



Environmental and Processing Contaminants from Paks and Furan

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ABSTRACT

Maintaining an adequate, safe food supply has been a major goal of the Federal Government since 1906, when the first Federal food and drug law was signed into law. Historically, chemicals such as salt, sugar, and wood smoke have been used to preserve foods. Modern food technology relies extensively on the use of chemicals not only for preservation but also to produce appealing colors, flavors, aromas, and textures. Most developed countries now have food laws designed to permit the use of such chemicals in food under conditions judged to be safe. These chemicals are not considered adulterants or contaminants and are classed as intentional additives. Other chemicals may enter food as a result of their use in food production, handling, or processing. Such substances maybe legally permitted if they are unavoidable under good manufacturing practices and if the amounts involved are considered safe. These chemicals are classed as incidental additives. The presence of both these classes of chemicals in food is controlled by regulation.

INTRODUCTION

Environmental contaminants include substances from natural sources or from industry and agriculture. Many of the naturally occurring contaminants in food are of microbiological origin and consist of harmful bacterial, bacterial toxins, and fungal toxins. (Aflatoxin, a contaminant of peanuts and grains, is an example of a fungal toxin or mycotoxin.) The second category of environmental contaminants includes organic chemicals, metals and their complexes, and radionuclides. Only those environmental contaminants introduced into food as a result of human activities such as agriculture, mining, and industry are considered in this assessment.

The environmental contamination of food is a result of our modern, high-technology society. We produce and consume large volumes of a wide variety of substances, some of which are

toxic. It is estimated that 70,000 chemicals may currently be in commercial production in the United States and that 50 of these chemicals are manufactured in quantities greater than 1.3 billion lbs per year.

Seven percent of this country's gross national product (GNP), \$113 billion per year, is generated by the manufacture and distribution of chemicals. During the production, use, and disposal of these substances, there are opportunities for losses into the environment. For example, the Environmental Protection Agency (EPA) estimates that there are more than 30,000 chemical and radioactive waste disposal sites. Of these, 1,200 to 2,000 are considered threats to human health (Wang et al., 2013).

Environmental contamination of food takes two forms: long-term, low-level contamination resulting from gradual diffusion of persistent chemicals through the environment, and relatively shorter term, higher level contamination stemming from industrial accidents and waste disposal.

Chemical contamination is a major problem in the food industry all around the world. There are numerous potentially harmful compounds in the environment that could contaminate human food. They can be both inorganic and organic, and they can come from a variety of places. The environment could be a source of pollutants. Metals like lead and mercury, dioxins, and polychlorinated biphenyls are examples of this (PCBs). Pesticide use in agriculture has the potential to contaminate food. Similarly, medications used in humans and animals may contaminate streams and endanger consumers' health. Furthermore, food packaging practices could be a source of contamination, with so-called "migrants" seeping from the packaging materials. These pollutants have the potential to cause acute or chronic toxicity. Toxicity can be related to the route of exposure and dose, and personal attributes such as age and health status might influence a person's vulnerability.

Some food products may be more polluted than others according to the nature of contamination. This could be due to a variety of causes, including pesticide exposure, plant uptake mechanisms from the environment, or pollutants from food packaging (Stasinis et al., 2014; Price et al., 2014). An individual's exposure to these pollutants is influenced by their dietary habits. Nursing neonates, for example, consume a large amount of pollutants that are excreted in breast milk (Leibson et al., 2018). Different harmful effects may result from exposure at different periods of life. Prenatal exposure to persistent organic pollutants, for example, has been associated to a rise in childhood obesity and blood pressure (Vafeiadi et al., 2015).

In Nigeria and all over the world, millions of people daily consume street foods that provide a wide range of essential energy needs and nutrients. According to a survey by the World Health Organization (WHO) in 1996, 74% of countries reported that street foods contribute significantly to urban food supply, defined as "ready-to-eat foods," processed or fresh, which are sold at stationary locations or hawked in streets and open places as opposed to stores and licensed establishments (Mwangi et al., 2001). These foods are mostly cheap and they are also easily accessible (FAO, 2007). There is a wide acceptability of street-vended foods among majority of the Nigerian population which cuts across social, cultural, and political class. The

inexpensive and convenient nature of these hawked foods has experienced an economic boom in the last decade.

