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Levels of some trace metals (Pb, Cd and Ni) and their possible health risks from consumption of selected fish and shellfish from Nigerian markets

[1*]Ihedioha, J. N., [1]Amu, I. A., [1]Ekere, N. R. and [1]Okoye, C.O.B.

[1]*Department of Pure and Industrial Chemistry, University of Nigeria, Nsukka, Enugu State, Nigeria*

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Abstract

The study assessed the levels of Pb, Cd and Ni in fish and shellfish from Nigerian markets and also evaluated any possible risk in their consumption. The dried fish and shell fish samples were predigested with 1mL HNO_3 and later ashed at 450°C. The ash was dissolved in 5 mL HCl and the solution was analysed with an atomic absorption spectrophotometer. Results show that mean metal concentrations (mg/kg) in fish were in the following ranges: Pb (3.48-8.93), Cd (0.23-0.68), and Ni (1.57-4.36) while in shell fish, the ranges were: Pb (4.05-6.35), Cd(0.43-0.61) and Ni (2.37-3.69). There was evidence of considerable contamination as most of the samples had values exceeding the standard permissible limits set by some regulatory bodies. The estimated daily intake of lead for adult and children were appreciable. The total hazard indices (THI) were less than one showing low risk from consumption of the studied fish and shellfish species.

Introduction

Fish constitute important sources of protein, essential minerals and vitamins. Moreover, the polyunsaturated n-3 fatty acids in fatty fish species are biologically important. It contains low cholesterol, and its consumption could reduce the risk of coronary heart disease, decrease mild hypertension and prevent certain cardiac arrhythmias (Kris-Etherton *et al*., 2002).However, due to increasing chemical pollution of the aquatic ecosystem, the concentrations of toxic chemicals in fish has become of considerable interest because of the danger of bio-accumulation (Marcus *et al.*, 2013). Thus, seafood consumption has become an important dietary route of human exposure to a variety of chemical contaminants (Llobet *et al.*, 2003; Usero *et al*., 2003).

Cadmium and lead are known environmental toxicants. Human adults may suffer headache, abdominal pain, memory loss, kidney failure, male reproductive problems, and weakness, pain, or tingling in the extremities due to exposure to lead. In children, loss of appetite, abdominal pain, vomiting, weight loss, constipation, anemia, kidney failure, irritability, lethargy, learning disabilities, and behavior problems may occur (Pearce, 2007). Cadmium accumulated in the kidney and liver over a long time can lead to health problems (McLaughlin *et al*., 1999; WHO, 2004). Its presence in nature and in human food could cause serious damage to kidneys, lungs, bone and could lead to anaemia and, sometimes, hypertension (Afshar *et al*., 2000).

According to the Institute of Medicine (2003), high level of nickel is carcinogenic and could cause respiratory problem. Public health risks from dietary exposure to pollutants continue to be the subject of research and regulation. For over two decades, progressively higher values of toxic trace metals have been reported in fish from Nigeria. Kakulu (1987) reported mean values of Pb (1.92 mg/kg) and Cd (0.12 mg/kg) in fish and shellfish from the Niger Delta. Higher values of lead (2.02-7.29 µg/g) were reported in fish and shellfish from Lagos lagoon (Okoye, 1991) while much higher levels Pb (8.99±4.20 mg/kg) and Cd (0.45±0.28 mg/kg) were reported for dried fish from Nigerian market (Okoye,1994). Subsequent reports have shown much more contamination; Akan and Abiola (2008) published concentration of Pb (10.81-152.42 mg/kg) in fish from Lagos lagoon while Obodo (2004) had reported lead level (61.32 mg/kg) in catfish and (62.79 mg/kg) in Tilapia from River Anambra, in Southeast Nigeria. This study was aimed at determining the present levels of lead, cadmium and nickel in some sea foods in Nigeria and investigating any possible health risk that could arise from the consumption of the studied fish samples.

