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Pollution Indices and Health Risk Assessment of Heavy Metal levels in *Oryza Sativa* (Rice) consumed in Southeastern, Nigeria

Victor U. Okechukwu[1], Daniel O. Omokpariola[1,*]

[1]*Department of Pure and Industrial Chemistry, Nnamdi Azikiwe University, Awka, Nigeria*

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ABSTRACT

The research investigates the concentrations of heavy metals, namely Lead (Pb), Cadmium (Cd), Zinc (Zn), Chromium (Cr), Copper (Cu), and Nickel (Ni), in rice grains obtained from Southeastern Nigeria. The study compares imported and locally cultivated rice varieties. The rice samples were acquired, processed, and subjected to analysis for heavy metals using atomic absorption spectrophotometry. To evaluate the data and assess human health risks, statistical analysis, USEPA models, and FAO/WHO standards were employed. The results revealed that Chromium (Cr) and Cadmium (Cd) concentrations were below detectable levels (0.001 mg/kg), with 27% of Lead (Pb) falling below 0.001 mg/kg. Copper (Cu) concentrations ranged from 1.03 to 3.43 mg/kg, Nickel (Ni) from 0.28 to 3.36 mg/kg, Zinc (Zn) from 1.13 to 23.5 mg/kg, and Lead (Pb) from 0.001 to 20.8 mg/kg. All samples adhered to limits for Cu, Cd, Zn, and Cr, while 60% and 66% exceeded FAO/WHO standards for Ni and Pb. The estimated daily metal intake varied significantly, and the human health risk assessment revealed hazard quotients (HQ) below one for adults but above for children in all samples. Hazard indices surpassed one for both populations due to cumulative health risks from Ni, Pb, and Cu. The incremental lifetime cancer risk (ILCR) for Pb and Ni ranged from 1.2E-05 to 7.5E-06 and 1.5E-04 to 3.2E-04, respectively. Despite potential health risks, continuous monitoring of toxic metal concentrations in rice samples is crucial to prevent adverse effects on consumers.

1. Introduction

Rice, recognized for its rich nutritional content, constitutes a major portion of the caloric intake for most Nigerians [1]. As a staple food for roughly half of the global population, its ease of production and widespread availability for sale make it indispensable [1]. While some regions have a long-standing tradition of rice cultivation, for many, rice was once considered a luxury reserved for special occasions. However, with increased accessibility, rice has become a daily dietary staple in Nigeria. Although heavy metals are natural components of the Earth's crust, they typically exist at non-toxic levels. However, due to their persistence and bioaccumulation, they become environmental contaminants. Factors such as fertilizer and pesticide use, industrial waste deposition, and water used for irrigation contribute to the heavy metal presence in soil, which plants absorb through the atmosphere [2,3,4]. Crops cultivated in soil contaminated with heavy metals accumulate elevated metal levels, posing a risk when consumed, introducing toxic metals into the human system through ingestion [2,5]. Excessive heavy metal accumulation in the environment poses toxicological risks to humans, plants, and animals, leading to various health issues, including decreased immunological defenses, intrauterine growth retardation, impaired psycho-social behaviors, malnutrition-related disabilities, and an elevated incidence of upper gastrointestinal cancer [6,7]. Studies have highlighted the close association between metal concentrations in plants and soil levels, a crucial aspect in human and

* Corresponding author.; e-mail: mariamkh681@gmail.com
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ecological risk assessment modelling [4,8,9]. Predictions of estimated daily intake rates derived from metal exposure through soil, water, air, and food are possible. The U.S. Environmental Protection Agency's (EPA) hazard quotient (HQ) is widely employed to evaluate potential health risks associated with prolonged metal exposure through various media [10-12]. Nigeria, the top producer and consumer of rice in West Africa and the second-largest global rice importer, annually acquires at least two million metric tons from countries like China, India, and Thailand [13]. Studies in China and Thailand have correlated human renal dysfunction with Cadmium (Cd) contamination in rice [14]. Common pollutants in arable soil, including toxic metals like Ni, Hg, Cr, and Cd, originate from mining, industrial activities, and waste effluents [15,16]. Research in Southeast China revealed widespread Cd contamination in local soils [17]. Given the paramount concern for food safety, the current study in Southeastern Nigeria aims to evaluate concentrations and health risks associated with Ni, Cu, Pb, Zn, Cr, and Cd in imported and locally cultivated rice grains to determine potential health risks to consumption.

2. Materials and Methodology

2.1. Sample Collection and Preparation

For this analytical study, thirty (30) rice brands were procured from diverse markets in the Southern part of Nigeria. Each sample was securely enclosed in a sampling bag and identified with the brand's name and a unique sample ID. Subsequently, 100 grams of each rice sample underwent grinding using an electric grinder to achieve a fine powder. The resulting powdered rice was sifted through a 2-micron mesh to remove any remaining large debris. These powdered samples were then stored in air-tight containers, awaiting further analysis.

2.2. Elemental Analysis

Dried samples, each weighing two grams (2g), were carefully measured into digestion flasks. To these samples, 4ml of perchloric acid and 8ml of nitric acid were added into the digestion flasks and underwent heating on a hot plate set at 550□ until complete digestion of the samples was achieved. After digestion, the samples were appropriately diluted with distilled water within the range of standards prepared from a metal stock solution, as defined by the American Public Health Association [18]. The concentrations of heavy metals in the samples were determined using atomic absorption spectrometry (Varian AA240, Agilent, USA), following the procedure outlined by Braid et al. [19].

2.3. Human Health Risk Assessment (HHRA)

2.3.1. Estimated Daily intake (EDI) of heavy metal

The estimated daily intake (EDI, mg/kg/day) of heavy metals through rice consumption was computed using the formula [20,21]:

$$EDI = \frac{C\ X\ IR\ X\ ED\ X\ EF}{BW\ X\ AT} \qquad (1)$$

Where: EDI represents the estimated daily intake, C is the metal concentration (mg kg–1), IR is the ingestion rate (409.7 g/day for adult Nigerians), EF is the exposure frequency (365 days yr–1), ED is the exposure duration (70 years), BW is the body weight of the consumer (assumed average of 70 kg for adults and 16.7 kg for children), and AT is the specific period of exposure for non-carcinogenic effects (30 years x 365 days = 10950 days) [22,23].

2.3.2. Non – carcinogenic risk

The non-carcinogenic risk was assessed using the hazard quotient (HQ), calculated as [21, 24]:

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

Where: HQ = hazard quotient (HQ) assesses and group each toxicant based on the non – carcinogenic adverse effects due to exposure. RfD is the reference dose (mg/kg/day) estimates the maximum permissible dose via exposure to the human population to cause health risk effects during a lifetime. The oral reference doses of Zn, Pb, Cd, Cu, Ni and Cr(III) is 0.3, 0.004, 0.001, 0.04, 0.02 and 1.5 [10, 23]. If HQ less than 1, it suggests no major health risk, while vice versa for HQ greater than 1 [24 – 26].

2.3.3. Hazard Index (HI)

The hazard index (HI), evaluating the potential risk of adverse health effects from a mixture of chemical elements, was calculated for the rice samples using the equation [24, 27, 28]:

$$HI = \Sigma\ HQ \qquad (3)$$

HI is the sum of HQ (assuming additive effects). If HI < 1, chronic risks are assumed unlikely to happen, while non-cancer risks are likely if HQ ≥ 1.

2.3.4. Carcinogenic Risk

The cancer risk (CR), representing the incremental probability of developing cancer for individuals exposed to a given chemical over a lifetime, was calculated using the equation [25,28]:

$$CR = CDI \times SF \quad (4)$$

The chronic daily intake of chemical carcinogen (CDI) was determined by

$$CDI = \frac{Edi\ X\ IR\ X\ ED\ X\ EF}{BW\ X\ AT} \qquad (5)$$

Where CDI is the chronic daily intake of chemical carcinogen, mg/kg BW/day which represents the lifetime average daily dose of exposure to the chemical carcinogen, SF is cancer slope factor (mg/kg day-1) for a substance. SF values for the metals were Pb= 0.085, Ni = 0.91, Zn= 0. A cancer risk of 10^-4 to 10^-6 is considered acceptable by the United States Environmental Protection Agency (USEPA) [25]. Cancer risk of 10-4 and 10-6 indicates a probability of 1 in 10,000 individuals and 1 in 1,000,000 individuals developing cancer during a lifetime.

2.4. Statistical analysis

Data from the rice samples were subjected to analysis using the Statistical Package for Social Science (SPSS) 20.0 and Microsoft Excel 2016 software. Results were reported as mean ± standard deviation (SD) and analyzed using one-way analysis of variance (ANOVA).

3. Results and Discussion

3.1. Heavy metal concentration in Rice samples.

The analysis of heavy metal concentrations (mg/kg) in 30 rice samples collected from various locations in the southern part of Nigeria is detailed in **Table 1**. Notably, the concentrations of Cr and Cd were below the detectable level of 0.001 mg/kg, and 27% of Pb fell below the detectable level of 0.001 mg/kg. Metal concentrations ranged from 1.03 to 3.43 mg/kg for Cu, 0.28 to 3.36 mg/kg for Ni, 1.13 to 23.5 mg/kg for Zn, and 0.001 to 20.8 mg/kg for Pb. Sample M1 (Thailand rice) displayed the highest zinc concentration at 23.41 mg/kg, whereas sample O1 exhibited the lowest zinc concentration at 1.134 mg/kg. Sample W1 (Thailand rice) had the highest lead concentration at 20.77 mg/kg, while eight samples were below the detectable level of 0.001 mg/kg. In terms of nickel concentration, Sample I1 (Nigerian rice) recorded the highest value at 3.36 mg/kg, and sample W1 (Thailand rice) had the lowest concentration at 0.282 mg/kg. Furthermore, Sample Y1 (Nigerian rice) had the highest copper concentration at 3.426 mg/kg, while Sample B2 (Indian rice) displayed the lowest copper concentration at 1.033 mg/kg. All rice samples adhered to limits for Cu, Cd, Zn, and Cr, but 60% and 66% exceeded recommended standards for Ni and Pb, respectively. Tables 2 and 3 outline the estimated daily intake (EDI) in mg/kg/day of Cu, Ni, Zn, and Pb through the consumption of rice samples for both the adult and child populations in Southeast Nigeria. The estimated daily intake of copper for the adult population ranged from 0.0146 to 0.0287 mg/kg/day, while for the child population, it varied from 0.061 to 0.120 mg/kg/day. Similar ranges were observed for Ni, Zn, and Pb. The study compared the concentrations of heavy metals (Zinc (Zn), Copper (Cu), Lead (Pb), Nickel (Ni), Cadmium (Cd), and Chromium (Cr)) in rice samples from the Eastern part of Nigeria. The mean concentration in rice grain followed the order: Zn > Pb > Cu > Ni > Cd > Cr. Comparisons with FAO/WHO standards, as shown in Table 1, revealed that the concentration of Cd, Cr, and 27% of Pb fell below the detectable limit. Cd, a non-essential element in foods and natural water, tends to accumulate in the kidneys and liver, and its persistence in the body has been linked to renal damage and abnormal urinary excretion of proteins [30, 31]. Pure chromium has no adverse effects, with toxicity mainly attributed to hexavalent compounds in large quantities. Hexavalent chromium compounds can cause bronchial carcinomas, gastroenteritis, and hepatocellular deficiency [32 – 34].

