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Research Article

ASSESSMENT OF THE CONTAMINATION OF VEGETABLE CROP BY POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) ON GARDENING SITES IN SOUTH BENIN

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Abstract

Vegetables produced in Benin's coastal towns play a major role in the supply of fruit and vegetables to households. These production areas are close to interstate highways that are busy with vehicles or trucks and sometimes with passing aeroplanes. However, these vehicles, trucks or aeroplanes can contaminate market garden produce with aromatic and polycyclic hydrocarbons (PAHs). This study aims to assess PAH levels in amaranths and carrots in the market garden areas of Houéyiho, Sèmè-Kpodji and Grand-Popo in southern Benin. A total of 10 vegetable samples (4 samples at the Houéyiho site, 4 samples at the Sème-Kpodji site and 2 samples at Grand-Popo) were collected. The analyses were carried out by gas chromatography-mass spectrometry. The data collected were subjected to an analysis of variance followed by a 5% Newman Keuls student test. The results of the PAH analyses showed the presence of twelve PAHs in the vegetables (Amaranth and Carrot) at all three sites. The levels of Benzo (a) Anthracene, Benzo (a) Pyrene and Dibenzo (a, h) Anthracene varied very significantly ($P<0.001$) both between the different vegetables and between the sites and were more concentrated in the amaranths than in the carrots. This study shows that there are potential health risks associated with the high consumption of the vegetables studied at the different sites.

Keywords: Market gardening, Vegetables, Contamination, PAH, South Benin.

INTRODUCTION

Urban and peri-urban vegetable production in Benin's coastal cities plays a major role in supplying households with fruit and vegetables. They provide the populations of Benin's coastal cities with 64% of their annual consumption of market garden produce (Adorgloh-Hessou, 2006; Adifon *et al.,* 2015). According to (Ouikoun *et al.,* 2019), amaranth, carrot and chilli are the main vegetable crops in the production sites of Houéyiho, Sèmè-Kpodji and Grand-Popo. However, market garden production in urban and peri-urban areas has specific features that are peculiar to rural agriculture, namely: new market and non-market functions, complexity of land tenure, heavy use of chemical inputs, the need for water control and control of marketing and supply channels for synthetic plant protection products (Moustier and Mbaye, 1999; Ogouwalé, 2007). Due to the lack of arable land, market gardeners in Benin's coastal cities produce in inappropriate areas such as roadsides with vehicles, rubbish dumps or next to aeroplane runways, next to factories. According to the work of (Crépineau-Ducoulombier and Rychen, 2003), on the assessment of PAH contamination of agricultural plots located near a motorway and an airport revealed a higher level of PAH contamination for the motorway site (188 µg/kg dry matter) than for the airport (69 µg/kg dry matter).

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The highest concentrations were observed in the vicinity of the national road, thus revealing the importance of the nature of the vehicles circulating (proportion of trucks) and their average speed (Crépineau-Ducoulombier and Rychen, 2003). Soot and smoke from all sources such as combustion engine exhaust and cigarette smoke are the source of Aromatic and Polycyclic Hydrocarbons (PAHs) such as benzo[a]pyrene (Dupuy, 2014). Barbecued foods are also a source of PAHs (Fismes *et al.,* 2002). PAHs consist of at least two to six fused aromatic rings. Strictly speaking, they contain only carbon and hydrogen atoms. The toxicity of PAHs varies greatly. Some have moderate effects (hepatic, haematological, immunological), such as anthracene and pyrene, while others, such as benzo(a)pyrene, are recognised as carcinogenic (Collins *et al.,* 2005). To date, eight compounds of the PAH family are classified as category 2 carcinogens by the European Union: benzo(a)pyrene, benzo(k)fluoranthene, benzo(j)fluoranthene, benzo(a)anthracene, benzo(b)fluoranthene (or benzo(e) acephenanthrylene), benzo(e)pyrene, chrysene and dibenzo (a,h) anthracene. According to (Meudec *et al.,* 2006), the health consequences of the presence of PAHs in humans are: severe disruption of vitamin A metabolism, modification of the immune system, decreases in the weight of newborns, growth rate and activity level of children, increase in cancers with the age of the individuals, etc. The present study aims to assess PAH levels in amaranths and carrots in the market gardens of Houéyiho, Sèmè-Kpodji and Grand-Popo in southern Benin.

