Archives of Ecotoxicology, Vol. 5, No. 2, pp. 61-65, 2023

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Archives of Ecotoxicology

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Contamination of Polycyclic Aromatic Hydrocarbons (PAHs) on Processed Food: A Review

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Article info
Received 2 September 2023
Revised 24 September 2023
Accepted 7 November 2023
Published online 10 November 2023

Mini review

Keywords:
Endocrine Disruptor;
Carcinogenic, Mutagenic,
Smoking; Roasting, Frying;
Barbecue

Abstract

Polycyclic Aromatic Hydrocarbons (PAHs) are wide range of environmental pollutants that commonly contaminate processed food as combustion by-products and constitute a wide range of toxic, carcinogenic, mutagenic adverse health risk. PAHs as an endocrine disruptor can also interfere with the production, release, metabolism, and elimination of unwanted substance in the body or can mimic the occurrence of natural hormones. PAHs formed during food processing largely depends on a number of factors such as type of the food, heating and processing methods. The major sources of PAHs in the environment includes petrogenic, pyrogenic and biological sources. PAHs contamination on processed food via processes such as smoking, roasting, frying or grilling (barbecued) is a function of both the fat content in the food substance and its proximity to the heat source. Studies revealed that eating contaminated food substances can cause reduced pregnancy, foetal reabsorption, gastric neoplasms, increased liver weight, fibrosis, having neurological and carcinogenic risk. PAHs can be removed from the environment via the following processes of either Biodegradation, Photolysis degradation, Dry deposition and/or Wet deposition.

1. Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) are wide range of environmental pollutants, which were found in air polluted areas (Bostrom *et al.*, 2002), processed food as combustion by-products and constitute a wide range of toxic, carcinogenic, (Ezike and Ohen, 2018; Akpambang *et al*, 2009), mutagenic, (Forsberg *et al*, 2011; Lee, *et al*, 2015) organic compounds, (Forsberg, 2011; Lee, *et al*, 2015) based on two or more aromatic rings and they belonging to the Food and Environment Contaminants, (Lee, *et al*, 2015. PAHs are reported to compromised sperm capacities and modified endocrine hormone levels (Jeng and Yu, 2008). Benzo[a]pyrene, a PAH substance was found to accumulated in the testis of an exposed rat (Ramesh *et al*., 2001), disturbed testicular steroidogenesis (Inyang *et al*., 2003) and actuated DNA harm (Ramesh *et al*., 2017). Incomplete combustion of petroleum derives was also reported to deplete and reduce sperm and androgen generation and changed the structure and functions of two major sorts of substantial testicular cells namely the interstitial Leydig cells, which are responsible for androgen hormone production, and the Sertoli cells of seminiferous tubules, which support spermatogenesis (Watanabe, 2005; Yoshida *et al*., 1999). Human studies of infertile populations (Xia *et al.,* 2009) and occupationally exposed individuals (Hsu et al., 2006) suggested a negative association between semen quality and PAH levels.
The environment exposure of PAH is reported to associate with sperm and DNA damage among men in the industrialized region of China (Han *et al.,* 2011). So also, smoking was also connected to a noteworthy decrease in sperm production and quality (Vine *et al.,* 1994).
Collectively, these make the testicles a coordinate target for antagonistic wellbeing impacts of PAHs. PAHs are well-recognized endocrine disturbing chemicals and regenerative toxicants (Ramesh *et al*., 2017). Most past studies on PAHs ecotoxicity has been essentially centered on the high atomic weight (HMW PAHs ≥5) rings. HMW PAHs likely intervene their endocrine-disrupting impacts acting through aryl hydrocarbon, estrogen or androgen receptors, with structure-dependent agonistic or antagonistic impacts (Zhang *et al.,* 2016).

