

# X-ray fluorescence spectrometric determination of heavy metals in selected rice samples sold on the Liberian market: a case study conducted in Paynesville City, Greater Monrovia

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## Abstract

Background: Environmental pollution with toxic heavy metals can lead to the possible contamination of rice. Rice is a staple food widely consumed in the urban and rural parts of Liberia daily. Rice is cultivated in approximately 113 countries and a fundamental source for energy and protein. Objective: The study assesses selected heavy metals (As, Pb, Cd, Se, and Cr) concentration in selected imported rice and traditionally grown rice and bulgur wheat in Liberia. Methods: Six grade of imported rice, Bulgar wheat, and traditionally grown rice were purchased from the Duport Road and Red-Light markets in Greater Monrovia and analyze using X-ray Fluorescence Spectrometer. All data were analyzed using XLSTAT, and data was used to calculate the risk factor of each rice sample. Results: The mean concentration of heavy metal found in the rice as follow: As, 1.31ppm; Cd, 9.42ppm; Cr, 12.3ppm; Se, 5.73ppm; and Pb, 1.75ppm. The estimated daily intakes (EDIs) were calculated in combination with the rice consumption data. The mean intakes of As, Cd, Cr, Se, and Pb through rice were estimated to be 1.32, 9.42, 12.4, 5.74, and 1.75 mg/kg BW/day. Chromium has the average estimated daily intake. The combined hazard index for the heavy metals in each sample and the total cancer risk for each sample contributed most significantly to a cancer risk of rice consumption during adult life expectancy.

Conclusion: The selected heavy metal concentration from the rice sample was above the FAO/WHO reference Standard but was within the range of the contaminant level except for chromium, which is above the accepted range. However, the hazard index and the total cancer risk indicate a potential non-carcinogenic and carcinogenic risk.

**Keywords:** Duport Road; Red-light; EDI; Greater-Liberia; XRF; Rice.

## 1. Introduction

Rice is more widely consumed food globally[1]. Rice is one of the significant staple grains worldwide, increasing its productivity and nutritional quality is one of the foremost priorities[2]. Rice requires nutrient elements (heavy metals) found naturally in the earth to maintain average growth and development[3], [4]. Heavy metal (Cd and Zn) contaminating the environment, especially the soil, has been one of the most challenging pollution problems because of the severe toxicity leading to various illnesses, and significantly reduced the dry weight of the shoots and roots [4], [5]. The presence of heavy metals in the soil leads to severe adverse effects on plants and has been a subject of concern for many decades. Heavy metals due to poisonous, accumulating traits, and extended longevity are potential environmental contaminants with the capability of causing human health problems [6], [7]. According to the Food and Agriculture Organization, rice is cultivated in approximately 113 countries and provides 27% dietary energy supply and 20% of dietary protein intake in developing nations [8]. Food safety concerns triggered by toxic trace heavy metals have considered environmental issues in underdeveloped and developing countries [9]. Too much accumulation of agrochemicals could lead to the contamination of agricultural soils and plants.

Rice is a staple food widely consumed in the urban and rural parts of Liberia daily but not grown commercially to serve the citizenry. It is commonly consumed in Liberia but imported from countries located in Europe, Asia, the United States of America, and Sub Saharan nations as well as traditionally grown in the north and north-east of Liberia. Imported or traditionally grown, rice could be prone to disease

due to the presence of heavy metals. There has not been substantial qualitative research on traditional grown and imported rice in Liberia. Previous studies have shown heavy metal contaminants of food such as rice, other food crops, and fruits [9].