MATERIALS AND METHODS

Study environment

Samples were collected in the plots of Houéyiho, Sèmè-Kpodji and Grand Popo in southern Benin (Figure 1). The market garden site of Sèmè-Kpodji is located on either side of the Cotonou-Porto-Novo interstate road between 6°21 and 6°22 North latitude and 2°34 East longitude. The Houéyiho site is located in the heart of the city of Cotonou between latitudes 6°21 and 6°22 North and 2°23 to 2°24 East and is situated in the 12th arrondissement of the city of Cotonou. The market garden site of Grand-Popo is located in southwest Benin between 6°12 and 6°27 North latitude and 1°37 to 1°55 East longitude (Ouikoun *et al.,* 2020). The biological material consisted of vegetables: carrot and amaranth. For the analyses, a total of 10 vegetable samples (four samples at the Houéyiho site, four samples at the Sème-kpodji site and two samples at Grand-popo) were taken.

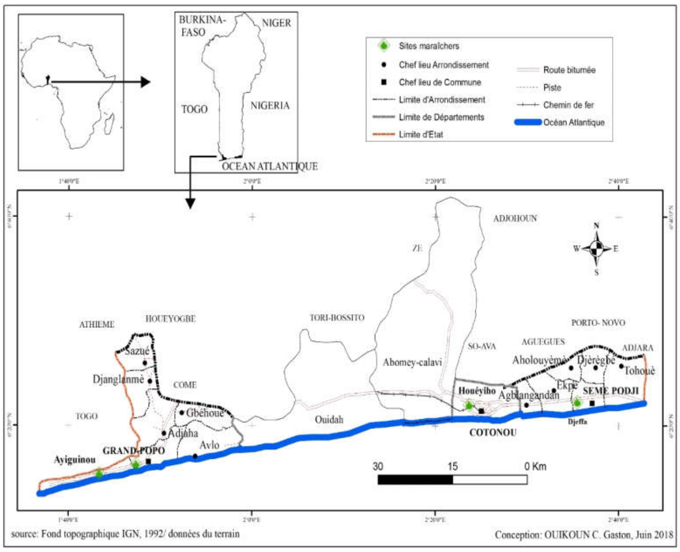

Figure 1. Location map of the study area

Method of collecting the different samples

Sampling was carried out following the overall experimental design of the entire study (Ouikoun *et al.,* 2019). In Cotonou (Houéyiho), the site was subdivided into three blocks B1, B2 and B3 (Figure 2a). The entire eastern zone of the Houéyiho market garden perimeter was considered as Block 3 and the western zone as Block 1. Between Blocks 1 and 3, Block 2 is located. Each of Blocks B1, B2 and B3 is composed of different plots numbered from P1 to P5. The market garden site of Sèmè-Kpodji (Vimas) was also subdivided into three blocks B1, B2 and B3. The blocks on this site were formed according to the slope or the heterogeneity of the site's topography, not forgetting the position of the market gardens in relation to the Benin-Nigeria interstate road and the Atlantic Ocean. The market gardens at the Sèmè-Kpodji site, located at the bottom of the slope and close to the Benin-Nigeria interstate road, were considered to be in Block 3. Those at the top of the slope and closer to the Atlantic Ocean were considered to belong to Block 1. Between these two blocks is Block 2 (Figure 2b). Each block is subdivided into two large plots on either side (east and west) of a main path or track. At the Grand-Popo site, the samples were taken in two districts (Grand-Popo Centre and Agoué) on market garden plots on either side of the Cotonou-Lomé interstate road (Figure 2c).