2. Polycyclic Aromatic Hydrocarbons (PAHs)

Polycyclic aromatic hydrocarbons (PAHs) are a group of chemicals that are formed as a result of incomplete burning of coal, gas, wood, garbage, or other organic substances, such as tobacco, which can bind and form small particles matter in the air, (CDC, 2009). They are large group of organic compounds containing two or more fused or condensed aromatic rings made up of carbon and hydrogen atoms, (Ezike and Ohen, 2018; Orodu and Samuel, 2018) formed as a result of incomplete combustion processes, and may originate from petrogenic or pyrogenic sources, (Forsberg, 2011) and also in natural processes, such as carbonisation such as volcano, (FSA, 2015). PAHs are found in smoked food as combustion by-products and constitute a wide range of toxic, carcinogenic, (Ezike and Ohen, 2018; Akpambang *et al*, 2009), mutagenic, (Forsberg *et al*, 2011; Lee, *et al*, 2015) organic compounds, (Forsberg, 2011; Lee, *et al*, 2015) based on

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https://doi.org/10.36547/ae.2023.5.2.61-65

two or more aromatic rings and they belonging to the Food and Environment Contaminants, (Lee, *et al*, 2015).

PAHs occurs mostly as a complex mixture not as a single compound and can occur naturally, but can also be synthesized industrially as an individual compound for research purposes; this however, is not as the mixtures found in combustion products, but as pure chemical compound. They are classified as relatively persistent organic environmental contaminants, (Mičuli *et al,* 2011). They exist either as colourless, white, or pale yellow-green solids. They possess a relatively pleasant odour (FSA, 2015). PAHs are used in medicines and making dyes, plastics, and some pesticides, or asphalt used in road construction (CDC, 2009). They can also be found in substances such as crude oil, coal, charcoal, coal tar, and roofing tar. They can also be found in the air, water, and soil. They can occur in the air, either attached to dust particles or be deposited as solids in soil.

3. Classification of PAHs

PAHs are broadly classified into two main groups based on their physical and biological properties as High molecular weight (HMW) and low molecular weight (LMW) PAHs.

The High Molecular Weight (HMW) PAHs: These are PAHs that contains 4 – 6 aromatic rings and has a lesser bio-degradability potential by microorganisms, hence they are said to persist in the aquatic ecosystem, by bio-accumulating in aquatic organisms like fish and mussels and are more carcinogenic to humans, (Orodu and Samuel, 2018).

The Low Molecular Weight (LMW) PAHs: This class of PAHs contains 2 – 3 aromatic rings and they possess a lesser carcinogenic degree to, but are also having toxic effect to many aquatic organisms, (Orodu and Samuel, 2018)

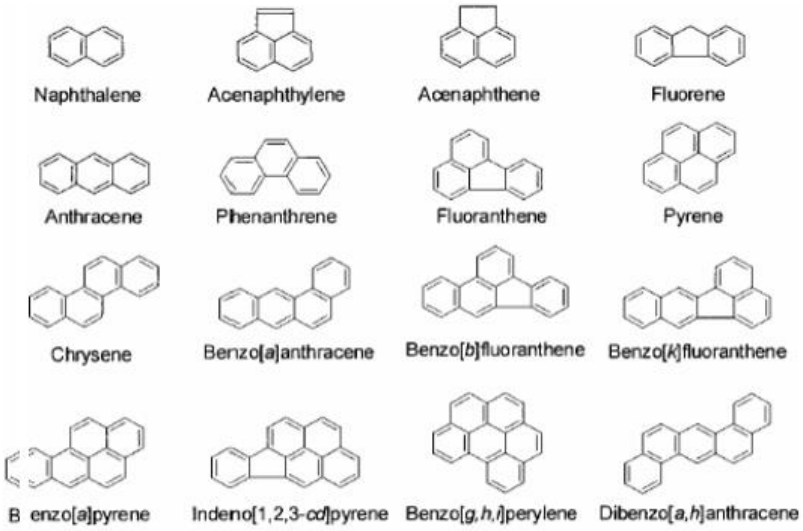

Fig 1 Structure of some PAHs **Sources**: culled from Amos-Tautua (2013)

Similarly, Abdel – Shafy and Mansour (2016) classified PAHs into "small" and "large" PAHs. Stating that PAHs containing up to six (6) condensed aromatic rings as small PAHs while those that contains more than six (6) condensed aromatic rings are categorized as large.

