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Evaluation of Polycyclic Aromatic Hydrocarbons Contamination in Fresh And Dried Sardinella Madarensis (Flat Sardine Fish) From the Sombriero River, Niger Delta, Nigeria

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Abstract

The Niger Delta is a region that has been significantly impacted by oil exploration and other anthropogenic activities, that have resulted in severe environmental pollution. Fish serves as a primary source of protein in the local diet; hence, assessing contamination levels in fish species commonly consumed is crucial for public health safety. This study evaluates the contamination levels of polycyclic aromatic hydrocarbons (PAHs) in composite samples of fresh and dried (traditionaly smoked) *Sardinella madarensis* fish collected from the Sombreiro River in Nigeria's Niger Delta. A total of 30 fish samples (15 fresh and 15 dried) were randomly collected from different vendors in locations along the riverbank and analysed as composites to provide an average contamination profile. PAHs were measured using gas chromatography-mass spectrometry (GC-MS), and the coefficient of variation (CV%) was used to evaluate the variability of contaminant concentrations. The results revealed significantly higher concentrations of 13 PAHs in dried fish, suggesting the influence of traditional smoking. Notably, the concentration of benzo(a)pyrene (BaP), a class I carcinogen, in dried fish (51.08 µg/kg), significantly exceeds the EU's maximum limit of 2 µg/kg for BaP in smoked foods. High concentrations of benzo(b)fluoranthene both fresh and dried fish samples, were also noted, suggesting its environmental prevalence. These findings revealed the associated health risks of fish consumption in the region, emphasizing the urgent need for continuous monitoring and improved processing methods to safeguard public health.

Keywords: Environmental Pollution, Niger Delta, Polycyclic Aromatic Hydrocarbons (PAHs), *Sardinella madarensis*.

Introduction

The Niger Delta area in Nigeria, rich is biodiversity as well as oil and gas deposits, covering over 20,000 square kilometers of wetlands, is home to over 20 million people from 40 ethnically diverse groups [1]. Unfortunately, several years of extensive oil exploration and related activities have led to significant environmental challenges in the region. It has been reported that over ten thousand oil spill incidents occurred in the Niger Delta between 2011 and 2022 [2]. These spill incidents, alongside illegal bunkering, industrial discharges, and urban development, have introduced harmful pollutants like polycyclic aromatic hydrocarbons (PAHs) into water systems[2, 3] such as the Sombreiro River, which serves as significant support for fishing and agriculture for nearby communities.

PAHs, a class of semi-volatile and volatile organic compounds with many fused aromatic rings, produced mostly during the incomplete combustion of organic biomass and fossil fuels are major contributors to environmental pollution[4, 5]. These hydrophobic compounds are known toxins that have the propensity to bioaccumulate in fish tissues, posing serious health risks for people who rely on fish as food [4, 6-8]. Usually, fish take in these PAHs through their gill, skin, or via ingestion [9]. When ingested, PAHs undergo metabolism and activation by the enzyme cytochrome P450, and then form electrophiles that covalently bind to DNA molecules, ultimately leading to mutations and possible genotoxic effects [10]. Smaller molecular weight PAHs, such as naphthalene and phenanthrene, are less toxic than the heavier ones, such as benzo(a)pyrene, which are more stable and possess greater toxicity [11]. The toxicity of PAHs increases synergistically with the number of aromatic rings present in their molecular structure [12]. Several PAHs have been classified as probable human carcinogens by the International Agency for Research on Cancer [13].

The fact that the oil-rich Nigeria's Niger Delta region is characterized by frequent oil spills from pipeline vandalism, operational mishaps, and inadequate infrastructural maintenance cannot be overemphasized, this represent a significant source of PAH contamination [14]. In addition, urban runoff and industrial effluents also contribute a substantial amount of PAHs to the rivers and coastal areas across Nigeria. This therefore raises concerns about the potential risk to aquatic and human life that rely on fish from that region for sustenance.

Fish, especially species like *Sardinella madarensis* (flat sardine fish), play a vital role in the marine food web and serve as important indicators of environmental health in the region. *S. madarensis* is an important fishery resource in the marine and brackish waters [15]. Reports from previous studies have shown that levels of PAHs, particularly the carcinogenic compound benzo(a)pyrene surpassing permissible safety limits have been reported in fish species and other seafood such as Clarias gariepinus, *Ethmalosa fimbriata*, *Chrysichthys nigrodigitatus*, *Macrobrachium macrobrachium* and *Tympanotonus fuscatus*, obtained from rivers in the Niger Delta [16,17]. Consumption of such PAH-laden fish can lead to a plethora of illnesses in humans, including neurological disorders, congenital anomalies, and increased risk of cancer [18,19].

Given the ecological and economic significance of the Sombreiro River, this study aims to quantify and compare the levels of 16 priority PAHs in fresh and dried *S. madarensis* samples from the Sombreiro River, Rivers State, Nigeria, and thus provide valuable insights into the ecosystem health and the health implications of consuming such fish.

