1 **Original Research Article**

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3 **ESSENTIAL ELEMENTS AND HEAVY METALS**
4 **IN SARDINE FISH FROM KIVUKONI, KUNDUCHI**
5 **AND BAGAMOYO FISH MARKETS IN**
6 **TANZANIA**
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11 **ABSTRACT**
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This study has assessed the concentrations of the essential elements Ca, Cu, Fe, Se and Zn as well as the toxic heavy metals Al, As, Cd, Cr, Ni and Pb in samples of sardine fish collected from Kivukoni, Kunduchi and Bagamoyo fish markets. The collected fish samples were oven dried, grinded to fine powder and compressed to pellets. The pellets obtained were analyzed for both essential elements and toxic heavy metals using Energy Dispersive X- rays Fluorescence Spectroscopy (EDXRF). The mean concentrations of the analyzed elements were compared to the maximum tolerable levels (MTLs) set by international organizations. The mean concentrations of Ca, Se, Zn, As, Cr, and Cd were higher than their MTLs while, the mean concentrations of Cu and Ni were lower than their MTLs for samples from all fish markets. The mean concentrations of Al and Fe were higher than their MTLs for samples from Kunduchi and Bagamoyo fish markets. The mean concentrations of Pb were below the detection limit (0.50) μg/g for the spectrometer used in this study. The MTLs for Ca, Fe, Cu, Se, Zn, Al, Cr, Ni, As, and Cd were 130,000 ; 200 ; 30 ; 40 ; 0.75 ; 100 ; 0.15 ; 2.00 ; 0.50 ; and 2.00 μg/g respectively, according to FAO/WHO 2002, WHO 1983, COMA 1998, CFS2009, WHO1985, COT 2003 and CODEX 2005 respectively.

13
14 Keywords: Aquatic Environments, Energy Dispersive X-rays Fluorescence (EDXR), Sardine
15 Fish, Kivukoni, Kunduchi and Bagamoyo fish markets
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17 **1. INTRODUCTION**
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19 Seafood's could be a good source of the essential elements for many African communities
20 especially the coast communities. Contamination of the seafood's and the general aquatic
21 environments by heavy metals is the problem of major concern worldwide [1]. All metallic
22 elements with an atomic number greater than that calcium (40.04) are referred as heavy
23 metals [2]. These include arsenic (As), Cadmium (Cd), Lead (Pb) and Mercury (Hg) which
24 are also of major concern because; they are extremely toxic even at low concentrations [3].
25 The above elements are brought into the aquatic environments as a result of the natural

26 activities such as seafloor and bedrock dredging, volcanic eruptions, soil erosion and
27 weathering as well as anthropogenic activities such as agriculture, municipal wastewater
28 discharges, mining, incinerations, domestic discharges and industrial discharges [4]. It is
29 reported that, more than 80% of the world's waste water and more than 95% of the least
30 developed countries wastewater are released into the aquatic environments untreated. The
31 wastewaters ultimately end up in the oceans and seas, thereby, introducing different
32 contaminants into the aquatic environments [5,6].

33 Heavy metals which enter the aquatic environments are taken up by fish and undergo
34 bioaccumulation in the liver, gills, kidney, intestine and other organs [7,8]. Since fish are
35 consumed by human beings, heavy metals are eventually consumed by human beings and
36 other organisms at the top of the food chain [1]. heavy metals therefore pose a potential
37 threat to the health of humans and other organisms at the top of the food chain [7]. Heavy
38 metals pollutions can trigger a wide array of health problems such as infertility, miscarriages,
39 cancer, damages to the vital organs such as kidney, brain, central and peripheral nervous
40 system leading to an ultimate death [2,3].

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42 Various studies indicate that, the accumulations of heavy metals in fish and other marine
43 foods have been increasing steadily in the recent years[9,10,11,12]. In Egypt, concentrations
44 of copper (Cu), Zinc (Zn), Lead (Pb) and cadmium (Cd) in fish samples collected from El –
45 Fayoun province were found to be higher than their maximum tolerable levels (MTLs) set by
46 WHO,2004 [13]. In Kenya, the study was conducted a study to investigate the levels of Lead
47 (Pb), Nickel (Ni), Manganese (Mn), Zinc (Zn), Cadium (Cd) and Chromium (Cr) in the gills of
48 the tilapia fish and water in the Athi –River by using Atomic Absorption Spectrometry[8]. The
49 study concluded that, the levels of the mentioned heavy metals in both fish and water, were
50 above their respective maximum tolerable levels set by WHO,2004 [8]. In Tanzania, the
51 assessment of the concentrations of heavy metals in the fish samples collected from the
52 downstream and upstream of the north Mara gold mining using Energy Dispersive X- rays
53 Fluorescence (EDXRF) spectrometry[14]. The samples of catfish and lungfish from both
54 sites had higher mean concentrations of sodium (Na), potassium (K), copper (Cu), chromium
55 (Cr) and nickel (Ni) compared to their maximum tolerable levels (MTLs) set by FAO, 1983
56 and WHO, 1985 [14,15]. Furthermore, a study conducted by Koleleni and Haji [15] to
57 determine the concentrations of heavy metals in the sea port of Zanzibar, reported that, the
58 concentrations of chromium (Cr), Lead (Pb), Nickel (Ni) and Arsenic (As) in the samples of
59 sardine fish collected from the sea port of Zanzibar were higher than their maximum
60 tolerable level set by WHO, 2004 [16]. In the best of my knowledge, there had been no study
61 that had assessed the concentrations of the essential and toxic elements in sardine fish
62 harvested from the Indian Ocean and sold at Kivukoni, Kunduchi and Bagamoyo fish
63 markets. The sardine fish are the most landed fish species globally and they could serve as
64 a good source of micronutrients for the coast communities [12]. The aim of this study
65 therefore, to assess the concentrations of both the essential and toxic elements in sardine
66 fish harvested from the Indian Ocean and sold at Kivukoni, Kunduchi and Bagamoyo fish
67 markets. The assessment will facilitate the government and the general public with
68 information to evaluate the quality of the sardine fish to the food value and nutritional
69 security to avoid the risk of heavy metals pollution to the coast communities.

2. MATERIAL AND METHODS

2.1 Description of the Study Area

This study was conducted at Kinondoni and Bagamoyo districts. The Kinondoni and Bagamoyo coast line are located between latitudes and longitudes $6^0 34`0``$ S, $39^0 06`0``$ E to $6^0 51`0``$ S and $39^0 19`0``$ E, respectively. The selected fish markets are located alongside the Indian Ocean. Kivukoni and Bagamoyo fish markets are near to Dar es Salaam and Bagamoyo harbors respectively, while, Kunduchi fish market is near to Kunduchi Beach Hotel. This coastline has a total population of 5,463,209 inhabitants. The major economic activities along this coastline include manufacturing industries, trade, agriculture, mining, tourism, and fishing [17]. Figure **1** is a description of the study area.
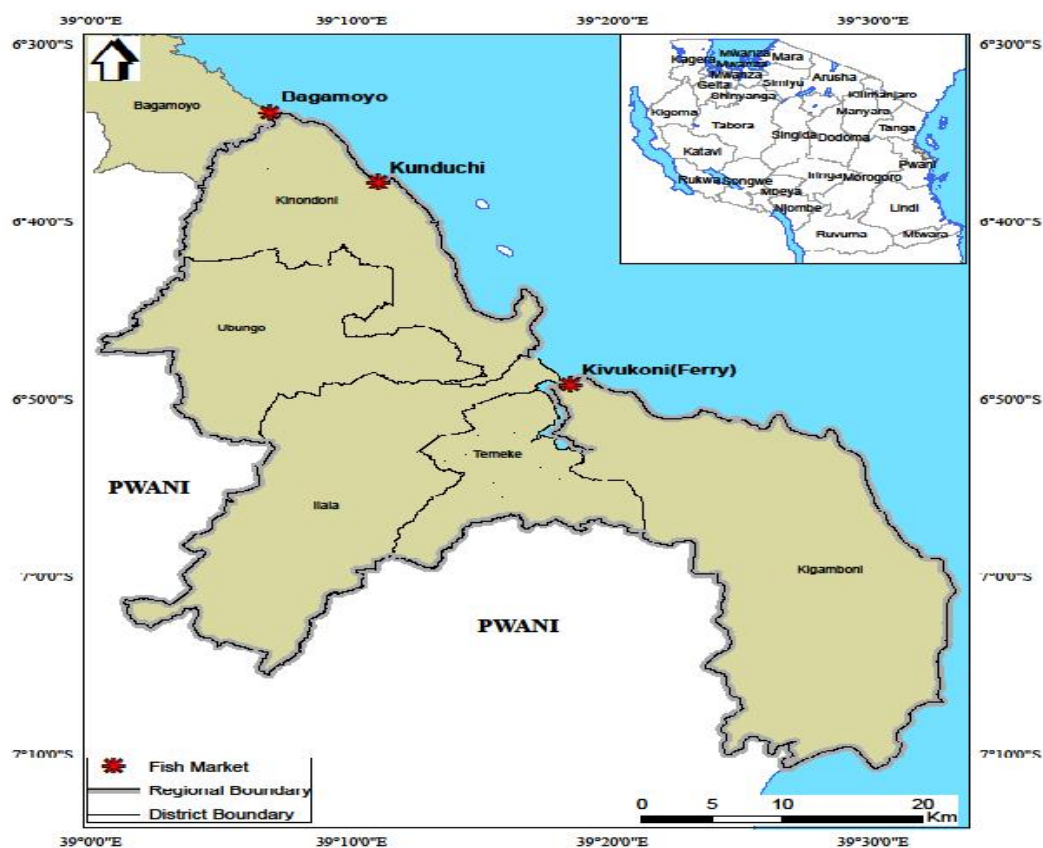
Source:Cartographic unit-University of Dar es Salaam

Fig. 1 Description of the study area

2.2 Sample Collection

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83 The samples of sardine fish were bought from the fishermen at Kivukoni, Kunduchi and
84 Bagamoyo fish markets. The purchased fish samples were kept in containers made of non-
85 wet table plastic materials. The containers with the fish samples were kept in a refrigerator till
86 the preparation day. Before the containers were used, they were preconditioned by a mineral
87 acid (ultra-pure HNO_3) and rinsed thoroughly with double distilled water.Figure **2** indicates
88 some of the collected fish samples.

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98   **Fig. 2 Pictures of some of the collected fish samples in a closed package**

99   **2.3 XRF Sample Preparation**

100   The collected fish samples were washed thoroughly using clean fresh water and oven dried
101   at the temperature of 45^0C to 50^0C for 48 hours. The samples were thereafter, ground and
102   sieved to obtain fine powders (<50 µm). For each sample, a mass of 4g was mixed with 0.9g
103   of starch (binder). The mixtures of the samples and the binders were homogenized using
104   Fritsch Pulverisette[TM] (Industriiesstrasse, 8-55743I dar Okerstein, German) homogenizer.
105   The homogenizer was set at speed of 120 revolutions per second for 10 minutes. After
106   homogenization, the samples were compressed to pellets using Restsch[TM] (Restch GmbH
107   Retsch-Allee 1-5, 42781Haan, German) pellet presser. A force of 15N was applied to the
108   pellet presser to produce pellets of 32mm outer diameter. Figure **3** indicates one of the
109   pelletized samples produced.