Although street food offers numerous advantages in improving food security, the safety of these foods should be properly monitored. Food safety is the assurance that food will not cause any harm or deleterious effect to the consumer when it is consumed (WHO, 1994). Street foods can pose a possible health risk to habitual consumers because food prepared and exposed to the open air for sale may become contaminated by pathogenic microorganism as well as chemical toxicant (Rane, 2011). Contamination of street food is as a result of so many factors such as preparation methods, poor packaging, vehicular exhaust emission, indiscriminate waste disposal, poor sanitation, industrial emission, and a list of all other pollution sources in the surrounding vending area (Omemu and Aderoju, 2008).

MATERIALS AND METHODS

Due to the nature of contamination, some food products may be more contaminated than others. This may be due to several factors such as varying exposure to pesticides, differences in plant uptake mechanisms from the environment, or contaminants from food packaging (Stasinou et al, 2014). Dietary make-up will affect an individual's exposure to these contaminants. For example, nursing neonates have a high intake of contaminants that are excreted in breast milk. Exposure at different life stages may result in different toxic effects as well. For example, prenatal exposure to persistent organic pollutants has been linked to an increase in childhood obesity and increased blood pressure.

For many food items including vegetables, fish, and other seafood human health risk assessment data is available after analysis of available foods (Mahmood and Malik, 2014). Urban farms and gardens may pose additional risks due to contaminants such as metals. Furthermore, drinking water may become contaminated.

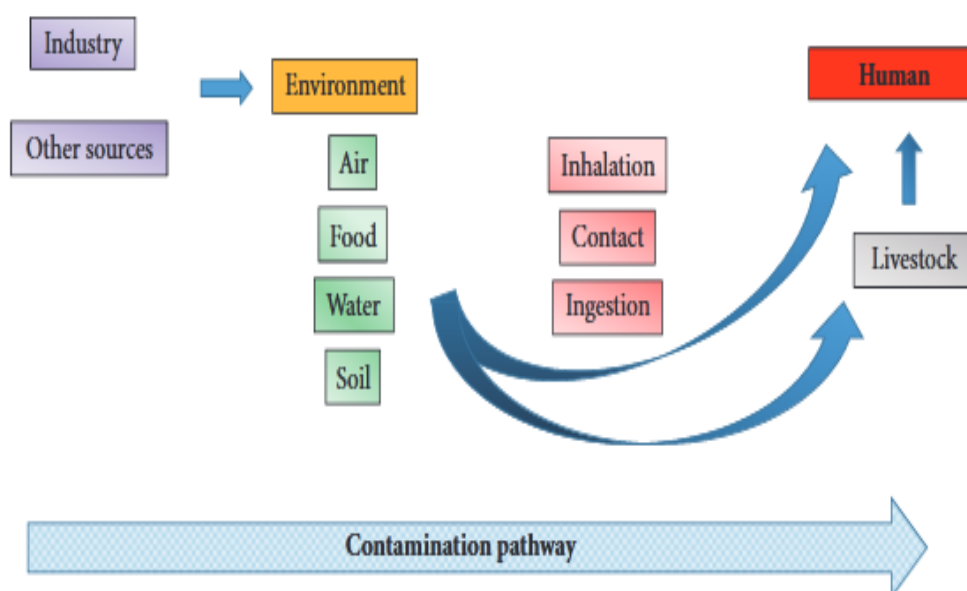


Figure 2.2: Source of Environmental Contaminants in Human Food (Mahmood and Malik, 2014).

Table 1: Example of Food Contamination with different chemicals around the world

Food contaminants	Food	Country
	Metal/metalloid	
Pb/Hg	Grain/Vegetable	China
Pb	Sheep livers	Spain
Pb, Cd, Cu, Zn	Agricultural crops	Romania
Cd	Locally produced foods	Belguim
Tl	Lettuce and chard	Germany
Pb, Cd	Soybeans	Argentina
Cu, Zn Mn, Fe	Fish	Turkey

Sources of Contaminants from the Environment to Food and Water (Smedley and. Kinniburgh, 2016)

Food contaminants	Food	Country
	Antimicrobials	
Antimicrobials	Pork meat	Madagascar
	eggs Table	Sudan
	Milk	Peru
	Beef	Nigeria
	Meat	Brazil

Sources of Contaminants from the Environment to Food and Water (Smedley and. Kinniburgh, 2016)

Likewise, in Spain, sheep near a mine were found to have lead contamination, with levels in 87.5% liver samples above European Union Maximum Residue Levels (MRL) (Pareja-Carrera et al., 2014). It is useful to consider the sources of contaminants in order to understand their pathway into food and water sources for consumption. Factors such as soil properties, activities by people, and point sources affect the accumulation of metals in the environment. For example, mining may result in release of substances such as arsenic and mercury [Xiao et al., 2017]. Once in the environment, these substances may contaminate food and water and result in human health hazards, with toxic effects varying depending on the contaminant(s) ingested (Table 1).