Materials and Methods

Materials

Fresh and dried *S. madarensis* fish, PAH reference standards (PAH-Mix 16, Merck), Dichloromethane (Sigma Aldrich), anhydrous sodium sulfate, silica gel, and deionized water were the reagents used in this study. The analysis was carried out using a Gas chromatograph – Mass Spectrometer system (HP 5890 Series II, Agilent/Hewlett Packard).

Description of the study area

The Sombreiro River, situated between approximately latitude 4.75°N and 5.25°N and longitude 6.75°E and 7.25°E, flows through six local government areas in Rivers State, Nigeria—Akuku-Toru, Degema, Asari-Toru, Abua-Odual, Ikwerre, and Emohua. A tributary of the River Niger, it drains into the Atlantic Ocean. Extensive fishing activities are carried out there, serving as a vital source of livelihood for local communities. The fish samples were obtained from vendors operating along the river's bank.

Sample collection and preparation

Thirty *Sardinella madarensis* fish samples were randomly obtained from five different vendors on the banks of the Sombreiro River, 15 of which were fresh and 15 were obtained dried by smoking. The sampling was conducted by random sampling. The fresh fish samples were filleted, and the muscle tissues were homogenized, while the dried fish samples were ground into a fine powder using a clean stainless-steel blender. For both fresh and dried samples, composite samples were prepared independently by thoroughly mixing the respective samples. The sample size was adapted from Öhrvik, *et al.* [20] who estimated that a composite sample should consist of a minimum of 12 specimens. The processed composite samples were stored in pre-cleaned, airtight polyethylene containers at -20°C until further analysis to prevent degradation of contaminants.

PAHs Analysis

To quantify the PAHs, a method adopted by Valet *et al*. [21] and Hassani and Farahani[22] were used with slight modification. 2.0±0.05 g of finely ground fresh and dried *Sardinella madarensis* composite samples were weighed into separate 100 mL beakers. Each sample was extracted with 20 mL of dichloromethane (DCM) and stirred for 25 minutes. The extract was subjected to solid-phase cleanup using a chromatographic column packed with 1 cm of glass wool, 2 g of activated silica gel, and 0.5 g of anhydrous sodium sulfate. The purified extract was concentrated using a rotary evaporator and transferred into labeled glass vials. A 1 µL aliquot of the eluate was injected into an HP 5890 Series II gas chromatograph mass spectrometer(GC-MS) using a 30 m capillary column (0.32 mm inner diameter, 0.25 µm film thickness). The GC oven was programmed for a temperature gradient, starting at 50°C (held for 1 min), increasing at 10°C/min to 250°C, and finally held for 10 min. The injection port and detector temperatures were set at 275°C, with helium, the carrier gas, at a flowrate of 1.2 mL/min. Calibration was performed using a seven-point standard curve, prepared from a certified PAH reference standard. The PAHs were identified based on their retention times within the column. All analyses were conducted in triplicate, and the mean values for fresh and dried fish samples were reported.

Statistical Analysis

Results were expressed as mean ± standard deviations, and coefficients of variation (CV%) were used to measure the variability in PAH concentrations in both fresh and dried fish samples. Statistical analyses were performed using Microsoft Excel.

PAH analysis

The results in Table 1 show clear differences in polycyclic aromatic hydrocarbon (PAH) concentrations between fresh and dried *S. madarensis* fish samples. Dried fish had higher concentrations of 13 out of the 16 PAHS identified. This could be explained by the fact that drying removes water, thereby concentrating the remaining compounds. Okenyi *et al*. [23] reported high concentrations of PAHs in fish dried with firewood. Drying methods like smoking or sun-drying have been reported to introduce new PAHs or cause existing ones to accumulate due to exposure to heat and smoke[24]. Locally, the smoke from firewood and charcoal is usually augmented with polyethylene materials, which could significantly increase the amounts of PAHs absorbed into the food material [23]. The use of chemically treated wood and prolonged smoking times could also exacerbate the accumulation of PAHs in fish dried by smoking [25].

Results and Discussion

Three PAHS, Acenaphthylene (Acy), acenaphthene (Acn), and fluorene (Flu), followed a different trend because they were found in higher amounts in fresh fish, suggesting that they may have evaporated or broken down in the course of drying. This was also observed in a previous study by Alomirah *et al.* [26]. Fluorene, a volatile PAH present at 20.66 µg/kg in fresh fish, completely disappeared in dried fish due to heat-induced degradation [27]. Similarly, benzo(b)fluoranthene (BbF) levels reduced significantly from 6514.09µg/kg in fresh fish to 4063.65µg/kg in dried fish, perhaps due to environmental factors such as thermal and photodegradation caused by smoking, breaking down the PAH into derivative compounds, reducing its concentration.

Additionally, fresh fish retains more moisture, which provides a medium that stabilizes PAHs, while drying alters the matrix, affecting their retention. Environmental factors like surface deposition or removal during drying also play a role in the differential buildup of these compounds [28,26].

