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1 Human health risks from metals and metalloid via consumption of food animals near Gold
2 Mines in Tarkwa, Ghana: Estimation of the daily intakes and target hazard quotients
3 (THQs)

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24

25 **Abstract**

26

27 Heavy metal and metalloid contamination in food resulting from mining is of major
28 concern due to the potential risk involved. Food consumption is the most likely route for
29 human exposure to metals. This study was therefore to estimate the daily intake and health
30 risk (based on target hazard quotients, THQ) from metals via consumption of free-range
31 chicken, goat and sheep near gold mines in Tarkwa, Ghana. The concentrations of Cr, Mn,
32 Fe, Co, Ni, Cu, Zn, As, Cd, and Pb were measured with an inductively coupled plasma-
33 mass spectrometer and Hg analysis was done using the mercury analyzer. The mean
34 concentrations of metals ranged from nd–542 mg/kg wet weight. Principal component
35 analysis of the results showed a clear separation between chicken, grouped on one side, and
36 the ruminants clustered on another side in both offal and muscle. Interestingly, As, Cd, Hg,
37 Mn and Pb made one cluster in the offal of chicken. Chicken muscle also showed similar
38 distribution with As, Hg and Pb clustered together. The daily intake of As ($\mu\text{g}/\text{kg}$ body
39 weight/day) were in the following ranges; [0.002 (kidneys of goat and sheep)–0.19 (chicken
40 gizzard)], Cd [0.003 (chicken muscle)–0.55 (chicken liver)], Hg [0.002 (goat muscle)–0.29
41 (chicken liver)], Pb [0.01 (muscles and kidneys of goat and sheep)–0.96 (chicken gizzard)]
42 and Mn [0.13 (goat kidney)–8.92 (sheep liver)]. From the results, daily intake of As, Cd,
43 Hg, Pb and Mn in these food animals were low compared to the provisional tolerable daily
44 intake guidelines. The THQs although less than one, indicated that contributions of chicken
45 gizzard and liver to toxic metal exposure in adults and especially children could be
46 significant.

47

48 *Keywords:* Offal, Muscle, Estimated daily intake, Target hazard quotient, Ghana

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51 **1. Introduction**

52 Heavy metals and metalloid (arsenic) are ubiquitous in the environment either naturally
53 or anthropogenically, and their concentrations were elevated promptly through waste
54 disposal, smelter stacks, atmospheric deposition, mining, fertilizer and pesticide use and the
55 application of sewage sludge in arable land ([Cui et al., 2005](#)). Mining and processing metal
56 ore can be a significant source of metal and metalloid contamination of the environment
57 ([Navarro et al., 2008](#); [Singh et al., 2005](#)). The increased occurrence of metal and metalloid
58 pollution in the environment has been associated with anthropogenic activities as effluents
59 and emissions from mines and smelters often contain elevated concentrations of toxic
60 metals including arsenic, cadmium, mercury and lead ([Tubaro and Hungerford, 2007](#)).

61 Food consumption has been identified as the major pathway of human exposure,
62 accounting for >90% compared to other ways of exposure such as inhalation and dermal
63 contact ([Loutfy et al., 2006](#)). In a review by [Yabe et al. \(2010\)](#), contamination of food
64 animals, fish, soil, water, and vegetables with heavy metals has reached unprecedented
65 levels over the past decade in some parts of Africa. As a result, human exposure to toxic
66 metals has become a major health risk.

67 Tarkwa (05°18'00"N; 01°59'00"W) is a town in the southwest of Ghana, located about
68 120 miles west of the capital city, Accra. As of 2010, Tarkwa was estimated to have a
69 population of 90,477 ([Ghana Statistical Service, 2010](#)). It is a noted centre of gold and
70 manganese mining. Tarkwa mine, which is a large open-cast gold mine, is situated to the
71 northwest of the town, and Nsuta manganese mine is situated to the east. Tarkwa has nearly
72 a century of gold mining history and has the largest concentration of mining companies in a
73 single district in Ghana and the West African sub-region ([Akabzaa and Darimani, 2001](#)). A
74 study conducted by [Hayford et al. \(2008\)](#) on the impact of the gold mining on soil and
75 staple foods collected around mining communities in Tarkwa showed high levels of some
76 toxic metals including arsenic and mercury in the soil and staple foods. In Tarkwa, the
77 extraction and processing of gold has given rise to various environmental related diseases
78 and accidents. According to the District Medical Officer of Health, the common mining-
79 related diseases observed in the area over the years include, but are not limited to: vector-
80 borne diseases such as malaria, schistomiasis and onchocerciasis; respiratory tract diseases,

81 especially pulmonary tuberculosis and silicosis; skin diseases; eye diseases, especially
82 acute conjunctivitis; accidents resulting from galamsey activities, and mental cases
83 ([Akabzaa and Darimani, 2001](#)). The aim of this study was therefore to determine the
84 concentrations of heavy metals and metalloid in offal and muscle of free-range chicken,
85 goat and sheep; to reveal the species differences in accumulation features of metals among
86 free-range chicken, goat and sheep; and to estimate the daily intake (EDI) and target hazard
87 quotient (THQ) (toxic metals) through consumption of these food animals in Tarkwa.

88

89 **2. Materials and methods**

90

91 *2.1. Sampling and pre-treatment*

92 In June 2012, liver, kidney and muscle samples were collected at slaughterhouses from
93 both male and female local goat and sheep ($n = 10$ species each) aged three years and above,
94 and stored in labeled plastic bags in cooler boxes. 10 free-range chickens were also bought
95 from townships surrounding the two major gold mines. All the townships were close to the
96 mine, and some households were within 1 km of the mine. The live adult chickens were
97 transported to the laboratory for dissection after exsanguination. Liver, kidney, muscle and
98 gizzard samples were collected. Samples were transported to the laboratory and kept frozen
99 at the Chemistry Department of the Kwame Nkrumah University of Science and
100 Technology, KNUST, Ghana before being transported and analyzed for metal
101 concentrations at the Laboratory of Toxicology, Graduate School of Veterinary Medicine,
102 Hokkaido University, Japan.

