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METAL CONTENTS IN FISH AND CRUSTACEANS FROM BRACKISH, FRESHWATER AND MARINE SYSTEMS IN SOUTH-WESTERN NIGERIA

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ABSTRACT

Present study investigated trace element concentrations in either muscles or whole samples of 37 finfish and crustaceans harvested with gillnet from fresh (Olomore), marine (Eti osa) and brackish (Lekki Lagoon) waters of Ogun and Lagos states, Nigeria between June and August 2013. Trace elements were determined using ICP-MS after digestion with a microwave autoclave system. The results revealed that the general trend of trace elements in the sampled finfish and crustaceans was marine > brackish > freshwater. If all the samples are considered, 16.2% and 13.5% exceeded the 20 μg g^{-1} and 30 μg g^{-1} limits for Cu and Zn, respectively, as set by the European Union, while 71%, 50%, 79% of marine, fresh and brackish samples, respectively, exceeded the limit of 48 mg Fe per 60 kg person as set by FAO/WHO. In addition, samples exceeded the acceptable limit set by WHO/FAO, thus adequate efforts should be placed on the monitoring of effluents that are being discharged into open water bodies in Nigeria.

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INTRODUCTION

Pristine freshwater and marine ecosystems are important for the survival of organisms living in them. However, various human activities both on land and from the sea have led to pollution of these pristine ecosystems (Mohammed et al., 2012; Rahman et al., 2012; SantÁnna et al., 2012). Heavy metals are one of the few but persistent pollutants known to occur both naturally and from anthropogenic sources (Nubi et al., 2008; Rahman et al., 2012). Pollution of water systems by heavy metals might present a potential risk factor due to the ability of aquatic organisms to assimilate, bioaccumulate and/or biomagnify contaminants from the surrounding waters and sediment (Sant´Ánna et al., 2012; Zeng et al., 2012). In the natural habitat, fishes are continually exposed to varying concentrations of inorganic and organic pollutants that threaten their survival (Olaifa et al., 2004). Fish and crustaceans are highly valuable and appreciated food items, and their capture and culture represent important sources of livelihoods for millions of people, particularly to those

residents along the world's freshwater and marine coasts. Previous studies on metal pollution in Lagos State had focused mainly on the Lagos Lagoon due to high-population and high anthropogenic activities (Don-Pedro et al., 2004; Williams and Edobor-Osoh, 2013). Smaller fishing communities were often neglected. Aside from the Lagos Lagoon, huge fishing activities are on-going on the Atlantic Ocean which involves the local communities around the Lekki Lagoon of Lagos State. Studies on trace element profiling of aquatic organisms such as fishes, shrimps or crabs from fresh, marine and brackish water systems in Nigeria are very scarce or not readily available. Studies on heavy metals profile of edible marine species are very scarce and are often restricted to the Niger-Delta zone (Eboh et al., 2006) or the inland water bodies (Adegbola et al., 2012; Oyakhilome et al., 2013). George et al. (2013) investigated heavy metal variations in two coastal fish species during wet and dry seasons in Nigeria, while Olojo et. al. (2012) reported heavy metal concentrations in relation to the seasonal changes in Oreochromis niloticus and Chrysichthys nigrodigitatus from the Lagos Lagoon. The present study aimed to assess the concentration of trace elements in commonly consumed freshwater, marine and brackish fish species in Nigerian waters.

MATERIAL AND METHODS

Study sites

The sampling sites include the Lekki Lagoon. The Lagoon lies between longitude 4° 14 4 and latitude 6° 25 4 N. The Lagoon is 6.4 m deep with a surface area of 247 km^2. Fish, crabs and shrimps from the Lekki Lagoon and marine ecosystems (Atlantic Ocean) were obtained through the efforts of local fishermen at two different locations at Okunraye village (about 30 km east of Lekki Free Trade Zone, Lagos State) and from the sea (Atlantic Ocean) overnight (Fig. 1). The freshwater fish samples were bought from fresh fish market at Olomore in Abeokuta, the Ogun State capital. The fish samples were identified by few members of the team and staff of the National Institute for Oceanography and Marine Research, Lagos State, Nigeria.

Collection of fish and crustaceans, and sample preparation

The analyzed aquatic species are presented in Table 1. The samples were collected between June and August 2013. The collected samples (17 marine, 6 fresh and 14 brackish samples) were frozen in prewashed polyethylene bags and brought to the laboratory in ice chests. In the laboratory, the samples were firstly allowed to thaw, then were washed with distilled water and oven-dried to obtain uniform weight

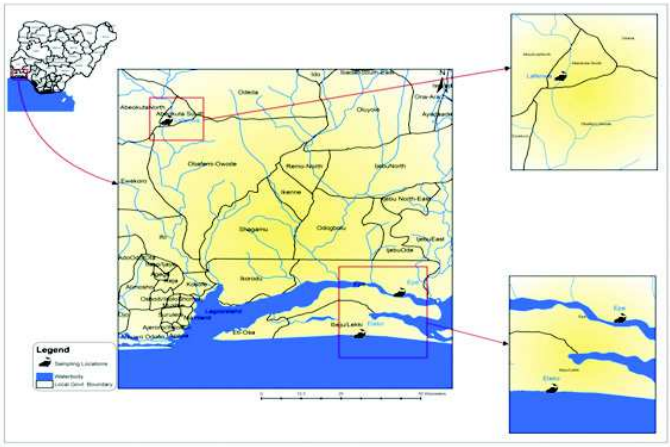

Fig 1. Map of the sampling locations

at 105°C. During the drying process, the samples were dissected to allow uniform heat distribution. After drying, the portion of fish muscle and whole samples of the crabs and shrimps were milled using pestle and mortar, and transferred into zip-lock polyethylene bags prior to chemical analysis.