4. Sources of Polycyclic Aromatic Hydrocarbons (PAHs)

Generally, the major sources of PAHs in the environment includes petrogenic, pyrogenic, and biological sources, (Abdel – Shafy and Mansour, 2016; Forsberg, 2011; FSA, 2015).

Petrogenic Sources: PAHs can contaminate the land, water and air via the process of crude oil extraction, transformation, transportation and storage, (Abdel – Shafy and Mansour, 2016). The major contaminated ecosystems include freshwater bodies and oceans as a result of oil spillage and air as a result of transformation processes.

Pyrogenic Sources: This can be unintentionally as a result of incomplete combustion, (Forsberg, 2011; FSA, 2015) or intentionally as a result of exposure of organic matter to a very high temperature under a low or no oxygen such as cracking of crude oil into lighter hydrocarbons, distillation of coal into tar and coke, (Abdel – Shafy and Mansour, 2016).

Biological Sources: PAHs are also said to be generated by some certain plants and bacteria and also through the degradation of vegetative matter in an ecosystem, (Abdel – Shafy and Mansour, 2016).

The mode and process of formation and introduction of PAHs into the environment can be either anthropogenic or natural, (Abdel – Shafy and Mansour, 2016; Forsberg, 2011; FSA, 2015).

5. Route of exposure to PAHs

Route of exposure include inhalation, ingestion and dermal contact, (Abdel – Shafy and Mansour, 2016; FSA, 2015; Lee *et al*, 2015; Kobayashi *et al*, 2007).

Inhalation: PAHs can bind and form small particles matter in the air. An individual becomes exposed living near hazardous waste sites or by breathing polluted air from cigarette smoke, wood smoke, coal smoke, and smoke from many industrial sites which mostly contains PAHs. Reported symptoms include reduced lung function, abnormal chest X – ray, cough, bloody vomit, throat and chest irritation

Ingestion: Eating contaminated food such as smoked, fried, roasted food substances and/or drinking contaminated water. Studies revealed the following effects reduced pregnancy, foetal reabsorption, gastric neoplasms, increased liver weight, fibrosis, having neurological, carcinogenic risk (Abdel – Shafy and Mansour, 2016; Forsberg, 2011; FSA, 2015).

Dermal contact: PAHs can also enter the human system through the skin, when the skin comes in contact with a contaminated soil (hazardous waste dumping site) or with used crankcase oil or robbing other products that contain PAHs. Observed symptoms include tumours, carcinomas, malignant

The major general routes of exposure to PAHs for non-smokers are from processed and/or preserved food involving thermal treatment having direct contact with the heating gas during such process and inhaled polluted air. (FSA, 2015).

The rate at which PAHs enter human system either via inhalation, ingestion or dermal contact can be influenced by the presence of other compounds an individual is exposed to, at the same time with the PAHs. PAHs can enter all the tissues of the human system containing fat because of their lipophilic chemical character, (Lee *et al*, 2015; Kobayashi *et al*, 2007). Larger amounts of PAHs are stored in the kidneys, liver, and fat with a smaller amount in the spleen, adrenal glands, and ovaries. PAHs are transformed in the body tissues into many different substances. Some of which substances are more harmful while others are less harmful than the original PAHs. Some can be eliminated in the body system, primarily in the faeces and urine. (CDC, 2009).

