could 644 result from a combination of several factors: 1) an atmosphere loaded with dust favoring 645 heterogeneous chemistry to obtain secondary aerosol and the rise of biomass burning emissions; 646 2) the increase of photochemical activity and higher concentrations of hydroxyl radicals in the 647 dry season, which can oxidize SO<sub>2</sub> from combustion (Arndt et al., 1997) to SO<sub>4<sup>2-</sup></sub> (Li et al., 648 2014); and 3) the wind transport of anthropogenic secondary particles from the industrial zone 649 located upstream from our sites. Finally, the proportion of Cl<sup>-</sup> relative to the total mass of ions 650 651 is highest for coarse particles at both traffic sites especially during the wet season, suggesting that Cl<sup>-</sup> at AT and CT sites if from natural origin and probably from sea salt emissions. 652

- 653 If we focus now on dust during the two wet seasons, concentrations are higher in 2016 than in 2015 at CT and AT sites for coarse particles (no data of fine particles are available in WS2016). 654 655 This is consistent with observed aerosol optical depth (AOD) values at CT, which increased by a factor of 2 between 2015 and 2016. No AOD value is given by Léon et al. (2019) at Abidjan 656 657 in WS2015 to allow such comparison in Abidjan. Moreover, during the wet season, an Angström coefficient (AE) on the order of 1 has been found at CT site, indicating smaller 658 659 particles that could be due to road resuspension. It is interesting to note that during WS2016, AOD and AE are respectively higher and lower at Abidjan than at Cotonou. Again, this is 660 consistent with our dust concentrations at CT site. In Abidjan, we could assume that another 661 source of  $Ca^{2+}$ , which is not taken into account in our dust calculations, may explain our dust 662 concentration data. That may be the result of anthropogenic Ca<sup>2+</sup> emissions from residential 663 in 2016 than in 2015 more important as shown earlier 664 combustion, (http://naei.beis.gov.uk/overview/pollutants?pollutant\_id=84). 665
- The relative contribution of dust generally peaks in the coarse mode and, to a lesser extent, in the fine mode, reflecting their natural origin. It is interesting to note that the dust contribution observed in this study for the year 2016 at the Abidjan site is in agreement with the results of Xu et al. (2019) which show a PM2.5 dust contribution of 35-50% compared to our values of 18-52%.

671

#### 672 **4.3. Abidjan Waste Burning site**

Concentrations measured at AWB site are slightly lower than values found in the other sites.
This can be explained by the larger distance of the site to the main studied source (here waste
burning source) than in the other sites. However, PM2.5 values are also higher than WHO
guidelines. +

Aerosol mass, EC and OC concentrations are higher in dry than in wet season, which suggests 677 678 less waste burning activities during the wet season or impacts of other local anthropogenic sources or long-range biomass burning sources. Highest values are found in DS2017 with the 679 lowest OC/EC ratio, as at AT site. OC/EC ratio is highly variable at AWB (1-10) which 680 confirms that AWB site may be impacted by different types of sources as well as by secondary 681 aerosol organic formation which can be detected for OC/EC higher than 2 (Turpin et al. 1990; 682 Hildermann et al. 1991; Chow et al. 1996). Note that OC/EC typical for waste burning source 683 684 is of the order of 8 (Keita et al., 2018).

- 685 It is also observed that at the AWB site, PM mass concentrations are mainly distributed in C 686 mode (30-44%) over the entire period of study, excepted during the WS2015, and to a lesser 687 extend in F mode (21-44%). EC and OC being mainly distributed in C and UF modes. Watersoluble fraction of organic carbon is important (32%) and on the order of the one found at AT 688 689 site. Same for WSI concentrations and WSI composition. At AWB, WSI values are globally slightly higher in wet than in dry season. However, it is interesting to underline that  $Ca^{2+}$  is 690 691 much higher in dry season than in wet season, especially in DS2017. This is in agreement with 692 dust concentrations and trace element concentrations, which have been found to be maximum 693 at AWB, reaching 35.8% of the total PM mass in the dry season. These maximum percentages 694 are due to the large contribution of both Al and Na crustal elements which account for about 26%. Also note that Cu/Sb of 0.08 in DS2017, which indicate an influence of re-suspended 695 particles. A Zn/Cd value of 56 is obtained for the AWB site which is in close agreement with 696 values reported for oil burning (Watson et al., 2001, Samara et al., 2003). That could indicate 697 that oil might be one of the waste burning materials. 698
- 699 Our result suggests that AWB aerosol mass is influenced by a mix of sources, including fuel 700 combustion and mineral salt from sources around the measurement site, associated to long-701 range source impact of dust and biomass burning which will be further discussed in the next 702 paragraph.
- 703