*Corresponding author.
Email: *janefrances.ihedioha@unn.edu.ng*

Table 1. Species of fish and shellfish and their habitats

Fish/Shellfish	Species		Habitat
Bony fish	i)	*Uraspis helvola*	An ocean species pelagic in surface waters as well as demersal
	ii)	*Ethmalosa fimbriata*	Coastal marine waters, estuaries, lagoons and rivers.
	iii)	*Trichinus haumella*	Muddy bottoms in coastal waters
	iv)	*Pseudotolithus elongates*	Found in brackish waters and estuaries
	v)	*Tilapia guineensis*	Muddy bottoms in coastal waters
Stockfish (Cod)	*Gadies species*		Cold or temperate northern seas at depth 130-360m. Many live near bottom
Cat fish	*Synodontis clarias*		Sandy area of water
Cray fish	i)	*Nematopaleamo hastatus*	Muddy bottom
	ii)	*Presiunika marita*	Sandy and muddy bottoms (300-700m depth) in estuaries
Crab	*Callinectas amnicola*		Lagoons and rivers
Periwinkle	*Tympanotonus fuscatus*		Mangrove areas

Materials and Methods

Sampling

Between March and July 2012, dried samples of seven species of fish and four of shellfish were purchased from Ogbete market in Enugu, Abakaliki market in Ebonyi State and Port Harcourt market in Rivers State, all in Southern Nigeria. Table 1 shows the different fish and shellfish species and their habitats.

Sample preparation and ashing:

The fish samples were dried to constant weight at 105°C and each specie was pulverised to make three composite samples using porcelain mortar and pestle. 2g of the composite samples were predigested overnight with 1 mL of conc. nitric acid and later charred on an electric hot plate in the fume chamber. The charred samples were ashed in a furnace at 450°C for 4 hrs. The resulting ash were dissolved in 5 mL 1:1 HCl. The solutions were transferred into 25 mL standard flasks and made up to mark with double distilled de-ionized water. Metal analyses were done with a 210 VGP Buck scientific atomic absorption spectrophotometer (USA) equipped with air-acetylene flame. 1mL HNO_3 and 5 mL 1:1 HCl in a 25 mL standard flask made up to mark with double distilled de-ionized water served as sample blanks.

Quality assurance procedures

Glass wares were soaked in detergent solution for two days and later in 5:2 mixtures of HCl and H_2O_2. They were later washed with deionized water. All reagents were of analytical grade (Merk, Germany) and double distilled de-ionized water was used to prepare solutions.

The LOD and LOQ were calculated statistically using AOAC (2002) method:

$$LOD = Xb + 3Sb; \quad LOQ = Xb + 10Sb$$

where *Xb* is the mean concentration of the blank and *Sb* is the standard deviation of the blank

Precision and accuracy of the analytical procedure were investigated by carrying out recovery experiment. Metal recovery experiment was investigated using standard addition method. 2 g triplicate samples of *T. fuscatus* were spiked with 25 mg/L mixed solution of the metals before charring and ashing. The sample solution was finally made up to 25 mL to obtain a spiking concentration of 1 mg/L of each metal. Results were compared to those of unspiked samples.

$$\% \text{ Recovery} = \frac{a-b}{c} \times 100$$

where a = concentration in the spiked sample, b = concentration in the un-spiked sample; c = amount of the metal ion added.

Health risk assessment

Possible health risks from consumption of the dried fish and shell fish samples were assessed using the model equation developed by Agency for Toxic Substances and Disease Registry for fish consumption (ATSDR, 2005).

Table 2. Mean percent recoveries, limit of detection and limit of quantification of trace metals from *T. fuscatus*

Trace metals	Spiking conc. (mg/L)	Conc. in unspiked sample (mg/L)	Conc in spiked sample (mg/L)	% Recovery	%Precision	LoD (mg/L)	LoQ (mg/L)
Pb	1.00	32.40	33.30	90	2.29		
	1.00	29.84	30.77	93		0.012	0.019
	1.00	34.88	35.77	89			
Mean ±SD				91± 2.08			
Cd	1.00	4.32	5.255	86	1.32		
	1.00	6.00	6.95	88		0.002	0.007
	1.00	3.84	4.77	86			
Mean ±SD				87± 1.15			
Ni	1.00	20.80	21.75	95	3.26		
	1.00	29.84	30.81	97		0.277	0.340
	1.00	27.60	28.51	91			
Mean ±SD				94± 3.06			