114   **Fig.3 A pelletized sample**

115   The samples so produced were properly labeled and taken to the Spectro Xepos[TM] (Sepetro
116   Analytical Instruments GmbH, Boschstr. 10, 47533 Kleve, German) spectrometer for heavy
117   metals analysis. Figure **4** indicate the picture of the Spectro Xepos[TM] spectrometer used in
118   this study.

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120   **Fig. 4 The picture of the Spectro Xepos[TM] spectrometer**

121   **2.4 Sample Elemental Analysis**
122   Each pellet was irradiated by x-rays from the x-rays tube so as to produce the characteristic
123   x-rays. The x-ray tube was operated at a maximum power of 50W and a maximum voltage of

124 50KV. A semiconductor detector (Silicon Lithium)　was used to detect and measure the
125 energy of the characteristic x-rays produced. A computer connected to the spectrometer was
126 used to display the spectra of the intensities of the characteristic x-rays against energies of
127 the elements in the sample. The spectrometer was calibrated weekly and set according to
128 manufactures specifications.

129 Elemental composition of each pellet was computed by the X- Lab Pro[TM] software with Turbo
130 quant (Tq 9232) algorithm. This software, corrected for matrix effects, interference effects
131 and background effects on the basis of fundamental parameter method. The software
132 converts the intensities of the characteristic x-rays into concentrations of the radiating
133 elements according to the equation 1[18]

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135 $C_i = K_i I_i M_a$　　　　　　　　　　　　　　　　　　　　　　　　　　　　(1)

136 Where, C_i is the concentration of the radiating element in the sample, M_a is the matrix
137 correction factor, K_i is the constant of proportionality and I_i is the intensity of the fluorescence
138 radiations from element (i).

3. RESULTS AND DISCUSSION

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141 **3.1 Minimum Detection Limits (MDL)**

142 Minimum detection limits refers to the minimum concentration of the analyzed elements that
143 can be detected by the instrument (spectrometer). It is the lowest concentration that can be
144 determined to be statistically significant from the analytical blank [19]. The concentration of
145 the analyzed element in the sample is considered to be statistically significant, if the net peak
146 intensity of its characteristic x-rays is at least thrice the standard deviation of the background
147 noise. The minimum detection limits can be very low when the background counts are not
148 significant and vice –versa is true.

149 The minimum detection limits for the analyzed elements were determined using the X-Lab
150 Pro[TM] software package according to the equation 2; [19].

151 $$MDL = \frac{3C_i}{I_i - I_b} \sqrt{\frac{I_b}{T_b}}$$　　　　　　　　　(2)

152 Where, C_i is the concentration of the analyzed element, I_b is the intensity of the background
153 counts, I_i is the intensity of the characteristic x-rays from the analyzed element and T_b is the
154 time used to measure the background counts. Table **1** indicates the minimum detection limits
155 of the spectrometer for the analyzed elements.

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157 **Table 1: The MDL (µg/g) for the analyzed elements**

ATOMIC NUMBER	CHEMICAL SYMBOL	ELEMENT	MDL (µg/g)
13	Al	Aluminum	35.2
20	Ca	Calcium	6.0
24	Cr	Chromium	5.2
26	Fe	Iron	2.6
28	Ni	Nickel	1.0
29	Cu	Copper	1.4
30	Zn	Zinc	1.0
33	As	Arsenic	0.3
34	Se	Selenium	1.0
48	Cd	Cadmium	3.8
82	Pb	Lead	0.5

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161 **3.2 Mean concentrations of the essential elements for the sardine fish from**
162 **Kivukoni, Kunduchi and Bagamoyo.**

163 The statistical package for social sciences (SPSS – Version 17) software was used
164 to compute the mean concentrations of the essential elements for samples from
165 each fish market. Tables **2** and **3** indicate the arithmetic means and geometric
166 means of the concentrations of the essential elements for each fish market.

167 **Table 2 Arithmetic means of concentrations ± standard error of the mean (SEM) (µg/g)**
168 **for the essential elements**

ELEMENTS	FISH MARKETS		
	KIVUKONI (n = 10)	KUNDUCHI (n = 10)	BAGAMOYO (n = 10)
Calcium (Ca)	37981.9 ± 5558.2	30388.5 ± 103.9	38310.9± 103.0
Iron (Fe)	149.9 ± 0.4	211.9± 1.0	234.8 ± 0.8

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Copper (Cu)	4.0 ± 0.1	2.6± 0.1	3.5 ± 0.1
Selenium (Se)	1.7 ± 0.0	2.2 ± 0.1	1.9 ± 0.0
Zinc (Zn)	159.8 ± 1.1	105.1 ± 0.6	130.9 ± 0.5

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175 **Table 3 Geometric means of the concentrations ×/÷ standard deviation (SD) (µg/g) for**
176 **the essential elements**

ELEMENTS	FISH MARKETS		
	KIVUKONI (n = 10)	KUNDUCHI (n = 10)	BAGAMOYO (n = 10)
Calcium (Ca)	25983.0 ×/÷ 17576.6	30405.3 ×/÷ 328.6	38282.3 ×/÷ 328.4
Iron (Fe)	149.8 ×/÷ 1.3	34.1 ×/÷ 3.3	34.1 ×/÷2.4
Copper (Cu)	4.0 ×/÷ 0.4	2.6 ×/÷ 0.2	3.4 ×/÷ 0.2
Selenium (Se)	1.7×/÷ 0.1	2.2×/÷ 0.2	2.0 ×/÷ 0.1
Zinc (Zn)	159.8 ×/÷ 3.5	105.3 ×/÷ 1.9	131.3 ×/÷ 1.5

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178 From the tables, it can be seen that, Calcium is the highest mean concentrations for samples
179 from all fish markets. Iron (Fe) recorded the 2nd higher mean concentrations for samples
180 from Kunduchi and Bagamoyo fish markets while Zinc (Zn) recorded the 2nd higher mean
181 concentration for samples from Kivukoni fish market. Copper (Cu) recorded the 3rd higher
182 mean concentrations followed by Selenium (Se) for samples from all fish markets. It can be
183 noted that, the mean concentrations of the essential elements were in the order; Ca > Zn >
184 Fe > Cu > Se > (for samples from Kivukoni fish market), Ca > Fe > Zn > Cu > Se (for
185 samples from Kunduchi fish market) and Ca > Fe > Zn > Cu > Se (for samples from
186 Bagamoyo fish market).

187 It is reported that, calcium is the 5th most abundant element in the earth's crust [20]. It exists
188 in the form of lime stones, calcite, marble, ice land spar, dolomite, stalactite, stalagmite,
189 gypsum and phosphate in the phosphoric rocks which are soluble in water (Xing and Liu
190 2011). Perhaps the higher mean concentrations of calcium obtained at all sampling areas
191 are due to the fact that, calcium compounds are highly soluble in water and therefore are
192 easily absorbed by the aquatic organisms. Iron is the 4th most abundant element in the
193 earth's crust. In most cases, it exists as oxides which are highly insoluble in water [21].
194 Concentrations of iron in water bodies due to natural activities are quite low due to its low
195 solubility in water; however, expansions of populations, rapid urbanizations and
196 industrialization lead to various ecological problems in the ecosystems of most water bodies.

197 Significant speciation, concentration and bioavailability of iron in water and sediments occur
198 as a result of the anthropogenic activities [22]. This suggests that, apart from the natural
199 activities such as precipitation, dust, as well as weathering of rocks and soils, the higher
200 mean concentrations of iron recorded for samples from all fish markets, are may be due to
201 industrial discharges, domestic discharges and municipal wastewater treatment plants.

202 Zinc exists naturally in rocks, soil and water and in most cases as zinc sulphide (ZnS). Most
203 of zinc compounds are water soluble, though; the metal itself is not [23]. The high
204 concentrations of zinc recorded for samples from all fish markets, might be due to the fact
205 that, most of zinc compounds are water soluble and hence are easily absorbed by aquatic
206 organisms. Also natural activities such as weathering and abrasion of rocks as well as
207 anthropogenic activities such as corrosion of zinc galvanized ships and boats, domestic and
208 industrial discharges are the factors which might have caused elevated levels of zinc for
209 samples from all fish markets.

210 The mean concentrations of copper and selenium were lower compared to those of the other
211 essential elements analyzed in this study. Selenium occurs naturally in rocks; soil and water
212 although; it's also associated with mining of coal, phosphates, as well as refinery and
213 combustion of fossil fuels [24]. Also agricultural drain water, sewage slugs, fly of ash from
214 coal-fired power plants are sources of selenium into the aquatic environments [25]. Industrial
215 sources of copper into the aquatic environments include; wood production, iron and steel
216 production, waste incinerations, coal combustion, oil and gasoline combustion. Also
217 agricultural runoffs and domestic use of fertilizers also contributes to raise the levels of
218 copper in the aquatic environments [26]. Since there was no evidence of elevated levels of
219 lead (Pb) which is more associated with fossil fuels, it implies that, the levels of copper and
220 selenium recorded at all fish markets, are not related to refinery and combustion of fossil
221 fuels; rather, they might be related to sewage slugs, agricultural drain water, industrial
222 discharges, domestic discharges and natural activities.

223 Furthermore, the results of this study indicate that, sardine fish from Kivukoni, Kunduchi and
224 Bagamoyo fish markets are rich in essential elements Ca, Fe, Cu, Se and Zn. This implies
225 that, they can be used as a good source of the essential elements for the coast communities,
226 provided that; they are prevented from being polluted by heavy metals. Calcium is essential
227 in muscle contraction, acolyte activation, building of strong bones and teeth, blood clotting,
228 nerve impulse transmission, heart beat regulation and fluid balance within cells [20]. Iron is
229 essential for almost all living organisms as it takes part in a wide range of metabolic
230 processes such as oxygen transport, deoxyribonucleic acid (DNA) synthesis and electron
231 transport. Iron can however form free radicals, thus; its concentration in blood must be in
232 proper levels because, excessive amounts of iron can damage the tissues [21].

233 Zinc is an essential mineral for normal fetal growth and development as well as milk
234 production during lactation [27]. Zinc deficiency triggers an array of health problems in
235 children, many of which become chronic, such as weight loss, stunted growth, weakened
236 resistance to infections and early deaths. In fact, zinc deficiency is linked to about 116,000
237 child deaths every year [11]. Selenium is a major component of many enzymes and it plays
238 important roles in ant oxidation, reproduction, muscle function and tumors prevention [28].