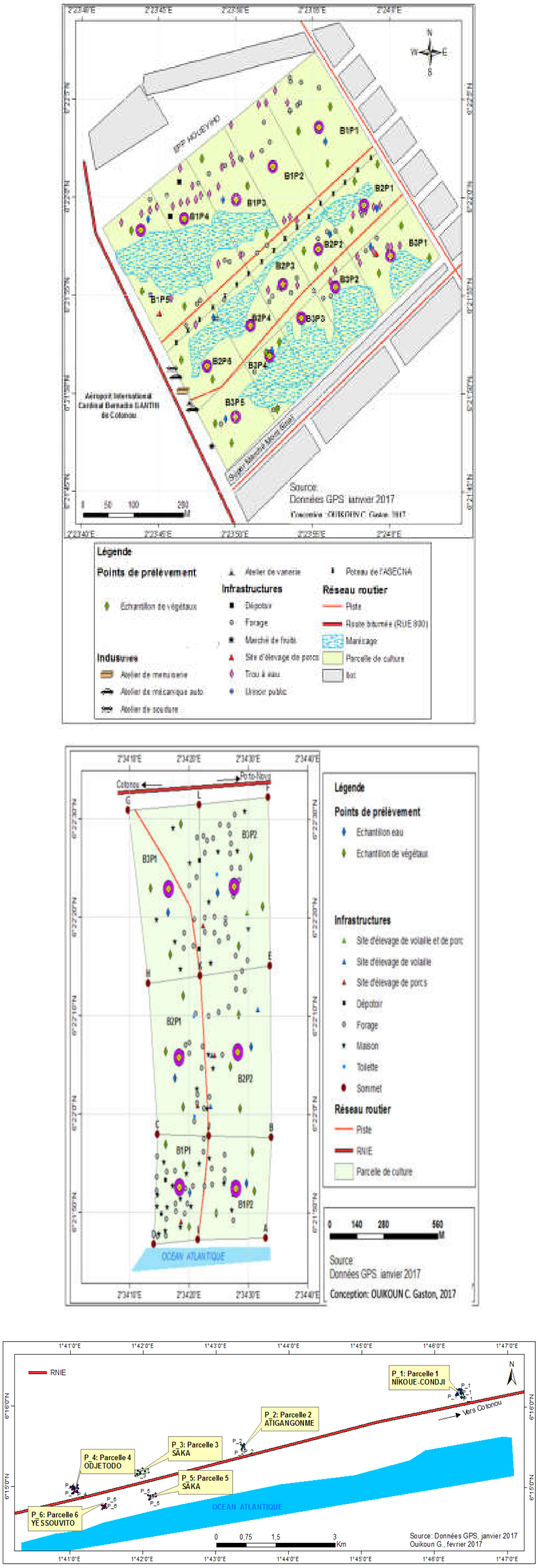

Figure 2. Market garden site a: Houéyiho; b: Sèmè-Kpodji site c: Grand-Popo site B = Block and P = Plot

Method of analysis of samples in the laboratory

All samples were wrapped with aluminium foil, labelled and transported to the laboratory using a cooler. The samples were processed according to the sampling guide for environmental analysis (Kipopoulou *et al.,* 1999). Analyses were performed by gas chromatography-mass spectrometry according to the analytical protocol developed by (Kipopoulou *et al.,* 1999).

Statistical analysis method

The collected data were subjected to a one-way analysis of variance (ANOVA) using the ANOVA procedure in SAS (Statistical Analysis System) version 9.2. Comparisons of means were made using the Student Newman-Keuls test (Dagnelie *et al.,* 1986). Similarly, the means of the different PAHs of the study sites obtained from the analysis of variance were subjected to a trend analysis according to the South-East-South-West gradient in order to assess the evolution of the levels. These trends were assessed using regression equations and the coefficient of determination R^2 (Rimi *et al.,* 2011). Correlations were also carried out using the MINITAB 14 software between the different PAHs.

RESULTS

Screening of PAHs in vegetables (amaranth and carrot)

Table 1 shows the results of the screening of PAHs in vegetable products from the different sites. From this table it appears that twelve out of sixteen different types of PAHs are present in amaranths and carrots from the three study sites. However, of these twelve PAHs present, fluorene, phenanthrene and anthracene are the least represented. Similarly, between the two speculations, all twelve PAHs are present in carrots compared to chilli. In general, fluorene is the only PAH absent in the Sèmè-kpodji and Grand-popo sites (Table 1).

