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Physicochemical Characterization of Air Pollution Particulate Matter (PM_{2.5} and PM_{>2.5}) in an Urban Area of Cotonou, Benin

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Abstract: PM_{2.5} and PM_{>2.5} samples were collected in Cotonou (Benin) using high volume cascade impaction air samplers. The samplings were based on continuous collection over twelve days. Physical and chemical characteristics of samples were determined by size distribution (laser granulometry), specific surface areas (BET method), inorganic elements (ICP-MS), water-soluble ions (IC), CHNS analysis and organic compounds (GC-MS). Average concentrations of air particulate matter were 180.9 µg/m³ and 94.5 µg/m³ in PM_{2.5} and PM_{>2.5}, respectively. The higher water-soluble ions recorded were Ca²⁺, SO₄²⁻, NO₃⁻, Na⁺ and Cl⁻ for both PM. Moreover, concentrations were almost two-fold higher for PM_{2.5} compared to PM_{>2.5}, with 10.7 µg/m³ of total metals found in PM_{2.5} versus 5.6 µg/m³ in PM_{>2.5}. Both PM samples under study presented similar repartition of elements considering their percentages. Results suggested that PM_{>2.5} samples contain agglomerates of fine particles. Identification tools of major pollution source as inorganic elements, paraffins, fatty acids ratios and PAHs ratios indicated that PM under study originated from traffic exhaust.

Keywords: particulate matter concentrations; size distribution; metals; PAHs ratio; Cotonou



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1. Introduction

Air pollution particulate matter (PM) is a complex mixture of metals, salts, organic chemicals and biological materials [1–3]. It is a serious environmental health risk factor for humans [4,5]. Environmental toxicology studies used chemical and physical characterization of PM (source, size, mass, surface area, organic composition, metals, etc.) to assess adverse health effects linked to particles [6–13]. These studies have shown that particle exposure could cause many diseases in both developed and developing countries [14–16]. However, the underlying mechanisms of action generating adverse health effects are still unclear. Airborne particulate matter, especially the fraction of particles with aerodynamic diameters below 2.5 µm (fine particles PM_{2.5}) had large surface areas and adsorbed compounds such as heavy metals, polycyclic aromatic hydrocarbons (PAHs), and many other organic compounds into their surface various. Their small diameters allow them to reach the alveolar compartment of lungs, where they are able to cause adverse effects on health [17–21]. It was shown that these particles cause more toxic effects than coarse particles [22].

Many studies have shown the influence of particle characteristics on biological responses that may cause many diseases [8,23,24]. The particles come from natural and anthropogenic sources such as industries, traffic, heating, etc. [25,26]. In a recent study, Yusuf et al. [27] found that transport activities are the major sources that emit heavy pollutants found in the environment. An urban area with mainly exhaust fumes is a strong source of PM, especially during the rainy season [28,29]. In developing

countries, there is little data concerning the physicochemical characterization of PM and assessment of their adverse health effects [20–30]. In Sub-Saharan Africa countries, little air quality data is available; it appears that having a better knowledge on the air pollution is important [31–33]. Over the past decade, the air quality of Cotonou, the economic capital of Benin, has worsened in the city due to the widespread use of motorbikes with that consume gasoline of doubtful quality as the main fuel source and the presence of old second-hand cars [29,32,34–37]. The city had more than 94,000 motorbikes and more than 350,000 old second-hand cars [34–38]. In such a situation, in order to examine the quality of ambient air, a pilot study has reported that ambient air in Cotonou contained lot of air pollutants, including volatile organic compounds (VOCs) the benzene and its derivatives and ultrafine particles (UFPs) associated with polycyclic aromatic hydrocarbons (PAHs) at high concentrations [39,40].

In this study, samples of ambient particulate matter $PM_{2.5}$ and fraction of particles with aerodynamic diameters above $2.5 \mu m$ ($PM_{>2.5}$) were collected in Cotonou where the road traffic is remarkably dense. The main objective was to determine physical and chemical characteristics of particles in order to choose specific physicochemical parameters for selection of possible tracers of urban activities.

2. Materials and Methods (See the Schematic Diagram, Figure S1)

2.1. Site Description and PM Sampling

Two PM ($PM_{>2.5}$ and $PM_{2.5}$) samples were collected in Cotonou, Benin (Figure 1) and samplings were based on continuous collection from 16 November 2010 to 8 December 2010. Samples were obtained from St Michel ($6^{\circ}22'2.3'' N/2^{\circ}25'46.4'' E$), a district which records dense road traffic. The sampling site (under urban-related emission sources) has SSW/SW as the prevailing wind direction. Other meteorological conditions were an average temperature of $31.4^{\circ}C$, 72% of relative humidity and $3.0 m/s$ average wind speed during the period. Both types of particles were collected using high volume cascade impaction air samplers (Staplex, New York, NY, USA), with the collection method described by Billet et al. [41]. Briefly, the impactor's plates were mounted without any filter and backup filter to maintain a constant aspiration flow rate (i.e., $80 m^3/h$). Two identical impacting systems have been used alternatively and were changed every three days. Samples were pooled after each collection. Sampling pumps were placed to approximately 50 m of the crossroad and about 1.5 m above the ground. ASECNA (Agence pour la Sécurité de la Navigation Aérienne en Afrique et à Madagascar) of the international airport Bernardin GANTIN supplied meteorological data (wind direction and speed, temperature and relative humidity). After PM sampling, impacting plates were dried under a laminar flow bench. Then, PM were removed from plates and divided into two size fractions: particles with aerodynamic diameters above $2.5 \mu m$ (coarse particles $PM_{>2.5}$) and particles with aerodynamic diameters below $2.5 \mu m$ (fine particles $PM_{2.5}$). The two types of particles were kept in cleaned vials and immediately stored at $4^{\circ}C$. An amount of 1 g of $PM_{>2.5}$ and more than 2 g of $PM_{2.5}$ were collected. Thereafter, samples were conveyed for analysis in Dunkerque, France.

2.2. Experimental Analysis

2.2.1. PM Physical Characterization

PM Size Distribution

Each sample was prepared with 20 mg of particle in 50 mL of distilled water and well mixed. Samples suspensions were ultrasonically dispersed before measurement. PM size distribution was studied by using laser granulometry. The laser granulometer (Beckman Coulter LS 13 320 MW with Universal Liquid Module) measures particle size distributions over the range from 0.017 to $2000 \mu m$ of sample suspensions, based on polarization intensity differential scattering (PIDS). In all measurements, beam obscuration was kept within the limits of 8% to 12%. Cumulative frequency and relative frequency size distribution were used for analysis of the results.

