**Review Paper**

**Polycyclic aromatic hydrocarbons (PAHs) in the environment, sources, effects and reduction risks**

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Polycyclic Aromatic Hydrocarbons (PAHS) are large group of environmental contaminant, originating from incomplete combustion or pyrolysis of organic matter (FAO/WHO, 2005). These compounds occur as contaminants in different kinds of foodstuffs including dairy products, vegetables, fruits, oils, cereals, and smoked meats (Camargo and Toledo, 2002; Dobrinas et al., 2008; Simko, 2002). The sources of PAH in food are mainly environmental pollution and food processing (drying, smoking) and cooking (roasting, grilling, and frying) (Zang et al., 2010). A number of PAHs have been found to have carcinogenic and mutagenic effects while some of them may act as synergists (Wenzl et al., 2006). One of the major routes of human exposure to PAHs in non-smoking people is food. These compounds can reach the food chain by different ways as PAHs have been found in different food products, such as dairy products, vegetables, fruits, oils, coffee, tea, cereals and smoked meat, therefore the analysis of PAHs in food is a matter of concern (Plaza-Bolanos et al., 2010). Over the years, different sources of PAH contamination of food have been found. Food items and products could be contaminated by soils, polluted air and water (WHO, 2005). Some aquatic food products, such as fish, can be exposed to PAHs present in water and sediments and the PAH content greatly depends on the ability of the aquatic organisms to metabolize them (Plaza-Bolanos et al., 2010). On the other hand, PAHs are also found in foods as a result of certain industrial food processing methods such as smoke curing, broiling, roasting and grilling over open fires or charcoal which permit the direct contact between food and combustion products (Silva et al., 2011). Furthermore, in the food processing industry, food additives such as smoke flavoring products (SFP), lubricants, solvents, propellants, glazing agents and protective coatings contribute to contamination of food items by PAHs (Moret and Conte, 2000). It has been found that raw foods do not usually contain high levels of PAHs, presence of PAHs in uncooked food, such as vegetables, seeds and grains have been found to accumulate on the waxy surface of many vegetables and fruits (Adetunde et al., 2012), Olabemiwo et al. (2013b) in their study showed that PAHs in the study samples were from pyrolytic source and that there exist a correlation between some PAHs and total PAHs in the soil in the vicinity of places where the plants were cultivated.

In general, PAHs are not present individually but in...
mixtures. Sixteen PAHs that have been extensively monitored are the compounds included in the United States Environmental Protection Agency (USEPA) list of priority organic pollutants (USEPA, 1994) are; naphthalene, acenaphthylene, acenaphthene, fluorine, anthracene, phenanthrene, fluoranthene, chrysene, benzo (a) anthracene, pyrene, benzo (k) fluoranthene, benzo (b) fluoranthene, benzo (a) pyrene (Marce and borrull, 2000), dibenzo (a,h) anthracene, dibenzo (b,c) fluoranthene and benzo (ghi) perylene (Marce and borrull, 2000). This study is therefore designed to look at various sources of polycyclic aromatic hydrocarbon in the environment, health effects and ways of reducing the risks associated with PAHs.

Sources/origin of PAHs and environmental fate

PAHs originate from incomplete combustion of organic matter. At high temperatures organic compounds undergo partial cracking and form small unstable fragments (pyrolysis), mostly radicals, which can recombine to give relatively stable PAHs (pyrosynthesis). All the compounds containing carbon and hydrogen can be optimal precursors of PAHs. Large amount of PAHs are emitted from both natural and anthropogenic sources, even though the latter are the main contributors to the environmental contamination. The main source of exposure to PAHs for the adult is food, which contribute to more than 90% of total exposure (WHO 1998, SCF 2002). However for smokers, significant contribution of PAHs exposure may be attributed to cigarette smoking. The additional intake of one of the PAHs, benzo[a]pyrene, for a person smoking 20 cigarettes per day was estimated to be 210 µg, which is in the same order of magnitude of the mean intake from food (the mean benzo[a]pyrene intake from food was about 110 µg per day(SCF 2002). Other minor routes of exposure to PAHs are inhalation of polluted ambient and indoor air, ingestion of house dust, and dermal absorption from contaminated soil and water (Gomma et al., 1993).

PAHs have solubility in water but are readily soluble in organic solvent or organic acids. Thus environments, PAHs are generally found adsorbed on particulates and on humic matter, or dissolved in any oily contamination that may be present in water, sediment soil. The solubility of PAHs in water is inversely proportional to the number of rings the PAHs molecule contains (EFSU, 2000).

PAHs are solids at room temperature. Since PAH tend to low vapour pressure, they are usually adsorbed on particulate matter in the atmosphere. The vapour pressure of PAH is inversely proportional to the number of rings contained and thus almost all five-ring PAHs compounds are particulate bound (Figure 1). While three-ring PAHs are also present as vapour in the atmosphere.

Low molecular weight PAHs (2 - 3) are volatile, therefore they are present in relatively small concentration (Olabemiwo et al., 2013a) in the
environment mainly in the vapour phase, while heavier PAHs (more than 5 rings) are predominantly adsorbed on organic particulate matter, usually on small particles (<2.5 μm). Four ring compounds have an intermediate behavior.

PAHs are chemically stable, poorly degraded by hydrolysis, but are susceptible to oxidation and photo-degradation. They are lipophilic compounds and they have a very poor aqueous solubility, which can be improved by the present of detergents or organic compounds (such as caffeine).

**Occurrence of PAHs in diet**

As PAHs are ubiquitous in the environment, it is not surprising that they are present in almost all food. For example, it has been reported that cereals were found to contain PAHs at levels of 6 - 14 μg/kg, fats and oils at 8 - 11 μg/kg and seafood at 7 - 8 μg/kg respectively (FSA, 2000; Falco et al., 2003). However, a high level of PAHs is not usually observed in raw food (WHO, 1998). Food processing or cooking steps such as roasting, grilling, barbecuing and smoking generate PAHs and increase the level of PAHs in the food being cooked (SCF, 2002). Charred food of almost any composition contains PAHs (Phillips, 1999), while only very low level of PAHs was detected when food was cooked by some cooking steps such as steaming. A study on the level of total PAHs formed in duck meat showed that levels as high as 130 and 320 μg/kg were found when the duck meat was cooked by roasting and charcoal grilling respectively, whereas only less than 8.6 μg/kg was detected when cooked by steaming (Chen and Lin, 1997). In overseas studies, cereals were found to be the main dietary source of PAHs, accounting for some 27 to 35% of total dietary exposure, a result probably due to the high amount of consumption. Although barbecued food only contributed a smaller part of the PAHs intake, people with a diet rich in roasted, barbecued or grilled, smoked food may have significant intake of PAHs (SCF Annex, 2002). The occurrence of PAHs in food is mainly due to:

i.) Environment contamination (through atmospheric fallout).

ii.) Food processing involving treatment at high temperature or direct contact with combustion fumes (grilling, toasting, smoking, e.t.c).

iii.) Vegetables are mainly contaminated by deposition of small airborne particulates. Broad-leaved vegetable in areas exposed to industrial emission and / or motor vehicle exhausts can be heavily contaminated.

iv.) Due to their polar nature, fat and oils are particularly prone to PAH contamination. Environmental contamination during plant development or manipulation during food processing is responsible for the "background contamination" observed in vegetable oils. The practice to dry seeds or olive pomace with system involving direct contact with combustion fumes, before oil extraction, always leads to high contamination levels that fortunately can be reduced during oil refining.

v.) Source of contamination for marine organisms can includes oil spills, run-offs from land and industrial effluents leaking from creosote wharfs and pilings. Aquatic organism present a degree of contamination that depends on the degree of contamination of the water where the live in and their capability to metabolize there xenobiotics.

vi.) Food processing involving direct contact with combustion gases, such as grilling and smoking, represents the main source of PAHs in foodstuffs. During grilling it important to avoid fat dripping onto the heat source responsible for fat hydrolysis producing large amount of PAHs that are then deposited onto the food surface by the smoke.

**Sources of food contamination**

They waxy surface of vegetable and fruit is able to concentrate low molecular mass PAHs through surface adsorption and particle-bound high molecular mass concentrate the surface due to atmospheric fallout. PAHs can also contaminate foods during industrial smoking, heating and drying processed that allow combustion products to came into direct contact with food. (Including seed oils and olive residue oils) oil (Speer et al., 1990).

PAHs are also formed as a result of certain home of food preparation methods, such as grilling, and smoking. High PAHs concentration have been reported charcoal grilled/barbecued foods (such as fatty meat and products grilled under prolonged and severe condition), in foods smoked by traditional techniques (fish in particular), and in mussels and other seafood from polluted water (Phillips, 1999). Smoked and grilled food is a large of the usual diet.