#### 704 4.4. Interannual variability of aerosols in Abidjan and Cotonou

EC and OC concentrations are generally higher in DS2017 than in DS2016 for all the sites. This is not due to the meteorological condition, which is similar in both years. This is also not due to biomass burning impacts. Indeed, when looking at MODIS burnt areas for our period of study (http://www.aeris-data.fr/redirect/MODIS-MCD64A1), burnt areas of west African savannas are higher in 2016 than in 2017. Therefore, carbonaceous aerosol concentrations should be higher in 2016. Then, this could be due to a counter effect between biomass burning emission

strength and air mass transport efficiency. As a result, biomass burning impact could not explain 711 712 the difference in EC and OC during the dry season between 2016 and 2017. Rather, this is due to the variability of local sources. In DS2016 in Abidjan, there was a general strike of civil 713 714 servants of the State with important consequences on urban activities. Lower activities were observed (lower fish smoking emissions, lower traffic ...) in DS2016 compared to DS2017, thus 715 explaining the lower EC and OC concentrations at Abidjan sites. In Cotonou, highest 716 carbonaceous aerosol values in DS2017 may be explained by backtrajectory patterns: Cotonou 717 would be impacted by air masses coming from the high polluted Lagos (Nigeria) area in that 718 719 period whereas from less polluted northern areas in DS2016. Such an assumption is validated by the AOD values at 550nm from MODIS satellite images (http://www.aeris-720 721 data.fr/redirect/MODIS-MCD64A1), which show very high particulate concentrations in the 722 Guinean Golf (Figure 14).

This figure also shows the AOD difference between Cotonou and Abidjan for DS2017, with higher values at Cotonou than in Abidjan for the campaign period, in agreement with our measurements of aerosol mass, EC, OC and dust. This is confirmed by the DACCIWA sunphotometer AOD and Angström coefficient (AE) measurements at Abidjan and Cotonou (Léon et al., 2019; Djossou et al., 2018). Indeed, in DS2017, during our period of measurements, mean AOD in Cotonou is of the order of 1.3 versus 0.9 in Abidjan for an AE of 0.6 for both sites, which clearly indicates the presence of coarse dust particles.

Finally, aerosol mass and dust concentrations have been seen to be higher in DS2016 than in DS2017 in Abidjan whereas values are on the same order of magnitude at Cotonou. Such high values at Abidjan in DS2016 can be explained by the back-trajectory pattern with air-masses all coming from northern dusty areas in DS2016 (Bodélé depression in Tchad, Prospero et al. (2002), Washington et al., (2003), Knippertz et al. (2011), Balarabe et al., (2016)) and/or from northern dusty countries (Mali, Niger) (Ozer, 2005), whereas in DS2017, contribution of southern marine clean air-masses may also be noted.

In the wet season, aerosol mass, EC and OC are higher in WS2015 than in WS2016. This may
be due to particulate wet deposition, more efficient in WS2016 which have been seen earlier to
be more rainy (4.7mm) than in WS2015 (2mm). Moreover, at AT site, dust concentrations are
higher for coarse particles in WS2016 than in WS2015. Such variations may be explained by
long-range dust sources and/or road dust resuspension processes. As no dust event has been
noticed, local source explanation seems to be more evident.
In AT, CT and AWB, OC/EC ratios are globally on the same order for WS2015, WS2016 and

744 DS2016, with values lower than for DS2017. This could be due to lower traffic activities linked

to the DS2016 strike and the wet season vacation periods. Indeed, much higher OC/EC ratios
measured in DS2017 are typical of those of diesel vehicles (Mmari et al., 2013; Keita et al.,
2018). Finally, it is interesting to note that OC/EC ratios measured in this study are in the range
of those previously reported for other megacities such as Agra in India with 6.7 (Pachauri et al.,
2013), Helsinki in Finland with 2.7 (Viidanoja, 2002), Cairo in Egypt with 2.9 (Favez, 2008),

- Paris in France with 3.5 (Favez, 2008), and Milan in Italy with 6.6 (Lonati et al., 2007).
- 751