103

104 *2.2. Analysis of total Mercury (Hg)*

105 The concentration of total mercury (Hg) was measured by thermal decomposition, gold
106 amalgamation and atomic absorption spectrophotometry (Mercury analyzer, MA-3000;
107 Nippon Instruments Corporation, Tokyo, Japan), after preparation of the calibration
108 standard. Recovery rates of Hg for the certified reference material, DOLT-4 (Dogfish liver,
109 the National Research Council, Canada) ranged from 92% to 103% ($94.3 \pm 4.2\%$). The
110 detection limit (Hg) was 2.0 pg of total Hg.

111

112 *2.3. Sample preparation and metal analysis*

113 Approximately 0.5 g of individual samples were dried in an oven at 40°C and digested
114 in a closed microwave extraction system, Speed Wave MWS-2 microwave digestion
115 (Berghof, Germany). Briefly, the dried samples were placed in prewashed digestion vessels
116 followed by acid digestion using 5 mL of (65%) nitric acid, HNO_3 (Kanto Chemical Corp.,
117 Tokyo, Japan) and 1 mL of (30%) hydrogen peroxide, H_2O_2 (Kanto Chemical Corp., Tokyo,
118 Japan). The digestion vessels were capped and placed into a 10-position turntable

119 conditions followed by a ramped temperature programme: ramp to 160 °C (5 min hold);
120 and increase to 190 °C (15 min hold). After cooling, samples were transferred into plastic
121 tubes and diluted to a final volume of 10 mL with milli Q water. A reagent blank was
122 prepared using the same procedure. An Inductively Coupled Plasma-Mass Spectrometer
123 (ICP-MS; 7700 series, Agilent technologies, Tokyo, Japan) was used for quantification.
124 The instrument was calibrated using standard solutions of the respective metals (to establish
125 standard curves before metal analysis). All chemicals and standard stock solutions were of
126 analytical-reagent grade (Wako Pure Chemicals). The detection limits ($\mu\text{g/L}$) of chromium
127 (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic
128 (As), cadmium (Cd), and lead (Pb) were 0.003, 0.025, 0.154, 0.0004, 0.024, 0.007, 0.226,
129 0.002, 0.001 and 0.001 respectively. Concentrations of metals were expressed in mg/kg wet
130 weight (mg/kg ww)

131

132 *2.4. Quality assurance and quality control*

133 For heavy metals, replicate blanks and the reference materials DORM-3 (Fish protein,
134 the National Research Council, Canada) and DOLT-4 were used for method validation and
135 quality control. Replicate analysis of these reference materials showed good accuracy
136 (relative standard deviation, RSD, $\leq 3\%$) and recovery rates ranged from 80% to 115%.

137

138 *2.5. Data analysis*

139 *2.5.1 Estimated daily intake (EDI) of toxic metals*

140 The estimated daily intake (EDI) of toxic metals (As, Cd, Hg, Pb and Mn) depended on
141 both the metal concentration in offal or muscle and the amount of consumption of the
142 respective food. The EDI of metals was determined by the following equation:

$$EDI = \frac{MC \times FDC}{BW}$$

143 where MC is average concentration of metal in food ($\mu\text{g/g}$, on fresh weight basis); FDC
144 represents the average food daily consumption of offal and muscle in this region
145 (g/person/d); BW is the average body weight. It was assumed that the local inhabitants
146 consumed an average liver, muscle and gizzard of 150 and 100 g/day for adult (60 kg in
147 BW) and children (30 kg in BW) respectively. For kidney, it was assumed that inhabitants

148 consumed 10 and 8 g/day for adults and children respectively ([Ihedioha and Okoye, 2013](#)),
149 because the size of kidneys are generally smaller and is not favorite compared with liver,
150 muscle, or gizzard. The metal intakes were compared with the tolerable daily intakes for
151 metals recommended by the [FAO/WHO \(2010\)](#), [WHO \(2000\)](#) and [FSA \(2006\)](#).

152

153 *2.5.2 Target Hazard Quotient (THQ)*

154 The health risks from consumption of offal and muscle of chicken, goat and sheep by
155 the local inhabitants were assessed based on the THQ. The THQ is a ratio of determined
156 dose of a pollutant to a reference dose level. If the ratio is less than 1, the exposed
157 population is unlikely to experience obvious adverse effects. The method of estimating risk
158 using THQ was provided in the U.S. EPA Region III risk-based concentration table
159 ([USEPA IRIS, 2007](#)) and in [Chien et al., \(2002\)](#), and is based on the equation below:

$$THQ = \frac{EFr \times ED \times FIR \times MC}{RfD \times BW \times AT} \times 10^{-3}$$

160 where THQ is target hazard quotient; EFr is exposure frequency (365 days/year); ED is
161 exposure duration (70 years); FIR is food ingestion rate (g/person/d); MC is average
162 concentration of metal in food (µg/g, on fresh weight basis); RfD is the oral reference dose
163 (mg/kg/d); BW is the average body weight, adult (60 kg); children (30 kg); AT is the
164 average exposure time (365 days/year × number of exposure years, assuming 70 years in
165 this study). Oral reference doses were based on 3E-04, 1E-03, 5E-04, 4E-03, 0.14 mg/kg/d
166 for As, Cd, Hg, Pb and Mn respectively ([USEPA, 1997](#); [USEPA IRIS, 2007](#)).