6. Processed Food and PAHs Contamination

Food substances can become environmentally contaminated in rare case as a result of exposure to atmospheric contaminated particulate matter (Kobayashi *et al*, 2007), most of the contamination comes through food preservation and processing methods involving high temperature having direct contact with

combustion gases, such as smoking, roasting, frying, (Ezike and Ohen, 2018; Abdel – Shafy and Mansour, 2016). When food particularly meat, fish and their products are being smoked, roasted, barbecued, or grilled; PAHs are then formed as a result of incomplete combustion or thermal decomposition of these organic matter (Amos-Tautu *et al*, 2013). Pyrolysis of the fats in the food substances generates PAH that become deposited on the such food item in question. PAH production by either smoking, roasting, frying or grilling (barbecued) is a function of both the fat content in the food substance and its proximity to the heat source, (Amos-Tautu *et al*, 2013; FSA, 2015, CDC, 2009). Food processing involving high temperature like smoking, frying, roasting is said to be the major source of food contamination with PAHs level of 130 - 200 µg/kg found in smoked food product against 0.01 – 1 µg/kg in the unsmoked food product, (Abdel – Shafy and Mansour, 2016)

Amos-Tautua *et al*, (2013) discovered that roasted beef meat (suya meat) and smoked mackerel fish were contaminated with PAHs but surprisingly it wasn't detected in roasted plantain. Smoking of meat and fish being one of the major sources of PAHs into the human food chain does not only gives special taste, colour and aroma to food substance, but also enhances preservation due to the dehydrating, bactericidal and antioxidant properties of smoked food substance, (Mičuli *et al*, 2011) and as such smoking becomes the commonest age longed method of food processing and preservation having the potential of exposing larger population across the different social strata to PAHs.

Foods of animal origins are also said to be another source of PAHs given that PAHS are lipophilic and as such fatty foods like eggs or dairy products like milk, yoghourt, butter, can easily be contaminated (Lee *et al* 2015). It is worthy of note, that PAHs can be found in some cow's milk probably not only because of the lipophilic character of PAHs but maybe as a result of grazing on polluted land. Some PAHs are having bio – accumulation potentials (Amos-Tautua *et al*, 2013) as they are found to accumulated in the tissues of some mollusc like clams, oysters, species that are exposed to various kind of PAHs following oil spillage in the ocean and be contaminated as well (Lee *et al*, 2015; Kobayashi *et al*, 2007) this increases the chances of contaminating food sourced from the marine as the marine food web can be contaminated. Due to stable structure and lipophilic character of PAHs, they are apt to concentrate and intensify in the food chain notably related to fat (Lee *et al* 2015).

Studies conducted by Lee *et al* (2015) on seafood and dairy products revealed that the total PAHs concentration was 1.06 µg/kg in seafood and 1.52 µg/kg in dairy products. Similarly, in another study by Mičulis *et al*, (2011) they concluded that the production of smoked food products (meat) with PAH4 levels less than 10 µg kg^{-1} can be possible in a non-intensely smoked product.

The Food Safety Authority of Ireland (FSA), (2015) quoted EU Scientific Committee on Food (SCF) in identifying 15 PAHs compounds which are regarded as carcinogenic genotoxic i.e. benz(a)antrhacene, benzo(b)fluoranthene, benzo(j)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene, chrysene, cyclopenta(c,d)pyrene, dibenz(a,h)anthracene, dibenzo (a,e) pyrene, dibenzo(a,h)pyrene, dibenzo(a,i)pyrene, dibenzo(a,l)pyrene, indenol (1,2,3-cd) pyrene, and 5-methylchrysene. Again, it was discovered that benzo (α) pyrene, has higher carcinogenic value than other PAHs compound, while benzo (α) pyrene contribute 120% from total carcinogenic that found in foodstuff such as smoked products (Swastawati *et al*, 2015).