Industrial regions often have extensive environmental contamination by metals. In Romania, lead, cadmium, copper, and zinc contaminated crops, exceeding maximum acceptable levels in some samples. In China, cadmium from a zinc smelter contaminated leaf and root vegetables particularly. Arsenic, selenium, lead, and other metal and metalloid contaminants were found near a coking plant in China, contaminating soil and food, and detected in blood samples from children. In that case, ingestion of food was determined to be the major exposure pathway for local children. In Belgium, cadmium was detected in locally produced food items grown near nonferrous metal plants. Thallium from a steel plant in south China was found to contaminate soil and thence vegetables, exceeding German standards for the maximum permissible level and showing hyperaccumulation in plants such as leaf lettuce, chard, and pak choi.

Many fruits and vegetables have been shown to be contaminated by metals. For example, cadmium in soil was detected in navel oranges in China and lead and cadmium in soybeans in Argentina (Cheng, et al., 2015). Also in China, various metals were detected in edible seeds, with levels of copper sufficiently high to show an increased health risk to people consuming them (Chen et al., 2010). On the contrary, contamination levels of mercury in rice samples from a city in eastern China were below levels likely to affect human health. In the global arena, methylmercury has been detected in fish and other seafood around the world (Peng et al., 2016). Fish tissues from Turkey were shown to be contaminated with copper, iron, zinc, and manganese. Various metals have also been detected in fish from Sicily, with some concentrations exceeding European regulation limits. In Asia, food species of turtles have been shown to contain mercury. With regard to water, endemic arsenism from contaminated drinking water has been reported in China. Monitoring has detected nickel in drinking water in New South Wales, Australia, but levels do not appear to pose any health risk for the local population (Alam et al., 2008).

SAMPLES COLLECTION

14 different food samples, purchased from food eateries in Port Harcourt sub-urbs Rivers State. Two different product categories were represented, namely cereal/maize-based and meat/fish-based samples. Four meat/fish-based samples, specifically smoked fish (catfish grilled and croaker grilled) and smoked meat (beef grilled and chicken grilled) labelled A, B, Q, and R respectively. While the second paper will investigate mainly on Cereal/maize-based samples, microwaved rice packaged with plastic container at different time interval (10 mins and 30 mins), barbecue fish and chicken processed with and without aluminium foil. And roasted corn and meat wrapped with and without paper labelled sample (X and Y), (Sx, Tx, S, and T), (Cx, Dx, C and D) respectively. The samples were stored unopened at +4°C until the analyses that were performed in triplicate

Determination of Heavy Metal Residue

According to Pequerul et al., (2016) the heavy metals were identified. Briefly, the samples were labeled and blended into smaller sizes after being appropriately labeled. After being oven dried for 15 minutes at 600°C to remove moisture, the samples were allowed to cool overnight. After cooling, the ash residue was soaked in water and digested. The various ash residues were placed in labeled crucibles, which were then filled with 2.5 mL sulfuric acid. After heating for 30 minutes, the crucibles were allowed to cool. Following digestion, 10 mL of HNO₃ was added drop by drop until a clear solution was obtained. After that, the contents were heated for one hour and then cooled. The contents of the crucibles were then filtered into a 50 ml volumetric flask and distilled water was added to bring it up to the required concentration. After 15 minutes of heating, the contents were allowed to cool. The resulting filtrate was then fed into an Atomic Absorption Spectrometer (UNICAM SOLAAR 32) to determine the concentrations of the chosen metals.

RESULTS

Concentrations of PAHs components in traditionally smoked fish (catfish grilled and croaker grilled) sample A and B, and traditionally smoked meat (beef grilled and chicken grilled) sample Q and R in various Eatery in Port-Harcourt Sub-urb, Rivers State as shown in Table 1. A total of 11 (eleven) fraction of PAHs were found in the samples which include; Anthracene, Naphthalene, Fluoranthene, Fluorine, Phenanthrene, Chrysene, Pyrene, Benzo(a) anthracene, ,

Benzo(a) pyrene, Benzo(b) fluoranthene and Benzo(k) fluoranthene, with plot of the concentration shown in figure 1. The total concentration PAHs in the samples ranged from (25.198 - 82.428 $\mu\text{g/kg}$) in the following decreasing order: $Q > R > A > B$. The smoked meat were found to contain high concentration of total PAHs than smoked fish.

The amount of PAHs decreases from Anthracene 15.370 $\mu\text{g/kg}$ to Chrysene and Benzo(b) Fluoranthene (which shows no detection) 0.000 $\mu\text{g/kg}$ in Sample A. and the amount of PAHs decreases form Anthracene 15.200 $\mu\text{g/kg}$ to Fluorene, Phenanthrene, Chrysene, Benzo(a) pyrene, and Benzo(b) Fluoranthene (which shows no detection) 0.000 $\mu\text{g/kg}$ in Sample B for smoked fish.

While the amount of PAHs decreases from Anthracene 32.200 $\mu\text{g/kg}$ to Phenanthrene and Benzo(a) pyrene 0.030 $\mu\text{g/kg}$ in Sample Q and the amount of PAHs decreases from Anthracene 24.400 $\mu\text{g/kg}$ to Benzo(a) pyrene (which shows no detection) 0.000 $\mu\text{g/kg}$ in Sample R for smoked meat.

Furthermore, in smoked fish, Catfish grilled (Sample A). Anthracene (15.370 $\mu\text{g/kg}$) has the highest value among the 3-rings PAHs present while Fluorene (0.040 $\mu\text{g/kg}$) has the lowest value. Pyrene (7.000 $\mu\text{g/kg}$) is the highest for 4-rings PAHs present and Chrysene (which shows no detection) 0.000 $\mu\text{g/kg}$ the lowest concentration. Among the 5-rings PAHs, dibenzo(k) fluranthene (3.000 $\mu\text{g/kg}$) has the highest concentration while Benzo(b) fluoranthene (0.000 $\mu\text{g/kg}$) the lowest concentration. Also, for Croacker fish grilled (sample B) Anthracene (15.200 $\mu\text{g/kg}$) has the highest value among the 3-rings PAHs present while Fluorene and phenanthrene (which shows no detection / concentration) 0.000 $\mu\text{g/kg}$ has the lowest value. Pyrene (4.650 $\mu\text{g/kg}$) is the highest for 4-rings PAHs present and Chrysene and Benz(a) anthracene (which shows no detection) 0.000 $\mu\text{g/kg}$ the lowest concentration. Among the 5-rings PAHs, Benzo(k) fluranthene (2.450 $\mu\text{g/kg}$) has the highest concentration while Benzo(b) fluoranthene and Benzo(a) pyrene (which shows no detection / concentration) 0.000 $\mu\text{g/kg}$ has the lowest concentration.

While, in smoked meat, Beef grilled (Sample Q). Anthracene (32.200 $\mu\text{g/kg}$) has the highest value among the 3-rings PAHs present while phenanthrene (0.030 $\mu\text{g/kg}$) has the lowest value. Chryene (6.700 $\mu\text{g/kg}$) is the highest for 4-rings PAHs present and Benz(a) anthracene (0.080 $\mu\text{g/kg}$) the lowest concentration. Among the 5-rings PAHs, Benzo(b) fluranthene (3.100 $\mu\text{g/kg}$) has the highest concentration while Benzo(a) pyrene (0.030 $\mu\text{g/kg}$) has the lowest concentration. Also, for Chicken grilled (Sample R). Anthracene (24.400 $\mu\text{g/kg}$) has the highest value among the 3-rings PAHs present while Fluorene (0.040 $\mu\text{g/kg}$) has the lowest value. Pyrene (4.000 $\mu\text{g/kg}$) is the highest for 4-rings PAHs present and Benz(a) anthracene (0.060 $\mu\text{g/kg}$) the lowest concentration. Among the 5-rings PAHs, Benzo(k) fluranthene (1.400 $\mu\text{g/kg}$) has the highest concentration while Benzo(a) pyrene (which shows no detection / concentration) 0.000 $\mu\text{g/kg}$ has the lowest concentration.

Table 4.1: A plot showing Concentrations of PAHs components in traditionally smoked fish (catfish grilled and croacker grilled) sample A and B, and smoked meat (beef grilled and chicken grilled) sample Q and R.

PAH COMPONENTS	A	B	Q	R
Anthracene	15.370 ± 0.0190	15.200 ± 0.050	32.200 ± 0.100	24.400 ± 0.050
Naphthalene	5.050 ± 0.040	3.500 ± 0.050	15.400 ± 0.210	5.500 ± 0.010
Fluoranthene	1.100 ± 0.003	0.068 ± 0.001	1.400 ± 0.005	0.070 ± 0.004
Fluorene	0.040 ± 0.003	Nd	0.060 ± 0.002	0.040 ± 0.001
Phenanthrene	0.056 ± 0.001	Nd	0.030 ± 0.002	0.070 ± 0.002
Chrysene	Nd	nd	6.700 ± 0.004	3.500 ± 0.002
Pyrene	7.000 ± 0.01	4.650 ± 0.050	6.000 ± 0.020	4.000 ± 0.004
Benzo(a) anthracene	0.030 ± 0.002	nd	0.080 ± 0.003	0.060 ± 0.001
Benzo(a) pyrene	0.020 ± 0.001	nd	0.030 ± 0.002	nd
Benzo(b) Fluoranthene	Nd	nd	3.100 ± 0.002	1.000 ± 0.001
Benzo (K) Fluoranthene	3.000 ± 0.02	2.450 ± 0.04	2.850 ± 0.020	1.400 ± 0.05
Total PAHs	28.290	25.198	82.428	47.802

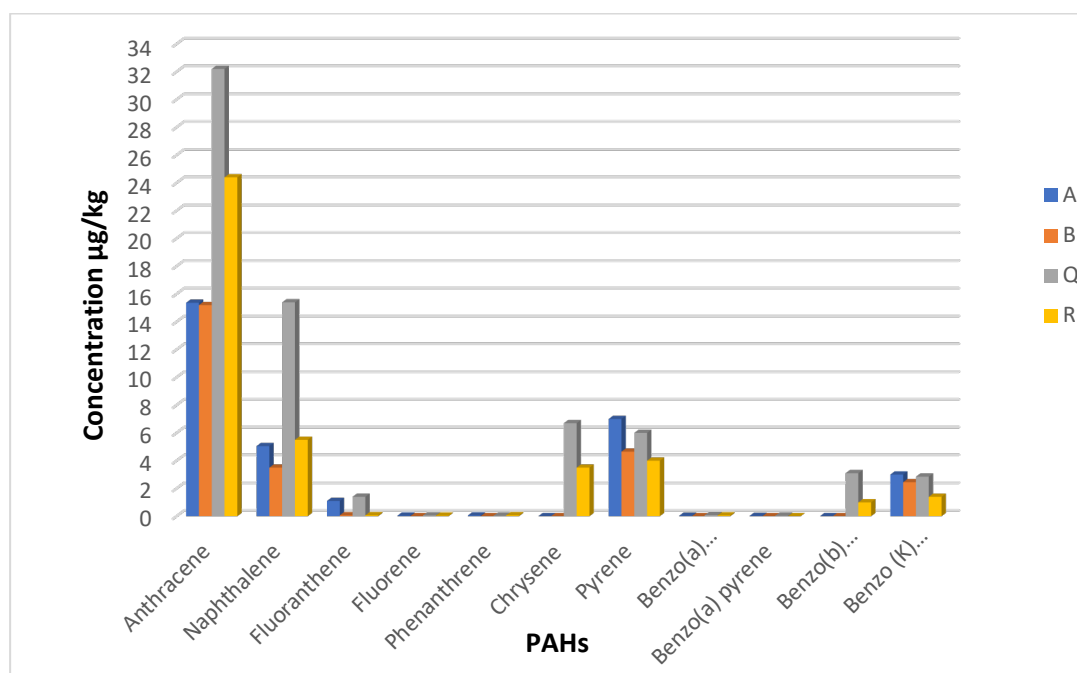


Figure 4.1: A plot showing Concentrations of PAHs components in traditionally smoked fish (catfish grilled and croacker grilled) sample A and B, and smoked meat (beef grilled and chicken grilled) sample Q and R.
Sources and Origin of PAHS in samples

PAHs come from two different sources, according to popular belief: Petrogenic (gasoline, kerosene, diesel, lubricating oil, and asphalt are examples of unburned petroleum and its products.) and Pyrolytic (organic matter incomplete combustion Combustion of fossil fuels, combustion of vehicles, smelting, waste incinerators, forest fires, and coal combustion) Several molecule ratios of polycyclic aromatic compounds, such as Phenanthrene/Anthracene (Phen/Anth), Fluoranthrene/Fluoranthrene + Pyrene (FI/FI + Py), and Fluoranthrene/Phenanthrene (FI/Phen), have been used to determine the source of PAHs. According to Table 4.2, the PAH ratio for Phen/Anth in this study is between 0.000932 to 0.0036. The value is much lower than 10 in all of the samples, including smoked fish and smoked meat, indicating a predominance of pyrogenic sources (Tolosa et al. 2003). The PAHs in this study are derived from petrogenic rather than pyrolytic sources, based on the Phen/Anth concentration ratios. The PAH isomer pair ratio Fluoranthene/ Fluoranthene + Pyrene (FI/FI + Py) has also been used as a separate chemical tracer to infer a possible PAH source in environmental samples (Yunker et al. 2002). According to Yunker et al. (2002), a PAH isomer ratio of less than 0.4 indicates petrogenic sources, 0.4-0.5 indicates pyrogenic sources, and more than 0.5 indicates coal, grass, and wood combustion. The FI/FI + Py ratio indicates that petrogenic sources play a role in the spread of these contaminants.

Table 4.2: Molecular PAHs indices ratio

PAHs Ratio	A	B	C	D
Phen/Anth	0.003643	0	0.000932	0.002869
Fl /Fl + Py	0.135802	0.014413	0.189189	0.017199
Fl / Phen	19.64286	0	46.66667	1

Phen = Phenanthrene; Anth = Anthracene; Fl = Fluoranthrene; Py = Pyrene

Identification of Furan and Furan Derivatives in Traditionally Smoked Fish (Catfish Grilled and Croacker Grilled) and Meat (Beef Grilled and Chicken Grilled)

The concentrations of the Furan found in sample A, B, Q AND R respectively. Total of 14 furan component were found in the samples which include; 2,3,7,8- Cl-substituted (2,3,7,8-TCDF, 2,3,7,8 TCDD, 1,2,3,7,8 PCDF, 2,3,4,7,8 PCDF, 1,2,3,7,8 PCDD, 1,2,3,4,7,8 HXCDF, 1,2,3,6,7,8-HXCDF, 1,2,3,4,7,8 HXCDD, 1,2,3,6,7,8 HXCDD, 1,2,3,7,8,9-HXCDD, 1,2,). Among the furan component 2,3,7,8-TCDD and 2,3,4,7,8-PCDF shows no detection in all samples. 2,3,7,8-TCDF and 1,2,3,7,8-PCDD were found in all. 1,2,3,7,8-PCDF, was found in all sample except for sample B. 1,2,3,4,7,8-HXCDF, was found only in sample Q. 1,2,3,6,7,8-HXCDF, was found in all sample except sample A. 1,2,3,4,7,8-HXCDD, was found only in sample Q. 1,2,3,6,7,8 HXCDD, was found only in sample A and B. 1,2,3,7,8,9-HXCDF, was found only in sample B and R. 1,2,3,7,8,9-HXCDF, was found only in sample Q and R. 1,2,3,4,6,7,8-HPCDF, was found only in sample B, 1,2,3,4,7,8 HPCDD, was found only in sample R. and 1,2,3,4,7,8,9 HPCDF, was found only in sample Q.

Total concentration of furan range from (14.91 to 37.81 µg/kg), with the concentration decreasing in the order A>B>R>Q. smoked fish (Catfish grilled and Croacker grilled) shows the highest total concentration of furan. And smoked meat (Beef grilled and Chicken grilled) shows the lowest concentration. Figure 4.2.

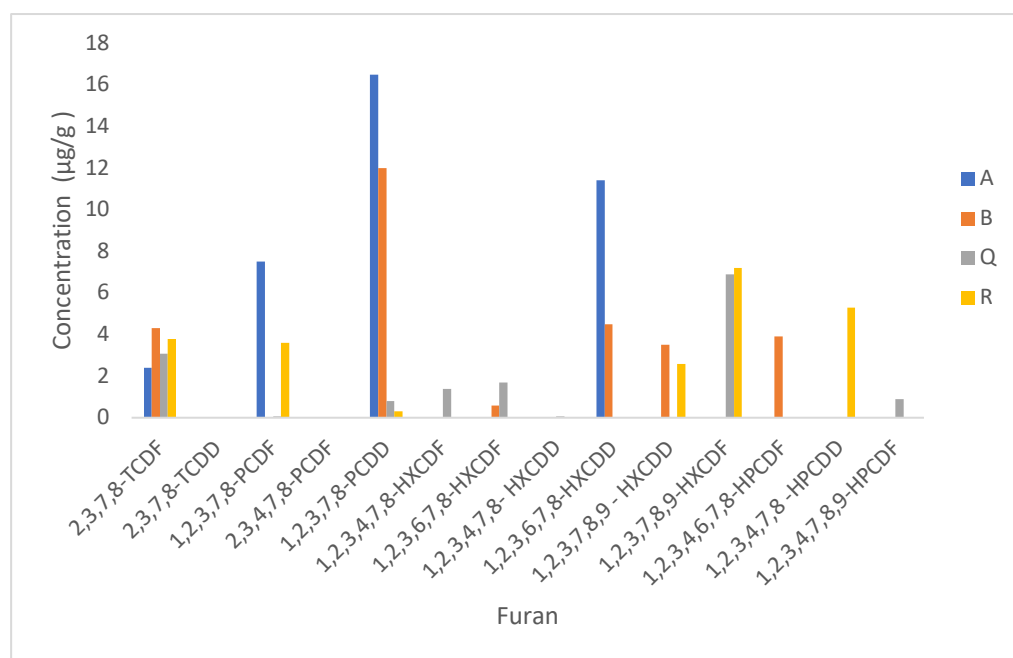


Figure 4.2: A plot showing concentration of all Detected Furan in in traditionally smoked fish (catfish grilled and croacker grilled) sample A and B, and traditionally smoked meat (beef grilled and chicken grilled) sample Q and R.

CONCLUSION

This study has shown that levels of PAHs in traditionally smoked fish as sold in Port Harcourt, Eatery sub-urbs, in Rivers state, exceeded the permissible limits which exceeds the 0.5 µg/kg maximum level for smoked fish established by the European Commission. Also, it seems evident from our results that the traditionally smoked fish and meat as presently consumed by the people of Port Harcourt may create high health risk and such there should be some level of restraints in their consumption by humans. It is generally accepted that the sources of PAHs are categorized in to two origins: Pyrolytic (incomplete combustion of organic matters combustion of fossil fuel, Vehicular engine combustion, smelting, waste incinerators, forest fire and coal combustion) and Petrogenic (unburned petroleum and its product gasoline, kerosene, diesel, lubricating oil and asphalt). The PAH ratio shows that Phen/Anth in this study is in the range 0.000932 - 0.0036 The value is much lower than 10 in all the samples i.e. in traditionally smoked fish and meat which is generally considered indicative of a predominance pyrogenic sources (Tolosa el al. 2003). The Phen/Anth concentration ratios indicate that the PAHs in this study are derived from petrogenic rather than pyrolytic sources. PAH isomer pair ratio Fluoranthene/ Fluoranthene + Pyrene (FI/FI + Py) has also been used as distinct chemical tracer to infer possible source of PAHs in environmental sample (Yunker el ta 2002). Yunker et al. (2002) stated that the PAH isomer ratio measurement FI/FI + Py of less than 0.4 implies petrogenic, 0.4-0.5 suggests pyrogenic sources and more than 0.5 imply combustion of coal, grass and wood. The FI/FI + Py ratio shows that petrogenic sources contribute to the distribution of these contaminants.

Furan in traditionally smoked fish (catfish grilled and croacker grilled) and meat (beef grilled and chicken grilled) Total concentration of furan range from (14.91 - 37.81 µg/kg), with the concentration decreasing in the order A>B>R>Q. smoked fish (Catfish grilled and Croacker

grilled) shows the highest total concentration of furan. And smoked meat (Beef grilled and Chicken grilled) shows the lowest concentration.

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