167

168 *2.6. Statistical analysis*

169 IBM SPSS 20.0 Statistical software (SPSS Inc., Illinois, USA) was used to perform
170 ANOVA analysis followed by Tukey's test. ANOVA and Tukey analyses were used to
171 compare metal concentrations in the liver, kidney, muscle and gizzard and differences were
172 considered statistically significant with *p* value < 0.05. Principal component analysis (PCA)
173 was done to determine the distribution pattern of metals in organs using JMP statistical
174 software v. 10 (SAS Institute).

175

176 **3. Results and discussion**

177

178 *3.1. Levels of heavy metals and metalloid*

179 Concentrations of Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb in the liver, kidney
180 muscle and gizzard of free-range chickens, as well as metal and metalloid levels in liver,
181 kidney and muscle of goat and sheep from Tarkwa were determined ([Table 1](#)). [Table 1](#)
182 shows that the mean Cd concentrations were in the range of nd–0.73 mg/kg ww in all
183 samples while the mean concentrations of Pb were in the range of nd–0.39 mg/kg ww, Cr
184 (0.01–0.24), Mn (0.15–3.57), Fe (6.62–542.31), Co (nd–0.11), Ni (0.01–0.55), Cu (0.35–
185 105.8), Zn (5.12–65.89), As (nd–0.14) and Hg (nd–0.12) mg/kg ww. Cu and Zn were
186 detected in 100% of all offal and muscle samples for all the food animal studied.

187 Ni can cause respiratory problems and is a carcinogen. The permissible limit of Ni in
188 food according to [WHO \(2000\)](#) standard is 0.5 mg/kg. In this study concentrations of Ni
189 ranged between 0.01–0.55 mg/kg ww, in chicken liver and muscle respectively. As shown
190 in [Table 1](#), the highest mean concentration of Ni was found in the local chickens while Zn
191 levels in goat muscle exceeded the permissible limit of 50 mg/kg in meat ([USDA, 2006](#);
192 [OJEC, 2001](#); [European Commission Regulation, 2006](#)).

193

194 *3.1.1. Arsenic*

195 As was detectable in 100%, 83%, 80% and 52% of all gizzard, liver, kidney and muscle
196 samples respectively. Concentrations of As in muscle and offal samples of free-range
197 chicken, goat and sheep ranged from nd to 0.14 ± 0.11 mg/kg ww in goat muscle and
198 chicken kidney respectively ([Table 1](#)). The As distribution in liver, kidney and muscle of
199 chicken was found to be higher and differ statistically with that of goat and sheep ($p < 0.05$).
200 However, there was no significant difference when As levels in goat offal and muscle was
201 compared with sheep ([Table 1](#), $p > 0.05$). Mean concentration of As in the kidney ($0.14 \pm$
202 0.11 mg/kg ww) and gizzard (0.08 ± 0.05 mg/kg ww) of free-range chicken from Tarkwa
203 were found to be higher than the [USDA \(2006\)](#); [OJEC \(2001\)](#); [European Commission](#)
204 [Regulation \(2006\)](#) and [SAC/MOHC, 2005](#) standard of 0.05 mg/kg ww in food. The high
205 levels of As in the free-range chicken may be due to the nature of the gold bearing ore

which is mineralized pyrites and arsenopyrates. Processing of the ore involves roasting and this results in the production of arsenic trioxide gas which is distributed throughout the study area by air current. As is a toxic substance and due to its non-biodegradable nature, it could accumulate in surface soil and water. It could also enter the food chain through plant assimilation ([Amonoo-Neizer et al., 1995](#); [Griffis et al., 2002](#)). This could be the possible reason for the high As concentrations in free-range chickens from Tarkwa as they continuously pick food particles and water from the ground.

213

214 *3.1.2. Cadmium*

Mean level of Cd in chicken kidney (0.73 ± 0.81 mg/kg ww) was within the recommended residual concentrations of 0.5 to 1.0 mg/kg ww in offal for human consumption ([European Commission, 2001](#)). Cd was detectable in 14% of all muscle, 100% of all liver and kidney, 90% of chicken gizzard. The concentrations of Cd in livers and kidneys of chicken were significantly higher compared to those of goat and sheep (Tukey test; $p < 0.05$). The higher frequencies of Cd in liver and kidney (100%) are likely due to their special functions; liver as storage organ and kidney as excretory organ ([Stoyke et al., 1995](#)). There have also been suggestions that animals exposed to Cd accumulate it in their kidneys because of the presence of free protein-thiol groups which leads to a strong fixation of heavy metals ([Pompe-Gotal and Crnic, 2002](#)). The likely unidentified sources of Cd which the animals could come in contact with include municipal waste, electroplating, plastic and paint waste. Other sources include leachates from nickel–cadmium based batteries and cadmium plated items which are so carelessly discarded by battery chargers and users in Ghana. Recently, electronic wastes are disposed and often burnt at refuse dumps.