Globally the environment is heavily polluted with heavy metals. Plants and soils, including other environmental constituents, may get contaminated with trace heavy metals through diverse artificial and natural sources of contaminants [9], and the amount entering the environment from different artificial sources is noticeably higher. Despite the low concentrations of natural heavy metals in the environment, they are considered as pollutants; simply because they cause chronic and acute toxicities due to their wide-ranging nature. Some heavy metals are probable environmental impurities with the potential of causing human health complications when excess quantities are discovered in the food we consumed [10]. Numerous agricultural practices usually are well thought out as one of the means of accumulating trace heavy metals to the surroundings. Agriculture methods, such as organic waste disposal, pesticide application, irrigation, and fertilization, are presumed as the paths of the entrance of toxic trace heavy metals in agronomic soils. The pollution of agronomic soil by toxic trace heavy metals turned out to be a severe environmental problem owing to the different ecological effects they triggered. The ingestion of contaminated foods has been considered as the principal conduit for human exposure to potentially toxic heavy metals [11].

This study is concerned with arsenic, lead, cadmium, selenium, and chromium contents in imported and traditionally grown rice. The persistence of these metals, either in rice or the soil, are known to be carcinogenic and pose many other health risks [11]. Heavy metal contaminants in the environment are concentrated as they move up the food chain through bio-magnification. Quality assurance of food is pivotal to ensure its level of safety. The concentrations of toxic trace heavy metals in both imported and traditionally grown rice were taken into consideration. The heavy metal values were compared with the permissible level endorsed by FAO and WHO to detect as to whether there exists any health vulnerability for local consumers. Therefore, the objectives of this research are to (1) determine the concentrations of arsenic, lead, cadmium, selenium, and chromium in imported and traditional grown rice purchased from the Duport Road and Red-Light markets, Paynesville City, and (2) risk assessment of the selected heavy metal.

## 2. Materials and methods

**Study Area.** A total of 7-rice samples and bulgar wheat were purchased from the Duport Road and the Red-light general market in Greater Monrovia. The rice sample was based on the country of importation, type of rice, and disregarding the price. A total of six samples of rice-based on their different brands of importation, one sample was traditionally rice grown in Liberia and the other wheat. All samples were prepared for elemental analysis using an X-ray fluorescence spectrometer.

### 2.1. Procedure

All samples were recorded based on the country of importation and the texture of rice, as shown in Table 2. The rice sampling was randomly purchased from seven different retailers within the Red light and Duport Road markets. Various brands of the rice samples were collected and stored in polythene bags, and subsequently transported to the Liberia Geological Survey laboratory for dry analysis or spectrographic analysis.

#### 2.1.1. Dry or spectrographic analysis

The selected samples were washed thoroughly with distilled water. After that, the samples were oven-dried at 50°C for 45-minutes using the LEEC Drying Cabinet and cooled. Each sample was pulverized using a clean laboratory mill to fine powder and sieve at -500mm and +500mm in diameter using the Madison Test Sieve. The +500mm in diameter of each sample was used for the X-ray fluorescence spectrometry analysis.

#### 2.1.2. X-ray fluorescence spectrometry (XRFS)

The instrument was calibrated through a system check, and each sample was added to a cuvette and inserted into the X-Ray Fluorescence Spectrometry equipment for elemental analysis, as depicted in Table 2. The reaction time to run each sample was 240-second at 400-nM wavelength. The selected toxic trace heavy metals used to estimate the daily intake for all the rice samples during this evaluation.

#### 2.1.3. Toxic trace heavy metals

The elements Arsenic (As), Cadmium (Cd), Chromium (Cr), Selenium (Se), and Lead (Pb) were selected for the elemental analysis for the detection of trace heavy metals in rice samples.

## 2.2. The maximum permissible limit of heavy metal in rice

The estimated exposure is the maximum contaminant level of heavy metal in rice that is permitted. According to the Joint FAO/WHO Expert Committee, the maximum permissible levels of testing heavy metals, in milligrams per kilogram, are given in Table 1. The estimated exposure values and the PMTDI were used to determine the permissible limit of the heavy metal in the rice.

**Table 1:** The Maximum Contaminant Level of Heavy Metal in Rice [12].

Heavy metal	Estimated Exposure ( $\mu\text{g}/\text{kg BW}/\text{day}$ )	PMTDI ( $\text{mg}/\text{kg bw}$ )
As	0.1 – 3.0	-
Pb	0.02 – 3.0	0.025
Cd	2.2 - 12	25 <sup>b</sup>
Se	-	11.0 [13]
Cr	20 <sup>a</sup> [6]	-

PMTDI = Provisional Maximum Tolerable Daily Intake; a = mg/kg (ppm); b =  $\mu\text{g}/\text{kg BW}/\text{month}$ .

## 2.3. Estimated daily intake (EDI) of As, Cd, Cr, Se, and Pb through rice consumption

The regular intake of metals depends on the level of heavy metal and the frequency of consumption. The EDI is a parameter used to quantify the oral exposure dosage during a specific period, expressed as a daily dose per unit body weight. The EDI is associated with the type of rice consumed and the level of heavy metal. The EDI was calculated using equation [3, 7]:

$$EDI = \frac{C_{\text{metal}} \cdot \text{DIR}}{\text{BW}} \quad (1)$$

Where  $C_{\text{metal}}$  is the concentration of the selected heavy metal ( $\mu\text{g}/\text{kg}$ ) in rice, DIR is the daily intake rate of rice in  $\text{kg}/\text{day}$ , and BW is the average body weight. The daily heavy metal dose depends on the heavy metal concentration. The DIR value is 0.13  $\text{kg}$  and a typical 60 $\text{kg}$  bodyweight in Nigeria [14] since there was an absence of data on dietary intake in Liberia. Nigeria and Liberia are part of the Economic States of West Africa, and rice is a staple food in Liberia. The exposure frequency was set at 64 years, the life expectancy in Liberia [15], and exposure duration of 365 days.

## 2.4. Human risk assessment

### 2.4.1. Non-carcinogenic risk (NCR)

The NCR was calculated using the EDI and reference dose (RfD) of the respective selected heavy metals. The ratio between the EDI and RfD is known as the Hazard Quotient (HQ).

$$HQ = \frac{EDI}{\text{RfD}} \quad (2)$$

HQs  $< 1$  indicate a safety level, whereas HQs  $> 1$  indicates potential carcinogenic effects. However, the sum of the HQ is the hazard index (HI). HI  $< 1$  and HI  $> 1$  indicate chronic risks are unlikely and likely to occur, respectively.

### 2.4.2. Carcinogenic risk (CR)

The slope factor (SF) and the EDI were used to estimate the probability of individuals consuming the rice to develop cancer over a lifetime. For instance, the CR of  $10^{-4}$  indicates the chance that 1 in 10,000 individuals will develop cancer.

$$CR = EDI * SF \quad (3)$$

The sum of the CRs of the heavy metals present in particular rice is used to calculate the cancer risk factor from consuming specific rice.

## 2.5. Statistical techniques of results

All the data were analyzed using XLSTAT, an analytical solution downloaded and integrated into Microsoft Excel 2016.

## 3. Results and discussion

The elemental analysis of the samples purchased from the Duport Road and Red-Light markets is presented in Table 2. From the table, the result shows that the high concentrations of cadmium (Cd), arsenic (As), Selenium (Se), lead (Pb), and chromium (Cr) were realized in imported rice from America, India, and traditionally grown rice, respectively. However, Indian parboiled rice (R-1 and R-3) has the highest concentrations of arsenic and lead. The parboiled rice samples imported from the United States of America (R-2) and India (R-1) contain the highest levels of cadmium (Cd) in the range of 22.03 ppm and 21.58 ppm, respectively. The locally grown rice (R-6) showed the highest concentration of chromium (Cr) of 16.46 ppm, followed by the white long-grain rice from China (R-5) with a high chromium concentration of 16.39 ppm.