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Table 1. Concentration (mg/kg) of Heavy metals in studied rice samples

S/No	Sample ID	Source Country	Cu (mg/kg)	Ni (mg/kg)	Zn (mg/kg)	Pb (mg/kg)	Cd (mg/kg)	Cr (mg/kg)
1	A1	Thailand Rice	1.449	0.833	7.422	BDL	BDL	BDL
2	F1	Thailand Rice	1.316	1.166	4.504	BDL	BDL	BDL
3	E1	Thailand Rice	1.283	1.849	5.659	BDL	BDL	BDL
4	G1	Thailand Rice	2.949	1.938	21.82	BDL	BDL	BDL
5	J1	Thailand Rice	3.256	2.209	23.47	7.808	BDL	BDL
6	K1	Thailand Rice	1.317	0.967	3.572	BDL	BDL	BDL
7	L1	Thailand Rice	1.531	1.948	2.568	1.995	BDL	BDL
8	M1	Thailand Rice	3.364	2.131	23.41	6.829	BDL	BDL
9	A2	Thailand Rice	1.754	2.325	5.437	6.731	BDL	BDL
10	B3	Thailand Rice	1.912	2.682	5.574	19.42	BDL	BDL
11	P1	Thailand Rice	1.233	1.367	2.120	15.67	BDL	BDL
12	Q1	Thailand Rice	1.416	2.343	1.929	13.49	BDL	BDL

13	C1	Thailand Rice	1.449	0.849	7.111	BDL	BDL	BDL
14	S1	Thailand Rice	1.331	1.781	2.202	15.48	BDL	BDL
15	T1	Thailand Rice	3.325	1.933	18.14	12.67	BDL	BDL
16	W1	Thailand Rice	3.173	0.282	18.95	20.77	BDL	BDL
17	D1	Indian Rice	1.267	0.483	4.382	BDL	BDL	BDL
18	B2	Indian Rice	1.033	1.550	3.618	BDL	BDL	BDL
19	R1	Indian Rice	1.399	2.633	2.434	14.33	BDL	BDL
20	B1	Nigerian Rice	1.514	1.397	3.884	BDL	BDL	BDL
21	H1	Nigerian Rice	3.249	1.949	22.62	BDL	BDL	BDL
22	I1	Nigerian Rice	1.816	3.364	2.655	0.833	BDL	BDL
23	C2	Nigerian Rice	1.549	1.449	6.090	16.49	BDL	BDL
24	N1	Nigerian Rice	1.283	1.567	2.315	13.50	BDL	BDL
25	O1	Nigerian Rice	1.233	1.832	1.134	13.83	BDL	BDL
26	V1	Nigerian Rice	1.064	2.494	3.677	11.47	BDL	BDL
27	U1	Nigerian Rice	2.077	1.479	3.584	10.14	BDL	BDL
28	Y1	Nigerian Rice	3.426	1.513	19.34	17.13	BDL	BDL
29	X1	Nigerian Rice	2.957	1.877	17.02	17.61	BDL	BDL
30	Z1	Nigerian Rice	3.000	1.767	15.09	10.17	BDL	BDL
	Minimum		1.033	0.30	1.13	0.001	-	-
	Maximum		3.426	3.70	23.50	20.84	-	-
	Mean		1.95	1.73	8.72	8.21	-	-
	Standard	deviation	0.85	0.67	7.80	7.34	-	-
WHO	permissible	limit [15]	-	1.5	50	0.2	0.1	1.0