PAH content in the different vegetables at the sites

Table 2 shows the average values of the different contents of the different PAHs. The analysis of this table shows that, according to the Student Newman Keuls test, the amaranths produced at the Houéyiho site contain the highest levels ($p<0.05$) of the different PAHs. Similarly, the crops from plots 4 and 5 contain high levels of the different PAHs compared to plots 1, 2 and 3, regardless of the crop at the Houéyiho site (Table 2). The same is true for the Sèmè-Kpodji and Grand-Popo sites. Generally speaking, according to the results of the Student Newman Keuls test, the Houéyiho site in the Littoral department has the highest PAH values ($p<0.05$) for all crops, while the Grand-Popo site in the Mono department has the lowest values.

Table 3 presents the results of the one-way analysis of variance performed on the PAH data. The analysis of the table shows that the levels of Benzo (a)Anthracene, Benzo (a) Pyrene and Dibenzo (a, h) Anthracene vary very significantly ($p<0.001$) both between the different crops and between the sites. Similarly, there is a highly significant difference ($p<0.001$) between the plots on the study sites with regard to the content of the different PAHs.

Table 1. Screening des HAP dans les produits maraîchers des sites

SITES	Pyrene	Benzo (a) anthracene	Chrysene	Benzo (b) fluoranthene	Benzo (k) fluoranthène	Benzo (a) pyrène	Indino (1,2, 3-Cd) pyrene	Dibenzo (a,h) anthracene	Benzo (g,h,i) pérylene	Fluorene	Phenanthrene	Anthracene	TOTAL
HOUEYIHO													
Amaranth 1	X	X	X	X	X	X	X	X	X				09
Amaranth 2	X	X	X	X	X	X	X	X	X	X			10
Carrot 1	X	X	X	X	X	X	X	X	X		X	X	11
Carrot 2	X	X	X	X	X	X	X	X	X	X	X	X	12
SEME-KPODJI													
Amaranth 1	X	X	X	X	X	X	X	X	X				09
Amaranth 2	X	X	X	X	X	X	X	X	X				09
Carrot 1	X	X	X	X	X	X	X	X	X		X	X	11
Carrot 2	X	X	X	X	X	X	X	X	X		X	X	11
GRAND-POPO													
Amaranth	X	X	X	X	X	X	X	X	X				09
Carrot	X	X	X	X	X	X	X	X	X		X		10

Table 2. Average content of different PAHs in vegetables (amaranth and carrot) by site

Sites	HAPs (µg/kg)		
	Benzo(a) Anthracène	Benzo(a)Pyrène	Dibenzo(a,h) Anthracène
GRAND-POPO			
Amaranth	1,01 ± 0,00 e	1,09 ± 0,01 f	0,690 ± 0,300 c
Carrot	0,22 ± 0,01 j	0,08 ± 0,00 j	0,007 ± 0,00 d
HOUEYIHO			
Amaranth 1	1,84 ± 0,00 b	2,62 ± 0,01 b	1,78 ± 0,01 a
Amaranth 2	2,76 ± 0,01 a	2,89 ± 0,01 a	2,01 ± 0,00 a
Carrot 1	0,53 ± 0,01 g	0,99 ± 0,00 h	0,49 ± 0,01 c
Carrot 2	0,65 ± 0,01 f	1,01 ± 0,01 g	0,51 ± 0,00 c
SEME-KPODJI			
Amaranth 1	1,22 ± 0,01 d	1,32 ± 0,01 e	1,16 ± 0,01 b
Amaranth 2	1,34 ± 0,01 c	1,42 ± 0,01 d	1,30 ± 0,01 b
Carrot 1	0,30 ± 0,01 i	0,32 ± 0,01 i	0,12 ± 0,01 d
Carrot 2	0,33 ± 0,01 h	1,86 ± 0,00 c	0,13 ± 0,01 d

The means with the same alphabetic letters are not significantly different ($P > 0.05$) according to the Newman-Keuls test.

Table 3. Results of one-factor analysis of variance (Fisher value) performed on PAH data

Source of variation	Valeur de Fisher			
	Degree of freedom	Benzo(a)Anthracene	Benzo(a)Pyrene	Dibenzo(a,h)Anthracène
Samples	9	45504.6***	77104.0***	56.46***
Repetition	2	11.04ns	16.71ns	0.81ns

ns : p >0.05 ; ***: p < 0.001

Table 4. Pearson correlation between the different PAHs

	Benzo(a)Anthacène	Benzo(a)Pyrene	Dibenzo(a,h)Anthracène
Benzo(a)Anthracène	1		
Benzo(a)Pyrene	0,836***	1	
Dibenzo(a,h)Anthracène	0,950***	0,811***	1

*** : P < 0,001

PAH content in vegetables

Figure 3 shows the levels of PAHs in pigweed and carrots at all study sites. The analysis of this figure shows that the levels of the different PAHs in amaranths are significantly ($p<0.05$) higher than in carrots (figure 1). Similarly, the levels of Benzo (a) pyrene in both vegetables are significantly ($p<0.05$) higher than the levels of Benzo (a) Anthracene and Dibenzo (a,h) Anthracene.

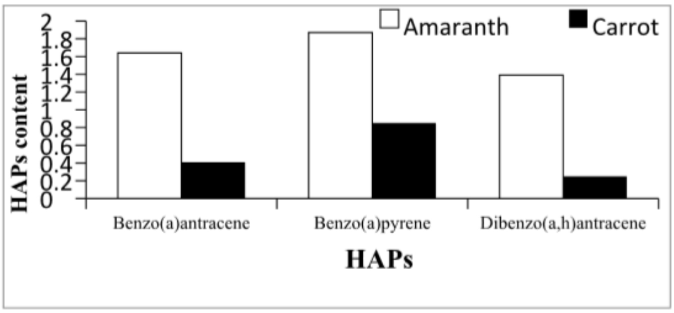

Figure 3. PAH contents in different vegetables

Trend analysis of the levels of the different PAHs according to the south-east/south-west gradient

The analysis of the gradient is justified by the fact that the Cotonou and Sème-Kpodji area has a high density of both human and rolling stock compared to Grand-Popo. It is therefore important to see the evolution of PAHs from a high density area to a less dense area. The averages from the analysis of variance were subjected to a trend analysis. The analysis of this figure shows that the values of the levels of the different PAHs show a regressive trend (with a regression rate ranging from 0.09% to 0.32%) from the south-east to the south-west (Figure 4) with a peak at the Houéyiho site in the Littoral department.

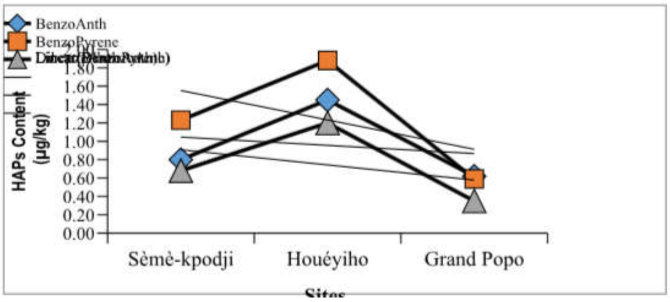

Figure 4. Trends in the levels of the various PAHs along the south-east/south-west gradient

As a result, the concentration of the different PAHs in the amaranths, cores and soils decreases as one moves towards the south-west of Benin. However, this decrease is not significant ($p >0.05$) and does not show any definite pattern with respect to the R^2 values, which are all low. The correlation analysis carried out between the different PAHs (Table 4) reveals that there is a positive and significant correlation ($p<0.001$) between all the different PAHs. This means that a crop containing one PAH also contains a high content of the other PAHs.