Chemical analyses

Reagents and chemicals

Milli-Q water (18.2 MΩ cm) was obtained by Millipore equipment (Bedford, USA) and used for analysis to prevent contamination. Nitric acid (65%) and mixed standard of trace elements were obtained from Merck (Darmstadt, Germany), while rare earth elements mix standard and Sn were bought from CPI, USA. Other metals (Na, Ca, Sb, Hg, K, P and Mg) were purchased from Roth, Germany. Prior to use, nitric acid was subjected to further purification through distillation using quartz sub-boiling distillation apparatus.

Sample digestion

Samples were digested using microwave autoclave system. For digestion, about 250 mg of the dried fish, crab or shrimp samples or the reference materials (DOLT-3 (Dogfish liver) and RM 8414 (bovine muscle powder) purchased from National Research Council, Canada) were transferred into 12 mL quartz vessels and the digestion was performed after addition of 5 mL of nitric acid. The digestion programme and analysis were performed according to Olujimi et al. (2015).

Metal determination

Measurement of metals was performed using 7700x ICP-MS (Agilent, USA). The accuracy of calibration was checked with SRM 1643a (trace elements in water) purchased from NIST, Gaithersburg, USA. The result accuracy was checked

Table 1. A list of finfish and crustaceans investigated for trace, toxic and rare earth elements

	Type of organism	Scientific name	Local name	Common name
B1	Fish	*Sphyraena afra*	Esun	Barracuda
B2	Fish	*Chrysichthys nigrodigitatus*	Chrysichthys	Silver catfish
B3	Fish	*Ethmalosa fimbriata*	Efolo	Bonga shad
B4	Fish	*Caranx senegallus*	Kunkun	Senegal jack
B5	Fish	*Elops lacerta*	Sugbon	West African ladyfish
B6	Fish	*Caranx hippos*	Kusukusu	Crevalle jack
B7	Fish	*Galeoides decadactylus*	Popo	African threadfin
B8	Fish	*Cynoglossus sp*	Iya abo	Tonguesole
B9	Fish	*Tilapia zilli*	Epiya	Tilapia
B10	Fish	*Cynoglossus lagoensis*	Abo	Nigerian tonguesole
B11	Fish	*Pteroscion peli*	Uke	
B12	Fish	*Strongylura senegalensis*	Lakoro	Senegal needle fish
B13	Crustacean	*Parapenaeopsis atlantica*	Ede	Guinea shrimp
B14	Crustacean	*Callinectes pallidus*	Akan dudu	Gladiator swimcrab
F1	Fish	*Clarias gariepinus*	Aro	Catfish
F2	Fish	*Alestes macrolepidotus*		Silverside
F3	Fish	*Mormyrus rume*		Trunkfish
F4	Fish	*Pellonula afzeliusi*	Sawa	Freshwater sardine
F5	Fish	*Chrysichthys nigrodigitatus*	Obokun	Silver catfish
F6	Fish	*Oreochromis niloticus*	Epiya	Tilapia
M1	Fish	*Mugi caphalus*	Atoko	Mugil
M1	Fish	*Sardinella marderensis*	Sawa	Sardine
M3	Fish	*Synatura cadenati*	Abo Olokun	Giunea sole
M4	Fish	*Scomberomous tritor*	Kote	Skip jack
M5	Fish	*Pomadasys jubelini*	Ikekere	Sompat grunt
M6	Fish	*Chloroscombus chrysurus*	Sayaganga	Atlantic bumper
M7	Fish	*Albula vulpes*	Omo eja (sugbon)	Bonefish
M8	Fish	*Arius latisculatus*	Kugbe	Rough-head sea catfish
M9	Fish	*Ilisha Africana*	Kafonran	West African Ilisha
M10	Fish	*Pseudotolithus senegakensis*	Apo	Cassava croaker
M11	Fish	*Pentanemus quinquarius*	Onirungbon	Royal threadfin
M12	Fish	*Cynoglossus brownie*	Abo olokunII	Tonguesole
M13	Fish	*Drepane Africana*	Akaraba	African sicklefish
M14	Crustacean	*Penaeus notialis*	Ede olokun	Pink shrimp
M15	Crustacean	*Penzeus sp*	Tanshoko	Shrimp
M16	Crustacean	*Panulirus regius*	Ede alaran	Royal spiny lobster
M17	Crustacean	*Portunus valids*	Akan pupa	Smooth swim crab