# 752 **4.5. Comparison with literature data**

Firstly, the comparison between our data and other DACCIWA results including other time 753 sampling focuses on PM2.5 levels, since these particle sizes are relevant for health impact 754 755 studies (Xing et al., 2016). In addition to our values, Figure 7 presents data from Xu et al. (2019) using personal samplers collected in the same area and at the same dates in 2016 during 12h on 756 757 women at the ADF site, students at the AWB site and drivers at the CT site, and from Djossou et al. (2018) study based on one week exposed filters collected at the same areas and for the 758 same periods as this study. We note that PM2.5 directly measured on women are 2.3 and 0.9 759 times our values obtained at the ADF site in dry and wet seasons, respectively, and 3.4 and 4.9 760 times higher on students than at the AWB site, and 1.6 and 2.1 times higher on drivers than at 761 the CT site. Also, our values are on average 1.6, 3, 5 and 8 times higher than weekly-integrated 762 values of Djossou et al. (2018) including our 3 days of measurements at the AWB, ADF, AT 763 764 and CT sites respectively. As it may be seen, the lowest concentrations are observed in Djossou 765 et al. (2018), whereas the highest concentrations are recorded in Xu. et al. (2019). This is valid 766 for all sites, seasons and campaigns. Differences between our values and Djossou values may be explained by the sampling times of the two studies. Indeed, as recalled, Djossou 767 768 measurements are weekly integrated, taking into account diurnal activities during all the week, including week-end and nights which have expected lower PM2.5 concentrations. Our study 769 770 includes only maximum pollution conditions for each site. The highest differences occur for 771 the traffic sites. This may be clearly understood since diurnal and weekly variations of traffic 772 sources are the most variable. Comparison between our values and Xu et al. (2019) values is 773 also interesting. Indeed, it is at the ADF site that on-site and women PM2.5 concentrations are 774 the closest, which shows that this site is the most representative of the pollution exposure to women. The biggest differences are found at the AWB site. As already mentioned, distance 775 from the site to the waste burning source is more important than for other sites, which explains 776 why concentrations obtained on students who are leaving close to the sources are much higher 777 778 than on-site concentrations. At the Cotonou traffic site, measurements taken from people are

also higher than on site measurements. Such differences can be explained by additional pollution exposure as people move around. Note that the sampling technique may also play a role in such a comparison. In terms of seasonal variation, our results are in agreement with longterm EC measurements obtained by Djossou et al. (2018) for the same sites and period. Finally, Table 4 compares our mean PM2.5 results obtained from 3-hour sampling for three consecutive

- days to literature data for different traffic sites in the world given at a daily scale. It is interesting
- to note that our values are situated at the higher end of the range of PM2.5 data observed from
- the other sites.
- 787 Secondly, Table 5 compares our OC and EC values to those obtained by Djossou et al. (2018) and Xu et al. (2019) as previously described for the same period and the same sites. Again, it is 788 789 interesting to note that Djossou's values are in general lower than ours. Indeed, for the wet and 790 dry seasons, our OC measurements are 4 and 1.4 times higher than Djoussou's at the AT site, 791 2.1 and 5.7 times higher at the CT site, and 2.5 and 2.5 times higher at the ADF site, respectively. As for PM2.5, this can be explained by the different sampling times between our 792 793 experiments that were performed at the peak of urban activities, while Djossou's dataset represents weekly integrated values. Differences at the ADF site are largely explained by the 794 795 temporal pattern of fish smoking activities which take place every day, only in the morning, as 796 such the associated pollution is not well represented in the weekly sampling. Finally, there are less differences at the AWB site between both datasets. As above explained, there are no marked 797 temporal variations of concentrations at AWB site. The predominant waste burning emissions 798 impacting our site can occur night and day, week-days and week-end since origin of such 799 burning can be either anthropogenic or from spontaneous combustion. It may be also recalled 800 that another reason of agreement between both datasets may come from the large distance 801 between the site and the local and regional sources. Comparisons made between our values and 802 those of Xu's personal data show that both OC and EC are of the same order at the ADF site, 803 804 whereas Xu values are higher than ours at the CT and AWB sites. This result is in agreement with what we found with PM2.5 concentrations as detailed above. Finally, Table 6 presents OC 805 806 and EC for the PM2.5 comparison between our values and other recent studies dealing with traffic sites in other regions of the world and with similar operational conditions. We find that 807 808 our values are situated in the middle of the range observed in these different studies. Briefly, as presented in Table 7, it is interesting to compare our WSOC concentrations to literature data 809 for different traffic sites of the world. We note that our values are on the same order as values 810 found in Asia and higher than those found in Europe. 811

- 812 Thirdly, the percentages of the total WSI to PM mass (15-20%) at the three Abidjan sites (ADF,
- AWB and AT) are in the same order of magnitude than the data from PM2.5 personal exposure
- samples collected at the same locations in 2016 by Xu et al. (2019). Our results also are very
- close to the ionic contribution of 9% of the PM10 mass found at the urban curbside site in Dar
- es Salaam in Tanzania during the wet season 2005 by Mkoma (2008).
- 817

## 818 **5.** Conclusion

- This paper presents the mass and the size-speciated chemical composition of particulate matter (PM) obtained during the dry and wet seasons in 2015, 2016 and 2017. During each campaign, 3-hour sampling at the peak period of pollution for three consecutive days was performed at three sites in Abidjan, representative of domestic fire (ADF), waste burning (AWB) and traffic
- 823 (AT) sources, and at one traffic site in Cotonou (CT).
- It is important to underline that our results and their temporal variations are very sensitive to (1) the source activities whose pollution levels are highly linked to socio-economic status of each city; (2) the impact of imported pollution (sea-salt, biomass burning, dust, anthropogenic emissions from neighboring countries), according to air mass origins; and (3) the particle wet deposition.
- The comparison between our results and other DACCIWA measurements underlines the importance of the distance of the chosen site to the sources. At the source level (such as ADF), pollution results at the site are in agreement with exposure of people living at this site. However, at the other sites, comparison is more difficult since the sites are under the influence of a mix of transported sources. That shows the key importance of exposure studies to estimate air quality and health impacts. That shows also the need of long-term studies to really understand role of imported sources in urban air quality.
- The main striking feature is that PM2.5 values are well above the annual and daily WHO 836 guidelines of 25 and 10 µg.m<sup>-3</sup>, respectively, whatever the site and the season. Also, measured 837 concentrations from this study are situated in the middle to the high part of the range of 838 839 worldwide urban aerosol concentrations given at a daily scale. In addition, we have stressed the importance of ultra-fine and fine particles in the studied aerosol and of species such as 840 841 particulate organic matter and water soluble organic carbon, which are well known to be particularly harmful. This is again a warning signal for pollution levels in African capitals if 842 843 nothing is done to reduce emissions in the future.
- 844 Our study constitutes an original database to characterize urban air pollution from 845 specific African combustion sources. The next step will be to cross such an exhaustive aerosol

- 846 chemical characterization to biological data in order to evaluate the impact of aerosol size and
- 847 chemical composition on aerosol inflammatory properties.
- 848

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# 855 Author Contributions

J.A. and C.L. conceived and designed the study. J.A., C.L. and E.T.D. contributed to the literature search, data analysis/interpretation and manuscript writing. J.A., C.L., A.B. and E.T.D. contributed to manuscript revision. J.A., C.L., J.F.L, H.C, V.Y., A.A, C.G, C.Z, E.C and S.K. carried out the particulate samples collection and chemical experiments, analyzed the experimental data.

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## 862 Additional Information

Figure S1 and Appendix A-D accompany this manuscript can be found inSupplementaryInformation.

865

### 866 **Competing financial interests**

- 867 The authors declare no competing financial interests
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- 870

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1253	Figure caption
1254	Figure 1: Map of the city of Abidjan reporting the geographical location of DACCIWA urban
1255	sampling sites.

Figure 2: Map of the city of Cotonou reporting the geographical location of DACCIWA urbansampling site.

- Figure 3: Pictures of the different sampling sites: (a) Traffic in Cotonou (Benin, CT station),
  (b) Waste burning in Abidjan (Côte d'Ivoire, AWB station), (c), Domestic fire, showing
  smoking activity in Yopougon, Abidjan (Côte d'Ivoire, ADF station), (d) " woro-woro
  and Gbaka " traffic in Abidjan (Côte d'Ivoire, AT station).
- Figure 4: Wind, pressure and temperature diagram at Abidjan and Cotonou during the differentcampaigns.
- Figure 5: Back trajectories arriving at Abidjan (a) and Cotonou (b) for each season (WS2015,
  WS2016, DS2016 and DS2017).

Figure 6: Aerosol Mass concentrations at the different study sites for each campaign and for the
different sizes (C in black, Fine in light Grey, Ultra-fine in grey). Bulk aerosol mass is
indicated in boxes.

- Figure 7: Comparison of PM2.5 mass concentrations in μg.m<sup>-3</sup> at the four sites with those
  obtained by Djossou et al. (2018) and Xu et al. (2019) for the same sites and periods.
  Data for the following weeks were then selected in Djossou et al. (2018): 20-27/07/2015
  for WS15, 4-11/01/2016 for DS16, 4-11/07/2016 for WS16 and 9-16/01/2017 for DS17.
- Figure 8: EC relative concentrations in each size classes (C in black, Fine in light grey, Ultrafine in grey) at the different study sites for each campaign. Bulk EC concentration for
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- Figure 9: OC relative concentrations in each size classes (C in black, Fine in light grey, Ultrafine in grey) at the different study sites for each campaign. Bulk OC concentration for
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- Figure 10: OC/ EC ratio for the different campaigns and sites for each aerosol size (C in black,
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- Figure 11: Water-soluble ionic species speciation for each site, each campaign and each aerosolsize.
- Figure 12: Dust concentrations at the different study sites for each campaign and for the
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<ul> <li>Figure 14: MODIS Aerosol optical depth regional distribution over W</li> <li>2017, focusing on our campaign date at Abidjan (a-c 01/11-12</li> <li>(b-d 01/6-7, right part).</li> </ul>	Vest Africa. Data are for 2-left part) and Cotonou
<ul> <li>1288 Figure 14. MODIS Actosof optical depth regional distribution over w</li> <li>2017, focusing on our campaign date at Abidjan (a-c 01/11-12</li> <li>1290 (b-d 01/6-7, right part).</li> <li>1291</li> <li>1292</li> <li>1293</li> <li>1294</li> </ul>	est Antea. Data are for e-left part) and Cotonou
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AFRICA CÔTE D'IVOIR	E
ABIDJAN DOMESTIC FIRES	ABIDJAN WASTE BURNING (AWB)

0 75 150 km



- 1301 Figure1



- 1308 Figure 2



# **Figure 3**





1346 Figure 5-a



- 1348 Figure 5-b











- 1364 Figure 8



- **Figure 9**

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1378 Figure 10



1382 Figure 11



1386 Figure 12

WSI POM EC Dust n.d.