7. Ecotoxicity of PAHs

Most studies on experimental animals on individual PAHs, most especially on benzo[a]pyrene, have revealed various degree of toxicological effects, such as haematological effects, reproductive and developmental toxicity, immunotoxicity, (FSA, 2015) and possessing a teratogenicity potential, (Hokkanen *et al*, 2018; Abdel – Shafy and Mansour, 2016). Their health implications such as carcinogenic and genotoxic (DNA-damaging) potential coupled with their ubiquitous nature in the environment has caused most concern among public health workers. Some PAHs have shown carcinogenicity, genotoxicity and mutagenicity in experimental animals (Amos-Tautua, *et al*, 2013). Some other PAHs have also been identified as being carcinogens with possible genotoxic properties but due to their lipophilic nature, some can be metabolised or broken down faster, both in human system and also in the environment, persistence for long periods of such group is not such a major problem. PAHs are said to have teratogenic, haematological, and immunotoxic effects, (Hokkanen *et al*, 2018; Abdel – Shafy and Mansour, 2016) exposing the yet unborn to health danger.

Ecotoxicity of PAHs in the aquatic ecosystems is influenced by metabolism and photo – oxidation and are said to be more toxic in the presence of ultraviolent light, possessing a moderate to high toxicity to aquatic organisms and birds (Abdel – Shafy and Mansour, 2016).

Terrestrial organisms are also adversely affected as it is said to be toxic to invertebrates inhabiting the soil, affecting their development, reproduction. Subsequently, plants are said to absorb and translocate PAHs from contaminated soil through their roots, while other plants are said to naturally synthesize PAHs which serves as their growth hormones, (Abdel – Shafy and Mansour, 2016). Mammals can absorb PAHs via ingestions, inhalation and dermal contact, (Abdel – Shafy and Mansour, 2016; FSA, 2015; Lee *et al*, 2015; Kobayashi *et al*, 2007) which also has a wide range of toxic, carcinogenic, (Ezike and Ohen, 2018; Akpambang *et al*, 2009), mutagenic, (Forsberg *et al*, 2011; Lee, *et al*, 2015).

Tab 1 PAHs considered by SCF 2002 to be mutagenic/genotoxic

S/N	Name	Abbreviation
1	Benz[a]anthracene	BaA
2	Benzo[b]fluoranthene	BbFA
3	Benzo[j]fluoranthene	BjFA
4	Benzo[k]fluoranthene	BkFA
5	Benzo[ghi]perylene	BghiP
6	Benzo[a]pyrene	BaP
7	Chrysene	CHR
8	Cyclopenta[cd]pyrene	CPP
9	Dibenz[a,h]anthracene	DBahA
10	Dibenzo[a,e]pyrene	DBaeP
11	Dibenzo[a,h]pyrene	DBahP
12	Dibenzo[a,i]pyrene	DBaiP
13	Dibenzo[a,l]pyrene	DBalP
14	Indeno[1,2,3-cd]pyrene	IP
15	5-Methylchrysene	5-MCH

FSA, (2015)

Tab 2 PAHs highest permissible limit in food

S/N	FOODSTUFFS	PAH	MAXIMUM LEVELS (µg/kg)
1	Dietary foods for special medical purposes intended specifically for infants	Benzo(a)pyrene	1.0
		Sum of PAH4	1.0
2	Oils and fats (excluding cocoa butter and coconut oil) intended for direct human consumption or use as an ingredient in food	Benzo(a)pyrene	2.0
		Sum of PAH4	10.0
3	Smoked meat and smoked meat products	Benzo(a)pyrene	2.0
		Sum of PAH4	12.0
4	smoked fish and smoked fishery products	Benzo(a)pyrene	2.0
		Sum of PAH4	12.0
5	Infant formulae and follow-on formulae, including infant milk and follow-on milk	Benzo(a)pyrene	1.0
		Sum of PAH4	1.0
PAH4 = benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoranthene and chrysene			

FSA, (2015): Toxicology Factsheet Series

8. Control and Management of PAHs

PAHs formed during food processing largely depends on a number of factors such as type of the food, heating and processing methods. The following measures can be applied to prevent PAHs contamination on food

I. Reduced contact of the food substance with combustion products during processing
II. Reduced food contact with flames most especially during barbecue and other food processing methods
III. Reduce the amount of fats and oil in grilling and processing food having direct contact with the combustion products
IV. Use low - medium heat and place the food substance far away from the heat source
V. Broiling having the source of the heat above the food substance, can significantly reduce PAH levels.
VI. Fat should not drip down onto an open flame, sending up a column of smoke that coats the food with PAHs.