B = Brackish water samples; F = Fresh water samples; M = Marine water samples

Table 2. Elemental concentrations of certified reference materials SRM 1640a, RM 8414 and DOLT-3 (Mean ± Standard deviation)

SRM 1640a				Bovine RM 8414				DOLT-3			
Metal	Certified	Measured	Accuracy (%)	Metal	Certified	Measure	Accuracy (%)	Metals	Certified	Measured	Accuracy %
Al	52.6 ± 1.8	41.0 ± 0.1	-22.05	Al	1.7 ± 1.4	1.50 ± 0.04	-11.8	As	10.2 ± 0.5	9.33 ± 0.24	-8.53
Sb	5.06 ± 0.05	5.01± 0.05	-0.99	As	0.009 ± 0.003	0.008 ± 0.003	-22.2	Cd	19.4 ± 0.6	19.5 ± 0.4	0.513
As	8.01 ± 0.07	7.62 ± 0.15	-4.87	Cd	0.013 ± 0.011	0.011 ± 0.006	-15.4	Cu	31.2 ± 1.0	34.1 ± 0.8	5.87
Ba	150.6 ± 0.74	151 ± 0.06	0.13	Cr	0.07 ± 0.04	0.08 ± 0.02	12.5	Fe	1484 ± 57	1466 ± 32	-1.21
Be	3.00 ± 0.03	2.01 ± 0.04	-33.00	Cu	2.84 ± 0.45	2.76 ± 0.06	-2.82	Pb	0.32 ± 0.05	0.29 ± 0.01	-9.38
B	300 ± 3	279 ± 1	-7.00	Fe	71.2 ± 9.2	71.3 ± 1.1	0.14	Hg	3.37 ± 0.14	3.39 ± 0.3	0.59
Cd	3.96 ± 0.07	3.96 ± 0.06	-0.25	Pb	0.38 ± 0.24	0.33 ± 0.02	-13.2	Ni	2.72 ± 0.35	2.99 ± 0.11	9.03
Cr	40.2 ± 0.3	40.3 ± 0.02	0.25	Mn	0.37 ± 0.09	0.32 ± 0.01	-13.5	Se	7.06 ± 0.48	7.15 ± 0.25	1.26
Co	20.1 ± 0.2	18.7± 0.03	-6.97	Mo	0.08 ± 0.06	0.06 ± 0.01	-25	Ag	1.20 ± 0.07	1.5 ± 0.03	20
Cu	85.1 ± 0.5	85 ± 1	-0.12	Ni	0.05 ± 0.04	0.02 ± 0.01	-60	Zn	86.6 ± 2.4	92.1 ± 0.6	5.97
Fe	36.5 ± 1.7	35.8 ± 0.1	-1.92	Rb	28.7 ± 3.5	27.8 ± 0.2	-3.14				
Pb	12 .0 ± 0.0	11.5 ± 0.1	-4.17	Se	0.076 ± 0.01	0.063 ± 0.01	-17.1				
Mn	40.1 ± 0.4	40.3 ± 0.1	0.50	Sr	0.052 ± 0.015	0.08 ± 0.01	35				
Mo	45.2 ± 0.6	44.5 ± 0.1	-1.55	Zn	142 ± 14	137 ± 3	-3.52				
Ni	25.1 ± 0.1	25.1 ± 0.1	0.00								
Se	19.9 ± 0.1	17.4 ± 0.2	-12.56								
Ag	8.02 ± 0.04	10.3 ± 0.1	22.14								
Sr	125 ± 1	133 ± 00	6.02								
Th	1.61 ± 0.02	1.60 ± 0.06	-0.62								
U	25.1 ± 0.3	22.1 ± 0.1	-11.95								
V	14.9 ± 0.2	14.1 ± 0.1	-5.37								
Zn	55.2 ± 0.3	53.8 ± 0.1	-2.54								

Table 3. Concentration of trace elements in fish and shell fish samples (Mean ± Standard deviation)