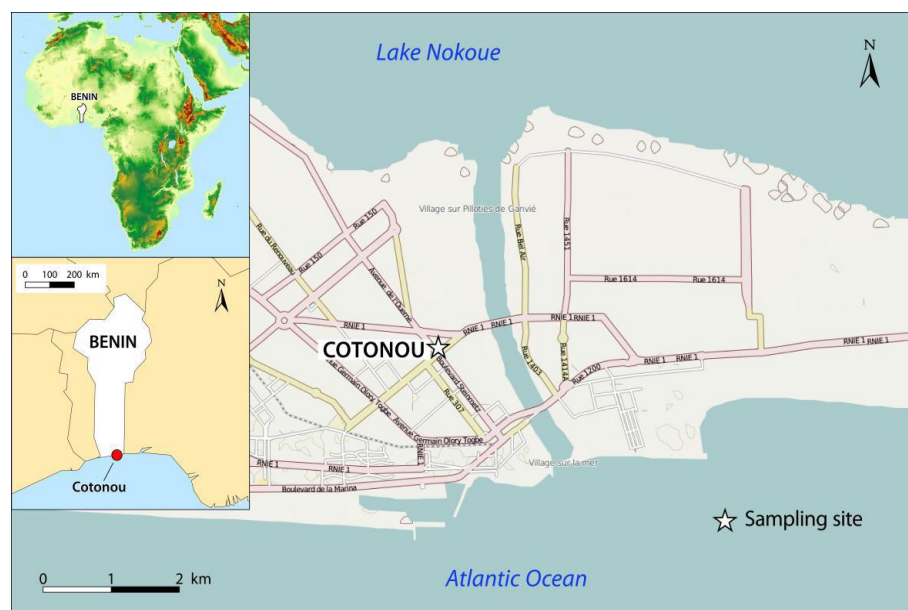


Figure 1. Map of urban sampling site in Cotonou, Benin.

Specific Surface Area

About 50 mg of each PM were placed in a small Pyrex U-shaped cell and analyzed by using a Quantasorb[®] Surface Area Analyzer (Quantachrome Corporation, New York, NY, USA), which is based on Brunauer–Emmett–Teller (BET) theory. Particle samples were outgassed at 200 °C for 1 h. Nitrogen was used as a calibration gas and helium as an inert carrier gas with a rate of 70% versus 30% nitrogen. Thereafter, volumes of pure nitrogen gas were adsorbed to their surface at −196 °C. The amount of nitrogen adsorbed at various partial pressures was used to calculate the sample's surface area.

2.2.2. Inorganic Elements and Water-Soluble Ions

Both PM were mineralized (about 20 mg) using HNO₃ and HClO₄ (ratio 1:2) by microwave digestion (MARS XPRESS, CEM Corporation) (the experimental conditions were 1600 W of power, 180 °C and 20 min of run time). Thereafter, the solutions were filtered by Whatman cellulose filters, diluted up to 25 mL with ultrapure water and analyzed by inductively coupled plasma-mass spectrometry (ICP-MS, Varian[®] 820 MS). Calibration solutions were prepared in a 1% HNO₃ matrix using a commercial 10 ppm multi-element standard (SCP33MS, SCP Science, Baie-d'Urfé, QC, Canada). The calibration range was from 0 to 50 ppb.

About 20 mg of particles collected was leached in 10 mL ultrapure water by ultrasonic agitation for 30 min. Extract solutions were filtered on cellulose acetate filter 0.45 µm. This step was repeated six times on the same aliquot to recover 95% of soluble ions and then 25 µL were injected. Soluble anions concentrations were determined by chemically suppressed ion chromatography (carbonate 1mM/bicarbonate 3.5 mM eluents and AG14A/AS14A columns, 1.2 mL/min) using a Dionex DX 100 ion chromatograph. Soluble cations concentrations were determined by chemically suppressed ion chromatography (methane sulfonic acid 20 mM eluents and CG12A/CS12A columns, 1 mL/min) using a Dionex ICS 900 ion chromatograph. The calibration ranges of soluble anions and soluble cations were from 0.5 to 100 ppm. Chromatographic data were analyzed by Chromeleon[®] software (Thermo Fisher Scientific, Waltham, MA, USA).

2.2.3. Element Composition

Element concentrations (carbon, nitrogen, hydrogen and sulphur) were detected by using CHNS/O Elemental Analysers (FlashEA 1112 equipped to 2 injectors MAS200R, Thermo Fisher Scientific). Carbon is converted by oxidation to carbon dioxide; hydrogen to

water; nitrogen to nitrogen gas/oxides of nitrogen; and sulphur to sulphur dioxide. CO₂, H₂O, N₂ and SO₂ obtained after oxidation and reduction of gases [42,43] were separated by a GC packed column (PTFE, 2 m, 6 × 5 mm), kept at 70 °C and detected by a thermo-conductivity detector. A quartz reactor was used to determine elements and kept at 950 °C with 140 mL/min of He flow as a carrier gas. For oxidation of the samples, a pressure of 250 kPa (He) and 220 kPa (O₂) were required. Cycle run time, sampling delay and oxygen injection end were 600, 25, 17 s, respectively. PM samples were previously introduced in tin capsules and packed carefully. Sulphanilamide (Thermo Fisher Scientific) was used as the standard of calibration. After first analysis, each sample was incinerated at 450 °C in order to remove organic carbon and then analyzed again to determine mineral carbon. The amounts of gases in column output were proportional to the carbon, hydrogen, nitrogen and sulfur contained in the samples.

2.2.4. Organic Compounds

The volatile organic compounds (VOCs) analytical method was based on the protocol described by Caplain et al. [44]. Samples were previously trapped in glass tubes and VOCs were desorbed by thermal treatment at 220 °C using GC/MS (Combi Injector/Desorber module-EM640 Brüker). Desorption time was 3 min to desorb VOCs and compounds from C₁₀ to C₁₅. These conditions prevented the desorption of the semi-volatile compounds subsequently extracted with solvents.

After thermal desorption, the two particle samples were extracted by soxhlet extraction with 100 mL of dichloromethane for 16 h to determine polycyclic aromatic hydrocarbons (PAHs), paraffins and carboxylic acids. The extracts were concentrated under nitrogen flux. An amount of 1 µL of each sample was injected in a gas chromatograph (VARIAN3800) coupled to a mass spectrometer (VARIAN1200 TQ). The capillary column is a factor four VF-5 ms (30 m × 0.25 mm × 0.25 µm) and used helium as the carrier gas. The total time for this analysis was 60 min; 40 °C for 5 min; 5 °C/min up to 310 °C; and 310 °C for 3 min. The parameters of the mass detector were an impact electronic ion current = 70 eV and a source temperature of 280 °C. The samples were analyzed between 40 and 350 mass units. Compounds were identified by comparing retention times of chromatographic peaks from samples with those from standard mixtures and by comparing mass spectra with those contained in NIST and/or WILEY libraries.