BDL – Below detectable level

The concentrations of nickel in rice grains fell within the FAO/WHO permissible limit of 1.5 mg/kg for human consumption, while zinc concentrations remained below the FAO/WHO limit [27,34]. However, 60% and 66% of the rice samples exceeded the FAO/WHO limits for nickel and lead, respectively [27,34]. This suggests that the concentrations of lead (Pb) and nickel (Ni) may have been influenced by various anthropogenic activities and the use of pesticides and chemical fertilizers [7,8]. The indiscriminate disposal of lead-acid batteries, vehicular emissions, and sewage water irrigation can contribute to the accumulation of lead in rice grains [35]. Lead has been associated with both plant and animal diseases, causing lipid peroxidation, loss of photosynthetic capacity in plants, and leading to renal impairment, immunotoxicity, reproductive organ toxicity, hypertension, and anemia [36]. Cadmium (Cd), copper (Cu), and zinc (Zn) are typically released through industrial emissions associated with acid rain, draining away from watershed soils, bedrocks, and lake sediments under acidic conditions [37]. Copper, a vital trace element with multiple biological roles, acts as a prosthetic group in several key enzymes. However, high intake of copper can result in headaches, dizziness, nausea, diarrhea, liver damage, and kidney damage [38]. The concentration of copper in the rice samples ranged between 1.03 and 3.426 mg/kg, with the highest level observed in sample coded Y1 and lowest in B2.

3.2. Estimated daily intake (EDI)

Tables 2 and **3** present the estimated daily intake (EDI) of Cu, Ni, Zn, and Pb through rice consumption for the adult and child populations in Southeast Nigeria. The EDI of copper for the adult population ranged from 0.0146 to 0.0287 mg/kg/day, while for the child population, it varied from 0.061 to 0.120 mg/kg/day. Similar ranges were observed for Ni, Zn, and Pb. The study suggests that while adverse health effects may be low for the adult population, individuals with lower body weight and higher consumption patterns may face a higher risk of adverse health effects from heavy metals. All metals considered were below the tolerable daily intake for an assumed average adult weight of 70 kg consuming 0.0679 kg of rice daily. However, the continuous ingestion of heavy metals from rice and other sources may pose health risks [39].

Table 2. Estimated dietary intake (EDI) (mg/kg/day) of Heavy metals for the Adult population through consumption of rice brands

S/N	Mean Conc. Rice	Cu	Ni	Zn	Pb
1	Thailand Rice	0.0146	0.0121	0.1313	0.1031
2	Indian Rice	0.0168	0.0212	0.0475	0.0652
3	Nigerian Rice	0.0287	0.0257	0.1209	0.1380
	Mean of 30 Samples	0.0266	0.0236	0.1191	0.1121

Table 3. Estimated dietary intake (EDI) (mg/kg/day) of Heavy metals for child population through consumption of rice brands

S/N	Mean Conc. Rice	Cu	Ni	Zn	Pb
1	Thailand Rice	0.061	0.051	0.550	0.432
2	Indian Rice	0.070	0.089	0.199	0.273
3	Nigerian Rice	0.120	0.107	0.506	0.578
	Mean of 30 Samples	0.112	0.099	0.499	0.470

3.3. Hazard Quotient/ Hazard Index

Tables 4 and **5** provide insights into the hazard quotient (HQ) and hazard index for the adult and child populations, respectively. HQ values are predominantly below one for Ni, Pb, Cu, and Zn in the adult population, except Ni in Indian rice samples, Nigerian rice, and the mean of 30 samples, which exhibited elevated quotient values. Conversely, HQ values surpass one for the child population, except for the mean of Indian rice. This suggests a potential risk from Ni, Pb, and Cu in rice samples, emphasizing the association of lead and nickel with health issues affecting the nervous, respiratory, cardiovascular, hematopoietic, immune, endocrine, hepatic, renal, and reproductive systems.

Nickel, being non-destructible in the body, undergoes alterations in its chemical form, and its metabolism is closely tied to its binding ability to form ligands and subsequent transport throughout the body [40]. The toxicity of nickel-containing substances is linked to the bioavailability of the metal ion (Ni^{2+}) at systemic or local target sites [41]. The hazard index (HI) resulting from the intake of heavy metals exceeds one for both the adult and child populations, indicating a likelihood of chronic health effects in both groups. This aligns with a similar study on diverse rice samples imported into Nigeria, which also reported HQ and HI values exceeding one [1].