B1	0.04 ± 0.01	7.8 ± 0.6	0.12 ± 0.01	1.0 ± 0.1	0.005 ± 0.001	<3.16⁺	52.2 ± 0.5	1.38 ± 0.04	18.8 ± 1.2
B2	0.02	49 ± 4	0.16 ± 0.01	2.5 ± 0.2	0.041 ±0.0065	174 ± 10	90.9 ± 11.5	0.93 ± 0.04	78.6 ± 5.2
B3	0.05 ± 0.02	163 ± 18	0.29 ± 0.12	39.5 ± 0.6	0.030 ± 0.009	236 ± 19	338 ± 7.2	2.656 ± 0.12	249 ± 13
B4	0.02	19.4 ± 0.6	0.81 ± 0.38	5.17 ± 0.03	0.012 ± 0.005	19.2 ± 1.2	255 ± 6	3.1 ± 0.2	66.8 ± 26.8
B5	0.02	26.4 ± 4.3	0.20 ± 0.01	6.37 ± 0.87	0.019 ± 0.002	58.9 ± 11.4	878 ± 114	1.95 ± 0.16	63.9 ± 8.6
B6	0.03	44.8 ± 2.8	0.36 ± 0.01	10.3 ± 1.5	0.009 ± 0.002	101 ± 6	106 ± 12	1.18 ± 0.02	69.5 ± 2.4
B7	0.02 ± 0.01	25.4 ± 29.9	0.26 ± 0.24	7.41 ± 0.72	0.042 ± 0.005	128 ± 1	473 ± 120	1.92 ± 0.32	46 ± 7
B8	0.02 ± 0.01	23.5 ± 2.1	0.16 ± 0.01	25.3 ±3.2	0.016 ± 0.001	36.3 ± 5.8	138 ± 18	1.34 ± 0.06	34.7 ± 4.2
B9	0.03 ± 0.01	29.5 ± 2.2	0.14 ± 0.01	7.61 ± 0.37	0.016 ± 0.002	192 ± 11	132 ± 23	1.14 ± 0.05	57.5 ± 9.2
B10	0.11 ± 0.01	40.6 ± 7.3	1.07 ± 0.03	41.6 ± 4.4	0.026 ± 0.004	164 ± 6	1292 ± 63	1.48 ± 0.07	89.2 ± 40.2
B11	0.02	5.6 ± 1.8	0.60 ±0.0 4	1.43 ± 0.19	0.025 ± 0.002	135 ± 7	56.8 ± 13.7	0.68 ± 0.03	21.3 ± 1.2
B12	0.02	6.65 ± 0.03	0.12 ± 0.01	5.28 ± 0.51	0.006 ± 0.001	<3.16⁺	359 ± 41	1.14 ± 0.04	29.1 ± 2.1
B13	0.30 ±0.02	80.9 ± 10.2	0.96 ± 0.38	500 ± 8	0.235 ± 0.015	312 ± 32	152 ± 22	230 ± 19	159 ± 32
B14	0.20 ± 0.02	64.4 ± 5.5	0.94 ± 0.36	268 ± 5	0.415 ± 0.023	507 ± 14	1158 ± 473	27.6 ± 0.8	125 ± 9
F1	0.03	5.35 ± 0.59	0.44 ± 0.16	1.94 ± 0.03	0.004 ± 0.001	<3.16⁺	237 ± 260	2.47 ± 0.04	59 .0 ± 0.5
F2	0.02	8.98 ± 0.7	0.09 ±0.01	0.58 ± 0.07	0.011 ± 0.001	<3.16⁺	25.6 ± 15	1.53 ± 0.07	25.9 ± 1.2
F3	0.011	82.4 ± 4.7	0.42 ± 0.01	2.26 ± 0.14	0.004 ± 0.001	322 ± 7	62.8 ± 14.6	1.75 ± 0.12	74.3 ± 3.4
F4	0.01 ± 0.01	4.99 ± 0.86	0.06 ± 0.01	0.80 ± 0.37	0.006 ± 0.001	<3.16⁺	6.14 ± 5.16	1.11 ± 0.00	22.2 ± 4.7
F5	0.03 ± 0.01	334 ± 51	0.15 ± 0.01	8.65 ± 1.11	0.012 ± 0.001	464 ± 63	648 ± 257	1.94 ± 0.06	44.8 ± 62.9
F6	0.03 ± 0.01	128 ± 71	0.06 ± 0.01	1.94 ± 0.76	0.005 ± 0.001	133 ± 13	337 ± 148	1.30 ± 0.09	93.5 ± 43.7
M1	0.012	38.1 ± 22.7	22.2 ± 0.5	0.41 ± 0.08	0.047 ± 0.026	<3.16⁺	66.8 ± 33.7	1.64 ± 0.23	70.2 ± 19.8
M2	0.050	13.7 ± 3.7	18.1 ±0.5	0.43 ± 0.05	0.017 ± 0.002	2.56 ± 1.79	13.2 ± 6.59	2.85 ± 0.17	43.2 ± 2.6
M3	0.04 ± 0.01	79.9 ± 7.4	6.93 ± 0.82	1.21 ± 0.23	0.017 ± 0.002	8.67 ± 1.59	241 ± 49	1.46 ± 0.04	165 ± 65
M4	0.01	13.3 ± 1.8	6.90 ± 0.12	0.19 ± 0.03	0.011 ± 0.004	3.36 ± 0.44	14.1 ± 11.7	2.56 ± 0.09	93.2 ± 8.5
M5	0.10 ± 0.01	44.6 ± 15	6.95 ± 0.33	2.56 ± 1.31	0.012 ± 0.002	37 ± 7	152 ± 61	25.3 ± 1.9	98.9 ± 26.7
M6	0.07	30.9 ± 6.2	7.24 ± 0.12	1.59 ± 0.07	0.039 ± 0.009	15.4 ± 7.6	161 ± 68	2.41 ± 0.02	84.4 ± 30.6
M7	0.01 ± 0.01	7.24 ± 1.81	18.2 ± 0.5	0.63 ± 0.03	0.021 ± 0.001	<3.16⁺	<2.15⁺	0.79 ± 0.04	25.8 ± 0.7
M8	0.09 ± 0.01	175 ± 36	19.6 ± 0.4	8.95 ± 2.73	0.012 ± 0.002	118 ± 7	289 ± 37	2.54 ± 0.13	146 ± 16
M9	0.02	24.1 ± 0.9	8.78 ± 0.37	0.37 ± 0.04	0.027 ± 0.002	8.77 ± 8.15	146 ± 126	1.31 ± 0.08	40 ± 4
M10	0.01	4.15 ± 0.31	23 ± 2	0.07 ± 0.02	0.004 ± 0.001	<3.16⁺	<2.15⁺	0.94 ± 0.06	14.8 ± 0.6
M11	0.01	138 ± 1	9.36 ± 0.20	2.86 ± 0.13	0.019 ± 0.002	39.1 ± 2.7	214 ± 51	1.39 ± 0.06	126 ± 20
M12	0.04 ± 0.01	291 ± 70	36.2± 1.40	2.01 ± 0.12	0.025 ± 0.004	80.9 ± 28.9	1303 ± 1295	1.47 ± 0.03	358 ± 170
M13	0.02 ± 0.01	15 ± 4	3.85 ± 0.12	0.25 ± 0.01	0.004 ± 0.001	<3.16⁺	28.9 ± 17.1	1.09 ± 0.07	20.9 ± 2.4
M14	0.43 ± 0.04	315 ± 83	34.9 ± 1.16	4.25 ± 0.18	0.091 ± 0.009	207 ± 15	400 ± 64	65.9 ± 2.6	222 ± 45
M15	1.25 ± 0.11	185 ± 25	35.9 ± 1.87	5.01 ± 0.12	0.855 ± 0.015	164 ± 13	348 ± 156	99.2 ± 4.6	143 ± 25
M16	0.28 ± 0.03	186 ± 60	95.3 ± 2.53	2.63 ± 0.01	0.311 ± 0.025	107 ± 0.04	338 ± 140	37.1 ± 2.4	190 ± 111
M17	0.97 ± 0.05	389 ± 14	64.0 ± 15	5.06 ± 0.13	0.171 ± 0.004	206 ± 14	887 ± 300	65.4 ± 2.6	301 ± 15

Table 3. Concentration of trace elements in fish and shell fish samples (Mean ± Standard deviation) continued

B1	0.251	3.43 ± 0.39	0.35 ± 0.02	0.57 ± 0.17	0.68 ± 0.14	1.01 ± 0.03	18.1 ± 1.2	24.5 ± 0.8
B2	0.170±0.001	9.38 ± 1.58	0.10 ± 0.01	0.58 ± 0.05	0.06 ± 0.01	1.74 ± 0.02	92.8± 4.1	34 ± 2
B3	0.090 ±0.003	97.6 ± 1.2	0.49 ± 0.03	0.83 ± 0.14	0.23 ± 0.01	.81 ± 0.03	816 ± 22	90 ± 7
B4	0.477 ± 0.032	12.9 ± 0.3	0.08 ± 0.01	4.41 ± 0.36	0.06 ± 0.01	1.38 ± 0.05	29.3 ± 2.4	49.8 ± 3.9
B5	0.239 ± 0.028	25.6 ± 2.6	0.40 ± 0.16	0.97 ± 0.23	0.13 ± 0.02	0.92 ± 0.06	85.9 ± 22.5	73.2 ± 5.5
B6	0.133. ± 0.013	49.5 ± 6.2	0.08 ± 0.01	0.79 ± 0.01	0.12 ± 0.01	1.03 ± 0.07	639 ± 69	102 ± 1
B7	0.191 ± 0.021	22 ± 1	0.018 ± 0.01	0.68 ± 0.16	0.05± 0.01	1.32 ± 0.05	168 ± 19	4.99 ± 2.5
B8	0.210 ± 0.008	73.9 ± 4.4	0.08 ± 0.01	0.35 ± 0.03	0.09 ± 0.02	1.17 ± 0.01	53 ± 19	60.7 ± 4.5
B9	0.070 ± 0.001	14.3 ± 0.6	0.07 ± 0.01	0.45 ± 0.06	0.07 ± 0.01	0.78 ± 0.04	240 ± 8	45.6 ± 2.2
B10	0.109 ± 0.012	83.8 ± 8.6	0.27 ± 0.04	0.84 ± 0.04	0.22 ± 0.03	1.48 ± 0.08	308 ± 97	49.2 ± 3.7
B11	0.193 ± 0.007	13 ± 2	0.03 ± 0.01	0.32 ± 0.03	0.02 ± 0.01	1.15 ± 0.04	70 ± 9	43.1 ± 1.8
B12	0.548 ± 0.024	5 ± 1	0.07 ± 0.01	0.39 ± 0.02	0.04	0.91 ± 0.06	43 ± 3	38.3 ± 2.5
B13	0.103 ± 0.009	12.9 ± 2.8	0.2 ± 0.01	0.70 ± 0.17	0.05 ± 0.07	0.94 ±0.02	152 ± 64	257 ± 108
B14	0.139 ± 0.008	569 ± 15	0.15 ± 0.01	0.82 ± 0.04	0.28 ± 0.05	0.96 ± 0.05	99 ± 6	93 ± 4
F1	0.034 ± 0.005	1.42 ± 0.07	0.09	0.79 ± 0.07	0.04 ± 0.01	0.61 ± 0.03	65.7 ± 0.6	36.8 ± 0.4
F2	0.252 ± 0.008	3.21 ± 0.64	0.01	0.30 ± 0.09	0.07 ± 0.01	2.02 ± 0.08	22.7 ± 2.3	27.8 ± 0.7
F3	0.174 ± 0.010	9.7 ± 0.9	0.03	2.54 ± 0.38	0.20 ± 0.01	2.11 ± 0.04	91.9 ± 5.8	26 ± 1
F4	0.070 ± 0.001	4.75 ± 3.05	0.06 ± 0.01	0.98 ± 0.12	0.11 ± 0.03	1.29 ± 0.05	25 ± 10	30 ± 1
F5	0.180 ± 0.015	135 ± 21	0.10	6.08 ± 0.42	0.11 ± 0.01	2.49 ± 0.07	927 ± 133	26.1 ± 0.7
F6	0.078 ± 0.016	3.03 ± 0.25	0.12 ± 0.01	0.78 ± 0.07	0.05 ± 0.01	1.23 ± 0.04	525 ± 65	30.9 ± 1.2
M1	0.301 ± 0.021	5.29 ± 3.14	0.24± 0.01	0.28 ± 0.10	0.04 ± 0.01	1.88 ± 0.03	124 ± 69	27.6 ± 1.6
M2	0.102 ± 0.006	3.56 ± 0.54	0.08 ± 0.01	0.30 ± 0.01	0.24 ± 0.06	3.78 ± 0.08	205 ± 33	46.2 ± 0.6
M3	0.130 ± 0.001	7.86 ± 2.33	0.04	0.73 ± 0.11	0.20 ± 0.03	1.25 ± 0.004	237 ± 34	27.2 ± 0.79
M4	0.101 ± 0.011	0.94 ± 0.02	0.04	0.12 ± 0.01	0.03 ± 0.01	2.36 ± 0.04	47.8 ± 3.9	29.6 ± 0.5
M5	0.213 ± 0.006	5.82 ± 1.32	0.06 ± 0.01	0.67 ± 0.20	0.43 ± 0.02	1.78 ± 0.05	176 ± 35	27.9 ± 0.9
M6	0.170 ± 0.003	1.88 ± 0.17	0.22 ± 0.01	0.73 ± 0.11	0.45 ± 0.07	2.59 ± 0.06	152 ± 35	55.1 ± 1.6
M7	0.323 ± 0.009	1.55 ± 0.02	0.06 ± 0.01	0.15 ± 0.02	0.02 ± 0.01	2.79 ± 0.05	21.8 ± 1.8	18.4 ± 0.5
M8	0.140 ± 0.008	9.66 ± 0.33	0.27 ± 0.01	1.25 ± 0.06	0.33 ± 0.05	3.69 ± 0.14	443 ± 3	65.2 ± 4.4
M9	0.163 ± 0.010	12 ± 6	0.02	0.43 ± 0.16	0.21 ± 0.04	1.57 ± 0.04	160 ± 45	53.4 ± 2.6
M10	0.145 ± 0.001	0.71 ± 0.16	0.01	0.41 ± 0.05	0.02	2.61 ± 0.04	10.9 ± 1.58	16 ± 1
M11	0.119 ± 0.018	20.4 ± 0.74	0.031	0.56 ± 0.18	0.22 ± 0.01	1.66 ± 0.14	290 ± 43	58.6 ± 2.3
M12	0.077 ± 0.012	24.6 ± 0.79	0.10 ± 0.01	1.14 ± 0.19	0.43 ± 0.03	1.87 ± 0.07	1016 ± 393	37.7 ± 3.3
M13	0.150 ± 0.012	2.13 ± 0.28	0.01	0.14	0.04 ± 0.01	2.73 ± 0.10	143 ± 7	16.9 ± 0.3
M14	0.126 ± 0.008	14.0 ± 0.7	0.32 ± 0.01	3.64 ± 3.68	0.19 ± 0.04	3.64 ± 0.21	549 ± 81	51.2 ± 3.2
M15	0.097 ± 0.013	15.7 ± 0.6	0.24 ± 0.01	1.08	0.43 ± 0.11	2.29 ± 0.13	397 ± 87	68 ± 4
M16	0.150 ± 0.010	6.32 ± 1.18	0.17 ± 0.01	1.24 ± 0.02	0.25 ± 0.09	2.27 ± 0.09	408 ± 214	60.4 1.6
M17	0.209 ± 0.021	95 ± 5	0.35 ± 0.03	1.09 ± 0.09	0.23 ± 0.01	2.06 ± 0.03	782 ± 35	88.7 ± 3.3

+ = $\mu g kg^{-1}$, B = Brackish water samples; F = Fresh water samples; M = Marine water samples

with the prepared drift standard (1 μg L^{-1} of all the analyzed trace and rare earth elements and 1 mg L^{-1} of Na, K, P, Ca and Mg). The concentrations and accuracy of measured trace and rare earth elements, and of the certified reference materials SRM 1643a, DOLT-3 (dogfish liver) and RM 8414 (bovine muscle powder) are presented in Table 2. The drift in measurement by the instrument was negligible (range from 0.79 to 1.01). The result of the SRM 1640a (trace elements in water) was acceptable to validate the calibration. The results of the DOLT-3 and SRM 8414 used for the digestion gave a good agreement between the certified and the measured concentrations as seen by the accuracy of the result obtained (Table 2).

RESULTS AND DISCUSSION

The elemental compositions (arithmetic mean ± standard deviation) of all the analyzed fresh, brackish and marine samples are presented in Table 3.

Essential elements in analyzed samples

The concentrations of essential elements within and among the three analyzed groups varied widely as shown in Table 3. The concentration range of Cu reported in this study shows that crabs and shrimps from marine and brackish waters accumulate more Cu than the fish species analyzed. The concentration ranges for most of the species analysed were similar to the concentrations reported in the literature for fish, crabs and shrimps (Zeng et al., 2012; Fung et al., 2013). Specifically, *P. atlantica*, *P. notialis*, *P. sp.*, *P. regius* and *P. Valids* were within the concentration range reported by Franca et al. (2005) and Tu et al. (2008).

According to the limits set by the European Community (EC, 2005), Ministry of Agriculture, Fisheries and Food (MAFF, 2000) and Turkish guidelines (Dural et al., 2007), 16.2% of the investigated species exceeded the 20 μgg-1 acceptable Cu concentrations in fish samples, while 13.5% exceeded the FAO/WHO (1989) limits of 30 μgg-1.

Co concentration is shown to vary within and between the three groups. The reported concentration is generally higher in freshwater fish samples than brackish and marine, except for the crabs and shrimps. Co concentration in this study is similar to the levels reported in literatures for fish muscles, i.e. in the range of <0.05 μg g^{-1} to 0.40 μgg^{-1} for the Black Sea coast, <0.01 μg g^{-1} to 0.37 μg g^{-1} for the Aegean and Mediterranean seas and 0.20 μg g^{-1} to 0.67 μgg^{-1} for Indian fish market (Turkmen et al., 2009).

Cr concentrations in brackish samples were generally higher than levels determined in freshwater and marine samples. Comparison with the literature data showed that Cr levels from this study were lower than the levels reported by Cacador et al. (2012) and Eca et al. (2013), but higher than the values (<0.023 μg g^{-1} to 0.061 μg g^{-1}) reported for muscles of lionfish (Fung et al., 2013).

The distribution of Fe in the analyzed species follows the order: marine > brackish > freshwater. The provisional tolerable daily intake for an average adult (60 kg body weight) set by FAO/WHO (1989) is 48 mg. However, from the study, about 71% of marine, 50% fresh and 79% brackish species exceeded the FAO/WHO limit. This might represent a serious problem in the fishing communities as they eat high amount of fish compared to non-fishing communities (Goma and Rana, 2007). The literature data on Fe concentrations in biota is scarce. However, the concentrations in the range of 24 μg g^{-1} to 90 μg g^{-1} by Moeller et al. (2003) and 9 μg g^{-1} to 136 μg g^{-1} reported by Turkmen et al. (2009) were similar to the concentration determined in this study. The reported concentration range of 458 μg g^{-1} to 1597 μg g^{-1} for crabs by Eca et al. (2013) was higher than the range reported in this study, while the concentration range of 0.163 μg g^{-1} to 1.58 μg g^{-1} (Fung et al., 2013) was lower compared to the Fe range found in all the three aquatic biotopes in our study.

Observed Mn concentrations in some species from our study were comparable to the published data, like 23.8 μg g^{-1} to 121.1 μg g^{-1} in fish muscle reported by Mohammed et al. (2012). Generally most of the recorded Mn levels were higher in organisms from the three habitats in our study compared to other reported data (Fung et al., 2013; Subotic et al., 2013). In comparison with Turkish guidelines, which proposed 20 μg g^{-1} acceptable limits in fish samples, 50% of brackish, 16.6% of fresh and 17.4% of marine species exceeded this guideline.

Ni concentrations were generally lower than 1 μgg^{-1} for most of the species except *C. senegallus, M. rume, C. nigrodigitatus* and *S. tritor*. In the literature, Ni was reported in the range from 0.581 μg g^{-1} to 0.681 μg g^{-1} (Mistra et al., 2004), <0.011 μg g^{-1} to 0.91 μg g^{-1} (de Mora et al., 2004), 0.691 μg g^{-1} to 4.341 μg g^{-1} (Rahman et al., 2012) and 3.491 μg g^{-1} to 3.861 μg g^{-1} (Taweel et al., 2013). Determined concentrations in all three aquatic biotopes from our study were within the reported ranges for Ni in edible fish.