Table 3. Mean concentrations (mg/kg, dry weight) of heavy metals in various fish and shellfish from some Nigerian markets

Fish samples	Lead	Nickel	Cadmium
Uraspis helvola	5.30±0.71	2.27±0.75	0.44±0.21
Ethmalosa fimbriata	5.63±0.22	1.57±0.09	0.23±0.08
Trichinus haumella	7.90±0.66	3.01±0.44	0.53±0.09
Pseudotolithus elongates	4.44±0.39	4.36±0.31	0.62±0.23
Tilapia guineensis	3.48±0.61	2.06±1.02	0.31±0.15
Gadies species	7.54±0.30	3.02±0.59	0.54±0.15
Synodontis clarias	8.93±1.17	3.18±0.30	0.60±0.06
Total mean content	**6.17±1.99**	**2.78±0.91**	**0.47±0.15**
Shellfish			
Nematopaleamo hastatus	5.48±0.40	2.74±0.65	0.49±0.16
Presiunika marita	6.35±0.70	3.69±0.77	0.61±0.09
Callinectas amnicola	5.41±0.31	2.37±0.91	0.43±0.17
Tympanotonus fuscatus	4.05±0.32	3.29±0.54	0.59±0.14
Total mean content	**5.32±0.95**	**3.02±0.58**	**0.53±0.08**

$$D = \frac{(C \times IR \times AF \times CF \times EF)}{BW}$$

D = exposure dose (mg/kg/day); C = contaminant concentration (mg/kg); IR = intake of contaminated medium (fish)(mg/day);. AF = bioavailability factor, unit less (actual amount of an ingested substance which enters the blood stream and available to cause possible harm to a person is always assumed to be unity for screening purposes) (ATSDR, 2005). EF = Exposure factor (unit less) was taken to be 1 as fish intake rate is a daily average; CF = conversion factor (10^{-6} kg/mg); BW = body weight (kg) 70 kg for adult and 16kg for a child (ATSDR, 2005).

The annual per capita fish consumption in Nigeria is 7.52 kg/year which translated to 20, 603 mg/day for adult and 1/3 of the value (6, 868 mg/day) for child (IBGE, 2004; Adewuyi, 2010) while daily shellfish consumption was 6500mg/day for adults and 1/3 of the value (2167mg/day) for child (IBGE, 2004; William *et al.*, 2013)

The Hazard Quotient (HQ) and Total Hazard Index (THI) were estimated as follows:

$$HQ = D/RfD$$

$$THI = HQ_{\text{toxicant 1}} + HQ_{\text{toxicant 2}} + HQ_{\text{toxicant n}}$$

RfD is the oral reference dose in mg/kg/day and refers to the maximum amount of toxicant which does not translate to adverse effect. The values were Pb= 0.004, Ni = 0.02, Cd =0.001 (USEPA, 2000).

Results and Discussion

Good recoveries of the spiked metals in the range of 87.00 to 94.00 were obtained while LoD and LoQ were quite low (Table 2). The mean concentration of the heavy metals in fish and shellfish were presented in Table 3. The range of mean concentrations (mg/kg) for fish were: Pb (3.48-8.93), Ni (1.57-4.36), and Cd (0.21-0.68). For the shell fish, the ranges were Pb (4.05-6.35), Ni (2.37-3.69), and Cd (0.43-0.61). Lead concentrations were generally high and could be

Table 4. Estimated exposure Doses (D) (x 10^{-3}) mg/kg/day; Hazard Quotient (HQ) and Total Hazard Index (THI) of fish consumption in Nigeria

Fish species	Exposures	Pb	Ni	Cd	THI
Uraspis	D Child	2.13	1.26	0.25	
helvola	HQ child	0.53	0.06	0.25	0.84
	D Adult	1.46	0.86	0.17	
	HQ Adult	0.37	0.04	0.17	0.58
Ethmalosa	D Child	2.42	0.67	0.10	
fimbriata	HQ child	0.61	0.03	0.10	0.74
	D Adult	1.67	0.46	0.07	
	HQ Adult	0.42	0.02	0.07	0.51
Trichinus	D Child	3.39	1.29	0.23	
haumella	HQ child	0.85	0.06	0.23	1.14
	D Adult	2.33	0.89	0.16	
	HQ Adult	0.58	0.04	0.16	0.78
Pseudotolith	D Child	1.91	1.87	0.29	
us elongates	HQ child	0.48	0.09	0.29	0.86
	D Adult	1.31	1.28	0.20	
	HQ Adult	0.33	0.06	0.20	0.59
Tilapia	D Child	1.49	0.88	0.13	
guineensis	HQ child	0.37	0.04	0.13	0.54
	D Adult	1.02	0.61	0.09	
	HQ Adult	0.26	0.03	0.09	0.38
Gadies	D Child	3.24	1.30	0.23	
species	HQ child	0 .81	0.07	0.23	1.11
	D Adult	2.22	0.89	0.16	
	HQ Adult	0.56	0.04	0.16	0.76
Synodontis	D Child	3.83	1.37	0.26	
clarias	HQ child	0.96	0.07	0.26	1.29
	D Adult	2.63	0.94	0.18	
	HQ Adult	0.68	0.05	0.18	0.91
Mean± SD	Child				0.93±0.26
	Adult				0.64± 0.18

favourably compared to the values reported between 1980's and 1990's by Kakulu *et al*. (1987) and Okoye (1991, 1994). Cat fish *(Synodontis clarias)* has the highest concentration of lead and *Tilipia guineenis*, a bony fish, the least. Among the shell fish, *P. martia* has the highest concentration of lead and cadmium. This is a bottom feeder and could feed on sediment component with high load of pollutant. The lead concentrations in all the samples exceeded the 0.05 mg/kg stipulated by FAO (1983) while 50% of the samples exceeded 0.5 mg/kg for cadmium (FAO, 1983) for fish. All the samples exceeded standard permissible limit of 0.5 mg/kg for nickel (WHO, 1991). High lead and cadmium concentrations reported in this study indicate considerable contamination of the Nigerian aquatic environment. This is no surprise in view of the high metal content reported in various studies in the country. Ugwu *et al.* (2011) reported enrichment of 185% in Pb, 53% in Cd and 73% in Ni in fermented cassava dried along roadside as compared to those dried far from the road and attributed the enrichment to vehicular emissions and dust particles raised by motor transport. Ihedioha and Okoye (2013) reported that 93% of muscle, 100% of kidney and intestine and 97% of tripe of cow meat contained Ni concentrations exceeding the guideline value. Ihedioha and Okoye (2012) also reported that 82% of meat samples exceeded the set limit of Cd in meat. Okoye *et al*. (2011) showed high lead (9.70-147.07 mg/kg) and cadmium (1.78-15.32 mg/kg) in local and exotic poultry meat.

The lead and cadmium values reported in this study are higher than (nd-3.31 mg/kg) for Pb and (0.01-0.1 mg/kg) for Cd reported in fish from Italy (Storelli, 2008). Also lower Pb (0.045-1.43 mg/kg) and Cd (0.01-0.145 mg/kg) levels were reported by Han *et al*. (1998) for fish in Taiwan. However, this study showed lower values in comparison to 1.10-2.238 mg/kg reported for shellfish in Taiwan (Chien *et al*., 2002)

Considering the intakes of these metals from consumption of fish by a 70 kg adult and a 16 kg child, using information on daily intake of fish and shell fish obtained from Adewuyi (2010) and William *et al.* (2013) respectively, the intakes for fish (µg/day) obtained were: Pb (126), Cd(10), and Ni (59) for a 70 kg adult while for a 16 kg child, they were: Pb (42), Cd (3), and Ni (20). The intakes for shellfish (µg/day) were Pb (35), Cd (3), and Ni (25) for a 70 kg adult while Pb (12), Cd (1), and Ni (9) were for a 16 kg child.