3. Results and Discussion

3.1. PM Physical Characteristics

In this study, the St Michel district (sampling site) recorded SSW as the dominant wind direction. During the sampling period, average concentrations of air particulate matter were 180.9 µg/m³ for PM_{2.5} and 94.5 µg/m³ for PM_{>2.5}. In Lagos (located in Nigeria, which is near to Benin), Adeleke et al. [45] reported extremely high levels of up to 272.8 µg/m³ for PM_{2.5}. Our PM_{2.5} and PM_{>2.5} concentrations were higher than those previously reported in Dakar (Senegal): 85 µg/m³ and 57 µg/m³, respectively [46]. Figure 2 shows the size distribution by cumulative frequency (%) and relative frequency (%). The total PM numbers with geometric diameter ≤ 2.5 µm were identical (97.5%) in both PM. The highest particle numbers detected in size classes were also identical (55.2%) for 0.5–1.0 µm. For the 0.33–0.5 µm class, proportions of 24.0% and 22.5% were found for PM_{2.5} and PM_{>2.5}, respectively.

Studies conducted in West Africa have also shown a significant level of PM_{2.5} fraction in ambient air of urban city. However, distributions found in this work were markedly higher than those recorded in Abidjan (88.3% of fine particle number, [20] and Dakar (94.7% of fine particle number, [9]). Other countries have found PM concentrations markedly lower than those recorded in this work (Table 1).

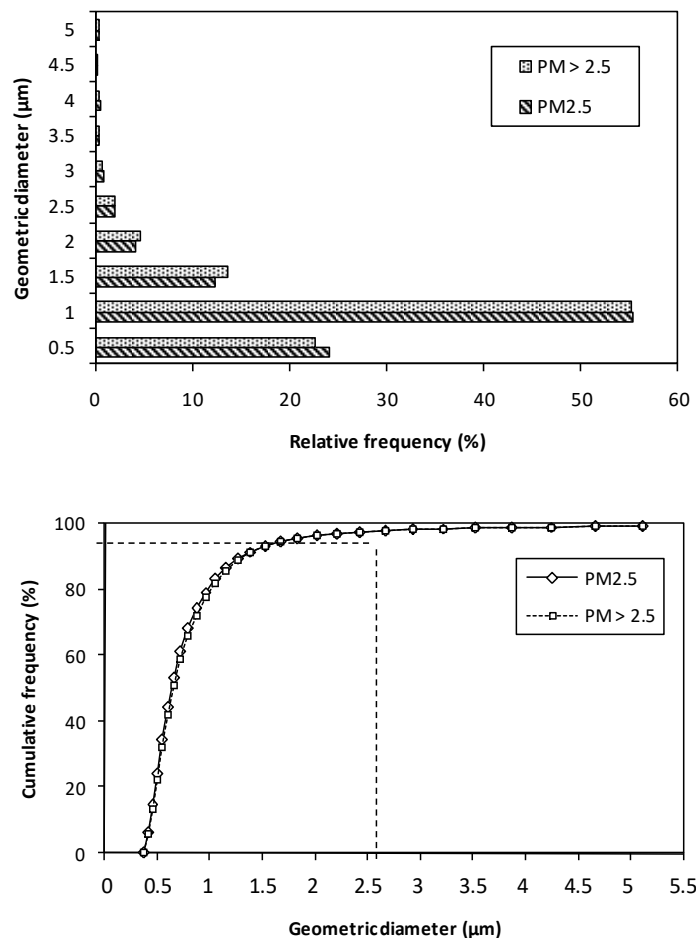


Figure 2. Size distribution by cumulative frequency (%) and relative frequency (%) of PM_{2.5} and PM_{>2.5} collected in an urban site, Cotonou.

Table 1. Concentrations of fine and coarse particles and inorganic water-soluble ions measured in different continents.

| Continent | City/Country | Concentration (µg/m ³) | | Sulphate | | | | Nitrate | | | | References |
|---------------|-------------------------|------------------------------------|------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|------------|
| | | | | PM _{2.5} | | PM ₁₀ | | PM _{2.5} | | PM ₁₀ | | |
| | | PM _{2.5} | PM ₁₀ | µg/m ³ | % | µg/m ³ | % | µg/m ³ | % | µg/m ³ | % | |
| Africa | Present study | | | | | | | | | | | |
| | Cotonou (Benin) | 180.9 | 94.5 | 1.4 | 21.5 | 0.5 | 16.6 | 0.8 | 13.0 | 0.2 | 5.8 | |
| | Tetouan (Morocco) | 18 | - | 3 | 17 | - | - | 1 | 6 | - | - | [47] |
| | Addis Ababa (Ethiopia) | 19–127 | - | 1.3–5.3 | 5.7 | - | - | 0.1–0.5 | 0.6 | - | - | [48] |
| | Dakar (Senegal) | 87 | 57 | 2.5 | - | 1.6 | - | 1.9 | - | 0.5 | - | [46] |
| | Dakar (Senegal) | 75.1 | - | 1.9 | 17.3 | - | - | 1.6 | 14.3 ^a | - | - | [9] |
| | Nairobi (Kenya) | 10.7–98.1 | - | - | - | - | - | - | - | - | - | [49] |
| | Abidjan (Côte d'Ivoire) | 105.1 | - | 1.1 | 21.6 | - | - | 1.6 | 31.7 ^a | - | - | [20] |
| | Conakry (Guinea) | 38–177 | 80–358 | 1.5 | 31.1 ^a | 2.2 | 21.3 ^a | 0.9 | 20.2 ^a | 3.4 | 32.9 ^a | [50] |
| Accra (Ghana) | 21–39 | 49–96 | - | - | - | - | - | - | - | - | [51] | |
| Asia | Xiamen (China) | 32.8 | 54.5 | 4.1 | - | 4.6 | - | 2.2 | - | 3.9 | - | [52] |
| | Delhi (India) | 125.5 | - | 13.1 | - | - | - | 10.8 | - | - | - | [53] |
| | Yokohama (Japan) | 20.6 | 9.6 | 3.8 | 48.3 ^a | 0.2 | 7.6 ^a | 1.0 | 12.2 ^a | 1.0 | 34.7 ^a | [54] |
| Europe | Athens (Greece) | 18.0 | 33.5 | 3.7 | - | 6.0 | - | 1.0 | - | 2.0 | - | [55] |
| | Palermo (Italy) | 29 | 42 | 2.5 | - | 2.3 | - | 2.9 | - | 4.1 | - | [56] |
| | Dunkerque (France) | 9.9 | - | 0.5 | 18.2 ^a | - | - | 0.8 | 32.5 ^a | - | - | [8] |
| | Barcelona (Spain) | 20.0 | 26.3 | - | 24 | - | 2.1 | - | 6.8 | - | 11 | [57] |
| | Duisburg (Germany) | 14.7 | 7.2 | - | 19 | - | 3.4 | - | 13 | - | 12 | |
| | Amsterdam (Netherland) | 25.4 | 8.4 | - | 14 | - | 5.3 | - | 18 | - | 14 | |
| America | São Paulo (Brazil) | 11–43 | 17–60 | - | - | - | - | - | - | - | - | [58] |

(-) = not available. ^a percentage is calculated considering raw data.

