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Assessment of polycyclic aromatic hydrocarbons contamination in urban soils from Niamey, Niger

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) are produced during incomplete organic matter combustion at high temperature and have potentially mutagenic and carcinogenic effects. No data on PAHs in soils from Niger have been reported despite the existence of numerous PAH emissions sources. This study provides data on PAH contamination of urban soils in Niamey. For this purpose, soil samples were collected from 10 urban sites and their PAHs levels were determined by high-pressure liquid chromatography (HPLC). PAH compound ratios such as phenanthrene/anthracene (Phen/Ant) were used to identify petrogenic or pyrogenic sources. The results showed that the total PAH concentrations in the soils ranged from 372 µg/kg to 4320 µg/kg and carcinogenic PAHs ranged between 23% and 53 % of the total PAHs. According to the evaluation criteria of PAH levels (Maliszewska – Kordubach 1996), soils of Niamey can be considered as contaminated at moderate to high degree by PAHs. Calculated values of Phen/Ant ratio varied from 0.72 to 16.55 and indicated that the PAHs in Niamey urban soil are mainly of pyrogenic origin such as incineration, open burning for domestic heating and cooking.

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds consisting of two or more aromatic rings. Their presence in all environment compartments result from both natural processes such as volcanic activity or forest fires and predominantly anthropogenic activities including waste incineration, burning wood, coal or garbage but also the operation of gasoline and diesel engines (Baek *et al.* 1991). This explains the ubiquitous nature of PAHs in the environmental matrices. PAHs include a few hundred molecules. However according to their toxicity, their abundance in the environment and current knowledge, the United States Environmental Protection Agency (US-EPA) has selected sixteen of them as priority pollutants including benzo(a)pyrene the most toxic and the most studied. PAHs are lipophilic compounds and are characterized by high melting and boiling, low vapor pressures and very low aqueous solubility values.

PAHs may largely contaminate all environmental matrices and raise toxicological, mutagenic and carcinogenic concerns (IARC 2003). The main routes of human exposure to PAHs are inhalation of fine particles, dermal contact and ingestion. The determination of PAH concentrations in environmental matrices such as soil or plants is necessary to characterize the levels of environmental pollution and finally to assess the risk of human exposure. Thus, several studies have been conducted in the world on the evaluation of PAH concentrations in plants (Tankari Dan- Badjo *et al.* 2008; Tankari Dan- Badjo *et al.* 2007; Crépineau *et al.* 2004) and soils (Motelay - Massi *et al.* 2004; Bryselbout *et al.* 2000; Maliszewska – Kordubach, 1996). In developing countries, particularly in Africa, studies on the assessment of PAH concentrations in soils are very rare and indicated total PAH concentrations of 120 µg/kg in Nigeria (Sojinu *et al.* 2010) and up to 39000 µg/kg in South Africa (Nieuwoudt *et al.* 2011).

In Niger, there is no data available on the levels of PAHs in environmental and biological matrices despite the presence of PAH sources. Indeed, with a continuous population growth and economic development, the percentage of the population living in Niamey city has increased significantly and therefore the quality of the environment is considered as a real issue in terms of safety. Mismanagement of waste based on uncontrolled burning of waste, formal and informal industries, poor roads and vehicles, and relatively high traffic density can significantly affect the environment in Niamey through the emission of chemical contaminants including PAHs.

The objective of this study was twofold: first to address in Niger the environmental pollution by PAHs, and secondly to increase our knowledge regarding the concentrations of PAHs and their distribution in urban soils. More specifically, the study is aimed at assessing the total PAH concentrations in Niamey urban soils, characterize their profiles and to identify the potential sources.

Material and methods

Study area and sample collection

The study area was Niamey, Niger's capital, located on the banks of the River Niger. With 1.5 million people in 2010, Niamey covers an area of 115 km^2 and is divided into five municipalities themselves subdivided into districts. Most activities including administrative, industrial, agricultural and commercial are located on the left bank. The uncontrolled urbanization of Niamey, the accelerated population growth and the lack or absence of adequate sanitation facilities, the development of informal industrial fabric, automobile traffic and waste incineration generate discharges or releases many pollutants including PAH which can contaminate environmental matrices. The choice of sites meets the need of representation of different pollution situations present in the study area. Ten sites were selected in urban areas (Fig.-1) in Niamey with one control site (T) away from polluting sources. The other nine sampling sites (S1 to S9) were selected not only based on their proximity to pollution sources but also the presence of agricultural activities: the site of the Niamey Hospital, which receives the raw sludge from the wastewater treatment plant and residue burning waste (S1); the sites Enitex (S2) and Braduni (S3) respectively receiving wastewater from the textile industry and the Niger brewery; the tannery Gamkallé (S4); the cooking center of cattle and sheep heads and legs, based on the combustion of wood and tires, located near a landfill (S5); Gounti Yena sites (S6 and S7), vegetable growing area surrounded by multiple pollution sources such as roads, raw sewage and finally the sites of the pond (S8) and discharge (S9) of the green belt receiving waste burned residues and wastewater. Sampling conditions were similar for all the ten sites. During March 2010, several samples were taken in order to assess the average PAH contamination of a site. Samples were collected from the surface layers of soil (0-10 cm) as close as possible to the pollution source. Sampling was done with a stainless steel tube of 10 cm high and 10 cm in diameter. In each site, a composite sample (0.5 to 1 kg of soil) was made from five primary samples. This sample was packaged in aluminium paper. In each sampling site, 3 composite samples were collected for a total of 30 composite soil samples. Soil samples were dried at room temperature (< 40°C), crushed and sieved to 2 mm.

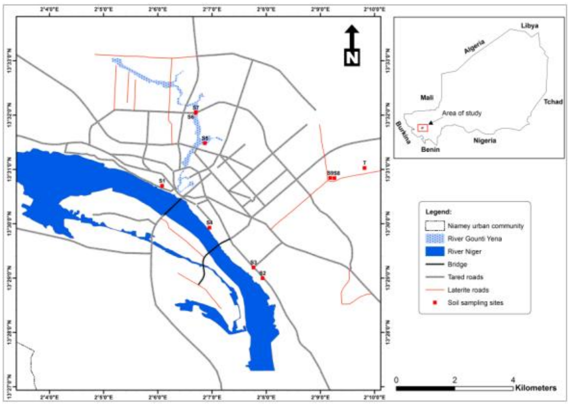

Fig. 1. Location of the soil sampling sites.

PAH extraction and analysis

Extraction and analysis of 15 PAHs were performed at Soil and Environment Laboratory. PAHs in the soil were extracted with an automated extractor Dionex ASE (Accelerated Solvent Extractor) 200 using a mixture of dichloromethane and acetone in isocratic mode as described by Ouvrard *et al.* (2011). PAH extracts were analyzed by HPLC. The HPLC system included an autosampler (Varian ProStar), a pump 9012 (Varian) with injection loop, column oven (Igloo-cil), fluorescence detector (Varian ProStar) coupled to UV detector (Varian ProStar) and integration software (Varian WS). HPLC analysis was performed using a column (ProSphere C18) with a program of elution gradient using acetonitrile and water. The mobile phase was acetonitrile/water, the flow rate 1 mL min^{-1} and the elution gradient: 10 min isocratic mode 50/50 (v/v); 30 min gradient mode from 50/50 to 100/0). For the following PAHs: naphthalene, acenaphthene, fluorene, phenanthrene, anthracene, we took into account the detection by fluorimetry. For fluoranthene, pyrene, benzo(a) anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h) anthracene, benzo(ghi) perylene, indeno(1,2,3-cd) pyrene, UV detection was favoured for better separation of peaks. The final result was expressed in µg/kg dry weight and the quantification limit was 0.2 µg/kg.

Statistical analysis

Analysis of variance using the SAS statistical software General Linear Model (GLM) procedure was applied to the total PAH concentrations and the individual PAH at the various sites. The threshold of significance was 5% ($P < 0.05$).

Results and discussion

Total PAH concentrations in different sites

The average of total PAH concentrations (∑15 HAP) determined in soils at various sites are given in Table I. Total PAH concentrations varied significantly from 372 µg/kg to 4320 µg/kg according to the site. The highest concentrations of ∑15 HAP were recorded in samples from Hospital wastewater station (S1: 4320 µg/kg) and Cooker of Katako (S5: 3347 µg/kg). The lowest concentration (372 µg/kg) was recorded at the control site (T) and was 11 times lower than that detected in S1. Indeed the road sites (S6 and S7), pond (S8) and discharge of the Green Belt (S9) showed relatively higher PAH total concentrations than the control site. Statistical data's analysis confirmed this result and showed a significant difference between two sites (S1 and S5) and the eight other sites. Indeed, unlike other sites, site S1 and S5 are distinguished mainly by the burning of waste in open air, this could explain their high PAH levels. These results revealed that urban soil could be affected by high PAH concentrations levels which are due to poor waste management, road traffic, domestic cooking, suggesting that PAH concentrations are strongly linked to the land use of the site. The total PAH concentrations obtained in this study are consistent with the upper range of data reported in the literature. For example, Sojinu *et al.* (2010) determined a maximum value 120 µg/kg in soil from oil exploration areas of the Niger Delta (Nigeria). Maliszewska-Kordybach (1996) measured an average of 264 µg/kg in agricultural soils in Poland. Moreover, the levels of PAH detected in samples of Niamey urban soil were lower than those reported for soils from some countries in the world. Indeed in soils and sediments collected from central South Africa, specifically targeting industrial, residential, and agricultural areas, the levels of PAHs had reached 39 000 µg/kg (Nieuwoudt *et al.* 2011). Liu *et al.* (2010) reported that the sum of 16 and 18 PAH concentrations in surface soil of Shangai (China) were very high and reached 31900 and 37400 µg/kg respectively.

Table 1. PAH concentrations and profiles in soils according different sites.

Sites	T		S1		S2		S3		S4		S5		S6		S7		S8		S9	
Compounds	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**	**µg/kg**	**%**
Naphtalene	32 b	8.6	228 b	5.3	94 b	9.1	40 b	5.0	98 b	9.7	560 a	16.7	122 b	9.2	16 b	1.6	160 b	9.2	118 b	10.5
Acenaphtene	6 b	1.7	0,2 b	0	nd	-	12 ab	1.5	12 ab	1.2	nd	-	17 ab	1.3	8 b	0.8	51 a	2.9	43 ab	3.8
Fluorene	15 a	4.0	25,0 a	0.6	14 a	1.3	8 a	1.0	5 a	0.5	41 a	1.2	22 a	1.7	13 a	1.3	14 a	0.8	19a	1.7
Phenanthrene	11 c	3.0	110 b	2.5	57 c	5.5	8 c	1.0	9 c	0.9	417 a	12.5	53 c	4.0	36 c	3.6	38 c	2.2	41 c	3.6
Anthracene	5 b	1.5	12 b	0.3	4 b	0.4	1 b	0.1	4 b	0.4	87 a	2.6	3	0.2	50 ab	5.0	4 b	0.2	4 b	0.4
Fluoranthene	7 c	2.0	352 a	8.1	13 c	1.2	6 c	0.8	9 c	0.9	237 b	7.1	23 c	1.7	12 c	1.2	40 c	2.3	51 c	4.6
Pyrene	140 b	37.7	741 a	17.2	341 b	32.7	121 b	15.3	317 b	31.0	627 a	18.7	260 b	19.6	182 b	18.2	289 b	16.7	209 b	18.7
Benzo(a) anthracene	84 b	22.6	573 a	13.3	106 b	10.2	58 b	7.3	23 b	2.2	411 ab	12.3	247 ab	18.6	66	6.6	362 ab	21.0	14 b	1.3
Chrysene	33 a	8.8	276 a	6.4	132 a	12.6	70 a	8.8	85 a	8.4	265 a	7.9	180 a	13.6	31 a	3.0	132 a	7.6	117 a	10.4
Benzo(b) fluoranthene	13 a	3.4	166 a	3.9	11 a	1.0	21 a	2.6	32 a	3.2	33 a	1.0	122 a	9.2	11 a	1.1	13 a	0.8	55 a	4.9
Benzo(k) fluoranthene	4 bc	1.1	144 a	3.3	7 bc	0.6	7 bc	0.8	9 bc	0.8	25 b	0.7	17 bc	1.2	3 c	0.3	18 bc	1.0	21 bc	1.8
Benzo(a) pyrene	14 b	3.7	260 a	6.0	28 b	2.6	28 b	3.5	35 b	3.5	104 b	3.1	116 b	8.7	20 b	2.0	79 b	4.6	67 b	6.0
Dibenzo(a,h) anthracene	4 a	1.1	94 a	2.2	237 a	22.7	400 a	50.4	346 a	34.0	169 a	5.0	129 a	9.8	554 a	55.1	399 a	23.1	81 a	7.3
Benzo(g,h,i) perylene	3 b	0.7	1048a	24.2	nd	-	8 b	1.1	15 b	1.5	346 b	10.3	16 b	1.2	2 b	0.2	76 b	4.4	242 b	21.6
Indeno(1,2,3-cd)pyrene	0,6 b	0.2	291 a	6.7	nd	-	6 b	0.8	20 b	2.0	26 b	0.8	nd	-	nd	-	53 b	3.1	37 b	3.4
∑15 PAH %	372 b	100	4320a	100	1043b	100	794 b	100	1020 b	100	3347a	100	1325 b	100	1004 b	100	1727 b	100	1117 b	100
Carcinogenic PAH	119	32	1512	35	386	37	517	65	469	46	770	23	636	48	653	65	915	53	279	25

Concentrations are given as mean values, expressed in µg/kg dry weight (a,b,c), for each line, mean values without the same letter differ significantly, $P < 0.05$, nd: not detected; Carcinogenic PAH : Benzo(a)anthracene, Benzo(b)fluoranthene, Benzo(k)fluoranthene, Benzo(a)pyrene, Dibenzo(a,h)anthracene, Indeno(1,2,3-cd)pyrene (IARC, 2003); T : control site; S1-S9: site 1 to site 9.

Due to their carcinogenic and mutagenic characteristics, concentrations of PAHs in the environment including soil and sediment matrices are regularly monitored and regulated. Maliszewska-Kordybach (1996) has established the criteria for the soils classification according to PAH levels. Indeed, the threshold values of this classification were derived from the results of determination of PAHs concentrations in European soils, as well as from estimation of risks of human exposure. According to this classification, levels of PAHs lower than 200 µg/kg are considered as not contaminated. Concentrations that fall within the range of 200 to 600 µg/kg imply weakly contamination whereas values between 600 and 1000 µg/kg indicate a significant contamination, and soils are heavily contaminated when the total PAH concentration exceeds 1000 µg/kg. According to Maliszewska-Kordybach (1996) criteria, Niamey urban soils were contaminated by PAHs because the total PAHs in all the sites were greater than 200 µg/kg. The control site was weakly contaminated, the site S3 was moderately contaminated and all the remaining sites are heavily contaminated.

PAH profiles in soil at various sites

The concentrations of individual PAHs and sum of carcinogenic PAHs determined in Niamey urban soils are also presented in Table I. Analysis of PAHs in soil showed different profiles at various sites. Indeed, concentrations of 11 on 15 PAHs like fluoranthene, pyrene, benzo(a)pyrene and acenaphtene, differed significantly according to the sites. All the 15 PAHs were detected on 6 sites. At the 4 remaining sites, 12 to 14 PAHs were detected. Indeed, benzo(g,h,i)perylene and acenaphthene were not detected respectively at S2 and S5, indeno(1,2,3-cd)pyrene was not determined on S2, S6 and S7. In general, considering the individual contribution of each PAH, pyrene, dibenzo(a,h)anthracene or benzo(g,h,i)perylene were the predominant constituents at different sites and theirs concentrations were up to 38%, 50% and 22 % of the total PAH concentrations respectively. Benzo(a)pyrene concentrations ranged from 2 µg/kg to 116 µg/kg and represented up to 8.7%. In the present study, percentages of carcinogenic compounds in each site were calculated and summarized in Table I. Carcinogenic PAHs ranged from 23 to 53 % of the total PAHs according to site. This result was similar to those found by Nieuwoudt *et al.* (2011) in Central South Africa soils (15 – 56%) and Motelay-Massei *et al.* (2004) in soils at the French Seine River (15 – 46 %).

In order to illustrate the PAH profile in soil, the PAHs were grouped according to their aromatic rings number : low molecular weight (LMW 2 to 3 rings: naphtalene, acenapthene, fluorene, phenanthrene, anthracene); medium molecular weight (MMW 4 rings: fluoranthene, pyrene, benzo(a)anthracene, chrysene); high molecular weight (HMW 5 rings or more: benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene), indeno(1,2,3-cd) pyrene). The distribution patterns of PAHs in soil collected at various sampling sites were shown in Fig. 2. In general in all cases MMW and HMW PAHs were the most abundant. The predominance of 4 to 6 ringed PAHs could be explained by the fact that these compounds generally emitted in particulate form and with relatively high molecular weight are deposited close to their emission source (Tankari Dan-Badjo *et al.* 2008). In 9 sites, the LMW PAHs group were the least abundant and represented from 9 to 33% of the total PAH concentrations. MMW and HMW PAHs represented from 29 to 71% and from 10 to 59% respectively, and suggesting those PAHs contributed significantly to the total PAH concentrations in all of ten investigated sites.

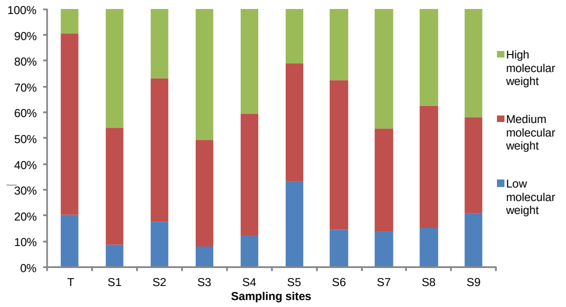

Fig. 2. PAH profiles in soils according to the number of aromatic rings.

T : control site; S1-S9: site 1 to site 9. Low molecular weight (PAHs with 2 to 3 rings: naphtalene, acenapthene, fluorene, phenanthrene, anthracene); medium molecular weight (PAHs with 4 rings:

fluoranthene, pyrene, benzo(a)anthracene, chrysene); high molecular weight (PAHs with 5 rings or more: benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene), indeno(1,2,3-cd)pyrene

PAH sources

The relative abundances or diagnostic ratios are valid indicators of PAH sources. Indeed diagnostic ratios are widely used to determine the origins of PAHs present in environmental samples like soil and sediment (Okedeyi *et al.* 2013; Sojinu *et al.* 2010; Liu *et al.* 2010). Several molecular ratios such as phenanthrene/anthracene (Phen/Ant), anthracene/anthracene + phenanthrene (Ant/Ant+Phen), fluoranthene/fluoranthene+pyrene (Flan/Flan+Pyr) are known to be used to identify whether the PAHs sources are petrogenic or pyrogenic origin. Phen/Ant ratio < 10 or Ant/Ant+Phen > 0.1 generally indicate that the PAHs were performed by coal combustion (pyrolytic) while Phen/Ant ratio >15 or Ant/Ant+Phen < 0.1 indicate that the PAHs source are petroleum combustion (Guo *et al.* 2011; Olajire *et al.* 2007). In this study, Phen/Ant and Ant/Ant+Phen ratios have been calculated in urban soils of Niamey and summarised in Table 2. In all the sites, except S2 (14.95) and S6 (16.55), the Phen/Ant ratios were < 10 suggesting that PAHs were emitted by pyrolytic sources. Considering Ant/Ant+Phen ratios, eight sites presented a ratio greater than 0.1, thus indicating that PAHs in these sites were of a pyrolytic source. In sites S2 and S6, this ratio was lower than 0.1 suggesting a petrogenic PAHs source. Taken together, these results clearly show that PAH contamination in Niamey urban soil mainly originates from pyrogenic sources such as incineration, open burning for domestic heating and cooking. However, the pyrolytic source contributes such as traffic emissions involved in presence of PAHs in soil of S6 for example.

Table 2. Ratios for pyrolytic and petrogenic orgin of PAHs according to sites.

PAHs	Pyrolytic	Petrogenic	PAH ratios according to sites									
			T	S1	S2	S3	S4	S5	S6	S7	S8	S9
Phenanthrene/anthracene[a,b]	<10	>15	2,07	9,32	14,95	6,83	2,34	4,81	16,55	0,72	9,44	9,06
Anthracene/anthracene + phenanthrene[a]	>0.1	<0.1	0,33	0,10	0,06	0,13	0,30	0,17	0,06	0,58	0,10	0,10

[a]Okedeyi *et al.* (2013) ; [b]Olajire *et al.* (2007).

T : control site; S1-S9: site 1 to site 9.

Conclusion

PAH concentrations analysed in Niamey urban soils varied significantly from 372 µg/kg to 4320 µg/kg. The highest total PAH concentrations were recorded in samples from Hospital wastewater station and were 11 times higher than that detected at the control site. According to the criteria established by Maliszewska-Kordybach (1996), soils of Niamey are contaminated at moderate to high degree by PAHs. Pyrene, dibenzo(a,h)anthracene or benzo(g,h,i)perylene were the most abundant compounds and represented up to 38%, 50% and 22 % of the total PAH concentrations respectively. Carcinogenic PAHs ranged from 23 to 53 % of the total PAHs according to site. In general, medium molecular weight and high molecular weight were the predominant contributors to the contamination level. The values obtained from different diagnostic ratios clearly indicated that the PAH in Niamey urban soil were mainly of pyrogenic origin such as incineration, open burning for domestic heating and cooking. In perspective, it seems appropriate to conduct this study in other regions of Niger in particular the operating sites of oil and coal.

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