Joint FAO/WHO Expert Committee on Food Additives (JECFA) stipulated a Provisional Tolerable Daily Intake (PTDI) of 3.57 µg/kg body weight for lead (WHO, 1993) and 0.83 µg/kg body weight for cadmium (JECFA, 2010). These translate to 250 µg/day and 57 µg/day lead for a 70 kg adult and 16 kg child respectively while for cadmium, they are 58

Table 5. Estimated exposure Doses (D) (x 10^{-4}) mg/kg/day; Hazard Quotient (HQ) and Total HazardIndex (THI) of shellfish consumption in Nigerian

Shellfish species	Exposures	Pb	Ni	Cd	THI
Nematopaleamo hastatus	D Child	7.42	3.71	0.66	
	HQ child	0.19	0.02	0.07	0.28
	D Adult	5.09	2.54	0.46	
	HQ Adult	0.13	0.01	0.05	0.19
Presiunika marita	D Child	8.60	5.38	0.83	
	HQ child	0.22	0.03	0.08	0.33
	D Adult	5.90	3.69	0.59	
	HQ Adult	0.15	0.02	0.06	0.23
Callinectas amnicola	D Child	7.33	7.33	0.55	
	HQ child	0.18	0.04	0.06	0.28
	D Adult	5.02	5.02	0.38	
	HQ Adult	0.13	0.03	0.04	0.20
Tympanotonus fuscatus	D Child	5.49	4.46	0.80	
	HQ child	0.14	0.02	0.08	0.24
	D Adult	3.76	3.06	0.55	
	HQ Adult	0.09	0.02	0.06	0.17
Mean± SD	Child				0.28±0.04
	Adult				0.20±0.03

µg/day for a 70 kg adult and 13 µg/day for a 16 kg child. The lead intakes obtained in this study for an adult were: fish (50%), shellfish (14%) while for Cd, fish (17%), shellfish (5%) of PTDI. For a child, the intakes were: fish (74%), shellfish (21%) for lead and fish (23%), shellfish (8%) for cadmium PTDI. JECFA has not evaluated nickel but WHO (1997) has given a Tolerable Daily Intake (TDI) of 5 µg/kg body weight. This translates to 350 µg/day for a 70kg adult and 80 µg/day for a 16 kg child. The Ni intake were fish (17%), shellfish (7%) TDI for adult while for child, it was: fish (25%) and shellfish (11%) TDI. These show appreciable intakes by children. Children are especially vulnerable to the acute and chronic effects of ingested chemical compounds since they consume more food per kilogram of body weight than adults, they are more exposed to chemical hazards in food than are adults (ENHIS, 2007).

Risk assessment is the process that evaluates the potential health effects from doses to humans of one contaminant received through one or more exposure pathways. The potential health hazard of the toxicants was interpreted based on the values of HQ and THI. Values less than one(HQ or THI < 1) mean no risk and the greater the values above one, the greater is the level of risk of the toxicant manifesting long term health hazard effects (Lemy, 1996, Lai *et al.*, 2010). The exposure dose (D), HQ and THI obtained are presented in Tables 4 and 5 for fish and shellfish respectively. Ingestion of daily dose of fish by adults leads to a potential accumulation of Pb ranging from 1.02×10^{-3} mg/kg *(T. guinenesis)* to 2.33×10^{-3} mg/kg *(S. clarias)* while for a child, it ranges from 1.49×10^{-3} mg/kg to 3.83×10^{-3} mg/kg (the same species). The methodology for estimation of hazard quotient (HQ) although does not provide a quantitative estimate on the probability of an exposed population experiencing a reverse health effect, but offers an indication of the risk level due to dietary exposure to the pollutant.

The HQ calculated for child and adult consumption of the fish and shellfish species studied indicates low risk (HQ<1). Also the mean total hazard indices (THI) of all the assayed heavy metals were lower than one (THI<1) for fish and shell fish. This shows low risk from consumption of the species. The HQ and THI calculated for child consumption of fish and shellfish were expectedly higher than those of adult. The relative contributions of the toxicants to THI were in the order Pb > Cd > Ni in all the fish and shellfish species under study.

Conclusion

The levels of Pb, Ni and Cd in the fish samples were appreciable as compared to limits set by FAO. These indicate considerable contamination by these metals in the Nigerian environment. The daily intakes of Pb, Cd and Ni from fish were appreciable compared to PTDI. The HI for the metals were less than 1 showing low risk arising from consumption of studied fish and shellfish species. The study does not suggest pollution yet. However, it is recommended that intake of these metals from other sources like meat and dairy products, grains and vegetables etc should be assessed to ascertain any possible health risk to the consumers.

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