PAHs can be removed from the environment via the following processes

Biodegradation: Certain bacteria like *Rhodococcus, Paracoccus, Sphingomonas* and *Nocardia* are said to degrade PAHs in both aerobic and anaerobic degradation process in the soil. The bioavailability of PAHs to bacteria is a function of their state (i.e. either dissolved or vapour state) as bacteria the ability to degrade PAHs in a solidified state, (Abdel – Shafy and Mansour, 2016).

Photolysis degradation: This involves the destruction of PAHs from a reaction initiated by light absorption. Coloured lighted particles like silica, aluminium is said to be more effective than dark particles such as carbon. Again, aqueous and vapour state PAHs are also more readily photo – degraded than its solidified state, (Abdel – Shafy and Mansour, 2016).

Dry deposition: Solidified PAHs tends to settle on the earth in the absent of any form of precipitation which depends on the properties of the PAHs, atmospheric conditions, temperature etc. (Abdel – Shafy and Mansour, 2016).

Wet deposition: This is the process whereby the solidified or vapoured PAHs are dissolve and removed from the atmosphere along with precipitation. This is influenced by the properties of the PAHs, temperature, climatic condition etc., (Abdel – Shafy and Mansour, 2016).

9. Endocrine Disruptors

Endocrine disruptors are exogenous compounds with the potential to disturb hormonal regulation and the normal endocrine system, consequently affecting health and reproduction in animals and humans (USEPA, 2006). Endocrine disruptors can interfere with the production, release, metabolism, and elimination of or can mimic the occurrence of natural hormones (Tabb and Blumberg, 2006).

Endocrine disruptors may also be derived from natural animal, human, or plant (phytoestrogen) sources; however, the current global concern is focused on synthetic endocrine-disrupting chemicals (EDCs) such as the PAHs (Peter *et al*,, 2017). This concern is further amplified by two factors, the wide range in chemical production and usage globally, and the increased pollution from these chemicals in the air, land and water. As such, the impacts of these synthetic chemicals on organisms through known or unknown effects on hormonal systems is a major concern to scientists.

Although EDCs can target various hormone systems, many observations concerning reproductive development and sex differentiation, together with early embryonic development and puberty, have focused on EDC interference with sex steroid hormones**.** Hormone-like chemicals had harmful effects on the organs and bodily functions of test animals (Gore, 2015).

Human exposure to PAHs occurs through both air (cigarette smoke, fossil fuel and wood combustion) and contaminated processed food involving thermal treatment of the food with direct contact with the combustion gases, methods such as toasting, smoking, roasting etc. For non-smokers, exposure to airborne PAHs is highest in densely populated urban areas and in rural areas where wood and coal are frequently used. Coal and biomass burning for cooking and heating in developing countries has been shown to lead to high indoor concentrations of PAHs. However, food appears to be the major source of PAH intake in industrialized countries, with grilled or charred meats, smoked food, contaminated cereals and vegetables as major sources (Srogi, 2007).

10. Conclusion

Most processed food having direct contact with the combustion gases can be contaminated with PAHs. PAHs as an endocrine disruptor can also interfere with the production, release, metabolism, and elimination of unwanted substance in the body or can mimic the occurrence of natural hormones. The consumption of PAHs contaminated food substances can cause reduced pregnancy, foetal reabsorption, gastric neoplasms, increased liver weight, fibrosis and it was also discovered by studies to have neurological and carcinogenic risk. PAHs can be removed from contaminated environmental media but hardly from the human system as it forms part of the synthetic endocrine-disrupting chemicals (EDCs).

Declaration of interest

The authors report no conflicts of interest.

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