In a nutshell, concentrations recorded in African countries, particularly Benin, were high and exceeded the PM concentrations of air quality guidelines revised on September, 2021 by the World Health Organization [59]. These guidelines promulgated that the highest level of PM_{2.5} should be 15 µg/m³ (24 h average concentration) and 5 µg/m³ (annual average). For PM₁₀, the concentrations are 15 and 45 µg/m³ for annual and 24 h average concentration, respectively.

On the other hand, the studied particulate matters had higher specific surface areas ranging from 10 m²/g to 12 m²/g for PM_{2.5} and PM_{>2.5}, respectively. Dieme et al. [9] and Kouassi et al. [20] had found similar values in urban cities (12 m²/g and 9 m²/g in Dakar and Abidjan, respectively). A recent study undertaken in Dakar found a specific surface area value of 10 m²/g for PM_{2.5} and 7 m²/g for PM_{>2.5} [46]. In reference materials, SRM1649a (mainly of coarse particles) and CRM8 (mainly of fine or ultrafine particles), values of specific surface areas were 35 m²/g for fine particles and 2 m²/g for coarse particles [60]. The high values of specific surface area recorded in this study were likely to be associated with carbonaceous particulate emissions from combustion processes, such as vehicular emissions. Similar results in PM_{2.5} and PM_{>2.5} suggest that PM_{>2.5} contain aggregates and/or agglomerates of fine particles within it. Particles having high specific surface areas are known to absorb chemical compounds such as volatile organic compounds and polycyclic aromatic hydrocarbons [41,61].

3.2. Inorganic Elements and Water-Soluble Ions

Water-soluble ions concentrations are presented in Figure 3. Ionic concentrations in PM_{2.5} were higher than those in PM_{>2.5}. The most abundant compounds were Ca²⁺ (2.5 µg/m³/39.2% for PM_{2.5} versus 1.1 µg/m³/37.7% for PM_{>2.5}), SO₄²⁻ (1.4 µg/m³/21.5% versus 0.5 µg/m³/16.6%) and NO₃⁻ (0.8 µg/m³/13.0% versus 0.2 µg/m³/5.8%). Na⁺ and Cl⁻ have nearly the same concentration values (0.6 µg/m³ for PM_{2.5} versus 0.5 µg/m³ for PM_{>2.5}). Na⁺ and Cl⁻ also presented similar percentages (9.8% and 9.7% for PM_{2.5}, respectively, versus 15.8% and 17.9% for PM_{>2.5}). Total water-soluble ions concentrations of PM_{2.5} were 6.3 µg/m³ versus 3.0 µg/m³ for PM_{>2.5}.

The most abundant water-soluble ions concentrations recorded were Ca²⁺, SO₄²⁻, NO₃⁻, Na⁺ and Cl⁻ for both types of PM. Na⁺, Cl⁻ and Mg²⁺ revealed sea salt influences [17,62]. While the high concentration of Ca²⁺ could come from natural erosion and/or dust from the cement industry, which is located approximately 1 km from the sampling site. Cazier et al. [17] found a similar result concerning amounts of CaCO₃ which are significant. Sulphates and nitrates clearly revealed anthropogenic activities [8,9,17,54,62]. In West Africa, values of sulphate concentration are often high. Sulphates present in the studied particulate matter could come from natural sources such as the surface of the Atlantic Ocean due to the proximity of the West African countries of the sea [63,64]. With regards to nitrate concentrations found in PM_{2.5}, they were also similar to those recently published [8,17,50,54]. Moreover, this nitrate value was lower than Dakar's nitrate concentration (Table 1). The PM_{>2.5} nitrate value was smaller than those reported in Japan (1.0 µg/m³/10.6%, [54]). However, the nitrate value observed in this work indicated an anthropogenic contribution. Nitrates in PM come primarily from NO_x (NO and NO₂) and are mostly of anthropogenic origins. They come from high temperature combustion processes and their gas-particle conversion in the atmosphere forms stable nitrates in the particulate state [65].

Otherwise, concentrations were almost two-fold higher in PM_{2.5} than in PM_{>2.5} with total metals of 10.78 µg/m³ found in PM_{2.5} versus 5.68 µg/m³ in PM_{>2.5} (Table 2). Among the main elements (Al, Fe, Mg, Mn, Na, Ti and Zn) found in particles, Al and Fe represent the highest concentrations in the two types of samples. Medium studied elements Ba, Cr, Cu, Pb and Sr showed concentrations ranging from 0.01 to 0.04 µg/m³ in PM_{2.5} and from 0.005 to 0.02 µg/m³ for PM_{>2.5}. Concentrations of other elements were relatively low (0.0005–0.001 µg/m³) in analyzed samples. Both PM samples under study presented similar repartition of elements as considering their percentage.

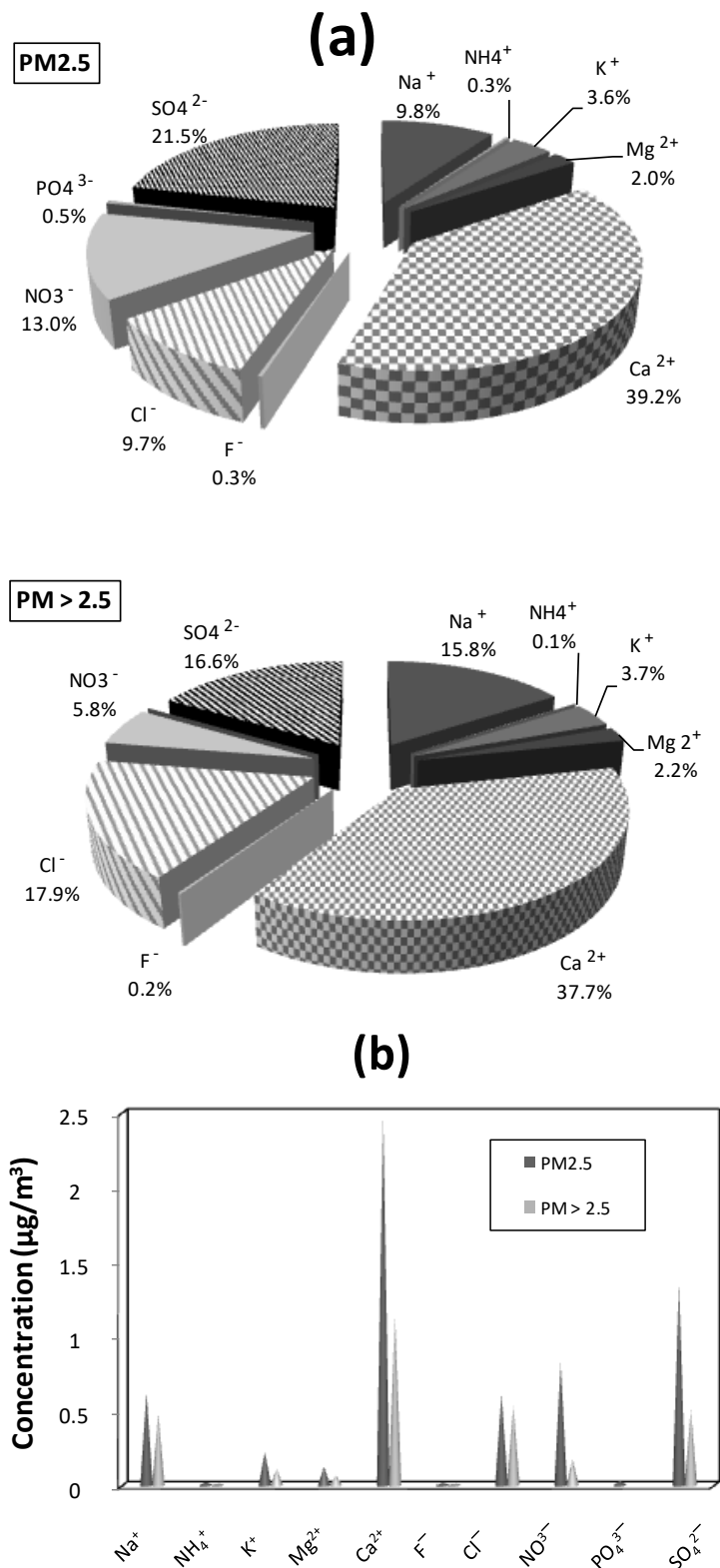


Figure 3. Proportion in % (a) and concentrations in µg/m³ (b) of water-soluble ions in the PM_{2.5} and PM_{>2.5} samples collected in an urban area of Cotonou (Benin).

Table 2. Inorganic elements of air pollution particulate matter collected.

| Metal and Trace | PM _{2.5} | | PM _{>2.5} | |
|-----------------|-------------------|-----|-----------------------|-----|
| | µg/m ³ | % | µg/m ³ | % |
| Al | 4.5 | 42 | 2.3 | 40 |
| Ba | 0.04 | 0.4 | 0.02 | 0.4 |
| Ce | 0.006 | 0.1 | 0.003 | 0.1 |
| Co | 0.001 | 0 | 0.0005 | 0 |
| Cr | 0.01 | 0.1 | 0.005 | 0.1 |
| Cu | 0.03 | 0.3 | 0.007 | 0.1 |
| Fe | 3.7 | 34 | 2 | 35 |
| La | 0.002 | 0 | 0.001 | 0 |
| Li | 0.001 | 0 | 0.001 | 0 |
| Mg | 0.8 | 7 | 0.4 | 7 |
| Mn | 0.07 | 0.6 | 0.03 | 0.6 |
| Na | 1.3 | 12 | 0.8 | 14 |
| Ni | 0.002 | 0 | 0.001 | 0 |
| Pb | 0.02 | 0 | 0.01 | 0.2 |
| Rb | 0.002 | 0 | 0.001 | 0 |
| Sn | 0.001 | 0 | 0.0005 | 0 |
| Sr | 0.03 | 0.2 | 0.01 | 0.2 |
| Ti | 0.13 | 1 | 0.06 | 1.0 |
| V | 0.003 | 0 | 0.002 | 0.0 |
| Zn | 0.1 | 1 | 0.07 | 1.3 |
| Total | 10.8 | | 5.7 | |

In literature, inorganic elements were usually associated with natural environment (e.g., Na, Mg, Ca, Ti) and anthropogenic origin (e.g., Fe, Al, Mn, Ba, Cr, Zn, Pb, Cu, Cd) [8,9]. Moreover, Cr, As, Ni, Pb, Zn, Cu, V and Cd are carcinogenic, Cd and As are potentially mutagenic and Pb and Hg are foetal toxic [66,67]. The main sources of atmospheric heavy metals are natural emissions (marine spray and Saharan dust), heavy fuel oil combustion, non-exhaust road traffic and industrial metallurgical processes [68–70]. Different researchers have attributed different groups of heavy metals to natural sources. Ni, Mn and Co concentrations were attributed to natural sources [71]. Kelepertzis [72] attributed Ni, Cr, Co and Fe to natural origin. In this paper, the main concentrations of Al, Fe, Mg, Mn, Na, Ti and Zn could come mainly from anthropogenic sources. Vehicular emissions, waste disposal, sewage and industrial emissions are known to be important sources of urban atmospheric heavy metals [73–75]. Inorganic elements in Cotonou were high during the sampling period. High concentrations of Fe and Al were often measured in African countries when sites were submitted to soil dust resuspension from arid regions [70,75]. Dieme et al. [9], Kouassi et al. [20] and Weinstein et al. [50] had shown similar results in Abidjan, Dakar and Conakry, respectively. According to regulatory standards set by the European Parliament and the Council of the European Union [76], lead levels in the two types of particles were higher than those detected by Dieme et al. [9]. Other results around the world are presented in Table 3. Metal concentrations of PM_{2.5} fraction are twice higher than those of PM_{>2.5}. Lee et al. [77] made a similar observation. This was likely because heavy metals are more easily accumulated in PM_{2.5} than in PM_{>2.5}. Metals as Cr and Cu are known to be derived from non-exhaust emissions (brake wear) [78]. The present study showed that Cu and Cr concentrations are more than twice as high in PM_{2.5} as in PM_{>2.5}. Wang et al. [79] had found similar concentrations of Cu for PM_{2.5} in China, but their Cr values were three times higher than our results. In addition, Zn is associated with tire wear [80]. Zn concentrations were 145 ng/m³ and 71 ng/m³ for PM_{2.5} and PM_{>2.5}, respectively. These Zn concentrations in PM_{2.5} were higher than those found in Ethiopia (3 ng/m³), Senegal (2 ng/m³) and Greece (4 ng/m³), which are countries that have similar values of Zn [9,48,55]. However, other studies carried out in China showed Zn concentrations of up to 465 ng/m³ and 590 ng/m³ for PM_{2.5} and PM₁₀, respectively [79]. On the other hand, Benin adopted unleaded gasoline, so the main sources of heavy metals of two PM samples from Cotonou are probably

influenced by road traffic and notably non-exhaust emissions and/or cement industry emissions [81,82].

