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Full Length Research Article

Heavy Metals and Carcinogenic Risk Assessment in Free-Ranged Livestock of Lead-Contaminated Goldmine Communities of Zamfara State, Northern Nigeria

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ABSTRACT

The consumption of meat is of great importance as it provides a good source of proteins and a significant amount of essential trace elements to the body. However, contamination of meat and meat products with heavy metals is becoming a serious threat to food safety and public health. Therefore, the present study aimed to evaluate the concentration of some heavy metals in the muscles and entrails of free-ranged cattle, sheep, and goats. A total of sixty (60) fresh samples of muscles, liver, kidney, small intestines, and stomach of free ranged cattle, sheep, and goats were collected from abattoirs of different goldmine communities of Anka, Bukkuyum, Maru, and Talata-Mafara Local Government Areas of Zamfara State, Nigeria. The samples were digested using 10 mL of a mixed 70% high-grade concentration of HNO_3 and 65% HCl (4:1 v/v); the mixture was heated until dense fumes disappeared forming a clear transparent solution and diluted to 50 mL with deionized water. Actual concentrations of Cd, Cr, Cu, Co, As, Ni, Mn, Pb, and Zn were determined using a Microwave Plasma Atomic Emission Spectrophotometer (MP-AES). Results indicate goat liver had the highest mean concentration of lead, arsenic, cobalt, and manganese (12.43± 0.31, 14.25±0.32, 3.47± 0.86, and 12.68± 0.92 mg/kg respectively), and the kidney had the highest concentration of copper and zinc (10.08±0.61 and 24.16±1.30 mg/kg respectively). Sheep kidneys had the highest bioaccumulation of cadmium and nickel (7.75± 0.65 and 2.08±0.10 mg/kg respectively) while chromium was observed to accumulate mostly in cattle muscles when compared with all other organs analysed. The target hazard quotients (THQs) for all the metals analyzed were below 1.0, but the risk indices for carcinogenicity (TR) predictably suggest exposed individuals were most likely to develop the disease. Therefore, intensive public health awareness of the risks associated with the consumption of heavy metal-contaminated meat should be prioritized.

Keywords: Contamination, Goldmine, Heavy Metals, Meat.

INTRODUCTION

Food safety and environmental pollution are matters of great concern worldwide. The ever-expanding population, industrialization, urbanisation, extensive agricultural practice, and mining activities have intensely contributed to the degradation of our natural environment. In recent years, mining industries have experienced a boom in operational activities due to the increased demand for metals and metalloid compounds by different manufacturing companies and the juicy price of gold in the world market. Several communities in both underdeveloped and developing countries have abandoned their usual occupation in search of gold (Rabiu *et al*., 2019b). In developing countries like Nigeria, mining activities and manual processing of metal ores have been observed in many parts of Nigeria (Lar *et al*., 2014; Santuraki *et al*., 2018; Rabiu *et al*., 2019b). Mining and metal ore processing in many countries have been observed to be responsible for the largest releases of heavy metals into the environment thereby constituting health risks to humans, animals as well as impact negatively on the quality of the environment (Ghanwat *et al*., 2015; Saeed *et al*., 2017). Heavy metals are ubiquitous and originate from both anthropogenic and natural processes. Although some heavy metals including Cobalt (Co), copper (Cu), nickel, iron (Fe), manganese (Mn), zinc (Zn), molybdenum (Mo), and selenium (Se) are essential trace elements but can cause serious health complications when consumed above recommended daily requirements Hassan Emami*et al*., 2023). Despite the nutritional needs of some heavy metals, arsenic (As), cadmium (Cd), chromium, mercury (Hg), and lead (Pb) are nonessential and detrimental to most higher plants and animals (Okoye and Ihedioha, 2010). These toxic metals have long been implicated to be associated with many health consequences including liver, kidney, reproductive, nervous system, cardiovascular, haematological, and immune dysfunctions, as well as intrauterine retardation (Rabiu *et al.,* 2019a).

Food chain heavy metal contamination occurs mainly via polluted air, water, and soil as well as through livestock and plant products grown in pollution-prone areas. Livestock production has been a significant source of animal protein worldwide. Meat from slaughtered cattle, goats, and sheep constitutes the largest source of animal protein for many Nigerian households (Seiyaboh *et al*., 2018). Moreover, most of the livestock are being raised in a commercial scale in the Northern parts of the country grazed on a free-range system throughout the year. In Zamfara State with reports of severe acute lead poisoning due to unsafe mining activities in many communities, free range grazing of livestock exposes the animals to heavy metal poisoning and bioaccumulation. The consumption of meat and meat products from heavy metal-contaminated livestock can pose a serious health implication to the human population. Toxic levels of lead and cadmium have been detected in the milk of cattle grazed around the Dareta community in Zamfara State (Sadiq *et al*., 2015). High blood levels of lead and cadmium were also reported in cows, goats, and sheep grazing in open fields in Nigeria (Okoye and Ugwu 2010; Akan *et al*., 2010). Ubwa *et al.* (2017) reported significant concentrations of lead, cadmium, chromium, and nickel in liver, kidney, chitterlings, and stomach) of cow, native goat, non-native goat and pig slaughtered at Wurukum abattoir Makurdi-Nigeria. Elsewhere, toxic and trace metals in calves and kids were reported in a polluted area of Northern Spain (Miranda *et al.,* 2005). Similarly, Jukna, *et al*. (2006) reported moderate concentrations of heavy metals in the viscera and muscles of Lithuanian cattle while Sharif *et al* (2005) reported high concentrations of heavy metals in cattle reared in the vicinity of a metallurgic industry. Alturiqi and Albedai (2012) in their study reported high concentrations of copper, zinc, iron, lead, cadmium, and mercury in certain fish, meat, and meat products in Saudi Arabian Markets. Food safety and environmental pollution are therefore, inextricably linked and hence should be given utmost attention. As a consequence, international organizations such as the World Health Organization (WHO), Food and Agricultural Organization (FAO) and US Environmental Protection Agency (US-EPA) have set permissible limits of heavy metal concentration expected in foodstuffs (FAO/WHO, 2011). This study was therefore, designed to assess the levels of some bioaccumulated heavy metals in tissues of cattle, sheep and goats grazed around active goldmine communities in Zamfara State

MATERIALS AND METHODS

Study Area

Zamfara State is currently faced with the problem of illegal mining activities. The most affected villages are Dareta, Bagega, and Yargalma of Anka and Bukkuyum Local Government Areas. In 2016, the CDC estimated that there were over 40 exposed to environmental heavy metal contamination with over 30,000 residents at risk of heavy metal toxicity. High lead concentrations in some wells and ponds in Dareta and ponds and streams in Abare villages in Anka Local Government Area were also documented (Okiei *et al.,* 2016). The soil in the affected communities was reported to have Pb levels up to 23 times the maximum acceptable levels in soils sat by USEPA.

Sampling

A total of sixty (60) fresh samples of muscles, liver, kidney, small intestines, and stomach of free-ranged cattle, sheep, and goats were collected from abattoirs of Dareta, Bagega, and Anka in Anka Local Government; Yargalma and Masamain Bukkuyum Local Government; Miyanchi and Kadauriin Maru Local Government and Sauna and Mafara of Talata-Mafara Local Government Areas of Zamfara State, Nigeria. The samples were collected in labelled zip-lock polyethylene bags and preserved in a liquid nitrogen ice container which was immediately transported to the laboratory at Usmanu Danfodiyo University Sokoto.

Sample Preparation

In the laboratory, the samples were washed with deionized water to remove adsorbed particulates and visible fat, and connective tissues were carefully removed. About two hundred milligrams (300 mg) of each of the samples were cut into small sizeable pieces using a clean stainless steel knife and oven dried at 80°C for about 24 hours until constant weight was achieved. The dried samples were ground using an acid pre-washed ceramic pestle and mortar and sieved through muslin cloth.

Sample Digestion

Exactly 200 mg subsample of each sample was weighed and placed in an acid cleaned beaker and then an acid mixture (10 mL, 70% high-grade concentration of HNO_3 and 65% HCl, 4:1 v/v) was immediately added to initiate the digestion. The content was then placed on a hot plate at 80^oC and continued heating until dense fumes disappeared forming a clear transparent solution. The solution was allowed to cool, filtered using Whatman no. 42 filter paper, and subsequently diluted to 50 mL with deionized water. Actual concentrations of Cu, Cd, Pb, Cr, Cd, Mn, Co, Zn, Ni, and Al were determined as μg/g dry weight tissue using a Microwave Plasma Atomic Emission Spectrophotometer (MP-AES).

Human Health Risk Assessment

Estimated Daily Intakes (EDIs)

The estimated daily intake (EDI) of heavy metal contaminated meat amongst the population of the study area was deduced using the relationship:

$$\text{EDIs (mg/kg)} = \frac{Cmetal \times Dintake}{Bweight}$$

C metal = metal concentration in the meat sample (mg/kg)

D intake = meat daily intake (0.345 kg/person/day) while

B weight = body weight (60 kg for adult)

Non-Carcinogenic Risk

The non-carcinogenic health risks in relation to meat consumption in the study area were determined using the calculated target hazard quotients (THQs) developed by the US Environmental Protection Agency (EPA). THQ is a ratio between the measured concentration and the oral reference dose (RfD) as determined by FAO/WHO. Calculated THQ values less than one (< 1), suggest the exposed population is safe while THQ ