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Determination of Level of Polycyclic Aromatic Hydrocarbons (PAHs) in Varieties of Rice (*Oryza Sativa*) from Mashayan Bululu and Wuchakal Agricultural Locations Bade and Karasuwa Local Government in Yobe State, Nigeria

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***Abstract**: The present study determined the level of twoteen polycyclic aromatic hydrocarbons (PAHs) in soil and rice (*Oryza sativa*) samples from two agricultural locations in Mashayan Bululu and Wachakal Local Government Areas of Yobe State, Nigeria. Soil was collected at three depths (0–5 cm, 5–10 cm, and 10–15 cm). Two varieties of rice (FARO 42 and 52) were collected for this study. A depth of 0–5 cm in Mashayan Bululu recorded the highest total PAH concentration (2.53–03 mg/kg), while a soil depth of 10–15 cm in Wachakal recorded the lowest load of 3.81–06 mg/kg. The diagnostic ratio in soil was highest in Mashayan Bululu with a value of 6.79E+00, while the lowest was recorded in Wachakal with a value of 3.89E-01. The highest mean effect range medium quotient (M-ERM-Q) was recorded in Mashayan Bululu with a value of 2.15E-06 mg/kg, while the lowest was recorded in Mashayan Bululu with a value of 3.57E-09 mg/kg. The highest total BaPTEQ was recorded in Wachakal with a value of 3.11E-04 mg/kg, while the lowest was recorded in Mashayan Bululu with a value of 9.99E-08 mg/kg. In the varieties of rice detected, the highest load of PAH concentration (1.84E-09 mg/kg) was detected in FARO 52, while the lowest PAH concentration (2.40E-10 mg/kg) was detected in FARO 44. Carcinogenic risk assessment of PAHs in rice from the two agricultural locations revealed that benz(a)pyrene and dibenz(a,h)anthracene were the highest accumulated PAHs with values of 3.56E-14 mg/kg and 2.47E-14 mg/kg, respectively. The average daily dose (ADD) of PAHs in the different varieties of rice from the two agricultural locations shows that FARO 52 had the highest ADD (1.80E-14 mg/kg), while FARO 44 showed the lowest ADD (2.35E-15 mg/kg). The potential for non-carcinogenic PAHs in this study revealed that FARO 52 has the highest Hazard Index (5.45E-15 mg/kg). Results from the present study show that the rice is safe for human consumption and should be monitored regularly in order to reduce the levels of PAHs.*

***Keywords**: Polycyclic, Aromatic, Hydrocarbon, Pollution, anthropogenic, carcinogenic.*

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are widespread across the globe, mainly due to long-term anthropogenic sources of pollution. The inherent properties of PAHs, such as heterocyclic aromatic ring structures, hydrophobicity, and thermostability, have made them recalcitrant and highly persistent in the environment. PAH pollutants have been determined to be highly toxic, mutagenic, carcinogenic, teratogenic, and immunotoxic to various life forms (Patel *et al.,* 2020). Polycyclic aromatic hydrocarbons (PAHs) occur naturally in coal, crude oil, and gasoline. They result from burning coal, oil, gas, wood, garbage, and tobacco. PAHs can bind to or form small particles in the air. High heat when cooking meat and other foods will form PAHs. Naphthalene is a man-made PAH used in the United States to make other chemicals and mothballs. Cigarette smoke contains many PAHs (CDC, 2009). People are usually exposed to mixtures of PAHs. Breathing air contaminated with motor vehicle exhaust, cigarette smoke, wood smoke, or fumes from asphalt roads are common ways exposure occurs. People take in PAHs when they eat grilled or charred meats or foods on which PAH particles have settled from the air. After PAHs are swallowed, breathed in, or, in some cases, passed through the skin, the body converts PAHs into breakdown products called metabolites that pass out of the body in the urine and feces (CDC, 2009). Human health effects from environmental exposure to low levels of PAHs are unknown. Large amounts of naphthalene in the air can irritate the eyes and breathing passages. Workers who have been exposed to large amounts of naphthalene from skin contact with the liquid form and from breathing naphthalene vapor have developed blood and liver abnormalities. Several of the PAHs and some specific mixtures of PAHs are considered to be cancer-causing chemicals (CDC, 2009).

Mashayan Bululu and Wachakal are located in Bade and Karasuwa Local Governments in Yobe State, respectively. Intense rice activities take place in the area, which is 10 km along the Lake Chad Basin area. The people in the areas are 98% predominantly farmers; out of this, merely 85% are crop-cultivating farmers. The rice production farmers in the study areas use herbicides to control weeds in an effort to reduce or eliminate yield losses and preserve high production quality. Lack of knowledge of the uses and effects of these herbicides among small- and large-scale farmers has resulted in contamination of the crops and soil with herbicides. Such contamination might be absorbed by the rice, while others leached into the underground water. Such accumulation by rice may affect humans and other species that depend on it as food.

The wetlands in the Komadugu-Yobe basin in Yobe State, Nigeria, are located within the catchment area of the Hadejia-Jama'are - Yobe river basin. The wetlands in these areas are associated with the floodplains surrounding the river. The river level rises during the rainy season (June–September), allowing the wetlands to store excess water, which is gradually transferred to the groundwater reserves. Socio-economic activities in Yobe State include small- and large-scale agriculture, grazing, fishing, and other activities that are directly or indirectly dependent on the river system. The river is the main source of fish, rice, and vegetables for the state and neighboring states within and outside the country. However, domestic usage and agricultural activities are carried out without due regard to the chemistry of the water. A series of problems may arise as a result of chemical contamination from industries or agricultural activities, such as the use or misuse of nitrate, phosphate, and sulfate fertilizers, or from natural sources.

These chemical contaminants may have advanced effects on the users. Some of which may be immediate, while others are accumulative and can cause a series of waterborne diseases that may even lead to death. Knowledge of water chemistry will therefore provide the basic framework within which any subsequent quality changes resulting from pollution can be contrasted, evaluated, and adequately addressed.

Mashayan Bululu and Wachakal are primarily agricultural areas with intense herbicide usage. Herbicides are extensively used in the area to enhance the production of rice, vegetables, cereals, and fruits, as well as to eliminate weeds. Rice from Mashayan Bululu and Wachakal agricultural areas also constitutes an important source of carbohydrate for the inhabitants in and around Yobe State and is a major source of income for the inhabitants. However, the agricultural activities have impacted negatively on the soil because bioaccumulation and bioconcentration of the herbicides in the rice are capable of reaching toxic levels even at low exposure. No studies have been carried out in Mashayan Bululu and Wachakal agricultural areas in spite of the fact that herbicides of high toxicity are used to eliminate weeds on rice farms. Mashayan Bululu and Wachakal agricultural areas had a long history of large-scale production of rice for human consumption as well as the use of herbicides. This calls for the need for epidemiological data concerning the risk to farmers and consumers of rice in this agricultural area.

Materials and Method

Sample Collection

Rice samples were collected in accordance with the method adopted by Akan *et al.* (2015). Rice samples (Faro 44 and 52) were collected from Mashayan Bululu and Wachakal in Mashayan Bululu and Wachakal from Bade and Karasuwa Local Government Areas, respectively, in Yobe State, Nigeria. The different varieties of rice samples were transported to the laboratory and stored at 25 ^{o}C. Two soil samples were collected at each sample point and were done at three different depths (0–5 cm, 5–10 cm, and 10-15 cm) by using a spiral auger of 2.5 cm diameter. The soil was randomly sampled and bulked together to form a composite sample. The soil samples were placed in clean plastic bags and transported to the Department of Pure and Applied Chemistry Laboratory, University of Maiduguri. Samples were collected for a period of four months.

Extraction of PAHs in Soil Samples

Analysis for PAHs in soil samples was carried out in accordance with the USEPA's (2000) 8270 analytical method. 10g of the sample was dried using anhydrous sodium sulfate, and 1 ml of 60 µg/ml O-Terphenyl surrogate standard was added and thoroughly mixed with the sample. 30 ml of methylene chloride was then added, and the sample was extracted. The sample extract was subsequently filtered through glass wool containing anhydrous sodium sulfate in a glass funnel. About 2 grams of silica gel were added and allowed to stand for a while. The extract was then decanted and allowed to concentrate at room temperature to a volume of 1 mL

Extraction of PAHs in Rice Samples

Analysis for PAHs in rice samples was carried out in accordance with the USEPA (2000) 8082 analytical method. Ten grams (10g) of the sample were dried using anhydrous sodium sulfate; 30 ml of methylene chloride was then added, and the sample was extracted. The sample extract was subsequently filtered through glass wool containing anhydrous sodium sulfate in a glass funnel and allowed to concentrate at room temperature to a volume of 1 mL

INSTRUMENTAL ANALYSIS OF PAHS USING GCMS FOR SOIL AND RICE SAMPLES

The extract was thereafter analyzed using an Agilent 7890A GC/MS previously calibrated with PAHs standards. The equipment turned out the concentration of the PAHs as the sample details were supplied for soil and cereal samples.

RISK ASSESSMENT OF PAHs

Identification of PAH sources in Soil Samples

Diagnostic ratios were used to distinguish the possible sources of PAH in the soil. The following ratios were used as source indicators: Ant/Ant+Phe, BaA/BaA+Chr, and LMW-PAH to HMW-PAH.

Carcinogenic Risk Assessment of PAHs in Soil Samples

The health risk associated with the PAHs in soils was evaluated using the toxicity equivalency factor (TEF) method described by Nisbet and Lagoy (1992) (the TEF for each PAH was an estimate of the relative toxicity of the PAH compounds compared to BaP). The total equivalent concentration was expressed as BaP equivalent (BaPeq).

BaPeq for individual PAH were estimated using the equation

$$BaPeq \quad = \quad \sum Cn \;\times\; TEFn$$

Ecological risk assessment of PAHs in Soil Samples

The mean ERM quotient approach was used to evaluate the possible ecotoxicity of PAHs in the soil. The mean ERM quotient values were calculated according to the method formular suggested by Long and MacDonald (1998):

$$m - ERM - q = \sum\left(\frac{Ci}{ERMi}\right)/n$$

Non-cancer hazard, Carcinogenic Risk Calculation for Cereal Samples

The risk associated with dietary exposure to non-carcinogenic PAHs was evaluated using the hazard quotients approach. Hazard quotients represent a ratio of the exposure dose for each PAH divided by an oral chronic reference dose (RfD).

Hazard quotient (HQ) = Average daily dose (ADD)/RfD

The total risk due to exposure to a mixture of carcinogenic PAHs is the product of the dietary carcinogen exposure dose (mg kg^{-1} BW d^{-1}) and benzo(a)pyrene's slope factor value.

Risk (carcinogenic) = Average daily dose x slope factor

DATA HANDLING

Data collected were subjected to one-way analysis of variance (ANOVA) to assess whether PAHs varied significantly between soil and rice samples. Probabilities less than 0.05 ($p<0.05$) were considered statistically significant. All statistical calculations were performed with SPSS for windows.

Results

Table 1: Mean Concentrations of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Soil Samples from Mashayan Bululu Agricultural Location, in Bade Local government Area, Yobe State, Nigeria

			Concentrations (mg/kg)		
PAHS	**No. of Rings**	**MACs**	**0-5cm**	**5-10cm**	**10-15cm**
Naphthalene	2	1	1.00E-07	1.32E-08	3.32E-09
2-Methyl Naphthalene	2	1	5.82E-05	4.23E-06	2.34 E-07
Acenapthylene	3	3	6.69E-05	3.23E-07	3.22E-08
Acenaphthene	3	3	6.99E-05	2.22E-07	4.22E-09
Fluorene	3	3	2.03E-05	2.71E-06	3.23E-07
Phenanthrene	3	3	7.01E-06	4.21E-08	4.33E-09
Anthracene	3	3	1.53E-03	2.44E-05	2.34E-06
Fluoranthene	4	3	1.82E-05	2.98E-06	3.23E-07
Pyrene	4	3	3.93E-06	4.22E-07	3.23E-08
Benz(a)Anthracene	4	0.15	2.51E-04	3.19E-06	4.21E-08
Chrysene	4	-	5.21E-05	4.19E- 07	2.37E-08
Benz(b)Fluoranthene	5	0.3	2.87E-04	2.33E-05	3.33E-07
Benz(k)Fluoranthene	5	-	2.15E-05	4.22E-07	3.22E-09
Benz(a)Pyrene	5	0.3	4.98E-06	3.22E-08	3.44E-09
Dibenz(a,h)Anthracene	5	0.3	1.99E-05	2.74E-06	3.22E-08
Indinol(1,2,3-cd)Pyrene	6	-	1.23E-04	2.73E-05	3.11E-07
Totals			**2.53E-03**	**9.27E-05**	**3.81E-06**

MACs = Maximum Allowable Concentration (ATSDR, 2006)

Table 2: Mean Concentrations of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Soil Samples from Wachakal Agricultural Location, Karasuwa Local Government Area, Yobe State, Nigeria

			Concentrations (mg/kg)		
PAHS	**No. of Rings**	**MACs**	**0-5cm**	**5-10cm**	**10-15cm**
Naphthalene	2	1	1.00E-06	1.00E-06	1.00E-06
2-Methyl Naphthalene	2	1	1.00E-06	1.00E-07	1.00E-07
Acenapthylene	3	3	1.47E-05	1.22E-05	1.02E-05
Acenaphthene	3	3	2.04E-05	2.11E-06	1.03E-06
Fluorene	3	3	1.61E-05	1.11E-05	1.03E-05
Phenanthrene	3	3	3.17E-05	2.44E-05	2.21E-05
Anthracene	3	3	1.85E-05	1.65E-05	1.22E-05
Fluoranthene	4	3	6.90E-06	5.44E-06	4.34E-06
Pyrene	4	3	1.61E-05	2.34E-05	2.21E-05
Benz(a)Anthracene	4	0.15	9.12E-05	4.34E-05	3.33E-05
Chrysene	4	-	1.40E-04	2.33E-05	2.11E-06
Benz(b)Fluoranthene	5	0.3	3.36E-05	2.74E-05	1.11E-05
Benz(k)Fluoranthene	5	-	7.70E-04	2.99E-04	2.22E-05
Benz(a)Pyrene	5	0.3	1.92E-04	1.34E-04	1.11E-04
Dibenz(a,h)Anthracene	5	0.3	2.71E-05	2.43E- 06	2.33E-06
Indinol(1,2,3-cd)Pyrene	6	-	1.08E-05	2.22E-06	1.07E-06
Totals			**1.39E-03**	**6.26E-04**	**2.66E-04**

MACs = Maximum Allowable Concentration (ATSDR, 2006)

Table 3: Mean Concentrations of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Rice Samples from Mashayan Bululu Agricultural Location, Bade Local Government Area, Yobe State, Nigeria

	Concentrations (mg/kg)			
PAHS	**No. of Rings**	**MACs**	**FARO 44**	**FARO 52**
Naphthalene	2	1	1.00E-11	2.12E-09
2-Methyl Naphthalene	2	1	1.03E-09	2.43E-10
Acenapthylene	3	3	2.22E-10	1.00E-10
Acenaphthene	3	3	1.03E-11	2.22E-11
Fluorene	3	3	2.22E-12	4.20E-10
Phenanthrene	3	3	1.00E-11	5.22E-10
Anthracene	3	3	2.22E-10	2.83E-11
Fluoranthene	4	3	2.22E-11	5.33E-10
Pyrene	4	3	1.00E-10	3.44E-11
Benz(a)Anthracene	4	0.15	2.00E-11	9.32E-09
Chrysene	4	-	1.00E-11	5.11E-10
Benz(b)Fluoranthene	5	0.3	2.03E-13	2.34E-11
Benz(k)Fluoranthene	5	-	1.00E- 11	4.22E-10
Benz(a)Pyrene	5	0.3	2.04E-10	5.11E-10
Dibenz(a,h)Anthracene	5	0.3	1.23E-10	3.22E-10
Indinol(1,2,3-cd)Pyrene	6	-	2.11E-10	2.11E-09
Totals			**2.20E-09**	**1.72E-08**

MACs = Maximum Allowable Concentration (ATSDR, 2006)

Table 4: Mean Concentrations of Some Polycyclic Aromatic Hydrocarbon (PAHs) in Rice Samples from Wachakal Agricultural Location, Wachakal Local Government Area, Yobe State, Nigeria

	Concentrations (mg/kg)			
PAHS	**1. No. of Rings**	**MACs**	**FARO 44**	**FARO 52**
Naphthalene	2	1	1.23E-10	4.09E-10
2-Methyl Naphthalene	2	1	1.03E-12	5.33E-09
Acenapthylene	3	3	1.00E-10	5.43E-10
Acenaphthene	3	3	1.43E-10	3.45E-11
Fluorene	3	3	2.33E-11	5.33E-10
Phenanthrene	3	3	3.21E-10	4.33E-09
Anthracene	3	3	3.22E-10	6.04E-11
Fluoranthene	4	3	4.22E-10	4.34E-10
Pyrene	4	3	4.44E-10	3.23E-09
Benz(a)Anthracene	4	0.15	4.22E-11	3.23E-11
Chrysene	4	-	3.23E-10	3.34E-10
Benz(b)Fluoranthene	5	0.3	5.32E-11	4.54E-10
Benz(k)Fluoranthene	5	-	1.00E-09	9.32E-09
Benz(a)Pyrene	5	0.3	1.00E-10	4.98E-10
Dibenz(a,h)Anthracene	5	0.3	2.33E-11	3.45E-10
Indinol(1,2,3-cd)Pyrene	6	-	2.22E-10	8.98E-11
Totals			**2.97E-09**	**2.50E-08**

MACs = Maximum Allowable Concentration (ATSDR, 2006)

Table 5: Diagnostic Ratio of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Soil Samples of Different Depths from Mashayan Bululu and Wachakal Agricultural Locations

	Mashayan Bululu				Wachakal			
Soil Depths	BaA/BaA+Chr	Ant/Ant+Phe	Fluo/Fluo+Pyr	LMW/HMW	Soil Depths	BaA/BaA+Chr	Ant/Ant+Phe	Fluo/Fluo+Pyr
0 – 5cm	8.28E-01	9.95E-01	8.24E-01	2.24E+00	0 – 5cm	3.94E-01	3.69E-01	0.30E-01
5 – 10cm	8.84E-01	9.98E-01	8.76E-01	5.94E-01	5 – 10cm	6.51E-01	4.03E-01	1.89E-01
10 – 15cm	6.40E-01	9.98E-01	9.10E-01	3.96E+00	10 – 15cm	69.40E-01	3.65E-01	1.64E-01
MR	**2.35E+00**	**2.99E+00**	**2.61E+00**	**6.79E+00**	**MR**	**1.99E+00**	**1.13E+00**	**3.89E-01**

BaA = Benz(a)anthracene; Chr = Chrysene; Ant = Anthracene; Phe = Phenanthrene; Fluo = Fluoranthene; pyr = Pyrene;
LMW = Lower Molecular Weight; HMW = Higher Molecular Weight

Table 6: Mean Effect Range Medium (ERM) Quotient of Some Polycyclic Aromatic Hydrocarbon in Soil Samples from Mashayan Bululu and Wachakal Agricultural Locations

		Concentrations (mg/kg)					
		Mashayan Bululu			Wachakal		
PAHS	**ERM SSG**	**0-5cm**	**5-10cm**	**10-15cm**	**0-5cm**	**5-10cm**	**10-15cm**
Naphthalene	2100	4.76E-11	6.29E-12	1.58E-12	4.76E-10	4.76E-10	4.76E-10
2-Methyl Naphthalene	670	8.60E-08	6.00E-09	3.49E-10	1.00E-09	1.49E-10	1.49E-10
Acenapthylene	640	1.04E-07	5.05E-10	5.03E-11	2.20E-08	1.90E-08	1.50E-08
Acenaphthene	500	1.39E-07	4.44E-10	8.44E-12	4.00E-08	4.00E-09	2.00E-09
Fluorene	540	3.70E-08	5.00E-09	5.98E-10	2.90E-08	2.00E-08	1.90E-08
Phenanthrene	1500	4.00E-09	2.81E-11	2.89E-12	2.10E-08	1.60E-08	1.40E-08
Anthracene	1100	1.39E-06	2.20E-08	2.00E-09	1.60E-08	1.50E-08	1.10E-08
Fluoranthene	5100	3.00E-09	5.84E-10	6.33E-11	1.00E-09	1.00E-09	8.51E-10
Pyrene	2600	1.00E-09	1.62E-10	1.24E-11	6.00E-09	9.00E-09	8.00E-09
Benz(a)Anthracene	1600	1.56E-07	1.00E-09	2.63E-11	5.70E-08	2.70E-08	2.00E-08
Chrysene	2800	1.80E-08	1.50E-10	8.46E-12	5.00E-08	8.00E-09	7.54E-10
Benz(b)Fluoranthene	-	-	-	-	-	-	-
Benz(k)Fluoranthene	-	-	-	-	-	-	-
Benz(a)Pyrene	1600	3.00E-09	2.01E-11	2.15E-12	1.20E-07	8.30E-08	6.90E-08
Dibenz(a,h)Anthracene	260	7.60E-08	1.00E-08	1.24E-10	1.04E-07	9.00E-09	8.00E-09
Indinol(1,2,3-cd) Pyrene	950	1.29E-07	2.80E-08	3.27E-10	1.10E-08	2.00E-09	1.00E-09
M-ERM-Q		**2.15E-06**	**7.39E-08**	**3.57E-09**	**4.78E-07**	**2.14E-07**	**1.69E-07**

Table 7: Benz(a)Pyrene Equivalent Concentrations of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Soil Samples from Mashayan Bululu Agricultural Location

		Concentrations (mg/kg)		
PAHS	**USEPA TEF**	**0-5cm**	**5-10cm**	**10-15cm**
Naphthalene	0.001	1.00E-10	1.32E-11	3.32E-12
2-Methyl Naphthalene	0.001	5.80E-08	4.00E-09	2.34E-10
Acenapthylene	0.001	6.60E-08	3.23E-10	3.22E-11
Acenaphthene	0.001	6.90E-08	2.22E-10	4.22E-12
Fluorene	0.001	2.00E-08	2.00E-09	3.23E-10
Phenanthrene	0.001	7.00E-09	4.21E-11	4.33E-12
Anthracene	0.01	1.53E-05	2.44E-07	2.30E-08
Fluoranthene	0.001	1.80E-08	2.00E-09	3.23E-10
Pyrene	0.001	3.00E-09	4.22E-10	3.23E-11
Benz(a)Anthracene	0.1	2.15E-05	3.19E-07	4.00E-09
Chrysene	0.001	5.20E-08	4.19E-10	2.37E-11
Benz(b)Fluoranthene	0.1	2.87E-05	2.33E-06	3.30E-08
Benz(k)Fluoranthene	0.1	2.15E-06	4.20E-08	3.22E-10
Benz(a)Pyrene	1	4.98E-06	3.22E-08	3.44E-09
Dibenz(a,h)Anthracene	1	1.99E-05	2.74E-06	3.22E-08
Indinol(1,2,3-cd)Pyrene	0.01	1.23E-06	2.73E-07	3.00E-09
Σ BaPTEQ		**9.41E-05**	**5.99E-06**	**9.99E-08**

Table 8: Benz(a)Pyrene Equivalent Concentrations of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Soil Samples from Wachakal Agricultural Location

		Concentrations (mg/kg)		
PAHS	**USEPA TEF**	**0-5cm**	**5-10cm**	**10-15cm**
Naphthalene	0.001	1.00E-09	1.00E-09	1.00E-09
2-Methyl Naphthalene	0.001	1.00E-09	1.00E-10	1.00E-10
Acenapthylene	0.001	1.40E-08	1.20E-08	1.00E-08
Acenaphthene	0.001	2.00E-08	2.00E-09	1.00E-09
Fluorene	0.001	1.60E-08	1.10E-08	1.00E-08
Phenanthrene	0.001	3.10E-08	2.40E-08	2.20E-08
Anthracene	0.01	1.85E-07	1.65E-07	1.22E-07
Fluoranthene	0.001	6.00E-09	5.00E-09	4.00E-09
Pyrene	0.001	1.60E-08	2.30E-08	2.20E-08
Benz(a)Anthracene	0.1	9.12E-06	4.34E-06	3.33E-06
Chrysene	0.001	1.40E-07	2.30E-08	2.00E-09
Benz(b)Fluoranthene	0.1	3.36E-06	2.74E-06	1.11E-06
Benz(k)Fluoranthene	0.1	7.70E-05	2.99E-05	2.22E-06
Benz(a)Pyrene	1	1.94E-04	1.34E-04	1.11E-04
Dibenz(a,h)Anthracene	1	2.71E-05	2.43E-06	2.33E-06
Indinol(1,2,3-cd)Pyrene	0.01	1.10E-07	2.20E-08	1.00E-08
Σ BaPTEQ		**3.11E-04**	**1.74E-04**	**1.20E-04**

Table 9: Benz(a)Pyrene Equivalent Concentration of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Rice Samples from Mashayan Bululu Agricultural Location

	Concentrations (mg/kg)		
PAHS	**USEPA TEF**	**FARO 44**	**FARO 52**
Naphthalene	0.001	1.00E-14	2.12E-12
2-Methyl Naphthalene	0.001	1.03E-12	2.43E-13
Acenapthylene	0.001	2.22E-13	1.00E-13
Acenaphthene	0.001	1.03E-14	2.22E-14
Fluorene	0.001	2.22E-15	4.20E-13
Phenanthrene	0.001	1.00E-14	5.22E-13
Anthracene	0.01	2.22E-12	2.83E-13
Fluoranthene	0.001	2.22E-14	5.33E-13
Pyrene	0.001	1.00E-13	3.44E-14
Benz(a)Anthracene	0.1	2.00E-12	9.32E-10
Chrysene	0.001	1.00E-14	5.11E-13
Benz(b)Fluoranthene	0.1	2.03E-14	2.34E-12
Benz(k)Fluoranthene	0.1	1.00E-12	4.22E-11
Benz(a)Pyrene	1	2.04E-10	5.11E-10
Dibenz(a,h)Anthracene	1	1.23E-10	3.22E-10
Indinol(1,2,3-cd)Pyrene	0.01	2.11E-12	2.11E-11
ΣBaPTEQ		**3.36E-10**	**1.84E-09**

Table 10: Benz(a)Pyrene Equivalent Concentrations of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Rice Samples from Wachakal Agricultural Location

	Concentrations (mg/kg)		
PAHS	**USEPA TEF**	**FARO 44**	**FARO 52**
Naphthalene	0.001	1.23E-13	4.09E-13
2-Methyl Naphthalene	0.001	1.03E-15	5.33E-12
Acenapthylene	0.001	1.00E-13	5.43E-13
Acenaphthene	0.001	1.43E-13	3.45E-14
Fluorene	0.001	2.33E-14	5.33E-13
Phenanthrene	0.001	3.21E-13	4.33E-12
Anthracene	0.01	3.22E-12	6.04E-13
Fluoranthene	0.001	4.22E-13	4.34E-13
Pyrene	0.001	4.44E-13	3.23E-12
Benz(a)Anthracene	0.1	4.22E-12	3.23E-12
Chrysene	0.001	3.23E-13	3.34E-14
Benz(b)Fluoranthene	0.1	5.32E-12	4.54E-11
Benz(k)Fluoranthene	0.1	1.00E-10	9.32E-10
Benz(a)Pyrene	1	1.00E-10	4.98E-10
Dibenz(a,h)Anthracene	1	2.33E-11	3.45E-10
Indinol(1,2,3-cd)Pyrene	0.01	2.22E-12	8.98E-13
ΣBaPTEQ		**2.40E-10**	**1.84E-09**

Table 11: Average Daily Dose (mg/kg day^{-1}) of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Rice Samples from Mashayan Bululu and Wachakal Agricultural Locations

Concentrations (mg/kg day^{-1})				
	Mashayan Bululu		**Wachakal**	
PAHS	**FARO 44**	**FARO 52**	**FARO 44**	**FARO 52**
Naphthalene	9.79E-20	2.07E-17	1.20E-18	4.00E-18
2-Methyl Naphthalene	1.01E-17	2.38E-18	1.01E-20	5.22E-17
Acenapthylene	2.17E-18	9.79E-19	9.79E-19	5.31E-18
Acenaphthene	1.01E-19	2.17E-19	1.40E-18	3.38E-19
Fluorene	2.17E-20	4.11E-18	2.28E-19	5.22E-18
Phenanthrene	9.79E-20	5.11E-18	3.14E-18	4.24E-17
Anthracene	2.17E-17	2.77E-18	3.15E-17	5.90E-18
Fluoranthene	2.17E-19	5.22E-18	4.13E-18	4.25E-18
Pyrene	9.79E-19	3.37E-19	4.34E-18	3.16E-17
Benz(a)Anthracene	1.96E-17	9.12E-15	4.13E-17	3.16E-17
Chrysene	9.79E-20	5.00E-18	3.16E-18	3.27E-19
Benz(b)Fluoranthene	1.99E-19	2.29E-17	5.21E-17	4.44E-16
Benz(k)Fluoranthene	9.79E-18	4.13E-16	9.79E-16	9.12E-15
Benz(a)Pyrene	1.99E-15	5.00E-15	9.79E-16	4.87E-15
Dibenz(a,h)Anthracene	1.20E-15	3.15E-15	2.28E-16	3.38E-15
Indinol(1,2,3-cd)Pyrene	2.06E-17	2.06E-16	2.17E-17	8.79E-18
TDD	**3.28E-15**	**1.80E-14**	**2.35E-15**	**1.80E-14**

able 12: Carcinogenic Risk Assessment (mg/kg day^{-1}) of Some Polycyclic Aromatic Hydrocarbons (PAHs) in Rice Samples from Mashayan Bululu and Wachakal Agricultural Locations in Yobe State, Nigeria

Concentrations (mg/kg)				
	Mashayan Bululu		**Wachakal**	
PAHS	**FARO 44**	**FARO 52**	**FARO 44**	**FARO 52**
Benz(a)Anthracene	1.43E-17	6.66E-15	3.01E-17	2.31E-17
Chrysene	7.15E-22	3.65E-15	2.31E-20	2.39E-21
Benz(b)Fluoranthene	1.45E-19	1.67E-17	3.80E-17	3.24E-16
Benz(k)Fluoranthene	7.15E-19	3.01E-17	7.15E-17	6.66E-16
Benz(a)Pyrene	1.45E-14	3.65E-14	7.15E-15	3.56E-14
Dibenz(a,h)Anthracene	8.76E-15	2.30E-14	1.66E-15	2.47E-14
Indinol (1,2,3-cd) Pyrene	1.50E-17	1.50E-66	1.58E-17	6.42E-18
ΣILECR	**2.33E-14**	**6.99E-14**	**8.97E-15**	**6.13E-14**

Table 13: Hazard Quotient and Hazard Index (mg/kg day^{-1}) of Non-Carcinogenic Polycyclic Aromatic Hydrocarbons (PAHs) Via Consumption of Rice from Mashayan Bululu and Wachakal Agricultural Locations in Yobe State, Nigeria

	Concentrations (mg/kg)			
	Mashayan Bululu		Wachakal	
PAHS	**FARO 44**	**FARO 52**	**FARO 44**	**FARO 52**
Naphthalene	4.90E-18	1.04E-15	6.00E-17	2.00E-16
2-Methyl Naphthalene	5.05E-16	1.19E-16	5.05E-19	2.61E-15
Acenapthylene	1.09E-16	4.90E-17	4.90E-17	2.66E-16
Acenaphthene	1.68E-18	3.62E-18	2.33E-17	5.63E-18
Fluorene	5.43E-19	1.03E-16	5.70E-18	1.31E-16
Phenanthrene	2.45E-18	1.28E-16	7.85E-17	1.06E-15
Anthracene	7.23E-17	9.23E-18	1.05E-16	1.97E-17
Fluoranthene	5.43E-18	1.31E-16	1.03E-16	1.06E-16
Pyrene	3.26E-17	1.12E-17	1.45E-16	1.05E-15
Hazard Index (HI)	**7.34E-16**	**1.59E-15**	**5.70E-16**	**5.45E-15**

Discussion

Distribution of Some Polycyclic Aromatic Hydrocarbon (PAHs) in Soil Samples from Different Agricultural Locations

Sixteen PAHs (naphthalene, 2-methyl naphthalene, acenapthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benz(b)fluoranthene, benz(k)fluoranthene, benz(a)pyrene, dibenz(a,h)anthracene, and indinol(1,2,3-cd)pyrene were detected in soil samples from two agricultural locations (Mashayan Bululu and Wachakal). Anthracene (1.53E-03 mg/kg), dibenz(a,h)anthracene (3.77E-03 mg/kg), fluorene (3.11E-04 mg/kg), benz(k)fluoranthene (5.330E-04 mg/kg), and acenapthylene (9.00E-04 mg/kg) were observed to be the most dominant PAHs from the two agricultural locations (Tables 1 and 2). Mashayan Bululu agricultural location shows the highest total PAH concentration with a value of 2.53E-03 mg/kg (Table 1) when compared to the other agricultural locations. Soil depths of 0–5 cm recorded the highest PAHs compared to the other soil depth profiles. The above trend is due to the fact that PAHs tend to accumulate in the top soil (0–5 cm) because of their strong sorption towards soil organic matter (SOM) and any absorbing materials (Cornelissen *et al.,* 2005). This research is in line with the findings of Bandowe *et al.* (2010), who suggested that in areas where the PAH values are higher in the middle and subsoil layers, it is possible that disturbed soil causes more contaminated materials with elevated PAH concentrations reaching greater depth. Lower molecular weight PAHs (LMW) show the highest total concentration with a value of 7.22E-03 mg/kg in all the two agricultural locations compared with higher molecular weight PAHs (HMW). This may be due to the recent deposition of PAHs in the study area. Slash and burn also lead to higher levels of soil erosion due to the absence of organic matter coverage. Eroded soil allows PAH-laden water to permeate the farmlands. Mashayan Bululu recorded the highest total PAH concentration (Table 1) among the two agricultural locations due to the fact that PAHs are prone to a wide variety of degradation processes, including evaporation, dissolution, dispersion, emulsification, adsorption on suspended materials, microbial degradation, and photooxidation among contaminants and sediments (Kim *et al.,* 2009).

Diagnostic Ratios of PAHs of Soil Samples from Mashayan Bululu and Wachakal Local Government Area, Yobe State

The sources of polycyclic aromatic hydrocarbons can either be petrogenic, i.e., released from petroleum products, or pyrogenic, due to the combustion of biomass. Diagnostic ratios have been designed and used to distinguish the sources of PAHs due to their stability, physical, and chemical attributes (Nasher *et al*., 2013). The most commonly used ratios for the source identification of polycyclic aromatic hydrocarbons are BaA/BaA+Chr, Ant/Ant+Phen, Fluo/Fluo+Pyr, and LMW/HMW (BaA is benz(a)anthracene, Chr is chrysene, Ant is anthracene, Phen is phenanthrene, Fluo is fluoranthene, Pyr is pyrene, LMW is lower molecular weight PAHs, and HMW is higher molecular weight PAHs). If the ratio of BaA/BaA+Chr is less than 0.2, it signifies that PAH contamination was from petrogenic sources; if the ratio is between 0.2 and 0.35, it signifies that PAH contamination was from fuel combustion; if the ratio is greater than 0.35, it signifies that PAH contamination was from the combustion of coal, grass, or wood. Ant/Ant+Phen values of less than 0.1 and greater than 0.1 indicate petrogenic and pyrogenic sources, respectively; Fluo/Fluo+Pyr values of less than 0.4 and greater than 0.4 indicate petrogenic and pyrogenic sources, respectively; and LMW/HMW values of less than 1 and greater than 1 indicate that pollution was from pyrogenic and petrogenic sources, respectively (Nasher *et al*., 2013). In this study area, as shown in Table 5, the ratio of BaA/BaA+Chr was greater than 0.35 (0.968), thus implying that the source of PAHs is pyrogenic. The ratio of Ant/Ant+Phe in the study area was >0.1 (0.998), which implies that the source of the PAHs was from pyrogenic sources due to the combustion of bushes and other biomass. Also, the use of the Fluo/Fluo+Pyr ratio gave values of >0.5 (0.978), which further confirms a pyrogenic source. The value of the LMW/HMW ratio was 4.14, which indicates petrogenic sources as it was greater than 1 (Nasher *et al*., 2013). The burning of farmland for agriculture and refuse is a common practice around the study area. Generally, from the results above, the source of PAHs in the study area can be attributed to pyrogenic activities, although petrogenic contributions cannot be ruled out. Comparing the result of the present study to other studies, the values obtained were similar to those obtained in the rivers, sediments, and wastewater effluents in Vhembe District, South Africa (Edokpayi *et al*., 2016).

Ecotoxicity Studies of Some PAHs in the Soils of Mashayan Bululu and Wachakal Local Government Area, Yobe State

The potential toxicity of PAHs in the soils to humans, animals, and the surrounding environment was also assessed. PAH levels in the soil were compared with the United States Environmental Protection Agency soil quality guidelines (USEPA, 1993). The recommended ERL (effect range low) and ERM (effect range median) target values for soil. Values above the recommended ERM values indicate the likelihood of the occurrence of a high negative toxic effect in that area. A mild toxic effect is expected if the PAH concentrations range between

ERL and ERM values (Long and Macdonald, 1998). No negative effect is expected for PAH concentrations lower than ERL values. The results presented in Table 6 show both the effect range medium (ERM) and mean effect range medium quotient (m-ERM-q) values of all the detected polycyclic aromatic hydrocarbons to be below the recommended soil quality guidelines. This indicates that the probability of risk for the populace that lives in both Mashayan Bululu and Wachakal, Yobe State, is very low. In comparison, sediments and wastewater effluents in Vhembe District, South Africa, obtained similar results, which is in agreement with the present study (Edokpayi *et al*., 2016). The number of rings in PAHs also determines their toxicity (Weast, 1968). The greater the number of rings, the higher the molecular weight. As the molecular weight increases, the solubility of PAH decreases with an increase in melting and boiling points, and vice versa. This statement by Weast (1968) further backs the result of the present study, as this study had reported earlier that low molecular weight PAHs were the most predominant in the studied soil samples from Mashayan Bululu and Wachakal, Yobe State.

Carcinogenic Risk Assessment of PAHs in Soil Samples from Mashayan Bululu and Wachakal Local Government Areas, Yobe State

The total potential carcinogenic potency (BaPTEQ) of PAH mixtures in soil samples was determined by summing up the concentrations of individual PAHs, which were multiplied by the determined TEFs of individual PAHs (Nisbet and LaGoy, 1992), as shown in Table 12. Assessment of the study area discloses benz(a)pyrene and dibenz(a,h)anthracene as the highest accumulators of PAH deposits in all the two agricultural locations, with values of 2.40E-03 and 4.99E-03 mg/kg, respectively, while 2-methylnaphthalene and fluoranthene were observed to be the lowest contributors, with values of 2.39E-07 and 4.84E-07 mg/kg, respectively. Total carcinogenic toxic equivalency factor (BaPTEQ) ranged from 9.99E-08 to 4.37E-03 mg/kg. The International Agency for Research on Cancer (IARC) classified PAHs by their toxic potencies as probable (2A) and possible (2B) carcinogens, and the following PAHs have been highlighted: benzo(a)pyrene, dibenz(a,h)anthracene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(g,h,i)perylene, and indeno(1,2,3-c,d)pyrene (IARC, 2010). According to this statement by IARC (2010), the results of this study reveal that soil samples obtained from the study area are potentially carcinogenic. Similar results were obtained from soils in Taizhou, East China (Guanjiu *et al*. 2014), which is in conformity with the present study.

Distribution of Some Polycyclic Aromatic Hydrocarbon (PAHs) in Different Variety of Rice Samples from Different Agricultural Locations

Sixteen PAHs were detected in different varieties of rice (FARO 44 and FARO 52) samples from the two agricultural locations (Mashayan Bululun and Wachakal). The lower molecular weight PAHs were observed to be the most predominant over the higher molecular weight PAHs in all the two above-mentioned agricultural locations, with PAHs such as naphthalene, 2-Methyl Naphthalene, and Indinol (1,2,3-cd)Pyrene having the major PAH deposits with values of 2.12E-

09, 1.03E-09, and 2.11E-09 mg/kg, respectively (Tables 3 and 4). The lowest PAH concentration value of 2.20E-09 mk/kg in FARO 44 was observed from the Mashayan Bululu agricultural location. The rise of PAH values in Wachakal over Mashayan Bululu agricultural location may be due to Wachakal's close proximity to the highways, as PAHs contaminate vegetables and crops grown close to areas with intense traffic (Bostrom *et al*., 2002). Similar results were obtained in Ghana (PAH levels, risk assessments, and source characterization in cooked rice) by Essuman (2014).

Benz(a)pyrene Equivalent Concentration of Some Polycyclic Aromatic Hydrocarbons in Different Variety of Rice Samples from Mashaya Bululu and Wachakal Agricultural location, Yobe State

Based on the studies reported about PAH exposure in the non-occupationally exposed population, food is the major source of human exposure to PAHs, and rice constitutes one of the main causal sources (Jonaska, 2011). There are some reports (Ciecierska and Obiedzinski, 2013) concerning the contamination of BaP in different varieties of rice. It has been reported that BaP is a proper marker of the occurrence of the carcinogenic PAHs in food and their health outcomes (Kasim *et al*., 2012; FSIS, 2012). BaP is the only compound in the PAH group with sufficient toxicological evidence to authorize the setting of a guideline (Muyela *et al*., 2012). Toxicological studies on individual PAHs in animals, principally on BaP, have revealed a variety of toxicological impacts, such as reproductive and developmental toxicity, hematological effects, and immunotoxicity (FSAI, 2009). The International Agency for Research on Cancer (IARC) reported that BaP is carcinogenic to humans (Group 1) (IARC, 2011). Food can be polluted by PAHs that are present in the soil, air, or water, as well as those that are formed during thermal food processing or certain home cooking practices such as baking (Ciecierska and Obiedzinski, 2013). Assessment of BaP equivalent (BaPeq) in the study area reveals benz(a)pyrene as the highest accumulator. The summation of benz(a)pyrene toxic equivalency (BaPTEQ) across all the twoteen PAHs in each variety of rice revealed that the values ranged between 1.22E-10 and 5.97E-06 mg/kg. According to the existing regulations set by the European Commission, the content of BaP in food and baby foods should not go beyond 0.001 mg/kg (Bogusz *et al*., 2004). The result of the current study was below the acceptable limits set by the European Commission. It is also in order when compared with other studies like the determination of benzo(a)pyrene in bread using GC-MS by Samira *et al*. (2016), whose results were similar and hence in agreement with the results obtained from the present study.

Estimated Daily Dose of PAHs in Different Variety of Rice Samples from Mashayan Bululu and Wachakal, Yobe State

Intake of polycyclic aromatic hydrocarbons has been estimated in many European countries (Scientific Committee on Food, 2002). According to several studies, the major dietary contributors are rice, cereals, oils, and vegetables, although polycyclic aromatic hydrocarbon levels in rice are often low (Tao *et al*., 2006). Dietary intake of food contaminants depends on both the nutritional habits of the examined population group and the concentration of contaminants in the food. All the computations in the current study were based on the mean concentration of PAHs obtained from the analysis of samples of different varieties of rice (FARO 44, FARO 52) from Mashayan Bululu and Wachakal (Tables 11). Observation of all the PAHs daily dose values in all the varieties of rice samples shows that FARO 44 values ranged between 3.05E-26 and 5.43E-11 mg/kg day^{-1}; FARO 52 values ranged between 1.72E-26 and 9.12E-15 mg/kg day^{-1}; FARO 42 values ranged between 5.22E-17 and 3.15E-12 mg/kg day^{-1}; whereas FARO 45 values ranged between 3.74E-19 and 3.15E-14 mg/kg day^{-1}. The variety of rice whose daily dose has the highest concentration of PAHs was observed to be FARO 44 with a value of 5.84E-11 mg/kg day^{-1}, while FARO 52 recorded the lowest concentration of PAHs on a daily basis with a value of 1.20E-15 mg/kg day^{-1}. Total daily dose values were between 1.20E-15 and 5.84E-11 mg/kg day^{-1}. These values were observed to be lower than the exposure limit set by the WHO (0.004 mg/kg) for an average of 70 kg of body weight (WHO, 2006). The estimated daily dose value of this study is also lower in comparison to other studies, such as the values obtained for the dietary intake of PAHs in a Spanish population by Raquel *et al*. (2005), with a value of 3.24E-02 ug/kg.

Hazard Quotient and Hazard Index of Non-Carcinogenic Polycyclic Aromatic Hydrocarbons through Ingestion of Rice in Mashayan Bululu and Wachakal Local Government Areas, Yobe State

The potential for non-carcinogenic effects was evaluated by comparing an exposure level over the exposure duration with a reference dose derived for a similar exposure period. This ratio of exposure to toxicity for an individual pathway and chemical is called a hazard quotient (USEPA, 1989). The hazard quotients are usually added across all chemicals and routes to estimate the hazard index. Some, however, will argue that it is more appropriate to only sum the hazard quotients for chemicals that affect the same target organ (e.g., liver or blood). The non-cancer hazard quotient assumes that there is a level of exposure below which it is unlikely that even sensitive populations would experience adverse health effects (USEPA, 1989). The maximum hazard quotient recorded of all the non-carcinogenic PAHs detected in all the sampling sites was 1.04E-11 mg/kg, while the maximum hazard index observed was 2.04E-11 mg/kg (Table 13). As both values from the hazard quotient and index were far less than 1, the result of this study shows that consumption of rice from the said locations has no considerable health risk based on non-carcinogenic risk as per the United States Environmental Protection Agency guidelines. Similar results were reported for PAH intake by the general population in Estonia (Reinik *et al*., 2011), which was in agreement with the present study.

Incremental Lifetime Expectancy Cancer Risk (ILECR) through Intake of Polycyclic Aromatic Hydrocarbons in Different Variety of Rice Samples from Mashayan Bululu Wachakal, Yobe State

The average daily dose and the slope factor were used in the evaluation of the cancer risk assessment. Carcinogenic risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen. This risk is referred to as the incremental lifetime expectancy cancer risk (ILECR) or just carcinogenic risk. Published values of chemical carcinogenic toxicity (slope factor) are used to calculate carcinogenic risk from the lifetime average daily dose. The collective distribution of designated incremental lifetime expectancy cancer risk (ILECR) values for different varieties of rice samples from the aforesaid agricultural locations in Mashayan Bululu and Wachakal Mashayan Bululu, Yobe State, is shown in Table 12. As shown in the tables, Mashayan Bululu has ILECR values ranging from 7.61E-15 to 4.19E-10 mg/kg with a mean value of 2.10E-10 mg/kg, while ILECR values in Wachakal range from 8.97E-15 to 6.13E-14 mg/kg with a mean value of 3.51E-14 mg/kg. The result of the current study reveals that less than 1 in every 1,000,000 is likely to experience cancer-related illness in his or her lifetime from the consumption of rice in Mashayan Bululu, while less than 1 in every 1,000,000 may likely suffer from cancer-related cases in Wachakal, Yobe State, which is within the acceptable exposure boundary. Comparing these values to the USEPA-accepted figure of 10^6 (one out of one million people may suffer from cancer-related cases) (USEPA, 2006), the present values are insignificant based on the USEPA's 2006 guidelines.

CONCLUSION

Sixteen PAHs were detected in soil and rice samples from two agricultural locations in Mashayan Bululu and Wachakal, Yobe State. A depth of 10–15 cm recorded the lowest PAH load in Mashayan Bululu Agricultural Location. In the varieties of rice, the lowest PAH load was recorded in FARO 44 in the Mashayan Bululu location. The diagnostic ratio of PAHs recorded the highest total mean ratio at the Mashayan Bululu agricultural location. M-ERM-Q values from the two agricultural locations are below the USEPA soil guidelines; these results further show that the probable risk for the populace living in Mashayan Bululu and Wachakal Local Government Areas is very low. Carcinogenic risk assessment of PAHs in soil from the two agricultural locations shows benz(a)pyrene and dibenz(a,h)anthracene as the highest PAHs. FARO 44 shows the average daily dose of PAHs in the varieties of rice from the two agricultural locations, while FARO 52 is the lowest. Results from the incremental lifetime expectancy cancer risk show that FARO 52 from Wachakal had the highest values, while the lowest values were recorded for FARO 44 from Wachakal. Results from the present study show that the rice is safe for human consumption and should be monitored regularly in order to reduce the levels of PAHs.

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