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- 1395 Figure 14

1405 List of table

- 1407 Table 1: Comparison of dust concentrations obtained from different methodologies
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Table 4: Comparison of PM2.5 concentrations with literature data. Only literature data given at a dailyscale have been selected.

- Table 5: PM2.5-EC and PM2.5-OC comparison with Djossou et al. (2018) and Xu et al. (2019) values.
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### 1442 Table 1: Comparison of dust concentrations obtained from different methodologies in µg.m<sup>-3</sup>

Dry 2017		Sciare et	Guinot et al.	Terzi et al.
		al. (2005)	(2007)	(2010)
ADF	С	18.5	11.2	86.9
	F	9.3	29.7	22.2
	bulk	27.7	40.9	109.1
AWB	С	12.3	48.5	126.4
	F	5.2	39.7	106.4
	bulk	17.6	88.2	232.8
AT	С	16.4	24.8	98.5
	F	4.3	13.4	34.2
	bulk	20.7	38.2	132.7
СТ	С	37.9	70.0	98.4
	F	23.4	45.6	55.8
	bulk	61.3	115.6	154.2

Wet 2	2016	Sciare et al., 2005	Guinot et al., 2007	Terzi et al.(2010)
ADF	С	35.34	21.5	27.9
AWB	С	13.46	8.6	21.1
AT	С	19.65	37.5	21.4
CT	С	42.98	26.2	52.5
	Bulk	42.98	26.2	52.5

each aerosol size

Site		Abidjan V	Abidjan Waste Burning		omestic Fire
Period	Size	WSOC	WSOC/OC	WSOC	WSOC/OC
Wet season	Coarse	1.3	24,6	8.2	32.5
2015	Fine	0.7	19.9	12.8	22.7
	Ultra fine	4.1	43.6	51.3	72.5
	PM2.5	5.5	33.7	69.5	47.2
Dry season	Coarse	0.4	12.3	4.4	18.8
2016	Fine	0.9	46.9	7.0	20.4
	Ultra fine	1.5	38.4	21.9	61.5
	PM2.5	2.7	32.7	31.0	32.0
Wet season	Coarse	1.3	42.5	16.5	44.3
2016	Fine	0.8	26.3	17.1	33.0
	Ultra fine	2.0	41.2	79.7	84.5
	PM2.5	3.5	37.1	106.0	52.0
Dry season	Coarse	1.9	32.9	12.1	36.0
2017	Fine	1.4	38.4	19.9	35.0
	Ultra fine	1.6	11.5	38.6	19.0
	PM2.5	4.0	30.0	65.8	29.0

- 1455
- 1456 Table 2 (suite): WSOC concentrations (µg.m<sup>-3</sup>) and WSOC/OC ratios (%) for each site, each campaign
- 1457 and each aerosol size

Site		Abidja	Abidjan Traffic		ou Traffic
Period	Size	WSOC	WSOC/OC	WSOC	WSOC/OC
Wet season	Coarse	2.4	39.6	1.1	23.3
2015	Fine	1.3	46.7	0.5	22.1
	Ultra fine	4.7	29.0	0.4	12.7
	PM2.5	6.9	34.0	2.2	18.0
Dry season	Coarse	1.4	43.0	2.3	64.1
2016	Fine	1.9	59.0	0.6	10.5
	Ultra fine	4.9	62.0	6.3	42.9
	PM2.5	7.5	49.4	8.0	29.0
Wet season	Coarse	1.1	23.1	1.2	34.7
2016	Fine	0.5	16.8	0.5	32.2
	Ultra fine	1.4	34.8	0.9	23.0
	PM2.5	2.4	26.0	1.9	28.0
Dry season	Coarse	0.9	24.0	3.5	37.8
2017	Fine	0.3	24.3	2.4	39.6
	Ultra fine	1.8	14.8	1.9	10.4
	PM2.5	2.6	16.0	6.0	18.2

