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ORIGIN, DISTRIBUTION AND IMPACT OF POLYCYCLIC AROMATIC HYDROCARBONS ON FOOD SAFETY AND HUMAN HEALTH

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Highlights:

- PAHs can be formed through two main mechanisms: pyrosynthesis and pyrolysis.
- Anthropogenic pyrolytic PAHs are the main source of PAHs in the environment.
- PAHs are ubiquitous compounds found in all environmental compartments
- Meat, meat products, oils, and fats are considered the main contributors to daily PAH intake through ingestion.
- Exposure to PAHs varies at an individual level depending on location and lifestyle habits.

ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are chemical compounds formed during incomplete combustion processes of organic materials such as coal, petroleum, wood, and certain industrial activities. The main sources of PAHs include emissions from the combustion of fossil fuels in vehicles, factories, and power plants. PAHs can also be released during waste incineration, wood burning, and food cooking. They are present in all environmental compartments, including air, water, soil, and living organisms. PAHs are associated with numerous adverse effects on human health and the environment. They are considered carcinogenic, mutagenic, and reprotoxic substances. Additionally, they can accumulate in the food chain, thereby affecting wildlife and flora. This article examines the origins, distribution and impact of PAHs on food safety and health and briefly presents the associated regulations.

Keywords: Polycyclic aromatic hydrocarbon, environment, health, food

1. INTRODUCTION

Polycyclic Aromatic Hydrocarbons (PAHs) are high molecular weight organic molecules. They belong to the subfamily of aromatic hydrocarbons, which are molecules composed of carbon and hydrogen atoms with at least two condensed aromatic cycles in their structure (Anietie et al., 2016). Due to their physicochemical characteristics, PAHs are detected in ecosystems ranging from polar to tropical regions. Consequently, they are found in the atmosphere, water, sediments, biota, and soils (Wilcke, 2007; Botta et al., 2014). Moreover, their lipophilic nature allows them to accumulate in the food chain (Shadi et al., 2012). PAHs are derived from incomplete combustion or pyrolysis of carbon compounds during natural processes (bushfires or volcanic eruptions), but mostly originate from anthropogenic activities (Akpambang et al., 2016; Kouadio et al., 2020). Among the 130 identified PAHs, a limited list is typically considered for environmental studies. These are the pollutants prioritized by the US Environmental Protection Agency (US-EPA) and include the most studied PAHs with environmental concerns (INERIS, 2005; Anietie et al., 2016). Indeed, PAHs are highly toxic pollutants due to their carcinogenic and mutagenic properties (Sparfel, 2018). In a rapidly developing context of industrialization and urbanization, PAHs pose a major challenge in terms of environmental pollution, food contamination, and public health. This literature review aims to provide an overview of the current knowledge on the origin, distribution, and impact of PAHs on food safety and human health. The following hypotheses can be considered:

- The main sources of PAHs in the environment are related to human activities.
- PAHs tend to accumulate in soil, sediments, and living organisms, which can result in contamination of terrestrial and aquatic food chains.
- Climate change can influence the distribution and biodiversity of PAHs.
- High-temperature cooking of food, such as barbecue and frying, can lead to the formation of PAHs in food.
- Chronic exposure to PAHs can increase the risk of cancer in humans.

2. ORIGIN

2.1. Pyrolytic PAHs

Pyrolytic PAHs originate from the incomplete combustion of organic matter. They are primarily formed during the combustion processes of coal, petroleum, and wood. Organic matter can be of natural origin, for example, from forest fires or volcanic eruptions, or anthropogenic, mainly from the combustion of fossil fuels, organic matter, or waste (Wang et al., 2001). Due to domestic and industrial emissions, anthropogenic pyrolytic PAHs are the main source of PAHs in the environment (Paris, 2017)."

2.2 Petrogenic PAHs

Petrogenic PAHs are organic compounds formed through the thermal degradation of organic matter in

sedimentary rocks such as petroleum and coal (Wang et al., 2001). They are typically emitted more directly into the environment, bypassing the atmosphere, through sources like road networks, oil spills, or the spreading of sludge from wastewater treatment plants. The introduction of petrogenic PAHs occurs on a restricted geographical scale, at a local level. Natural oil seepage and accidental oil spills at sea are the main causes (Crespo, 2009).

2.3 Diagenetic PAHs

Diagenetic PAHs are molecules that naturally form during the diagenesis process, which involves chemical and physical changes in sediments and rocks over time. They are formed in situ in sediments during the thermal maturation of organic matter (Wang et al., 2014). Due to their natural origin, diagenetic PAHs can be present in the environment even in the absence of human activities. Their impact on human health and the environment is considered less concerning than that of pyrolytic PAHs as they seem to only affect relatively small areas (Botta et al., 2014).

3. Formation of PAHs

PAHs can be formed through two main mechanisms: pyrosynthesis and pyrolysis.