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239   The analysis of the variances of the means (Turkey HSD) indicate that, samples from
240   Kivukoni fish market had significantly higher ($p < 0.05$) mean concentrations of Ca, Cu, and
241   Zn when compared to samples from Kunduchi fish market while samples from Bagamoyo
242   fish market had significantly higher ($p < 0.05$) mean concentrations of Ca and Cu when
243   compared to samples from Kunduchi fish market. On the hand, samples from Kunduchi fish
244   market had significantly higher ($p < 0.05$) mean concentration of Se when compared to
245   samples from Kivukoni and Bagamoyo fish markets. Since, there are many human activities
246   around Kivukoni and Bagamoyo fish markets compared to Kunduchi fish market, these
247   variations suggest that, apart from natural activities, human activities plays a significant role
248   to rise the levels of Ca, Cu and Zn among the sampling markets. Furthermore, variations of
249   the mean concentrations of the essential elements might indicate that, sardine fish sold at
250   the three fish markets have different feeding areas and hence different diets.

251   **3.3 Mean Concentrations of the Toxic Heavy Metals for the Three Fish Markets**
252   The statistical package for social sciences (SPSS - Version 17) software was used to
253   compute the mean concentrations of the toxic heavy metals for samples from each fish
254   market. Tables **4** and **5** indicate the arithmetic means and geometric means of the
255   concentrations of the toxic heavy metals for samples from each fish market.

256   **Table 4 Arithmetic means of concentrations ± standard error of the mean (SEM) (µg/g)**
257   **for the toxic heavy metals**

ELEMENTS	FISH MARKETS		
	KIVUKONI (n = 10)	KUNDUCHI (n = 10)	BAGAMOYO (n = 10)
Aluminum (Al)	52.8 ± 3.6	105.4 ± 4.5	129.3 ± 6.7
Chromium (Cr)	2.0 ± 0.2	21.7 ± 0.4	27.2 ± 1.0
Nickel (Ni)	0.9 ± 0.1	0.3± 0.0	0.8 ± 0.1
Arsenic (As)	3.8 ± 0.1	7.2 ± 0.0	5.39± 0.1
Cadmium (Cd)	0. 6 ± 0.1	2.6 ± 0.1	0.7± 0.7
Lead (Pb)	BDL	BDL	BDL

258   BDL for Pb = 0.50 µg/g
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266 **Table 5 Geometric means of the concentrations ×/÷ standard deviation (SD) (µg/g) for**
267 **the toxic heavy metals**

ELEMENTS	FISH MARKETS		
	KIVUKONI (n = 10)	KUNDUCHI (n = 10)	BAGAMOYO (n = 10)
Aluminum (Al)	51.2 ×/÷ 11.4	102.7 ×/÷ 14.3	130.0 ×/÷ 21.3
Chromium (Cr)	2.1 ×/÷ 0.4	22.0 ×/÷ 1.4	27.3 ×/÷ 3.1
Nickel (Ni)	0.9 ×/÷ 0.2	0. 3 ×/÷ 0.7	0.7×/÷ 0.3
Arsenic (As)	3.8 ×/÷ 0.6	7.2 ×/÷ 0.1	5. 3 ×/÷ 0.2
Cadmium (Cd)	0.6×/÷ 1.0	2.6 ×/÷ 0.1	0.7×/÷ 0.2
Lead (Pb)	BDL	BDL	BDL

268 BDL for Pb = 0.50 µg/g
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270 From the table 5, it can be seen that, aluminum recorded the highest mean concentration
271 followed by arsenic for samples from all sampling areas. The mean concentrations of
272 aluminum were (52.8 ± 33.6; 105.4 ± 4.5 and 129.3 ± 6.7) µg/g for samples from Kivukoni,
273 Kunduchi and Bagamoyo fish markets respectively. Aluminium is the third most abundant
274 element in the earth's crust and it occurs naturally in rocks, soil and water. It is not essential
275 for life; however, aluminium compounds are widely used in various industrial applications or
276 consumer products such as antacids, food additives and antiperspirants [29]Apart from
277 natural activities in rocks and soils, the high levels of aluminium for samples from all fish
278 markets, might be related to coagulants used in water treatment process and drugs
279 (medicines) such as antacids or antiperspirants which are brought to the ocean by rain
280 water, municipal wastewater treatment plants as well as industrial and domestic discharges.

281 The mean concentrations of arsenic were (3.8 ± 0.1; 7.2 ± 0.0 and 5.3 ± 0.1) µg/g for
282 samples from Kivukoni, Kunduchi and Bagamoyo fish markets respectively. It is reported
283 that, arsenic concentrations are normally higher in marine fish compared to fresh water fish
284 [30]. Arsenic in fish muscles is mainly found in organic form, which is less toxic compared to
285 inorganic arsenic [31]. Chromium recorded the 3rd higher concentrations after aluminium and
286 arsenic. The mean concentrations of chromium were (2.0 ± 0.2; 21.8 ± 0.4 and 27.2 ± 1.0)
287 µg/g for samples from Kivukoni, Kunduchi and Bagamoyo fish markets respectively. Rarely
288 chromium occurs naturally in the earth's crust as an element (metal), but, in most cases it
289 occurs in compound forms or ions in water [32]. Many chromium compounds are used in
290 painting pigments. Almost all chemical laboratories (academic, research, industry) discharge
291 considerable amounts of chromium into the environment every day [33]. This suggests that,
292 natural activities and industrial discharges are the main sources of chromium for samples
293 from all fish markets.

294 Cadmium recorded lower means concentrations for samples from all fish markets, compared
295 to aluminium, arsenic and chromium. The mean concentrations of cadmium were (0.6 ± 0.0;
296 2.6 ± 0.1 and 0.7 ± 0.7) µg/g for Kivukoni, Kunduchi and Bagamoyo respectively. Cadmium
297 occurs naturally in the earth's crust. It is vastly used in batteries, coating, plating and various
298 industrial applications [34]. Cadmium from various industrial applications enters into air and
299 binds with small particles which combine with water and soil, thereby, causing contamination
300 of fish, animals and plants. Spills from hazardous waste sites and improper wastes disposal
301 can cause cadmium leakages into the aquatic environments [35]. On the other hand, the

302 mean concentrations of lead were below the detection limits of the spectrometer used in this
303 study for samples from all fish markets. The minimum detection limit of the spectrometer was
304 0.50µg/g for lead. The mean concentrations of mercury were not included in this analysis
305 due to the fact that, EDXRF is not suitable for determination of concentrations of mercury.

306 Analysis of the variances of the means (Turkey HSD) for the toxic heavy metals indicate that,
307 samples from Bagamoyo fish market had significantly higher (p< 0.05) mean concentrations
308 of Al, As, Cd and Ni when compared to samples from Kivukoni. The samples from Kunduchi
309 fish market had significantly higher (p< 0.05) mean concentration of Al, As and Cd when
310 compared to samples from Kivukoni fish market. On the other hand, samples from Kivukoni
311 fish market had significantly higher (p < 0.05) mean concentration of Ni when compared to
312 samples from the other fish markets. Since Bagamoyo and Kunduchi fish markets are more
313 close to human settlements compared to Kivukoni fish market, this suggests that, large
314 percentages of the toxic heavy metals Al, As and Cd originates from domestic discharges
315 and agricultural drain waters.

3.4 Correlations Analysis for the Toxic Heavy Metals

317 Spearman correlation tests were carried out to determine the nature, magnitude and
318 directions of correlations among the toxic heavy metals. At Kivukoni fish market, a strong
319 and a statistically significant correlation was recorded between arsenic and nickel (r = 0.63; p
320 = 0.05). This correlation suggests that, arsenic and nickel originates from similar sources,
321 probably municipal wastewater treatment plants, domestic and industrials discharges. Also,
322 at this fish market, a moderately positive but not a statistically significant correlation was
323 recorded between arsenic and chromium (r = 0.37; p = 0.30). This correlation suggests that,
324 arsenic and chromium for samples from this fish market, originates from sources with weak
325 similarities. Probably, large or less amounts of arsenic could originate from agricultural and
326 domestic discharges while large or less amounts of chromium could originate from industrial
327 discharges. Also some of these heavy metals might be brought to the ocean by inorganic
328 and, or organic matters brought to the ocean by rain water and surface road run offs.

3.5 Correlations Analysis for the Essential Elements

330 Spearman correlation tests were carried out to determine the nature, magnitude and
331 directions of correlations among the essential elements. At Kunduchi fish market, strong,
332 positive but not statistically significant correlations were recorded between calcium and zinc
333 (r = 0.56; p = 0.095) and between copper and zinc (r = 0.59; p = 0.072). At Kivukoni fish
334 market, moderately positive but not statistically significant correlations were recorded
335 between copper and iron (r = 0.33; p = 0.33) and between copper and zinc (r = 0.44; p =
336 0.19). Furthermore, at Bagamoyo fish market, moderately positive but not statistically
337 significant correlations were recorded between calcium and selenium (r = 0.32; p = 0.36) and
338 between calcium and zinc (r = 0.38; p = 0.28). These correlations suggests that, these
339 essential elements do not originate from common sources, rather, they originate from
340 diversified sources, each source contributing more or less to the levels of the essential
341 elements. These sources could include natural activities such as seafloor and bedrock
342 dredging, soil erosions, weathering and rocks disintegrations. Also some essential elements
343 could be related to organic matters brought to the ocean by agricultural drain water; surface
344 road run offs and rain water.

345   **3.6 Comparison of the mean Concentrations of the Essential Elements to the**
346   **Maximum Tolerable Levels (MTLs) Sets by International Organizations**
347   Essential element can have adverse health effects when they exceed the required levels in
348   the body [21,36]. The table **6** indicates the mean concentrations of the essential elements
349   with their respective (MTLs) set by international organizations.

350   **Table 6 Mean concentrations of the essential elements with their (MTLs) set by**
351   **international organizations**

FISH MARKETS	CHEMICAL SYMBOL	ATOMIC NUMBER	ELEMENT	MEAN CONC. (µg/g)	MTLs (µg/g)	REFFERENCES
KIVUKONI	Ca	20	Calcium	37,981.88	130,000	[37]
	Fe	26	Iron	149.99	200	[38]
	Cu	29	Copper	4.02	30	[39]
	Zn	30	Zinc	159.84	40	[39]
	Se	34	Selenium	1.73	0.75	[40]
KUNDUCHI	Ca	20	Calcium	303,800.50	130,000	[37]
	Fe	26	Iron	211.91	200	[38]
	Cu	29	Copper	2.57	30	[39]
	Zn	30	Zinc	105.06	40	[39]
	Se	34	Selenium	2.19	0.75	[40]
BAGAMOYO	Ca	20	Calcium	38,310.85	130,000	[37]
	Fe	26	Iron	234.78	200	[38]
	Cu	29	Copper	3.45	30	[39]
	Zn	30	Zinc	130.96	40	[39]
	Se	34	Selenium	1.96	0.75	[40]

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353

354   From the table above, it can be seen that, the mean concentrations of zinc and selenium are
355   higher than their respective (MTLs) set by various international organizations. The mean
356   concentrations of zinc for samples from all fish markets are at least 3 times higher than the
357   recommended MTL for zinc (40µg/g) according to FAO, [39]. The mean concentration of
358   selenium is 2times higher than its MTL (0.75µg/g) according to COMA, [40]. On the other
359   hand, the mean concentrations of iron for samples from Kunduchi and Bagamoyo fish
360   markets are just above their respective MTL (200µg/g) according to FAO/WHO, [39]. The
361   mean concentrations of iron for samples from Kivukoni fish market are just below its MTL of
362   200µg/g according to FAO/WHO, [38]. The   implication of these findings is that, diets must
363   be properly controlled due to the fact that, excessive amounts of essential elements can
364   have adverse health effects.

365   **3.7 Comparison of the mean concentrations of the toxic heavy metals to the**
366   **maximum tolerable levels (MTLs) sets by international organizations**

367   Bearing in mind a wide array of adverse health effects that can arise from heavy metals
368   pollution, this study has compared the mean concentrations of the toxic heavy metals to
369   MTLs set by international organizations. Table **7** shows the mean concentrations of the toxic
370   heavy metals with their respective MTLs set by international organizations.

371   **Table 7 Mean concentrations of the toxic heavy metals with their (MTLs) set by**
372   **international organizations**

FISH MARKETS	CHEMICAL SYMBOL	ATOMIC NUMBER	ELEMENT	MEAN CONC. (µg/g)	MTLs (µg/g)	REFFERENCES
KIVUKONI	Al	13	Aluminum	52.75	100	[41]
	Cr	24	Chromium	2.09	0.15	[15]
	Ni	28	Nickel	0.90	2.00	[42]
	As	33	Arsenic	3.79	0.50	[36]
	Cd	48	Cadmium	0.58	0.5	[36]
	Pb	82	Lead	BDL	2.00	[43]
KUNDUCHI	Al	13	Aluminium	105.37	100	[41]
	Cr	24	Chromium	21.71	0.15	[15]
	Ni	28	Nickel	0.27	2.00	[42]
	As	33	Arsenic	7.22	0.50	[36]
	Cd	48	Cadmium	2.64	0.50	[36]
	Pb	82	Lead	BDL	2.00	[43]
BAGAMOYO	Al	13	Aluminium	129.33	100	[41]
	Cr	24	Chromium	27.15	0.15	[15]
	Ni	28	Nickel	0.84	2.00	[42]
	As	33	Arsenic	5.29	0.50	[36]
	Cd	48	Cadmium	0.70	0.50	[36]
	Pb	82	Lead	BDL	2.00	[43]

373   BDL for Pb = 0.50 µg/g
374
375   From the table 7, one can see that, the mean concentrations of arsenic and cadmium are
376   higher than their respective MTL of (0.50µg/g) according to [37] for samples from all fish
377   markets. The mean concentrations of arsenic are more at least 7times greater than their

14

378   respective MTL for samples from all fish markets. The mean concentrations of cadmium are
379   5times greater than their MTL for samples from Kunduchi fish market and are just above
380   their MTL for samples from Kivukoni and Bagamoyo fish markets. The mean concentrations
381   of aluminium for samples from Kunduchi and Bagamoyo fish markets are just above their
382   MTL, while, the mean concentration of aluminium for samples from Kivukoni fish market are
383   below their MTL of (100μg/g) according to [41]. Inorganic arsenic is very toxic even in low
384   concentrations. At higher concentrations it can cause infertility, heart disruptions, brain
385   damage and ultimate death. Actually inorganic arsenic is classified as a human carcinogen
386   [3]. Aluminium is a toxic heavy metal with no beneficial effects in the human body. When
387   ingested, it stays in the body with a biological half life of 50 years. Furthermore, cadmium is
388   classified as a human carcinogen [18,34]. When ingested, cadmium is efficiently retained in
389   the kidney and liver with a biological half life of 30 years [18].

390   These findings suggest that, a further detailed research is needed to determine the factors
391   which caused elevated levels of arsenic; cadmium and aluminium for samples from these
392   fish markets and how to prevent pollutions of the sea foods by heavy metals in order avoid
393   the adverse health effects to consumers of sea foods.

394   **3.8 Comparison of the mean concentrations of the analyzed elements from different**
395   **parts of the world**

396   The mean concentrations of the elements analyzed in this study were compared to the mean
397   concentrations of the same elements in samples of sardine fish collected from the different
398   parts of the world. Table **8** shows the summary of this comparison.

399   **Table 8 Comparisons of the mean concentrations (mg/kg) of the analyzed elements**
400   **from different parts of the world**

SAMPLING AREA	ANALYZED ELEMENTS											REFFERENCE
	Al	Ca	Cr	Fe	Ni	Cu	Zn	As	Se	Cd	Pb	
KIVUKONI	52.75	37,981.88	2.01	149.99	0.92	4.02	159.84	3.79	1.73	0.58	BDL	This study
KUNDUCHI	105.37	3038.45	21.71	211.71	0.27	2.57	105.06	7.20	2.19	2.64	BDL	This study
BAGAMOYO	129.33	3831.85	27.15	234.78	0.84	3.45	130.96	5.29	1.96	0.70	BDL	This study
ZANZIBAR PORT	-	-	22.30	311.90	86.80	6.6	-	-	-	BDL	1.70	[19]
ABUJA	-	-	-	3.99	-	0.311	1.89	-	-	-	BDL	[44]

FUKUSHIMA	-	-	0.60	-	0.1	9.6	130	7.5	6.6	0.01	-	[31]
EGYPT	-	-	0.22	-	-	-	2.37	-	-	0.048	-	[45]
TURKEY	-	-	22.16	-	-	23.27	-	-	0.01	-	0.20	[46]

401   BDL for Pb = 0.50 µg/g

402 As for the toxic heavy metals, it can be seen from the above table that, the mean
403 concentrations of cadmium (Cd) for samples from Kivukoni, Kunduchi and Bagamoyo fish
404 markets are higher than the values reported in the literatures reviewed in this study.
405 Furthermore, samples from Bagamoyo fish market had higher mean concentration of
406 chromium (Cr) compared to the values reported in different literatures reviewed in this study.
407 In general, the comparisons of the mean concentrations of the toxic heavy metals were in
408 the order;

409 Cr: Bagamoyo > Zanzibar port > Kunduchi > Kivukoni > Fukushima > Egypt
410 As: Fukushima > Kunduchi > Bagamoyo > Kivukoni
411 Ni: Zanzibar port > Kivukoni > Bagamoyo > Kunduchi > Fukushima
412 Cd: Kunduchi > Bagamoyo > Kivukoni > (Turkey and Fukushima) > Egypt

413 The toxic heavy metal (Pb) have not been involved in this comparison because, its
414 concentrations were below the detection limits of the spectrometer used in this study. On the
415 other hand, the mean concentrations of aluminium (Al) were not reported in the literatures
416 reviewed in this study.

417 As for the essential elements, it can be seen from the table above that, samples from
418 Kivukoni fish market had higher mean concentrations of zinc compared to the values
419 reported in the literatures reviewed in this study. Generally, the mean concentrations of the
420 essential elements were in the order:

421 Fe: Zanzibar port > Bagamoyo > Kunduchi > Kivukoni > Abuja > Turkey
422 Zn: Kivukoni > Bagamoyo > Fukushima > Kunduchi > Turkey > Egypt > Abuja
423 Se: Fukushima > Kunduchi > Bagamoyo > Kivukoni
424 The essential element (Ca) was not reported in the literatures reviewed in this study and
425 hence was not covered in this comparison.
426
427 **4. CONCLUSION**
428
429 This study has assessed the concentrations of the essential elements (Ca, Cu, Fe, Se and
430 Zn) as well as the toxic heavy metals (Al, As, Cd, Cr, Ni and Pb) in samples of sardine fish
431 collected from Kivukoni, Kunduchi and Bagamoyo fish markets. The mean concentrations of
432 the analyzed elements were compared to their respective maximum tolerable levels (MTLs)
433 set by international organizations. The mean concentrations of the essential elements (Ca
434 and Cu) were found to be lower than their respective MTLs while the mean concentrations of
435 Zn were higher than their MTLs for samples from all fish markets. The mean concentrations
436 of Fe and Se were higher than their respective MTLs for samples from Kunduchi and
437 Bagamoyo fish markets. The respective MTLs for these essential elements were; 130,000
438 µg/g (for Ca), 200 µg/g (for Fe), 30 µg/g (for Cu), 0.75 µg/g (for Se), and 40 µg/g (for Zn)
439 according to FAO/WHO [38], WHO [15] , COMA [40] and FAO/WHO [37]

440 Zinc (Zn) and Selenium (Se) occurs naturally in rocks, soil and water due to natural activities
441 such as weathering and abrasion of rocks [23,35]. Most zinc compounds are soluble in
442 water, though, the metal itself is not [23]. The elevated levels of zinc recorded for samples
443 from all fish markets may be due to the fact that, most of zinc compounds are soluble in
444 water and hence, are easily ingested by marine organisms. Also elevated levels of zinc may
445 be associated to anthropogenic activities such as corrosions of zinc galvanized ships and
446 boats, painting of ships and boats, improper damping of zinc containing compounds such as
447 fertilizers and batteries as well as domestic and industrial discharges.

448 Selenium occurs naturally due to erosion of rocks containing salinities and serenades which
449 are associated with sulphide minerals. Its abundance in the earth's crust is very low,
450 estimated as 0.05µg/kg to 0.1µg/kg [48]. Since, the abundance of selenium in the earth's
451 crust is very low, large percentages of selenium are brought to the ocean by glaciations,
452 floods, agricultural drain waters, industrial discharges and domestic discharges.

453 The mean concentrations of the toxic heavy metals arsenic and cadmium for samples from
454 all fish markets were found to be higher than their respective MTL of 0.50µg/g[36]. Arsenic in
455 marine fish is normally in higher concentrations than in fresh water fish. Arsenic in fish is
456 mostly found in organic form which is less toxic compared to inorganic arsenic [31].
457 Combinations of natural activities such as weathering, volcanic eruptions and biological
458 activities are the main sources of arsenic into the aquatic environments [49]. Also arsenic is
459 brought into the aquatic environments by floods, domestic and industrial sewages and from
460 human activities such as mining, agriculture (use of arsenic based pesticides) and improper
461 damping of arsenic based compounds [49]. Cadmium is widely used in batteries, coating,
462 and plating [33]. Cadmium levels in the aquatic environments are raised mainly due spills
463 from hazardous waste sites and industrial discharges [34].

464 The mean concentration of aluminium for samples from Kunduchi and Bagamoyo fish
465 markets were found to be higher than its respective MTL of 100µg/g according to CFS 2009.
466 Aluminium is the third most abundant element in the earth's crust and it occurs naturally in
467 rocks, soil and water. Apart from natural activities, aluminium is brought into the aquatic
468 environments by surface road run offs, domestic and industrial discharges carrying
469 chemicals such as antacids or antiperspirants and coagulants used in water treatment
470 processes [29]. On the other hand, the concentrations lead was found to be below the
471 detection limits of 0.50µg/g for the spectrometer used in this study.

472 It can be fairly concluded that, sardine fish from Kivukoni, Kunduchi and Bagamoyo fish
473 markets are rich in essential elements Ca, Cu, Fe, Se and Zn. The sardine fish therefore can
474 contribute significantly to the food and nutritional security of the coast inhabitants provided
475 that, measures are taken to prevent them from being polluted by heavy metals.

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