230

231 *3.1.3. Mercury*

Hg is a neurotoxic poison that causes neurobehavioral effects (especially on psychomotor coordination), neuroendocrine, and renal damage and gastrointestinal toxicity ([SAC/MOHC, 2005](#)). Hg was detectable in 38% of all muscle, 69% and 97% of all liver and kidney respectively, and 100% of chicken gizzard. The levels of Hg in free-range

236 chicken from Tarkwa were higher compared to that of goat and sheep. The result of this
237 study indicated that chicken kidney (0.12 ± 0.08 mg/kg ww) and liver (0.11 ± 0.07 mg/kg
238 ww) contained the highest concentrations of Hg followed by sheep kidney (0.07 ± 0.07
239 mg/kg ww). The concentrations of Hg in liver and muscle of free-range chicken from
240 Tarkwa was found to be higher and differed statistically with that of goat and sheep (Tukey
241 test; $p < 0.05$). However, there was no significant difference when Hg levels in goat offal
242 and muscle was compared with sheep (Tukey test; $p > 0.05$). The lowest and highest mean
243 concentrations of Hg in chicken were 0.01 ± 0.01 mg/kg ww in muscle and 0.12 ± 0.08
244 mg/kg ww in kidney, respectively; goat (nd– 0.03 ± 0.02 in muscle and kidney
245 respectively); sheep (nd– 0.07 ± 0.07 in the muscle and kidney respectively) (Table 1). The
246 [USDA \(2006\)](#); [OJEC \(2001\)](#); and [European Commission Regulation \(2006\)](#) permissible
247 limit for Hg is 0.05 mg/kg and the Standardization Administration of China/Ministry of
248 Health of China established maximum limits of Hg in food to be 0.01 to 0.05 mg/kg
249 ([SAC/MOHC, 2005](#)). The high levels of Hg especially in free-range chicken could be
250 problematic, as concentration levels exceeded the maximum values permitted in food. In
251 Ghana, amalgamation is the preferred gold recovery method employed by almost all
252 artisanal gold miners because it is a very simple, inexpensive and an easier to use technique.
253 These activities by local and small scale miners are widespread, and Hg is introduced into
254 the environment (via amalgamation process popularly known as "galamsey") ([Amonoo-](#)
255 [Neizer et al., 1995](#)). The high levels of Hg in offal of free-range chicken could therefore be
256 due to contamination from soil, feed or water.

257

258 3.1.4. Lead

259 Excess Pb is known to reduce the cognitive development and intellectual performance in
260 children and to increase blood pressure and cardiovascular disease incidence in adults
261 ([Commission of the European Communities, 2001](#)). Pb was detectable in all samples of
262 liver, kidney, 80% of chicken gizzard and 38% of muscle. The result of this study indicated
263 that chicken gizzard (0.39 ± 1.17 mg/kg ww) contained the highest concentrations of Pb
264 followed by chicken kidney (0.26 ± 0.31 mg/kg ww) and liver (0.13 ± 0.14 mg/kg ww).
265 This is because, to be toxic, Pb must be in a form which will be retained in the gizzard, thus

resulting in continuous absorption over a prolonged period ([Salisbury et al., 1958](#)). In another study by [Kendall et al. \(1996\)](#), analyses of field and laboratory data indicated that two scenarios can be envisioned when Pb is ingested: (1) the ingested grit/pellets containing Pb might be eliminated (via regurgitation or passage through the gastrointestinal tract) before any significant dissolution or absorption of Pb occurs; (2) the grit/pellets may be partially or totally dissolved in the gizzard, in which case causes Pb accumulation resulting in the occurrence of a range of toxicologic effects.

The levels of Pb in organs of free-range chickens may emanate mainly from contamination of soil, feeds and/ or water sources. Tukey's test was used to compare the levels of Pb in the three food animals, and results showed no statistical significance in liver and muscle when chicken was compared with goat and sheep ($p > 0.05$). In this study, concentration of Pb in some organs of chicken exceeded the [USDA \(2006\)](#); [OJEC \(2001\)](#); and [European Commission Regulation \(2006\)](#) standard of 0.5 and 0.1 mg/kg ww for Pb in offal and muscle respectively ([Table 1](#)). Generally, the low levels of Pb in the offals and muscles of the mentioned food animals could be due to the fact that, Pb accumulates in bone and the metal concentration increases with age ([Caggiano et al., 2005](#); [Rubio et al., 1998](#)).

283

284 *3.1.5. Manganese*

285 The role of Mn in neuro-psychiatric disorders is also documented ([Schild, 1980](#); [Jackson and Morris, 1989](#)). Mn was detectable in 100% of all offal and muscle samples. This study 286 showed that mean concentrations of Mn ranged from 0.15–3.51 (chicken); 0.62–2.43 (goat) 287 and 0.71–3.57 mg/kg ww (sheep) ([Table 1](#)). The concentration of Mn in kidney of free- 288 range chicken was found to be higher and differed statistically with that of goat and sheep 289 ($p < 0.05$). However, there was no significant difference when Mn levels in the muscle of 290 the three food animals were compared ([Table 1](#), $p > 0.05$). In the liver, the levels of Mn 291 between free-range chickens and sheep was statistically insignificant with mean values of 292 3.50 ± 0.92 and 3.57 ± 1.02 mg/kg ww respectively ([Table 1](#), $p > 0.05$). The mean 293 concentrations of Mn in offal and muscles of all three food animals were above the [WHO](#) 294 ([1996](#)) reference standard of 0.5 mg/kg except in chicken muscle ([Table 1](#)). The high Mn 295

296 levels in these food animals could be due to the fact that the sampling site at Tarkwa was
297 close to the Nsuta Mn mine (which is a famous site for Mn mining).

298 This study was compared with similar studies in different countries ([Table 2](#)). Results
299 from this study was found to be comparable with study by [Husain et al. \(1996\)](#), in goat and
300 sheep livers and kidneys, in Kuwait, but higher than similar study by [Villar et al. \(2005\)](#) in
301 the liver of poultry from the Philippines. Study done by [Yabe et al. \(2013\)](#) near a Pb and Zn
302 mine in Kabwe, Zambia indicated that the levels of Cd and Pb in the livers and kidneys of
303 free-range chickens were higher compared to this study but levels in muscle were
304 comparable. On the other hand, the levels of Pb and Cd in chicken gizzard from this study
305 was higher than the study by [Yabe et al. \(2013\)](#). From [Table 2](#), the mean concentrations of
306 Zn in this study was comparable in all organs except for gizzard when compared to the
307 study by [Yabe et al. \(2013\)](#).

308

309 *3.2. Distribution of metals*

310 ANOVA was used to compare the accumulation and distribution of metals in the organs
311 of free-range chicken, goat and sheep. The metal distribution in offal and muscle of chicken
312 was statistically significant ($p < 0.05$) except for Cr, As and Pb. There was also statistical
313 significance ($p < 0.01$) in the distribution of Mn, Co, Cu, Zn and Cd when the organs of
314 goat were compared. On the other hand metal distribution in organs of sheep were
315 significant except for As ($p > 0.05$). The liver and kidneys are target tissues for monitoring
316 metal contamination in animals because both organs function in removing toxic metals
317 from the body ([Husain et al., 1996](#); [Abou-Arab, 2001](#)). Similarly, organs of all species in
318 this study showed that the liver and kidney contained the highest levels of metals ([Table 1](#)).

319 Distribution of metals in offal and muscle samples in the food animals was analyzed
320 using PCA. PCA results ([Fig. 1](#)) showed a clear separation between chicken (C), grouped
321 on one side, and the ruminants, goat (G), and sheep (S), clustered on another side in both
322 offal and muscle. Interestingly, As, Cd, Hg, Mn and Pb made one cluster or group in the
323 liver and kidney of free-range chicken ([Fig. 1](#)). Chicken muscle also showed similar
324 distribution pattern, with As, Hg and Pb clustered together ([Fig. 1](#)). This could be attributed
325 to the difference in their feeding habits and/ or the different levels of metallothionein (MT)

326 from the sampled food animals (species differences in hepatic MT content). MT is a low
327 molecular weight protein in various mammalian and non-mammalian tissues and is
328 involved in the detoxification of metals ([Margoshes and Valle, 1957](#); [Kagi and Vallee, 1960](#)). According to study by [Bryan-Henry et al. \(1994\)](#), the hepatic MT levels in these
329 three food animals studied decreased in order of livestock > non-mammals; i.e. goat >
330 sheep > chicken.
331

332

333 *3.3. Dietary intake of metals and target hazard quotient*

334 The estimated daily intakes (EDI) of toxic metals for adults in the vicinity of Tarkwa
335 mine through consumption of offal and meat of free-range chicken, goat and sheep are
336 presented in [Table 3](#). The highest EDIs of As (0.19 µg/kg bw/day), Pb (0.96 µg/kg bw/day),
337 Cd (0.55 µg/kg bw/day) and Hg (0.29 µg/kg bw/day), through consumption were from the
338 gizzard and liver of chicken ([Table 3](#)). Meanwhile, the highest EDI of Mn (8.92 µg/kg
339 bw/day) was through consumption of sheep liver ([Table 3](#)). The calculated EDIs of As, Cd,
340 Hg, Pb and Mn in offal and muscle samples of the 3 food animals from Tarkwa were low
341 compared to the [FAO/WHO \(2010\)](#), [WHO \(2000\)](#) and [FSA \(2006\)](#) tolerable daily intakes
342 ([Table 3](#)). Children are especially vulnerable to acute, sub-acute and chronic effects of
343 ingestion of chemical pollutants, since they (children) consume more (twice of the amount)
344 of food per unit of body weight as adults ([ENHIS, 2007](#)). As a result intakes of these toxic
345 metals through food could be higher for children from Tarkwa.

346 The health risk from consumption of offal and muscle from the three food animals were
347 assessed based on the THQ. THQ value less than 1 means the exposed population is
348 unlikely to experience obvious adverse effects ([USEPA, 2000](#)). Although the calculated
349 individual THQ were below 1.0 ([Fig. 2](#)), attention should be paid particularly to some high
350 individual THQ values approaching 1.0 (As Cd, and Hg). The THQ for As in chicken
351 gizzard and liver which is largely consumed was higher in children (0.85 and 0.84
352 respectively) than in adults (0.63 in both cases). Similarly, the THQ for Cd in the liver of
353 chicken was 0.74 for children compared with 0.55 for adults ([Fig. 2](#)). As shown in [Fig. 2](#),
354 THQ for Hg in the liver of chicken was 0.76 and 0.57 in children and adults respectively. In
355 spite of this, caution must be taken since perennial intake of these contaminated food

356 animals is likely to induce adverse health effects arising largely from As, Cd, Hg and Pb
357 exposure. Meanwhile, the THQ for Mn was very low among all the three food animals
358 (data not shown) ranging from 0.001–0.06 and 0.001–0.008 in adults and children
359 respectively. The THQ for As, Cd, Hg and Pb showed that contributions of chicken gizzard
360 and liver to toxic metal exposure in adults and especially children could be significant.

361

362 **4. Conclusion**

363 The dietary intakes of As, Cd, Hg, Pb and Mn from the offal and muscle of free-range
364 chicken, goat and sheep in Tarkwa, Ghana were low when compared to the [FAO/WHO](#)
365 ([2010](#)), [WHO \(2000\)](#) and [FSA \(2006\)](#) tolerable daily intakes. Children are especially
366 vulnerable to acute, sub-acute and chronic effects of ingestion of chemical pollutants, since
367 they (children) consume more (twice of the amount) of food per unit of body weight as
368 adults ([ENHIS, 2007](#)). As a result intakes of these four toxic metals through food could be
369 higher for children from Tarkwa. THQ of As, Cd and Hg showed that there could be
370 potential health risk for inhabitants through consumption of larger quantities of
371 contaminated offal of free-range chicken. Prolonged consumption of these toxic metals in
372 the offal and muscle may lead to accumulation and cause toxicity and therefore there is a
373 clear need to avoid consumption of these contaminated food animals, as well as restrict
374 them from roaming and scavenging for food and/or water near the mining areas.
375 Furthermore, continuous monitoring of As, Cd, Hg and Pb residues (in these food animals)
376 in the vicinities of gold mines in Tarkwa is recommended in the interest of consumers.