Table 3. Concentration of heavy metals (ng/m³) in the ambient air of urban areas in different countries around the world.

| City/Country | | Pb | Zn | Ni | Cu | Cd | As | Cr | V | Reference |
|------------------------|-----------------------|-----|-----|----|-----|-----|-----|-----|----|------------|
| Present study | PM _{2.5} | 22 | 145 | 2 | 29 | - | - | 11 | 3 | This study |
| Cotonou (Benin) | PM _{>2.5} | 11 | 71 | 1 | 7 | - | - | 5 | 2 | |
| Xiamen (China) | PM _{2.5} | 19 | 87 | 7 | 7 | 0.5 | 2 | 8 | 11 | [52] |
| | PM ₁₀ | 22 | 116 | 8 | 11 | 0.6 | 2 | 11 | 12 | |
| Addis Ababa (Ethiopia) | PM _{2.5} | 8 | 3 | - | - | 0.5 | 1 | - | - | [48] |
| Athens (Greece) | PM _{2.5} | 11 | 4 | 4 | 8 | 0 | 1 | 4 | 9 | [55] |
| | PM ₁₀ | 15 | 6 | 7 | 18 | 1 | 1 | 7 | 13 | |
| Delhi (India) | PM _{2.5} | 2 | 13 | - | 4 | - | 7 | 8 | - | [53] |
| Baoshan (China) | PM _{2.5} | 133 | 465 | 27 | 29 | 3 | - | 31 | - | [79] |
| | PM ₁₀ | 137 | 590 | 32 | 41 | 3 | - | 56 | - | |
| Beijing (China) | PM _{2.5} | 261 | 0.2 | 29 | 117 | 13 | 51 | 86 | 18 | [74] |
| Dakar (Senegal) | PM _{2.5} | 7 | 2 | 1 | 5 | - | - | 2 | 2 | [9] |
| Dunkerque (France) | PM _{2.5} | 4 | 17 | 2 | 9 | - | 0.2 | 4 | 1 | [8] |
| Punta Sabbioni (Italy) | PM ₁₀ | 21 | 57 | 4 | 10 | - | 2 | 5 | 4 | [83] |
| Navarra (Spain) | PM _{2.5} | 2 | 18 | 1 | 12 | - | 0.2 | 2 | - | [84] |
| | PM ₁₀ | 3 | 29 | 2 | 27 | - | 0.2 | 3 | - | |
| Conakry (Guinea) | PM _{2.5} | 36 | 69 | 8 | 179 | - | - | 24 | 14 | [50] |
| | PM ₁₀ | 42 | 37 | 18 | 10 | - | - | 144 | 22 | |

(-) not available.

3.3. Element Composition

Total carbon was the highest rate of elements in the two analyzed aerosols (Table 4). Total carbon was found to 22.97% in PM_{2.5} versus 14.09% for PM_{>2.5}. Percentage of hydrogen were higher than nitrogen and sulfur in the two samples.

Table 4. Element composition of two aerosols collected.

| Element | Percentage | |
|-----------|-------------------|-----------------------|
| | PM _{2.5} | PM _{>2.5} |
| Total C | 22.97 ± 0.11 | 14.09 ± 0.02 |
| Organic C | 21.21 ± 0.10 | 12.81 ± 0.02 |
| Nitrogen | 0.94 ± 0.04 | 0.85 ± 0.01 |
| Hydrogen | 3.04 ± 0.01 | 1.81 ± 0.01 |
| Sulphur | 1.16 ± 0.00 | 0.96 ± 0.01 |
| EC/TC | 0.076 | 0.09 |

EC/TC = Elemental carbon/Total carbon.

The total carbon in PM is the sum of OC and EC; the EC levels are generally lower than the OC values [31]. In urban area, the contribution of TC in PM_{2.5} was 40% in Ashwaina, Ghana [85], 59% in Dar es Salaam, Tanzania [86] and is 23% in the present study. The reported EC for PM_{2.5} (7.6%) and PM_{>2.5} (9.0%) are quite similar. The EC/TC ratio provides insight to the sources of the carbonaceous aerosol including biomass combustion and traffic [31]. EC/TC ratios between 0.1 and 0.2 have been associated with biomass-burning activities in Morogoro, Tanzania [86]. These results are similar to values found in India [87]. In Ashaiman, Ghana, this ratio was 0.21 [85]. Our results are far above those observed in Ghana and Tanzania. In this paper, the OC level was 21% and 13% for PM_{2.5} and PM_{>2.5},

respectively. In Dar es Salaam, Tanzania reported OC percentages of 36% for PM_{2.5} and 24% for PM₁₀; these values are attributed to traffic emissions [86].

In Eastern Spain, organic carbon (OC) + elemental carbon (EC) were higher than those of this study. These percentages were 32% and 22% for PM_{2.5} and PM₁₀, respectively [62]. In Yokohama (Japan), OC + EC can reach 28% and 16% in PM_{2.5} and PM₁₀, respectively [54]. A recent study in Delhi (India) reported a value of 22% of OC + EC [53]. The ratio of (OC + EC) PM_{2.5}/(OC + EC) PM₁₀ were similar in Spain, Japan and Benin. Results of study conducted by Aldabe et al. [84] in Spain were contrary to those in the present work; OC + EC concentrations were rather abundant in PM₁₀ compared to PM_{2.5}. In accordance with Rodriguez et al. [62], OC + EC associated with phosphate and ozone should be due to the influence of bio-mass burning. In the present study, phosphate was practically absent and lead, nitrate, OC + EC, nickel were relatively high. Thus, the two PM could mainly come from vehicle exhaust. Moreover, both PM presented similar repartition of elements as compared to the total carbon. This confirms the same source of PM found in metal analysis. The uppermost concentrations of organic carbon were often linked with organic compounds.

3.4. Organic Compounds

In this study, the volatile organic compounds (VOCs) detected in analyzed samples were benzene and its derivatives. VOCs concentrations were more concentrated (seven times) in PM_{2.5} (8.9 ng/m³) than in PM_{>2.5} (1.2 ng/m³) (Table 5). However, these concentrations were lower in PM_{2.5} than those reported by Dieme et al. [9] and Dergham et al. [8] in the Dakar urban site (37.4 ng/m³) and Dunkerque (51.3 ng/m³), respectively. The toluene/benzene (T/B) ratio found in PM_{2.5} samplings was 3.9 and 1.4 in PM_{>2.5}. According to the literature, the T/B ratio ranges from 1.5 to 4.0 in urban areas indicated traffic emissions [88–91].

Table 5. Organic compounds in the two air pollution PM samples.

| VOCs | Concentration (ng/m ³) | | |
|-------------------------|------------------------------------|-----------------------|-------|
| | PM _{2.5} | PM _{>2.5} | |
| Benzene | 0.5 | 0.05 | |
| Toluene | 1.8 | 0.07 | |
| Ethylbenzene | 0.8 | 0.10 | |
| o-Xylene | 3.5 | 0.07 | |
| 123Trimethylbenzene | 1.2 | 0.71 | |
| Trimethylbenzene | 0.5 | <LQ | |
| Tetramethylbenzene | 0.3 | 0.20 | |
| Pentamethylbenzene | 0.3 | <LQ | |
| Total | 8.9 | 1.2 | |
| PAHs | | | |
| Naphtalene | Nap | 0.01 | 0.01 |
| Acenaphtylene | Ace | <LQ | 0.004 |
| Phenanthrene | Phe | 0.02 | 0.01 |
| Fluoranthene | Flu | 0.1 | 0.03 |
| Pyrene | Pyr | 0.3 | 0.06 |
| Benzo[a]Anthracene | BaA | 0.2 | <LQ |
| Chrysene | Chr | 0.3 | 0.01 |
| Benzo[b]Fluoranthene | BbF | <LQ | 0.02 |
| Benzo[k]Fluoranthene | BkF | 0.3 | <LQ |
| Benzo[a]Pyrene | BaP | 0.2 | 0.01 |
| Dibenzo[a.h]Anthracene | DahA | 0.2 | <LQ |
| Indeno[1.2.3-c.d]Pyrene | Ind | <LQ | 0.07 |
| Benzo[g.h.i]Perylene | BghiP | 0.4 | <LQ |
| Total | | 2 | 0.2 |

<LQ = Under limit of quantification; VOCs = Volatile Organic Compounds, PAHs = Polycyclic Aromatic Hydrocarbons.

Polycyclic aromatic hydrocarbons (PAHs) concentrations were far more concentrated in PM_{2.5} than in PM_{>2.5}. Reche et al. [92] and Li et al. [93] have found a similar result in Barcelona (Italy) and Beijing (China), respectively. The total PAHs levels in this work were lower than those found in Europe [94], Germany [95] and France [8]. However, our PAHs concentrations were higher than those reported by Pindado et al. [96] (in Spain). The quantity of benzo[a]pyrene, a leader of PAHs and classified as a carcinogenic B2 pollutant by the EPA, was smaller in PM_{>2.5} and greater in PM_{2.5} than those of recent studies performed in Spain [92,97]. Generally speaking, most of the PAHs have anthropogenic origins (vehicle exhaust, residential heating and power plants) and are usually distinguished by their emission sources using diagnostic ratios [93,98–100]. According to PAHs ratios in Table 6, the two types of PM are presented same ratio. BaP/(BaP + Chr), Flu/(Flu + Pyr), BaP/BghiP, BaA/(BaA + Chr) and Σ COMB/ Σ PAHs were 0.5, 0.3, 0.6, 0.4 and 0.9, respectively, indicating that PAHs adsorbed on PM can come from gasoline, diesel emissions, incomplete combustion and mainly road traffic emissions.

Table 6. Characteristic ratios of PAHs used for source identification.

| PAH Ratio | Value Range | Origin | Reference |
|------------------------------|-------------|-----------------------|-----------|
| Σ COMB/ Σ PAHs | ~1 | Combustion | [101] |
| BaP/BghiP | <0.6 | Non-traffic emissions | [102] |
| BaP/(BaP + Chr) | >0.6 | Traffic emissions | [103–105] |
| BaP/(BaP + Chr) | 0.5 | Diesel emissions | [103–105] |
| BaP/(BaP + Chr) | >0.5 | Gasoline emissions | [103–105] |
| BaA/(BaA + Chr) | 0.2–0.35 | Coal combustion | [106] |
| BaA/(BaA + Chr) | >0.35 | Vehicle emissions | [106] |
| Flu/(Flu + Pyr) | >0.5 | Diesel emissions | [107] |
| Flu/(Flu + Pyr) | <0.5 | Gasoline emissions | [107] |

Σ PAHs = sum of total non-alkylated PAHs. Σ COMB = Σ BaP, Chr, Flu, Pyr, BaA, Ind, BkF, BbF and BghiP.

Traffic emissions usually produce long chain *n*-alkanes. Concentrations of these compounds were higher in PM_{2.5} than in PM_{>2.5} (Figure 4). However, PM_{>2.5} were more concentrated in heavy paraffins (C₂₉ to C₃₆), with the exception of C₃₃.

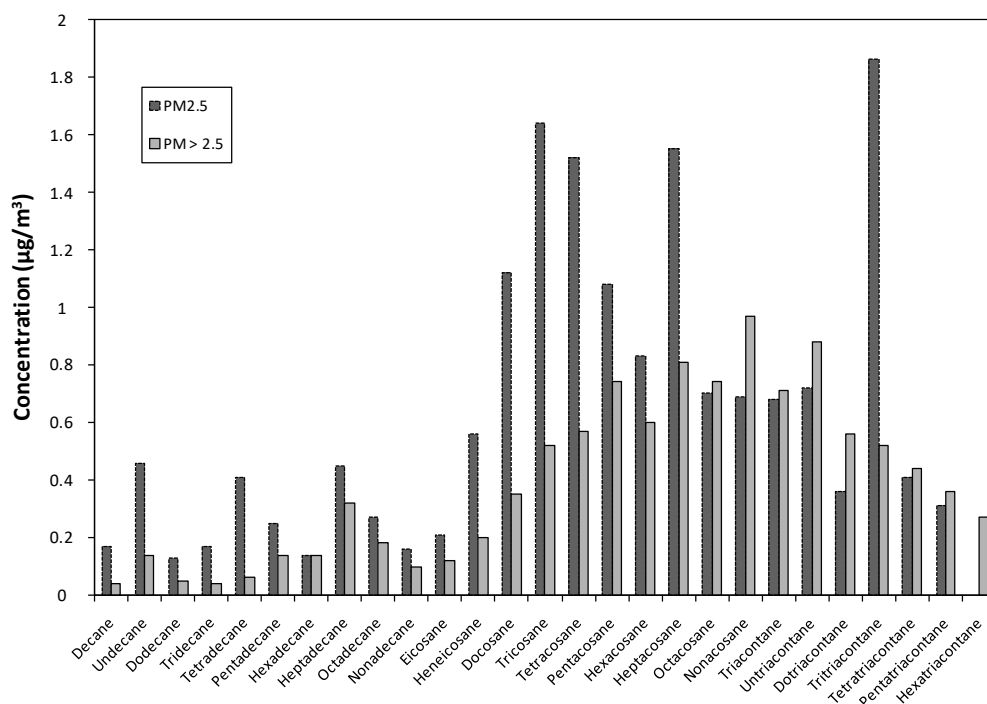


Figure 4. Paraffins concentrations in the two air particulate matter.

In this work, total paraffins were 16.8 ng/m^3 and 10.6 ng/m^3 for $\text{PM}_{2.5}$ and $\text{PM}_{>2.5}$, respectively. For $\text{PM}_{2.5}$, paraffin concentration was higher than those found at Dakar (Senegal) [9], Augsburg (Germany) [95] and Chapineri'a (Spain) [96]. Tefera et al. [33] showed $60.01 \pm 16.91 \text{ ng/m}^{-3}$ for concentrations of C28-31 n-alkanes. These values can reach concentrations fifty times higher in Beijing city (China) [93]. Indeed, C₁₃-C₁₉ indicated microbiota and diesel origin. On the other side, C₂₀-C₃₇ were attributed to fossil fuel rubbish and plant waxes [17,95,108]. In accordance with Kotianová et al. [109], *n*-alkanes up to C₂₀ and C₂₅ are used as sources in gasoline powered vehicles and heavy duty diesel trucks, respectively. This study showed that paraffins in PM samples were mainly resulting from gasoline and diesel emissions, thus traffic emissions. These observations are in agreement with results obtained in the PAHs study.