Aromatic hydrocarbons can be formed through the dehydrogenation of aliphatic hydrocarbons and the polymerization of alkenes and alkynes. Monocyclic hydrocarbons can then undergo pyrosynthesis to become PAHs. Carbon-carbon and carbon-hydrogen bonds are broken, forming free radicals at temperatures exceeding 500°C. The combination of acetylene with these radicals creates aromatic structures (Kislov, 2013).

Regarding the pyrolysis of aromatic hydrocarbons, the primary products are mainly phenolic compounds. The pyrolytic degradation reaction of phenol involves the initial removal of carbon monoxide from the carbon carrying the hydroxyl group. The cyclopentadienyl radicals then combine in pairs to form tetralin, which thermally decomposes into naphthalene and most aromatic hydrocarbons (Cypres, 1975).

4. Distribution of PAHs in the Environment

Due to their physicochemical properties, PAHs are ubiquitous compounds found in all environmental compartments: the atmosphere, water, sediments, biota, and soils. Their widespread distribution in the environment is a result of multiple abiotic and biotic processes controlled by their physicochemical properties: volatilization, long-distance transport, atmospheric deposition, adsorption onto particles, sedimentation, bioaccumulation, microbial activities, chemical oxidation, and photooxidation (Botta et al., 2014).

4.1 Air

PAHs present in the atmosphere can be in a gaseous state or associated with particles. Heavier PAHs with lower vapor pressures bind quickly and almost exclusively to the particulate phase. Compounds with 2 aromatic rings are exclusively present in the gaseous phase, while compounds with at least 6 aromatic rings are mainly present in the particulate phase. PAHs with 3 to 6 aromatic rings are distributed between

the two phases proportionally to their degree of aromaticity (Albinet, 2006).

PAHs are predominantly found in the environment in particulate form: percentages range from 79% to 97% depending on the compound studied (Botta et al., 2014; Kafilzadeh, 2015). The distribution of PAHs between the gaseous and particulate phases also depends on temperature, which affects vapor pressure. High temperatures increase the representation of PAHs in the gaseous phase, while lower temperatures increase the level of PAHs in the particulate phase (Li et al., 2006).

The concentrations of particulate PAHs vary greatly. They range from a few tens of pg/m^3 in remote regions like Greenland (Jaffrezo et al., 1994) to a few hundred ng/m^3 near roads or in urban areas like Santiago, Chile, where they reach $750 \text{ ng}/\text{m}^3$ (Didyk et al., 2000). Volatile PAHs can travel several tens of kilometers before depositing on the ground, unlike heavy PAHs (more than 4 rings), which are less mobile and deposit near their emission source (Wania, 1996).

Aromatic hydrocarbons can also be removed from the atmosphere through dry deposition (particles and dust) and wet deposition (precipitation), contaminating water, soil, and plants, thereby raising concerns about transfer in the food chain and food products (Desalme, 2011).

4.2 Soil

Atmospheric humidity affects the concentrations of pollutants found at ground level. Precipitation helps to carry dust and pollutants from the atmosphere to the soil, with greater efficiency for larger particles (Dégremont, 2009). Temperature also affects the physical properties of pollutants by increasing the solubility and vapor pressure of PAHs, thereby reducing their adsorption in the soil at higher temperatures. Lower temperatures lead to decreased volatility of compounds, causing them to accumulate in the soil (Kipopoulou, 1999).

When introducing PAH molecules into soil, they can interact with different soil constituents and undergo processes such as volatilization, reaction with the solid phase of the soil, leaching or washing away by precipitation, degradation, or bioaccumulation by living organisms (Semple et al., 2003).

Individual PAH concentrations in soils from natural sources range from 1 to $10 \mu\text{g}/\text{kg}$ (Edwards, 1983). The presence of plant roots promotes the removal of aromatic hydrocarbons from the soil (Liste, 2000). PAH concentrations in soils increase with increasing industrial impact, traffic, or domestic heating. The order of increasing PAH concentrations in soils is: arable soils < mineral soils under forests < permanent grasslands < urban soils < contaminated industrial soils (Wilcke, 2007). PAH concentrations in agricultural soils significantly increase with the application of sewage sludge, manure, and compost. In fact, sewage sludge often contains PAH concentrations ten times higher than the soil (Wilcke, 2007).

4.3 Water

Wind, rain, and soil leaching are responsible for high concentrations of aromatic hydrocarbons found in

water and sediment in oceans and rivers (Cavalcante, 2010). In an aquatic environment, the less hydrophobic compounds, such as BTEX and light PAHs (2 and 3 aromatic rings), are mainly found in the water. The more hydrophobic PAHs (4 to 6 aromatic rings) preferentially accumulate in the organic fraction of sediments (Kafilzadeh, 2015).

The PAH content in water is greatly influenced by pollution from anthropogenic activities near sampling locations (Cavalcante, 2010). In Boston Harbor, which is the most polluted port in the United States, concentrations of PAHs (sum of 16) in surface sediments (approximately 10 cm) range from 7300 to 358000 $\mu\text{g}/\text{kg}$ (Wang et al., 2001).