Table 4: Hazard Quotient (HQ) and Hazard Index (HI) of Heavy Metals for Adult Population through Consumption of Rice brands.

S/N	Mean Conc. Rice	HQ				
		Cu	Ni	Zn	Pb	HI=$\sum HQ$
1	Thailand Rice	0.365	0.605	0.437	0.70	2.1
2	Indian Rice	0.42	1.06	0.15	0.45	2.0
3	Nigerian Rice	0.717	1.28	0.403	0.96	3.4
	Mean of 30 Samples	0.665	1.18	0.397	0.783	3.0

Table 5: Hazard Quotient (HQ) and Hazard Index (HI) of Heavy Metals for Child Population through Consumption of Rice brands.

		HQ				
S/N	Mean Conc. Rice	Cu	Ni	Zn	Pb	HI=$\sum HQ$
1	Thailand Rice	1.525	2.55	1.83	3.02	8.925
2	Indian Rice	1.75	4.45	0.663	1.90	8.763
3	Nigerian Rice	3	5.35	1.686	4.04	14.076
	Mean of 30 Samples	2.8	4.95	1.663	3.28	12.693

3.4. Incremental Lifetime Cancer Risk

Table 6 presents the results of the incremental lifetime cancer risk (ILCR) for lead (Pb) and nickel (Ni), which are the carcinogenic metals among the analyzed elements. The ILCR values for Pb range from 1.20E-5 to 7.50E-6, while those for Ni range from 1.50E-4 to 3.20E-4 across all the rice samples. The ILCR of Pb falls within the recommended acceptable limit of (1.00E-04) to (1E-06). However, the ILCR of nickel exceeds the recommended limit of (1E - 4) to (1E- 6) [25] in all the rice samples. Consequently, there is no associated cancer risk within the USEPA cancer range of 1.0E-06 – 1.0E-04 for lead, but the ILCR for nickel suggests a potential cancer risk exceeding the recommended limits in all the analyzed rice samples.

Table 6: Incremental Lifetime Cancer Risk (ILCR) through Consumption of Rice.

S/N	Mean Conc. Rice	ILCR	
		Lead (Pb)	Nickel (Ni)
1	Thailand Rice	1.2E-05	1.5E-04
2	Indian Rice	7.5E-06	2.6E-04
3	Nigerian Rice	1.6E-05	3.1E-04
	Mean of 30 Samples	1.3E-05	3.2E-04

*(ILCR=10^{-6}) is the level of risk considered acceptable or inconsequential.
*(ILCR=10^{-4}) is considered serious and of great public health concern.

4. Conclusion

The current investigation focused on assessing both locally produced and imported rice samples across the southern part of Nigeria. Notably, 60% and 64% of lead and nickel concentrations, respectively, in both imported and locally produced rice samples exceeded the FAO/WHO permissible limits of 5.0 and 1.5 mg/kg for rice. However, concentrations of cadmium, zinc, copper, and chromium were found to be below the FAO/WHO recommended limits of 0.3, 60, 40, and 20 mg/kg, respectively. The Estimated Daily Intake (EDI) conducted in this study for a 70 kg body weight suggested that adverse health effects may be low. Nevertheless, individuals with lesser body weight and high consumption patterns are at a heightened risk of adverse health effects from heavy metals. The Hazard Quotients (HQ) of the studied metals were consistently above one in all the rice samples, indicating potential health risks and adverse effects associated with rice consumption. Continuous monitoring of rice samples is essential to mitigate exposure to toxic metals through rice ingestion in the region. Environmental pollution resulting from industrialization, along with the use of fertilizers, pesticides, herbicides, and insecticides, has been identified as contributing factors to the accumulation of heavy metals in plants and soil. Therefore, continuous, and thorough monitoring of both imported and locally produced rice is imperative to mitigate exposure to toxic metals through rice ingestion. This underscores the importance of stringent measures and regulatory frameworks to ensure the safety of rice products in the region.

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