### Bulk ng.m<sup>-3</sup> (%) DRY 2017 WET 2016 ADF AWB AT СТ ADF AWB СТ AT Al 10050.8 (1.8) 25186.1 (13.7) 14015.8 (12.26) 15480.4 (5.7) 1370.5 (0.4) 1990.1 (3.5) 2191.4 (2.4) 4010.5 (4.4) Κ 8634.3 (1.5) 6093.7 (3.3) 3677.7 (3.22) 5068.9 (1.9) 1105.0 (0.3) 472.0 (0.8) 275.9 (0.3) 1076.0 (1.2) 2070.6 (0.6) Na 6847.8 (1.2) 23430.5 (12.8) 15372.1 (13.44) 11529.3 (4.3) 3735.4 (6.6) 2861.5 (3.1) 5310.2 (5.8) Ca 4321.2 (0.8) 2923.7 (1.6) 4117.6 (3.60) 6233.5 (2.3) 4124.7 (1.1) 447.5 (0.8) 374.7 (0.4) 4954.02 (5.4 Mg 1940.6 (0.3) 384.0 (0.2) 410.3 (0.36) 823.2 (0.3) 1524.7 (0.4) 294.9 (0.5) 283.5 (0.3) 619.2 (0.7) 3807.9 (2.1) 709.3 (1.3) 987.3 (1.1) Fe 1709.9 (0.3) 1628.1 (1.42) 3406.8 (1.3) 1314.0 (0.4) 1549.4 (1.7) Р 1521.9 (0.3) 696.0 (0.4) 147.8 (0.13) 207.4 (0.1) 605.4 (0.2) 13.2 8.6 81.4 (0.1) Ti 457.9 (0.17) 170.8 (0.05) 75.7 (0.13) 96.8 (0.11) 488.9 (0.1) 2270.3 (1.2) 282.8 (0.25) 154.7 (0.17) Zn 189.7 (0.03) 80.9 (0.04) 57.9 (0.05) 149.4 (0.06) 60.3 (0.02) 1.9 41.1 (0.04) 36.2 (0.04) Zr 217.9 (0.19) 145.3 (0.05) 22.4 (0.04) 36.7 (0.04) 31.2 (0.03) 172.1 (0.03) 390.3 (0.21) Pb 87.1 (0.02) 11.0 (0.01) 4.8 11.5 8.3 2.1 2.3 9.3 (0.01) 37.4 (0.01) 0.77 0.09 0.0006 Sn 79.7 (0.01) 38.4 (0.02) 21.6 (0.02) 9.ç (0.01) Mn 74.2 (0.01) 35.2 (0.02) 33.7 (0.03) 160.6 (0.06) 48.9 (0.01) 12.01 (0.02) 9.1 (0.01) 41.41 (0.05) Rb 52.4 (0.01) 8.7 5.9 (0.01) 8.5 4.47 0.71 0.85 1.9 Sb 59.9 (0.01) 201.2 (0.11) 123.6 (0.11) 149.04 (0.06) 24.4 (0.01) 0 0.0006 2.9 Ba 37.3 (0.01) 53.3 (0.03) 47.4 (0.04) 65.8 (0.02) 18.5 8.02 (0.01) 9.9 (0.01) 32.0 (0.03) Ni 36.5 (0.01) 34.5 (0.02) 27.9 (0.02) 50.2 (0.02) 18.00) 33.1 (0.06) 9.7 (0.01) 14.9 (0.02) Cr 29.4 (0.01) 53.8 (0.03) 35.8 (0.03) 28.6 (0.01) 41.9 (0.01) 47.7 (0.08) 24.3 (0.03) 29.7 (0.03) 21.2 (0.02) 28.1 (0.01) 34.02 (0.01) 17.02 0 0.19 8.1 (0.01) Sr 15.5 (0.01) Cu 24.0 12.3 (0.01) 3.6 9.6 3.99 0.26 0.87 2.8) 0.22 Sr 12.6 17.1 8.9 (0.01) 7.8 (0.01) 0.23 Li 7.3 15.5 (0.01) 7.39 0.36 0.32 0.75 Cd 6.1 0.83 1.18 0.05 0.02 0.17 1.6 1.012.4 (0.01) v 5.5 5.1 10.62 2.14 1.84 2.0 3.35 Mo 5.5 8.0 4.9 3.19 4.56 6.84 (0.01) 2.04 3.2 Cs 5.4 0.9 1.2 0.94 0.11 0.12 0.01 0.17 0.97 Hf 4.5 10.8 (0.01) 6.8 (0.01) 4.63 0 0.67 1.03 4.2 0 0 (0) 0.05 0.60 4.5 1.22 As 3.1 Li 4.0 9.8 5.9 (0.01) 5.82 0.27 0.37 0.16 0.93 0.13 35.67 (0.01) 0.49 0.33 Co 3.8 1.1 2.1 0.86 3.7 6.0 (0.01) 9.85 1.06 0.50 0.42 2.03 Ce 6.8 0.92 2.9 4.78 0.54 0.24 0.25 La 1.8 3.5 0.98 0.5 0.63 Nb 1.5 2.6) 1.4 2.48 0.46 0.05 0.40 1.5 2.5 2.4 4.15 0(0)Nd 0.02 Sc 0.69 1.4 1.1) 1.31 0,00 0.03 0.13 0.28 0.003 Be 0.19 0.2 0.37 Ga 0.61 1.15 0.8 0.98 0.2 0.11 0.12 0.11 Ge 0.42 1.02 0.8 0.68 0.01 0.07 0.02 0.91 0.02 0.59 0.20 0.18 Se Rh 0.02 0.02 0.00002 0.002 0.002 0.0002 0.05 Te 0.06 0.08 0.08 0.02 0.02 0.02 0,00 Pr 0.40 0.74 0.7 1.13 0.06 0.01 0.004 0.15 0.27 0.46 0.45 0.76 0.01 0,00 0.07 Sm 0.05 0.08 0.01 0.003 0.0 Eu 0.08 0.15 0.05 Gd 0.31 0.57 0.54 0.86 0.02 0.001 0.15 0.04 0.07 0.07 0.005 0.02 Tb 0.10 0.24 0.42 0.40 0.57 0.001 0.05 Dy Но 0.05 0.09 0.09 0.12 0.01 0.008 0.01 0.03 Er 0.16 0.31 0.28 0.35 0.02 0.03 0.02 0.09

### 1458 Table 3: Trace element concentrations for bulk aerosol for each site and for DS2017 and WS2016.

Tm	0.02	0.05	0.05	0.05	0.002	0.003	0.001	0.01
Yb	0.18	0.38	0.32	0.34	0.01	0.02	0.03	0.09
Lu	0.03	0.06	0.08	0.06	0.003	0.005	0.005	0.02
Та	0.07	0.14	0.09	0.16	0.06	0.02	0.02	0.03
W	0.80	1.63	0.69	0.54	0.26	0.41	0.4	0.3
Tl	0.22	0.01	0.03	0.06		0.009		
Bi	0.32	0.26	0.02	0.08	0.06	-	-	0.08
Th	0.41	0.88	0.79	1.29	0.15	0.09	0.09	0.24
U	0.22	0.43	0.49	0.51	0.03	0.03	0.02	0.09
Total	36459.9	65817.6	40312.2	44159.2	12562.9	7874.7	7227.2	18001.1
Mass(µg/m <sup>3</sup> )	558.8	183.6	114.4	270.0	374.7	56.3	91.6	91.9

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	Location	PM2.5 (µg.m <sup>-3</sup> )	Reference
	Abidjan, Côte d'Ivoire	142	This work
	Cotonou, Benin	154	This work
	Beijing, China	81.4	Xie et al., 2019
	Christchurch, New Zealand	9.2	Tunno et al., 2019
	Pune, India	$98\pm28$	Pipal et al., 2019
	Delhi, India	123	Guttikunda and Calori, 2013
	Lahore, Pakistan	91	Colbeck et al., 2011
	Ahvaz, Iran	69	Shahsavani et al., 2012
	Hong Chong, Hong Kong	$54.7\pm25.6$	Cheng et al., 2015
	Lecce, Italia	16	Cesari et al., 2016
	Libreville, Gabon	35.8	Ngo et al. $2019$
	Port Gentille, Gabon	60.9	Ng0 et al., 2019
	Kenitra, Morocco	51.3	Zghaid et al., 2009
	Bilecik, Turkey	247	Gaga et al., 2018
	Algiers, Algeria	34.8	Bouhila et al., 2015
	Shobra, Egypt	216	Lowenthal et al.,2015
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Table 4: Comparison of PM2.5 concentrations with literature data. Only literature data given at a daily scale have been selected.

Table 5: PM2.5-EC and PM2.5-OC comparison with Djossou et al. (2018) and Xu et al. (2019) values.
Units are μgC.m<sup>-3</sup>.