**Table 2:** Elemental Analysis of Selected Heavy Metals (Ppm) in Selected Rice Bought from the Retailers in the Duport Road and Red-Light Markets

Rice	Country	Type of Rice	As	Cd	Cr	Se	Pb
R-1	India	Parboiled	1.17	21.58	7.16	1.05	1.59
R-2	America	Parboiled	1.14	22.03	6.82	1.25	1.87
R-3	India	Parboiled	1.50	21.20	6.90	1.17	1.95
R-4	Thailand	White Long Grain	1.24	2.08	16.25	1.14	1.69
R-5	China	White Long Grain	1.27	2.02	16.39	1.12	1.65
R-6	Liberia	Traditionally grown	1.26	2.06	16.46	1.10	1.68
R-7	America	Burger Wheat	1.36	2.31	13.43	1.26	1.84
R-8	America	Nobel Long Grain	1.31	2.11	15.38	1.18	1.74

The result in Table 3 shows that chromium (Cr) was in the range of 6.82 – 16.46 (mean of  $12.3 \pm 4.6$ ) ppm and cadmium in the range of 2.02 – 22.03 (mean of  $9.40 \pm 10.1$ ) ppm respectively. The concentrations of Arsenic (As) and lead (Pb) were low in concentration, with averages of  $1.32 \pm 0.11$  ppm and  $1.75 \pm 0.12$  ppm, respectively.

**Table 3:** Descriptive Analysis of Heavy Metal in the Rice Sample Sold on the Liberian Market

Variable	Observations	Minimum	Maximum	Mean	Std. deviation
As	8	1.170	1.500	1.315	0.105
Cd	8	2.020	22.030	9.424	10.088
Cr	8	6.820	16.460	12.349	4.567
Se	8	1.050	1.26	1.159	0.025
Pb	8	1.590	1.950	1.751	0.123

From the level of heavy metals analyzed in the selected rice, the prediction of the daily intake of heavy metal was calculated from the estimated daily intake formula using Equation 1. The estimated daily intake of the selected heavy metals measured from the selected rice samples is shown in Table 4.

The highest amount of the EDI (Table 4) of chromium (Cr) calculated to be 0.097  $\mu\text{g}/\text{kg}\cdot\text{day}\cdot\text{bw}$ , which corresponds to 9.7% of the total daily intake (TDI) of the Chinese White Long Grain consumed in Liberia. The maximum daily intake for Cr is 0.20 ppm. The maximum intake of cadmium from rice is 1.13  $\mu\text{g}/\text{day}\cdot\text{kg}\cdot\text{bw}$ . It is important to note that the daily intake of rice (DIR) in Nigeria is calculated to be 0.128  $\mu\text{g}/\text{day}\cdot\text{kg}\cdot\text{bw}$  (Indian Parboiled Rice), which corresponds to 12.8% of the tolerable daily intakes (i.e., 1  $\mu\text{g}/\text{day}\cdot\text{kg}\cdot\text{bw}$ ). The dietary intake for cadmium (Cd), Arsenic (As), Selenium (Se), and lead (Pb) ranges from 0.012 to 0.014 (TDI 1.2 – 1.4%); 0.007 – 0.009 (TDI 0.7 – 0.9%); 0.030 – 0.038 (TDI 3.0 – 3.8%); and 0.009 – 0.012 (0.9 – 1.2%)  $\mu\text{g}/\text{day}\cdot\text{kg}\cdot\text{bw}$ , respectively[7].

**Table 4:** Estimated Daily Intake of Heavy Metal from Selected Rice Sample Sold on the Liberian Market

	Country of Importation	Type of Rice	As (ppm)	Cd (ppm)	Cr (ppm)	Se (ppm)	Pb (ppm)
R - 1	India	Parboiled	0.0025	0.0468	0.0155	0.0023	0.0034
R - 2	America	Parboiled	0.0031	0.0477	0.0148	0.0027	0.0041
R - 3	India	Parboiled	0.0033	0.0459	0.0150	0.0025	0.0042
R - 4	Thailand	White Long Grain	0.0027	0.0045	0.0352	0.0025	0.0037
R - 5	China	White Long Grain	0.0028	0.0044	0.0355	0.0024	0.0036
R - 6	Liberia	Traditionally grown	0.0027	0.0045	0.0357	0.0024	0.0036
R - 7	America	Burger Wheat	0.0029	0.0050	0.0291	0.0027	0.0040
R - 8	America	Nobel Long Grain	0.0028	0.0046	0.0333	0.0026	0.0038