Potentially toxic elements in analyzed samples

Metals known for their toxicity to fish and human are: Ag, As, Be, Cd, Pb, Hg, Al, V, Sr, Sn and Ba. In this study, concentrations of Be, Sb, Sn, Tb and Tl were generally below the detection limit in most tissues of analysed samples. Thus, these elements were not considered for further discussion. Metals of concern due to their negative health effects in human are arsenic, cadmium, mercury and lead. The concentration range observed for As in this study was marine > brackish > freshwater species. As widely reported in the literature, marine organisms bioaccumute As as arseno-betaine that is considered nontoxic (Slejkovec

et al., 2004). For the fresh and brackish species, the range reported in this study was lower compared to the range of 1.35 μg g^{-1} to 22.4 μg g^{-1} reported for other fishes (Shah et al., 2009; Fung et al., 2013). However, Mn ranges were higher than the levels reported in China and Niger/Delta in Nigeria (Yi et al., 2011; Eboh et al., 2006).

Cadmium is known to accumulate in human kidney for a relatively long time of about 20 to 30 years, and at high doses might damage the respiratory system, induce renal and hepatic toxicity, and its association with bone disease, poor reproductive health, hypertension, tumor and hepatic dysfunction has been reported (Waalkes, 2000). Cd ranges in three groups of organisms were generally similar except for crustaceans in brackish and marine ecosystems, in which higher Cd levels were recorded. The determined Cd values in this study fitted well with the range reported for other locations (Turkmen et al., 2009; Zeng et al., 2012; Subotic et al., 2013). George et al. (2013) reported Cd concentrations higher than the international acceptable limit of 0.05 μg g^{-1} for two marine species from Nigeria. In the present study, Cd levels in all species except crustaceans were below limits set by the European Community and European Union (0.05 μg g^{-1} and 0.01 μg g^{-1}).

The concentration range found for Pb in selected organisms followed the order marine > brackish >freshwater. The observed accumulation trend in crustaceans indicated that *S. afra, C. pallidus, P. jubelini, C. chrysrus* and *C. brownii* accumulated more Pb compared to other crustacean species. Aside from the listed species, the reported concentration ranges for crustaceans were similar to the values reported in other studies (Al-Busaidi *et al.*, 2011; Fung *et al.*, 2013). Previous studies in Nigeria had reported lower or somewhat similar concentrations in fishes from Nigerian waters (George et al., 2013). If comparison with the international tolerable limits of 0.2 μg g^{-1} (EC, 2005) is made, 28.6% 16.7% and 58.8% of brackish, freshwater and marine species exceeded this limit, thus, indicating potential threat to human consumption.

Hg concentration ranges reported in this study for the three aquatic biotopes were generally higher than the values reported in Jamaica and Vietnam (Fung et al., 2013), but similar to the majority of other published data (Sankar et al., 2006; Cirillo et al., 2010). Based on European tolerable consumption limit of 0.5 μg g^{-1} in fish, only *S. senegalensis* exceeded this limit.

CONCLUSION

In the present study, levels of essential and toxic trace elements in marine, brackish and freshwater organisms, which are used for human consumption in South-western Nigeria, were compared. The general concentration trend of heavy metals in the finfish and crustacean samples followed the order: marine > brackish > freshwater. In addition, the data from the present study was compared with metal levels from already published data and with permissible metal levels given by international agencies with the final aim to make conclusion on pollution impact in the open water bodies in Nigeria. Since for Cu, Zn, Fe, Mn and Pb in almost all the organisms sampled and for Cd in crustaceans and Hg in *S. senegalensis* permissible metal levels were exceeded, strict environmental monitoring and water quality assessment should be performed.

Sažetak

SASTAV METALA U RIBAMA I RAKOVIMA BOĆATIH, SLATKOVODNIH I MORSKIH SUSTAVA U JUGOZAPADNOJ NIGERIJI

Prikazanom studijom istražene su koncentracije elemenata u tragovima u mišićima ili cijelim uzorcima 37 riba i rakova sakupljenim mrežama stajačicama u slatkovodnim (Olomore), morskim (Eti osa) i boćatim vodama nigerijskih država Ogun i Lagos, od lipnja do kolovoza 2013. Elementi u tragovima utvrđeni su pomoću ICP-MS nakon digestije u mikrovalnom autoklavnom sustavu. Rezultati su otkrili kako je opći trend elemenata u tragovima u uzorkovanim ribama i rakovima morski > boćati > slatkovodni. Uzimajući u obzir sve uzorke, 16,2% i 13,5% prekoračilo je granice bakra i cinka (20 μg g^{-1} i 30 μg g^{-1}), propisane od strane Europske Unije, dok je 71%, 50%, 79% morskih, slatkovodnih i boćatih uzoraka prekoračilo granicu od 48mg željeza na 60kg čovjeka, koju je postavila FAO/WHO. Uzorci su premašili prihvatljivu granicu postavljenu od strane FAO/WHO, čime je dokazano kako je potrebno uložiti trud u monitoring otpadnih voda koje se ispuštaju u otvorene vode Nigerije.

Ključne riječi: teški metali, ribe, rakovi, vodeni ekosustav, Nigerija

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