377

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387

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- 503

504 **Figure captions:**
505
506 **Fig. 1.** Distribution patterns of metals in food animals characterized by PCA (C: chicken;
507 G: goat and S: sheep).
508
509 **Fig. 2.** Target Hazard Quotient (THQ) of As, Cd, Hg and Pb in children and adults (C:
510 chicken; G: goat; S: sheep; G: gizzard; K: kidney; L: liver and M: muscle).
511
512
513

Table 1

Mean concentrations and range of metals (mg/kg ww) in various organs of free range chicken (C), goat (G) and sheep (S).

Sample		Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Cd	Hg	Pb
CG(n=10)	Mean±SD	0.06±0.08	0.64±0.46	81.4±87.2	nd	0.10±0.04	1.92±0.81	30.3±3.5	0.07±0.05	0.08±0.09	0.01±0.01	0.38±1.17
	Range	0.01-0.27	0.20-1.75	37.1-321.4	nd-0.01	0.06-0.19	0.74-3.19	24.3-34.9	0.02-0.16	nd-0.30	0.01-0.03	nd-3.70
CK(n=10)	Mean±SD	0.24±0.40	2.44±0.55 ^a	94.4±33.2	0.03±0.01	0.07±0.14	2.82±0.37	28.4±3.1	0.14±0.11 ^a	0.72±0.81 ^a	0.12±0.08 ^a	0.25±0.31 ^a
	Range	0.01-1.35	1.65-3.59	59.7-150.5	0.01-0.07	nd-0.46	2.01-3.38	25.2-35.6	0.02-0.37	0.16-2.26	0.02-0.28	0.02-1.05
GK(n=10)	Mean±SD	0.01±0.00	0.77±0.25 ^b	88.8±72.8	0.07±0.03	0.14±0.07	2.89±0.46	20.0±3.9	0.01±0.01 ^b	0.06±0.04 ^b	0.03±0.02 ^b	0.03±0.04 ^b
	Range	nd-0.02	0.50-1.35	39.2-278.5	0.01-0.12	0.05-0.27	2.42-3.76	15.0-27.5	nd-0.04	0.01-0.13	0.01-0.07	0.01-0.14
SK(n=10)	Mean±SD	0.01±0.00	1.14±0.27 ^b	49.1±15.4	0.03±0.02	0.09±0.04	3.170±0.45	22.2±4.1	0.01±0.01 ^b	0.15±0.15 ^b	0.07±0.08 ^{ab}	0.03±0.01 ^b
	Range	nd-0.01	0.87-1.70	24.8-80.6	0.01-0.06	0.04-0.15	2.58-3.90	18.1-28.1	nd-0.03	0.01-0.43	nd-0.23	0.01-0.06
CL(n=10)	Mean±SD	0.02±0.01	3.50±0.92 ^a	542.3±604.0	0.03±0.01	0.01±0.02	3.67±0.67	65.8±50.6	0.07±0.07 ^a	0.22±0.18 ^a	0.11±0.07 ^a	0.13±0.14 ^a
	Range	nd -0.04	1.57-4.84	72.0-1629.3	0.02-0.05	nd-0.06	2.72-4.74	32.5-182.2	0.01-0.21	0.05-0.56	0.05-0.25	0.01-0.41
GL(n=10)	Mean±SD	0.02±0.01	2.42±0.71 ^b	89.9±47.7	0.11±0.05	0.20±0.05	60.3±44.7	47.1±26.6	nd ^b	0.02±0.01 ^b	0.01±0.01 ^b	0.02±0.04 ^b
	Range	0.01-0.04	1.06-3.41	44.3-212.6	0.04-0.21	0.15-0.29	3.3-140.7	21.9-103.3	nd-0.01	0.01-0.04	nd-0.05	0.01-0.13
SL(n=10)	Mean±SD	0.02±0.01	3.57±1.02 ^a	126.3±71.3	0.07±0.03	0.33±0.13	105.8±74.3	43.6±9.6	0.01±0.01 ^b	0.05±0.02 ^b	0.03±0.03 ^b	0.06±0.03 ^{ab}
	Range	0.01-0.04	1.50-4.92	55.4-294.1	0.04-0.12	0.22-0.68	7.9-224.3	31.7-63.3	0.01-0.05	0.02-0.09	nd-0.09	0.02-0.11
CM(n=10)	Mean±SD	0.05±0.01	0.14±0.08 ^a	6.6±1.6	nd	0.55±0.30	0.34±0.03	5.1±1.7	0.04±0.03 ^a	nd ^a	0.01±0.01 ^a	0.01±0.03 ^a
	Range	0.02-0.09	0.09-0.38	4.7-9.5	nd	0.24-1.29	0.28-0.41	3.8-10.0	nd-0.13	nd	0.01-0.02	nd-0.10
GM(n=10)	Mean±SD	0.05±0.08	0.6±0.81 ^a	31.7±17.0	0.02±0.05	0.14±0.05	1.50±0.76	51.3±17.6	nd ^b	nd ^a	nd ^b	nd ^a
	Range	0.01-0.28	0.21-2.83	19.2-76.8	nd-0.15	0.08-0.23	0.91-3.38	17.0-78.2	nd-0.01	nd-0.02	nd	nd-0.01
SM(n=10)	Mean±SD	0.03±0.03	0.71±0.55 ^a	37.8±10.9	0.01±0.00	0.46±0.16	2.09±1.19	45.9±20.8	0.01±0.01 ^b	0.01±0.01 ^a	nd ^b	0.01±0.00 ^a
	Range	0.01-0.09	0.16-2.02	22.7-58.9	nd-0.01	0.15-0.66	0.84-4.85	27.0-83.4	nd-0.02	nd-0.03	nd-0.02	nd-0.01

n: number of samples; C: chicken; G: goat; S: sheep; K: kidney; L: liver; M: muscle.

different letters (a and b) between animals in the same organ indicates significant difference (Tukey's test; $p < 0.05$).

nd: not detected.