Table 1: Concentrations of PAHs in fresh and dried *S. madarensis* Fish Samples

PA H	SM Fresh (µg/kg)	SM Dried (µg/kg)	Mean SMF+SM D (µg/kg)	CV (%)
Naphthalene (Nap)	0.25 ± 0.003	39.52 ± 2.30	19.89	139.64
Acenaphthylene (Acy)	0.19 ± 0.002	ND	N/A	–
Acenaphthene (Acn)	0.38 ± 0.05	ND	N/A	–
Fluorene (Flu)	20.66 ± 1.12	ND	N/A	–
Phenanthrene (Phe)	38.53 ± 2.70	227.27 ± 14.40	132.90	100.42
Anthracene (Ant)	13.78 ±1.10	2806.33 ± 85.00	1410.06	140.04
Fluoranthene (Fln)	0.29 ± 0.01	3.11 ± 0.25	1.70	117.30
Pyrene (Pyr)	8.14 ± 0.65	246.44 ± 2.00	127.29	132.38
Benzo(a)anthracene (BaA)	8.95 ± 0.55	153.89 ± 12.50	81.42	125.88
Chrysene (Chr)	0.001 ± 0.0002	1.44 ± 0.10	0.72	141.23
Benzo(b)fluoranthene (BbF)	6514.09 ± 50.00	4063.65 ± 33.00	5288.87	32.76
Benzo(k)fluoranthene (BkF)	0.02 ± 0.003	853.75 ± 40.00	426.88	141.41
Benzo(a)pyrene (BaP)	0.03 ± 0.004	51.08 ± 4.20	25.56	141.26
Indeno(1,2,3-cd)pyrene (IcdP)	878.48 ± 12.00	1073.38 ± 33.00	975.93	14.12
Dibenz(a,h)anthracene (DahA)	2.22 ± 0.25	49.90 ± 3.70	26.06	129.37
Benzo(g,h,i)perylene (BghiP)	1.36 ±1.2	2.61 ± 0.20	1.98	44.53
PAH4 (Total)	6523.07	4269.06		

ND-Not detectable; N/A-not applicable

The highest concentration of PAH obtained was Benzo(b)fluoranthene (BbF) in both fresh (6514.09 µg/kg) and dried fish (4063.65 µg/kg), which suggests it may be a predominant PAH in the environment.

Statistical Analysis and Variability

The variations in PAH concentrations between fresh and dried composite fish samples were further emphasized by the coefficient of variation (CV). Compounds with the highest CV values of greater than ≥ 140% were Ant, BaP, BkF, and Chr. This reveals that their concentrations changed intensely after drying, indicating that drying had a drastic effect on their levels. On the contrary, indeno(1,2,3-cd)pyrene, IcdP, and BbF had a very low CV of 14.12 and 32.76%, respectively, indicating that their concentration remained fairly stable and consistent, most likely due to their high molecular weight and resistance to evaporation [29,30].

Health implications and Mitigation strategies

Given the well-established health concerns surrounding PAHs, regulatory agencies such as the European Food Safety Authority (EFSA) focus on four particularly dangerous PAHs collectively referred to as PAH4—benzo(a)pyrene, benzo(b)fluoranthene, chrysene, and benzo(a) anthracene [31,32,13]. The PAH4 levels in this study, which are the sum of the four aforementioned PAHs, were significantly higher than the European Union's safety threshold of 30 µg/kg for food products [31]. PAH4 levels in fresh fish were 6.514.09 µg/kg, and in dried fish, 4.154.27 µg/kg. These figures raise concern about a major threat to food safety, especially given that benzo(a)pyrene is classified as a Group 1 carcinogen by the International Agency for Research on Cancer [13]. BaP concentration in dried fish(51.08 µg/kg) far exceeds the EU maximum permissible level of 2 µg/kg for BaP in smoked fish, indicating a potential food safety risk and a need for improved processing methods [33]. These findings emphasize the possible health risks associated with PAH accumulation in dried fish, especially in communities where it is used as a dietary staple. Drying, by smoking, though necessary for preservation, increases exposure to harmful compounds and is not the safest option. This finding aligns with previous studies on PAHs in smoked fish from Senegal, reinforcing the impact of traditional drying methods[7]. According to Okenyi *et al*. [23], the oven drying method is safer; however, its availability or affordability to the average African fishmonger or homes is challenging. Adequate and proper monitoring and public health interventions are thus paramount to lessen the risks of PAHs contamination and protect communities reliant on local fish resources.

Conclusion

This study evaluated the occurrence and concentraion of polycyclic aromatic hydrocarbons in fresh and dried *Sardinella madarensis* collected from the Sombreiro River in the Niger Delta region of Nigeria. The results revealed higher amounts of 13 PAHs in dried fish compared to fresh fish. There was significant contamination of PAHs in the fresh and dried fish, with elevated levels of a known carcinogen, BaP, in dried fish (51.08 ± 4.20 µg/kg**),** suggesting a potential health risk to consumers. It was evident that, in addition to concentrating the already existing PAHs, drying introduced additional ones. These findings highlight the need for safer techniques in fish processing, stringent pollution control, frequent environmental monitoring and remediation, and public health initiatives to reduce contamination risks and safeguard communities that depend on local fish resources.

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