Otherwise, saturated fatty acids indicate the presence of agricultural activity. Therefore, Figure 5 reports significant saturated carboxylic acids in studied PM samples. Hexadecanoic (palmitic) acid (C₁₆) and octadecanoic (stearic) acid (C₁₈) were most abundant. A similar remark was published by Li et al. [93]. The concentrations were $120.45 \text{ } \mu\text{g/m}^3$ and $31.89 \text{ } \mu\text{g/m}^3$ in $\text{PM}_{2.5}$ for C₁₆ and C₁₈, respectively. The $\text{PM}_{>2.5}$ had two times lower C₁₆ ($52.83 \text{ } \mu\text{g/m}^3$) and C₁₈ ($13.94 \text{ } \mu\text{g/m}^3$) than $\text{PM}_{2.5}$. The total fatty acids were $330.76 \text{ } \mu\text{g/m}^3$ for $\text{PM}_{2.5}$ and $196.31 \text{ } \mu\text{g/m}^3$ for $\text{PM}_{>2.5}$.

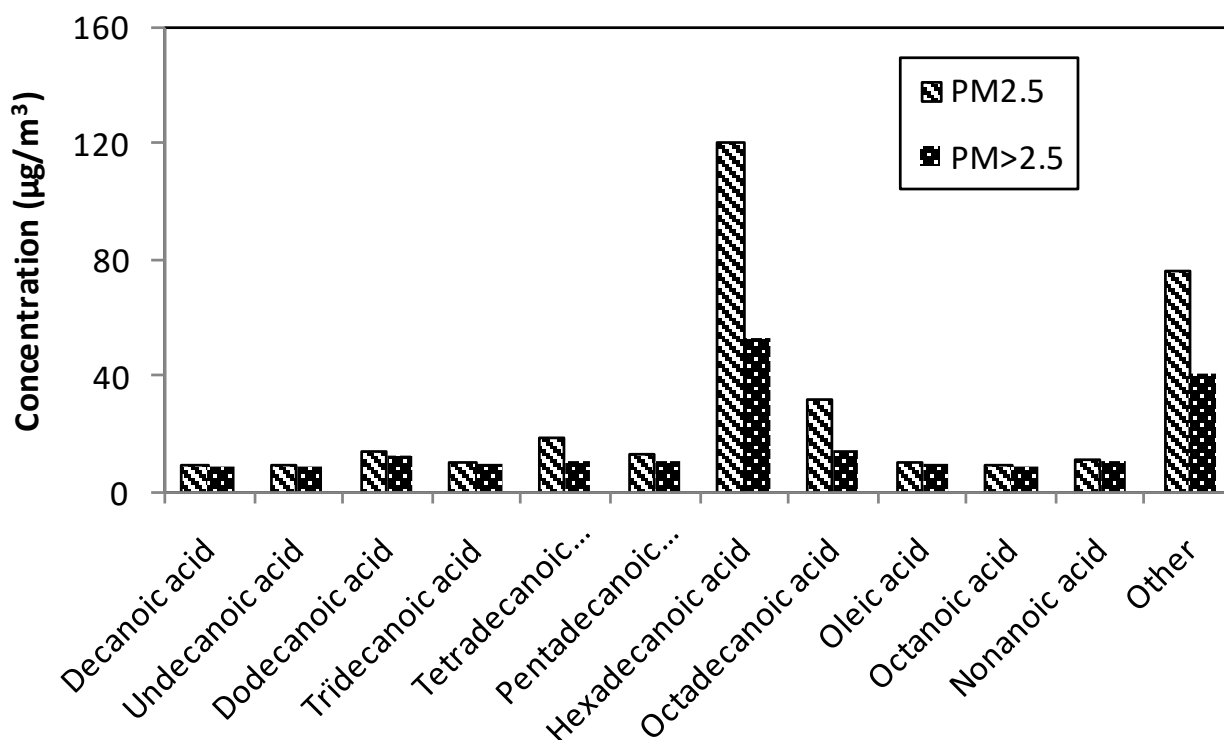


Figure 5. Carboxylic acids with a long aliphatic tail of two air pollution particulate matter.

According to Oliveira et al. [110], fatty acids with even-numbered carbons of less than 20 carbons are considered to be derived from microbial activities and cooking meat. The ratio of oleic acid/stearic acid is a tool to identify aerosol aging [93]. Rogge et al. [111] suggested that when ratio of stearic acid/palmitic acid < 0.25 , it originates from wood smoke and foliar vegetation combustion; between 0.25 and 0.5, it stems from car and diesel truck emissions; and is road dust when ranging from 0.5 to 1.0. In the present work, stearic acid/palmitic acid ratios were 0.26 in the two analyzed PM. Oleic acid/stearic acid ratios were 0.32 and 0.7 in $\text{PM}_{2.5}$ and $\text{PM}_{>2.5}$, respectively. Li et al. [93] reported a similar result in $\text{PM}_{2.5}$ according to the collection period in Beijing. In light of these results, fatty acids concentrations indicated mostly vehicle emissions and rejoined conclusions of previous analyses.

4. Conclusions

Physicochemical characterization of air pollution particulate matters PM_{2.5} and PM_{>2.5} collected in Cotonou revealed astounding identical granulometry profile and specific surface areas between particles samples. This suggests that particle PM_{>2.5} samples contain agglomerates of fine particles. However, concentrations of different inorganic and organic compounds were higher in PM_{2.5} than in PM_{>2.5} every time. Tools to identify sources were used to detect pollutants origin. Inorganic elements, PAHs, paraffins and fatty acids levels studies clearly indicated that both PM came from natural and/or anthropogenic emission sources, particularly, vehicle exhaust influence and traffic emissions. These compounds have adverse health effects, and it is necessary to complete research with toxicological studies in order to establish a monitoring plan of air quality in African countries. Moreover, the results of this characterization study of airborne particulate matter can be used as relevant data for establishing air quality management policies. Policy makers should focus on anthropogenic sources with a high impact on PM concentration to achieve a better air quality in highly populated cities of Benin.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/atmos14020201/s1>, Figure S1. Schematic diagram of the whole study on Cotonou's air pollution impact on human's health.

Author Contributions: Conceptualization, F.B.C., F.C., D.C. and A.V.; methodology, F.B.C., D.D., P.G., A.D., A.V. and F.C.; software, F.B.C.; validation, F.C., A.V. and D.C.; formal analysis, F.B.C. and A.V.; investigation, F.B.C., D.D., P.G., A.D., F.C., D.C. and A.V.; resources, F.B.C. and A.V.; data curation, F.B.C. and A.V.; writing—original draft preparation, F.B.C. and A.V.; writing—review and editing, F.C., A.V. and D.C.; visualization, F.B.C., F.C., A.V. and D.C.; supervision, F.C., A.V. and D.C.; project administration, L.A.-F., F.A., A.S., F.C., A.V. and D.C.; funding acquisition, L.A.-F., F.A. and A.S. All authors have read and agreed to the published version of the manuscript.

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