Table 5 represents the mean of heavy metals of branded rice imported into the country and cultivated locally. The results of the analysis showed that chromium (Cr) has the highest mean value, followed by cadmium (Cd), Selenium (Se), lead (Pb), and arsenic (As). In Table 5, the mean distributions of heavy metals in the traditional rice were calculated. Cr shows the highest value of the mean, followed by Se, Cd, Pb, and As. Comparing the mean concentration of the selected heavy metals to the estimated exposure level, as depicted in Table 1 for FAO and WHO, only chromium was below the maximum contaminant level. Table 5 also reveals that EDI of chromium (Cr) content was higher and has a tolerable daily intake of 7.30% as compared to the arsenic content with TDI of 0.78%. It is visible in Figure 1.

**Table 5:** Mean of the Estimated Daily Intake of Heavy Metal from A Rice Sample

	As	Cd	Cr	Se	Pb
Mean	1.32	9.42	12.4	5.74	1.75
EDI	0.0078	0.0559	0.0730	0.0340	0.0104
Recommended Dose [1]	0.13	0.06	0.20	0.50	0.21
Chinese Standard (mg/kg) [14]	0.50	0.20	-	10	0.20
FAO/WHO [11]	0.1-3	2.2-12	0.20	-	0.02-3
Slope Factor (SF) mg/(kg/day)-1	1.5a	15b	0.50e		0.0085e
RfD (mg/kg-day)	0.003	0.001c	0.003d	0.005c	0.0036b

a = RAIS (The Risk Assessment Information System); b = [16]; c = US EPA; d = IRIS; e = OEHHHA (California Office of Environmental Health Hazard Assessment).

The findings of this study as it relates to the selected heavy metals slightly differ with the concentrations of all of the chosen metals, namely, Cr, Cd, Se, Pb, and As, were within the maximum contaminant level except for Chromium, Cr, which was above the permissible level [14]. In the study, Lead (Pb), has the highest concentration of permissible limit for human consumption. But this can be validated by the fact that the allowable level of heavy metals in rice is directly proportional to the environmental surroundings where the rice was cultivated. This statement was validated in the study, in which they analyzed the correlation between rice, heavy metal concentrations, agricultural products, and the environmental surroundings[11]. They found out that As and Cd concentrations in the rice root were found to be several times larger than in the surrounding soil and fertilizer used, while Pb concentrations were lower. But, these concentrations were lower in the final rice grain.[17] Found out that the levels of elemental mercury and arsenic were higher than the limits prescribed by the Food and Drug Administration (FDA) and (FAO).

Health Risks to Residents from Heavy Metal Exposure in Brown Rice. The EDI values for As, Cd, Cr, Se, and Pb varies for all selected rice consume by an individual in Liberia. The EDI for Cd and Cr was high in all the rice and Bulgar wheat samples, which suggested a large adult daily intake of Cd and Cr in rice. The EDI of Pb, As, and Se was lower, and adult daily intake of these contaminants from the rice was not significant[16], [18]. For the risk assessment for consuming either of the selected rice over an individual life expectancy, the non-carcinogenic and carcinogenic risk assessment was calculated.