5. Food Contamination

Contamination of raw food (animals and plants) occurs during growth, as well as during transportation, storage, and processing (drying, grilling, roasting, smoking, or frying). PAH concentrations are generally lower in fruits (ranging from 0.7 $\mu\text{g}/\text{kg}$ for bananas to 1 $\mu\text{g}/\text{kg}$ for peaches) and vegetables (ranging from 0.7 $\mu\text{g}/\text{kg}$ for onions to 4.1 $\mu\text{g}/\text{kg}$ for cauliflower) compared to fatty products (ranging from 18.1 $\mu\text{g}/\text{kg}$ for butter to 19.2 $\mu\text{g}/\text{kg}$ for margarine), dairy products (ranging from 0.6 $\mu\text{g}/\text{kg}$ for yogurt to 12.8 $\mu\text{g}/\text{kg}$ for cheese), and meat (ranging from 1.2 $\mu\text{g}/\text{kg}$ for pork loin to 364.9 $\mu\text{g}/\text{kg}$ for salami).

Meat, meat products, oils, and fats are considered the main contributors to daily PAH intake through ingestion, accounting for 57-71% and 8-12% respectively in adults (Martorell et al., 2010). This can be explained by the lipophilic nature of aromatic hydrocarbons, which have a higher affinity for lipids (Martí-Cid et al., 2008).

6. Effects on Human Health

Exposure to PAHs varies at an individual level depending on location (continent, country, region, rural/urban/industrial area, traffic density) and lifestyle habits (consumption of grilled meats, smoking). According to Lee et al. (2007), for a non-smoker, diet is the main route of exposure to PAHs.

In terms of diet, consumption of products processed through drying, smoking, grilling, or high-temperature cooking can contribute to the intake of pyrolytic PAHs and Heterocyclic Amines (HCAs) (Kouadio et al., 2020).

Epidemiological studies on PAHs are predominantly conducted in occupational settings (coke plants, refineries, or foundries). They reflect the effects of mixtures of PAHs through inhalation (Kim, 2013; Okona-Mensah, 2005). These studies report an increased risk of lung, colon, stomach, and bladder cancers among workers in these professional sectors (Kim, 2013; Sparfel, 2018). Among PAHs: benzo[a]pyrene is recognized as a proven human carcinogen, dibenz[a,h]anthracene as a probable carcinogen, and chrysene as a possible carcinogen. Other effects of PAHs on the cardiovascular system, as well as their mutagenic, reproductive toxicological, or immunosuppressive effects, are only partially known and have mainly been documented through animal studies (Sparfel L., 2018; Wenzl et al., 2006). These findings have led to increased regulations on PAHs.

7. Regulations and Control Measures for PAHs

Due to their recognized adverse health effects, regulatory tools have been put in place to establish target values for PAH concentrations in ambient air, drinking water, and food.

Regarding ambient air, the target value is set by Directive 2004/107/EC (2004) only for Benzo(a)pyrene at 1 ng/m³ as an annual average calculated based on the PM10 fraction of suspended particulate matter.

The World Health Organization (WHO) imposes a maximum limit of 0.2 µg/L for the following 6 PAHs (Fluoranthene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, benzo(g,h,i)perylene, indeno(1,2,3,cd)pyrene) in drinking water (WHO, 2008).

The European Food Safety Authority (EFSA) has recommended 4 PAHs as markers in food products: benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), and benzo[a]pyrene (BaP), and their occurrence in food indicates a cancer risk (EFSA, 2008; Zelinkova et al., 2015).

EU Regulation No. 835/2011 of August 19, 2011, amending Regulation No. 1881/2006 with regard to maximum levels for polycyclic aromatic hydrocarbons in foodstuffs also defines maximum levels of PAHs in certain food products (European Commission, 2011).

The Association of Vegetable Oil and Protein Industries in Europe (FEDIOL) sets an upper limit for total PAH content at 25 µg/kg, as well as an upper limit for heavy PAH content at 5 µg/kg (Benzo(a)pyrene, dibenzo(a,h)anthracene, Benzo(g,h,i)perylene, Benzo(e)pyrene, perylene, anthanthrene, coronene). It also states that when the benzo(a)pyrene content in crude oil exceeds 1 µg/kg, activated carbon treatment during refining is necessary (Lacoste et al., 2003).

8. CONCLUSION

The origin of PAHs is mainly attributed to anthropogenic activities related to incomplete combustion of organic matter. Their distribution is influenced by population density, industrial activities, fossil fuel use, and climate change. These molecules tend to accumulate in soil, sediments, and living organisms, posing a risk to terrestrial and aquatic food chains and therefore food security and human health. High-temperature cooking, such as barbecuing and frying, can indeed result in the formation of PAHs in food. Some PAHs have been identified as potential carcinogens, and chronic exposure to these substances poses a significant health risk.

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