Location	Period	PM2.5 OC	PM2.5 EC	References
	July 2015	$22.6\pm3.4$	$4.3\pm0.2$	
	January 2016	$15.2 \pm 5.3$	$7.0 \pm 2.6$	This Work
	July 2016	$9.3 \pm 1.3$	$2.2\pm0.1$	THIS WORK
	January 2017	$16.1 \pm 1.7$	$18.9 \pm 1.4$	
Traffic Abidian	July 2015	$3.3 \pm 0.2$	$2.3\pm0.2$	
Cote d'Ivoire	January 2016	$7.7 \pm 0.0$	$3.9\pm0.0$	Dioscou et al. 2018
	July 2016	$7.6\pm0.2$	$4.9\pm0.0$	Dj08800 et al. 2018
-	January 2017	$19.1\pm6.2$	$13.9\pm5.5$	
	July 2015	$13.1 \pm 1.2$	$3.5 \pm 0.7$	
	January 2016	$27.8 \pm 11.3$	$10.9\pm2.6$	This Work
	July 2016	$6.7\pm1.9$	$2.0\pm0.5$	THIS WORK
	January 2017	$33.1 \pm 4.6$	$27.3\pm0.9$	
	July 2015	$4.2\pm0.7$	$1.5 \pm 0.1$	
Traffic Cotonou.	January 2016	$3.0\pm0.3$	$1.5 \pm 0.2$	51 1 6640
Benin	July 2016	$6.7 \pm 0.2$	$1.6 \pm 0.1$	Djossou et al. 2018
	January 2017	$14.5 \pm 0.8$	$4.4 \pm 0.7$	
	2			
	January 2016	$49.5 \pm 12.5$	$13.6 \pm 3.6$	Vu at al. 2010
	July 2016	$37.0\pm3.5$	$9.3\pm0.8$	Au et al. 2019
-	July 2015	$147.2\pm14.5$	$16.1 \pm 1.6$	
	January 2016	$56.5\pm51.5$	$7.4 \pm 3.1$	
Domestic fire	July 2016	$172.3 \pm 39.0$	$17.9 \pm 4.8$	This Work
	January 2017	$283.9\pm34.9$	37.9 ± 4.3	
Cote d'Ivoire	July 2015	$80.5 \pm 1.1$	$32.2 \pm 1.6$	
	January 2016	$76.3 \pm 13.7$	$11.4 \pm 0.2$	
	July 2016	$68.4 \pm 16.5$	$17.4 \pm 2.1$	Djossou et al. 2018
	January 2017	$66.4\pm7.5$	$21.1\pm 6.6$	
	January 2016	72.4 + 24.6	10.5 + 7.2	
	July 2016	$12.4 \pm 24.0$	$19.5 \pm 7.5$	Xu et al. 2019
-	July 2015	$169.3 \pm 197.8$	$11.3 \pm 10.8$	
	July 2015	$14.0 \pm 1.1$	$4.4 \pm 0.1$	
	January 2010	$7.7 \pm 1.5$	$5.0 \pm 0.3$	This Work
	July 2016	$10.0 \pm 2.4$	$1.5 \pm 0.5$	
HL D	January 2017	$21.9 \pm 4.2$	$19.2 \pm 2.4$	
Waste Burning Abidian	July 2015	$3.7 \pm 2.2$	$4.3 \pm 0.3$	
Cote d'Ivoire	January 2016	$13.9\pm9.0$	$3.6 \pm 1.8$	
	July 2016	$9.8\pm4.4$	$2.8 \pm 0.9$	Djossou et al. 2018
	January 2017	$22.4\pm7.8$	$8.7 \pm 3.0$	
	January 2016 July 2016	$85 \pm 57.4$ $65.2 \pm 65.2$	$15 \pm 4.7$ 12.3 ± 11.4	Xu et al. 2019

- 1512 Table 6: EC and OC comparison with literature values. Only literature data given at a daily scale have1513 been selected.

Location	OC (µg.m <sup>-3</sup> )	BC (µg.m <sup>-3</sup> )	Reference
Abidjan (Côte d'Ivoire)	16	8.1	This study
Cotonou (Benin)	20.2	11	This study
Bilecik (Turkey)	49.6-62.8	38.8-58.1	Gaga et al., 2018
Pune (India)	30	5	Pipal et al., 2019
Shanghai (China)	4.9-13.1	1.9-5	Ding et al., 2017
Lahore (Pakistan)	85.7-152	13.8-21	Stone et al., 2010
Agra (India)	25.4-70	3.3-9.5	Satsangi et al., 2012, Pipal et al., 2014
Delhi (India)	34.1-50	5.3-10.6	Bisht et al., 2015a, Pipal et al., 2014
Ahmedabad (India)	18.3	3	Rengarajan et al., 2011
Yokohama (Japan)	4	2	Khan et al., 2010
Beijing (China)	2.9-28.2	1.2-16.3	Guinot et al., 2007

- 1536 Table 7: Comparison of WSOC concentrations with literature data. Only literature data given at a daily
- 1537 scale have been selected.

Location	WSOC (µg.m <sup>-3</sup> )	Reference
Abidjan, Côte d'Ivoire	2-8	This work
Cotonou, Benin	2-8	This work
Beijing, China	9-27	Yu et al., 2018
Beijing, China	4-6	Xiang et al., 2017
Beijing, China	8-12	Tang et al., 2016
Beijing, China	7	Du et al., 2014
Beijing, China	6-8	Feng et al., 2006
Shanghai, China	2-7	Feng et al., 2006, Huang et al., 2012
Guangzhou, Hong Kong	2	Huang et al., 2012
Guangzhou, Hong Kong	5-10	Feng et al., 2006
Gwangju, Korea	2-3.5	Park et al., 2015
Tokyo, Japan	3-23	Sempere and Kawamura, 1994
Cairo, Egypt	3	Favez et al., 2008
Amsterdam, Netherland	1-2	Feng et al., 2007
Barcelone, Spain	1-2	Viana et al., 2007 and 2008
Brindisi, Italy	1.5	Genga et al., 2017
Saint Jean de Maurienne, France	1-5	Sullivan et al., 2004, Jaffrezo et al., 2005a