**Table 6:** Non-Carcinogenic Risk Assessment (Hazard Quotient for the Selected Heavy Metals) of Sample

	Country of Importation	Type of Rice	As	Cd	Cr	Se	Pb	HI.
R - 1	India	Parboiled	0.85	46.76	5.17	0.46	0.96	54.18
R - 2	America	Parboiled	1.02	47.73	4.93	0.54	1.13	55.34
R - 3	India	Parboiled	1.08	45.93	4.98	0.51	1.17	53.68
R - 4	Thailand	White Long Grain	0.90	4.51	11.74	0.49	1.02	18.65
R - 5	China	White Long Grain	0.92	4.38	11.84	0.49	0.99	18.61
R - 6	Liberia	Traditionally grown	0.91	4.46	11.89	0.48	1.01	18.75
R - 7	America	Burger Wheat	0.98	5.01	9.70	0.55	1.11	17.34
R - 8	America	Nobel Long Grain	0.95	4.57	11.11	0.51	1.05	18.18

The HQs (Table 6) for Cd and Cr were significantly higher. The HQ for cadmium 46.76, 47.73, and 45.93 was the highest for sample R-1, R-2, and R-3, respectively. Samples R-4, R-5, R-6, and R-8 have higher HQ values. Therefore, HQs of Cd and Cr for all the rice samples exceeded 1, indicating that the two metals pose potential non-carcinogenic risks for individuals consuming the rice. The HQs values for selenium in all the sample was less than unity indicating an unlikely chronic risk. Due to high HQs for Cd and Cr, significantly affected the Hazard Index (HI). Therefore, the HI for each sample was higher, indicating a high potential non-carcinogenic health risk and chronic toxicity due to the combined exposure of the selected heavy metals over a life time[16], [18].

**Table 7:** Carcinogenic Risk Assessment

	Country of Importation	Type of Rice	As (ppm)	Cd (ppm)	Cr (ppm)	Se (ppm)	Pb (ppm)	CRt
R - 1	India	Parboiled	0.0038	0.7014	0.00776	-	0.000029	0.7129
R - 2	America	Parboiled	0.0046	0.7160	0.00739	-	0.000034	0.7280
R - 3	India	Parboiled	0.0049	0.6890	0.00748	-	0.000036	0.7014
R - 4	Thailand	White Long Grain	0.0040	0.0676	0.01760	-	0.000031	0.0893
R - 5	China	White Long Grain	0.0041	0.0657	0.01776	-	0.000030	0.0876
R - 6	Liberia	Traditionally grown	0.0041	0.0670	0.01783	-	0.000031	0.0889
R - 7	America	Burger Wheat	0.0044	0.0751	0.01455	-	0.000034	0.0941
R - 8	America	Nobel Long Grain	0.0043	0.0686	0.01666	-	0.000032	0.0895

The CR values for cadmium in all the samples of the rice was high but higher in R-1 and R-2 (Table 7). The cadmium value exceeded  $10^{-4}$ , and the CRt values for R-1 and R-2 was 0.7129 and 0.7280, respectively indicating a higher potential carcinogenic risk for individual consuming R-1 and R-2 rice over a life expectancy. Finally, each rice sample CRt values exceeded the  $10^{-4}$  benchmark indicating carcinogenic risk over adult life expectancy [16] [19].

## 4. Conclusion

During the evaluation, branded imported and traditionally grown rice were analyzed for the determination of arsenic, lead, cadmium, selenium, and chromium contents. Taking into account the mean concentrations of the trace heavy metals that were evaluated in all of the imported and traditional examined branded rice samples, the Indian parboiled rice has the highest levels of arsenic and lead. Based on the results obtained from this study, it can be concluded that the concentration of selected heavy metals, namely, Cr, Cd, Se, Pb and As, in the brand of rice samples, be it imported or locally grown, sold on the Liberian markets are within the permissible limit for human consumption except that of Chromium, Cr, and Cadmium, Cd is above the allowable threshold for use according to FAO/WHO. Health Risks indicates the EDI values for As, Cd, Cr, Se, and Pb varies for all selected rice consume by an individual. The EDI for Cd and Cr was high in all the rice and bulger wheat samples, which suggested a large adult daily intake of Cd and Cr in rice and lower for Pb, As, and Se in each sample. Because of the high concentration of chromium analysis in locally grown rice, it is imperative to conduct a Geospatial study. Finally, from the HI and CRt values for all samples, it indicated a potential non-carcinogenic and carcinogenic risk effect over adult life expectancy.

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