DISCUSSION

According to (Kummerová *et al.,* 2013), PAHs are organic molecules, exclusively composed of carbon and hydrogen atoms and have at least two benzene rings. PAHs include several hundred molecules and can have two origins: petrogenic and pyrolitic. PAHs of petrogenic origin are formed during the geological transformation of organic matter and are present in fossil organic matter (coal, oil). The individual concentrations of PAHs of natural origin are in the order of 1 to 10 $\mu g.kg^{-1}$ of dry soil with total PAH concentrations of 500 to 1500 $\mu g.kg^{-1}$ of dry soil (Wilcke, 2000). Generally speaking, the amaranths produced at the Houéyiho site contain the highest levels of the different PAHs, i.e. on average 2.32 ± 0.15 mg/kg. This suggests that the aerial part absorbs more PAHs. The results of the different PAHs in the crops reveal that the sites of Houéyiho in the Littoral department contain the highest values (1.51± 0.12mg/kg) of PAHs for amaranth and carrot while the site of Grand-Popo in the Mono department contains the lowest values (0.52±0.01 mg/kg). A similar study by (Gao and Zhu, 2004) on maize, revealed PAH levels around 0.08 mg/kg; 0.51 mg/kg and 0.02 mg/kg. The fact that the Houéyiho site was identified as containing the highest PAH content in amaranth and carrot is not surprising. Indeed, (Gao and Zhu, 2004) also noted that on some lands, accumulation in the aerial system is the highest, with a total PAH content of 1.27 mg/kg compared to (1.51±0.12 mg/kg) for Houéyiho. Similar concentrations are observed in the aerial systems of maize grown on the other lands, varying between 0.01 and 0.06 mg/kg. According to the same author, the proportion of PAHs quantified in the aerial parts corresponds on average to less than 1% of the PAH concentrations in the roots and indicates a very low translocation between these two compartments of the plant. As a result, in our conditions, the risk of contamination of the environment and humans is to be feared in that the roots still concentrate a high content of PAHs compared to the leaves. From the results of our work, it follows that high levels of the different PAHs were recorded in amaranth compared to carrot. In environmental terms, studies have also observed the accumulation of PAHs in the aerial parts of plant species grown on contaminated land (Tao *et al.,* 2004; Gao and Zhu, 2004; Xu *et al.,* 2005). However, it is recognised that PAHs with low to medium molecular weight show a higher accumulation in the aerial system after root removal than molecules with high molecular weight (Gao and Zhu, 2004; Xu *et al.,* 2005). On the other hand, high molecular weight PAHs would have a higher adsorption potential and could not pass through root tissues (Kipopoulou *et al.,* 1999). However, this hypothesis is contradicted by the work of (Meudec *et al.,* 2006), who found other PAHs in aerial parts after root sampling. However, in terms of accumulation, (INRS, 1997) showed that passive transport has long been accepted for organic molecules such as PAHs, but complementary active transport has recently been suspected (Zhan *et al.,* 2010). The authors had also demonstrated a competition for uptake in wheat between a two-ring PAH (Benzo (a) pyrene) and a three-ring PAH (Dibenzo (a,h) anthracene), characterised by a reciprocal inhibition of root uptake of these two PAHs. This negative control of uptake between two- and three-ringed molecules is not evident here since the relative abundance of two-ringed PAHs in roots is twice as high as in soil. However, the differences in relative abundances in soil and roots may suggest competition between two- and three-ring PAHs (+ 2% and - 3% respectively) and four- and five-ring PAHs (+7% and -6% respectively). This observation reinforces the hypothesis of distinct PAH uptake mechanisms, depending on the number of cycles in this case. These results show that the environment is really polluted as these sites are close to the interstate highways and therefore the fumes are constantly present; they come alive almost every 24 hours. The aerial part of the plant is more exposed to the environment than the underground part.

Conclusion

Generally speaking, we found 12 out of 16 internationally recognised hazardous PAHs on the various vegetables from the three study sites. Also, there are three (03) PAHs, namely Benzo (a) pyrene, Benzo (a) anthracene and Dibenzo (a,h) anthracene, whose toxicity is no longer to be demonstrated. These three PAHs are considered to be the most toxic because of their probable carcinogenic potential (group 2A). The various vegetables are contaminated by PAHs and present risks of toxicity to human health. It can therefore be concluded that the environment of the production sites is polluted by different sources of organic and inorganic pollutants.

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