Table 2

Published data of mean metal levels (mg/kg dw) in organs of different animal groups from different regions.

Study/Country	Pollution Factor	Animal	Organ	Cr	Co	Ni	Cu	Zn	As	Cd	Hg	Pb
Yabe et al., 2013* , Zambia	mining	C (broiler)	L	0.06	0.05	0.03	56.80	28.60	0.00	0.00	0.06	0.06
		C(free-range)	L	0.07	0.09	0.07	3.40	30.90		1.60		4.15
			K	0.08	0.16	0.08	2.12	24.70		3.50		7.62
			M	0.08	0.01	0.02	nd	4.16		0.01		0.23
Uluozlua et al., 2009* , Turkey	markets		G	0.10	0.04	0.03	0.13	30.50		0.02		0.23
			L	0.04	0.02	0.01	12.10	22.50	0.06	2.24		0.12
Demirbas, 1999* , Turkey	control		L				2.95	28.10		0.04	0.08	0.09
Villar et al., 2005 , Phillipines			L							0.03		0.08
Husain et al., 1996 , Kuwait	wholesalers	G	L							0.05		0.13
			K							0.44		0.43
Okoye and Ugwu, 2010 , Nigeria	Industrial & urban		L				134.02	120.44		0.35		0.65
Abou-Arab, 2001* , Egypt	Industrial & urban		L				46.6	32.6		0.26		0.46
	industrial & urban		K				2.4	34.1		0.91		0.68
	industrial & urban		M				1.2	41.4		0.04		0.08
Husain et al., 1996 , Kuwait	wholesalers	S	L							0.04		0.13
			K							0.30		0.15
Caggiano et al., 2005 , Italy	urban		L							0.33		1.50
			K							6.71		2.00
			M							0.16		1.60
Abou-Arab, 2001* , Egypt	Industrial & urban		L				48.6	36.8		0.26		0.42
			K				3	34.2		0.82		0.54

C: chicken; G: goat; S: sheep; L: liver; K: kidney; M: muscle; G: gizzard.

* means mg/kg ww.

Table 3Estimated Daily Intake ($\mu\text{g}/\text{kg bw/day}$) of As, Cd, Hg, Pb and Mn.

Sample name	As	Cd	Hg	Pb	Mn
Chicken Gizzard	0.19	0.21	0.03	0.96	1.61
Chicken Kidney	0.02	0.12	0.02	0.04	0.41
Chicken Liver	0.19	0.55	0.29	0.34	8.77
Chicken Muscle	0.12	0.003	0.03	0.03	0.37
Goat Kidney	0.002	0.01	0.005	0.01	0.13
Goat Liver	0.02	0.05	0.03	0.07	6.07
Goat Muscle	0.01	0.01	0.002	0.01	1.55
Sheep Kidney	0.002	0.03	0.01	0.01	0.19
Sheep Liver	0.04	0.13	0.07	0.14	8.92
Sheep Muscle	0.02	0.02	0.01	0.01	1.77
TDI by FAO/WHO/FSA	2.0	1.0	0.71	3.57	200

FAO: Food and Agricultural Organization.

WHO: World Health Organization.

FSA: Food Safety Agency.

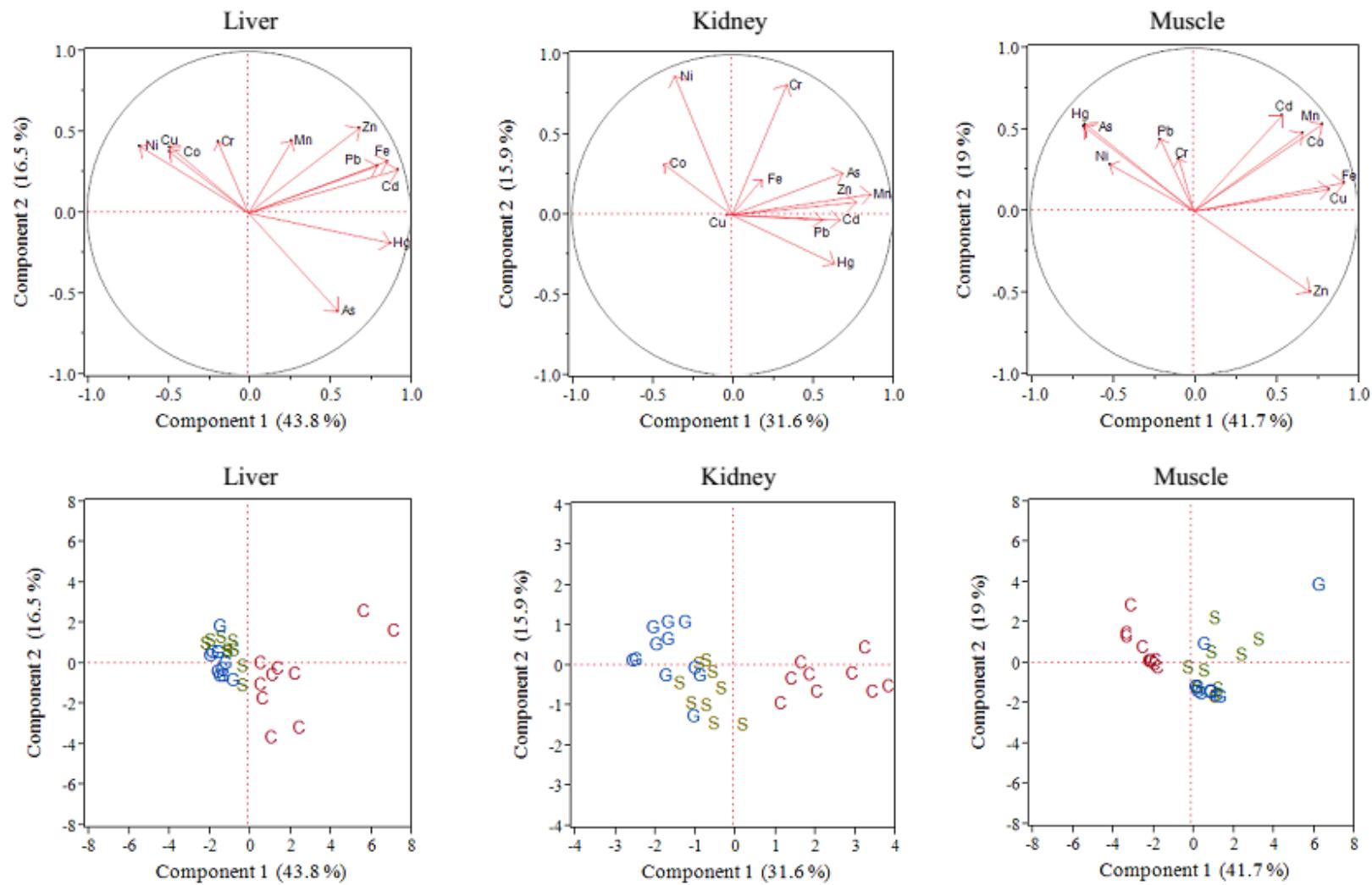


Fig. 1.

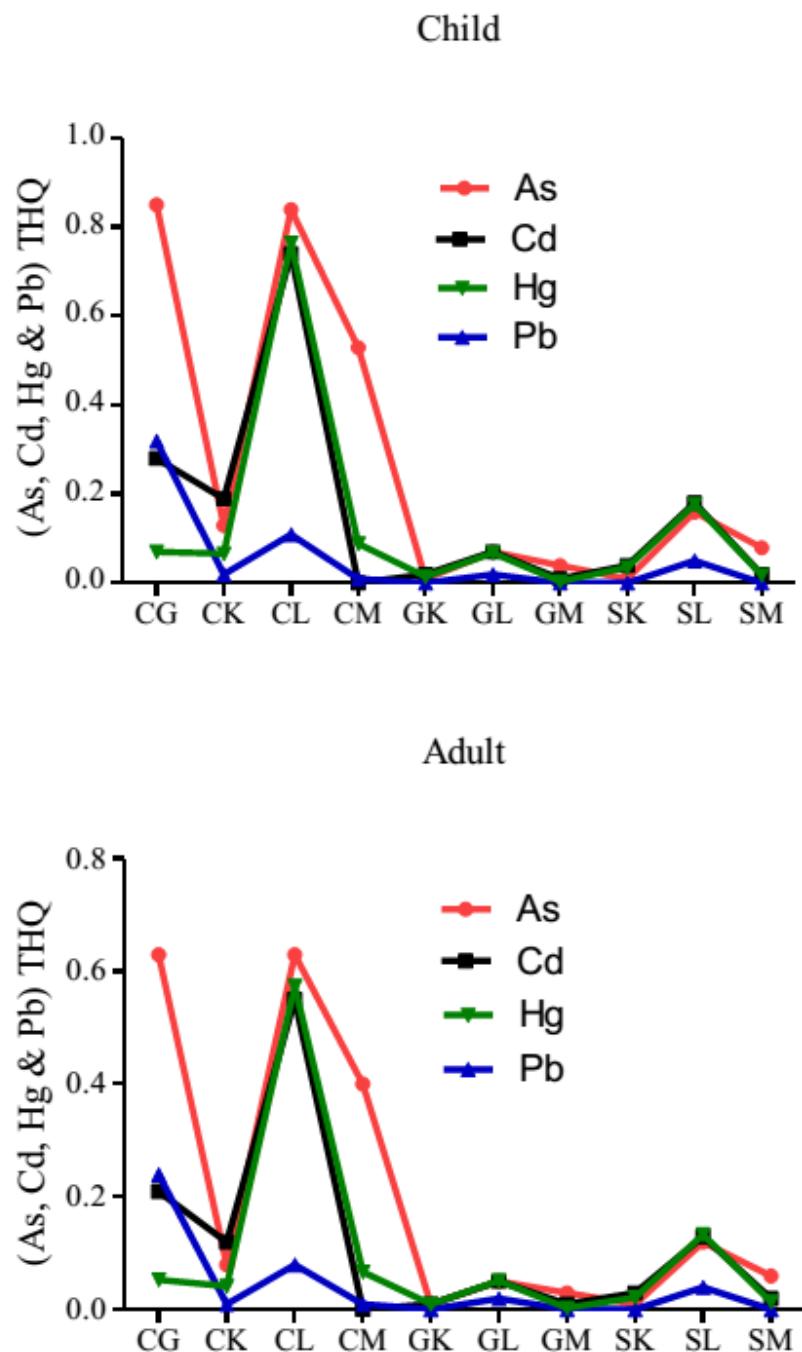


Fig. 2.