The presence of PAHs on coffee has also been reported and it has been suspected to be due to either a contamination of green coffee beans during the drying step or an endogenous formation in the coffee beans during the roasting (Houessou et al., 2007).

**Harmful health effects and severity of PAHS**

Several factors will determine whether harmful health effects will occur and what the type and severity of those health effects will be, the effect on human health depend mainly on the extent of exposure, the amount one is exposed to (or concentration), the innate toxicity of the polycyclic aromatic hydrocarbon (PAHs) and whether exposure occurs via inhalation, ingestion or skin contact.
Table 1. Genotoxicity and Carcinogenicity of some PAHs (IARC, 1987).

<table>
<thead>
<tr>
<th>Common name</th>
<th>Genotoxicity</th>
<th>IARC Classification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Acenapthene</td>
<td>Questionable</td>
<td>not yet evaluated</td>
</tr>
<tr>
<td>Acenaphthylene</td>
<td>Questionable</td>
<td>not yet evaluated</td>
</tr>
<tr>
<td>Anthracene</td>
<td>Negative</td>
<td>3</td>
</tr>
<tr>
<td>Benz(a)anthracene</td>
<td>Positive</td>
<td>2A</td>
</tr>
<tr>
<td>Benzo(b)fluoranthene</td>
<td>Positive</td>
<td>2B</td>
</tr>
<tr>
<td>Benzo(k)fluoranthene</td>
<td>Positive</td>
<td>2B</td>
</tr>
<tr>
<td>Benzo(g,h,i)Perylene</td>
<td>Positive</td>
<td>3</td>
</tr>
<tr>
<td>Benzo(a)Pyrene</td>
<td>Positive</td>
<td>2A</td>
</tr>
<tr>
<td>Chrysene</td>
<td>Positive</td>
<td>3</td>
</tr>
<tr>
<td>Dibenzo(a,h)anthracene</td>
<td>Positive</td>
<td>2A</td>
</tr>
<tr>
<td>Fluoranthene</td>
<td>Positive</td>
<td>3</td>
</tr>
<tr>
<td>Fluorene</td>
<td>Negative</td>
<td>3</td>
</tr>
<tr>
<td>Indeno(1,2,3-cd)Pyrene</td>
<td>Positive</td>
<td>2B</td>
</tr>
<tr>
<td>Phenanthrene</td>
<td>Questionable</td>
<td>3</td>
</tr>
<tr>
<td>Pyrene</td>
<td>Questionable</td>
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</tr>
</tbody>
</table>

IARC Classification

Group 1: The agent is carcinogenic to humans
Group 2A: The agent is probably carcinogenic to humans
Group 2B: The agent is possibly carcinogenic to humans
Group 3: The agent is not classifiable as to its carcinogenicity to humans

Intake of polycyclic aromatic hydrocarbon (PAHs) from contaminated soil may occur via inhalation of polycyclic aromatic hydrocarbon vapours, SCF 2002. According to WHO (2005), the occupational exposure to high levels of pollutant mixtures containing polycyclic aromatic hydrocarbon resulted in symptoms such as eye irritation, vomiting, convulsion and nausea. However, it is not known which of the mixture components were causative for these effects. Mixtures of polycyclic aromatic hydrocarbon are known to cause skin effects in animals and humans such as irritation and inflammation.

Health effects from long term exposure to polycyclic aromatic hydrocarbon may include liver damage, kidney problem, cataracts and jaundice. Repeated contact with skin may induce redness and skin inflammation. Naphthalene, a specific polycyclic aromatic hydrocarbon can cause the breakdown of red blood cells if inhaled or ingested in large amounts (SCF, 2002). Long-term studies of workers exposed to mixtures of polycyclic aromatic hydrocarbon and other places chemicals have shown an increased risk of skin, lung, bladder and gastrointestinal cancers. Studies have also reported asthma like symptoms, lung function, abnormalities, chronic bronchitis and decrease in immune function.

Some PAHs have been shown to have genotoxic effects both in vivo in rodents and in vitro in mammalian (including human) cell lines and prokaryotes (WHO, 1998). On the other hand, some PAHs do not appear to be genotoxic. The genotoxicity of PAHs is summarised in Table 1.

The International Agency for Research on Cancer (IARC) of the World Health Organization has evaluated the carcinogenicity of some PAHs based on evidence in human and experimental animals. The IARC’s classification of some PAHs are summarised in Table 1. Most of the PAHs evaluated are classified as Group 2B (possibly carcinogenic to humans) or Group 3 (not classifiable as to its carcinogenicity to humans). Three PAHs, namely, benzo[a]pyrene, benz[a]anthracene and dibenzo[a,h]anthracene, cause greater health concerns since they are classified as Group 2A (probably carcinogenic to humans). None of the PAHs are classified as Group 1 (carcinogenic to humans).

The carcinogenicity of the three PAHs of more concern is summarised as follows. Benzo[a]pyrene, when administered by the oral route, produced tumours of the gastrointestinal tract (forestomach), liver, lung and mammary glands of mice and rats (WHO, 1991; NTP, 2002). Dibenzo[a,h]anthracene and benz[a]anthracene produced tumours of the gastrointestinal tract (forestomach), lungs and liver in mice (SCF, 2002; NTP, 2002). Benz[a]anthracene also induced papillomas of the forestomach in mice when administered by gavage, and induced lung adenomas and hepatomas in mice in another gavage study (NTP, 2002).

Save level of polycyclic aromatic hydrocarbons intake

Prudent public health practice is to minimize and reduce the exposure to any agent that may have cancer causing potentials. Estimation of save PAHs intake levels are problematic because of the complexity of such mixture, regarding cancer risk, this is complicated by the need to high dose benzo (a) pyrene animal studies and by differences in risk estimation. Since carcinogenicity is the
critical end point of toxicity of PAHs and that some PAHs are genotoxic, it is not possible to define a level of intake which is without possible risk. According to Miller (1997), benzo(a)pyrene was the only PAH compound that has been evaluated by the joint FAO/WHO report committee on food additive (JECFA), but the committee was unable to establish a level of tolerable intake for benzo(a)pyrene. The committee however commented that there was a large difference between estimated human intake of benzo(a)pyrene and the doses producing tumours in animals. Thus any effects on human health are likely to be small. Despite this, JECFA is of the opinion that efforts should be made to minimize human exposure to benzo(a)pyrene as far as practicable.

Advice to trader on how to reduce PAHs exposure

(a) For preparing roasted food, gas grilling or electric oven roasting is preferred to charcoal grilling. For making dried meat, electric grilling is preferred to gas grilling.
(b) Heating chamber should be properly designed to avoid fat dripping onto the heat source.
(c) When roasting meat:
   i. Direct contact of meat with flame should be avoided. This could be achieved by placing the meat further from the heat source.
   ii. Fat dripping onto the heat source should be avoided. This could be achieved by trimming the visible fat from meat before roasting.
   iii. Meat could be cooked at lower temperature and avoid overcooking. However, the meat should be cooked thoroughly to destroy foodborne pathogens.
   iv. Prior to grilling or roasting, the meat could be cooked partially by a method which employs a lower cooking temperature such as boiling.

Advice to the public on PAHs exposure reduction

(a) Do not overindulge in roasted food, particularly charcoal grilled “Siu Mei” and the “skin and fat” portion.
(b) Remove the charred parts of food.
(c) Have a balanced diet and eat more fruits and vegetables.
(d) When going for roasting food, members of the public are advised to
   i. Trim visible fat from meat before roasting;
   ii. Partially cook the meat, e.g. by boiling, before roasting;
   iii. Avoid dripping fat onto the charcoal when roasting meat by putting the charcoal on sides of the stove and grilling food in the centre;
   iv. Consider to grill meat in foil packets to avoid contamination from smoke and flame; and
   v. Place the meat further from the heat source when roasting meat and avoid overcooking of meat. However, the meat should be cooked thoroughly to destroy foodborne pathogens.

Conclusion

Polycyclic aromatic hydrocarbons are present in the water, air, soil and food, both raw and processed foods but higher in the processed food than the raw foods. Contamination of food with PAH via environmental contamination should be controlled either by source-directed measures like filtering the smoke from relevant industries (e.g. cement work, incinerator and metallurgy) and limiting the exhaust fumes of PAH from cars. Also efforts should be made by the traders to reduce contamination with PAHs during drying and smoking processes e.g. by replacing direct smoking (with smoke developed in the smoking chamber, traditionally in smoke houses) with indirect smoking.

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Scientific Committee on Foods of EC (SCF), (2002). Opinion of the Scientific Committee on Food in the risk to human health of PAHs in food. Brussels: SCF.


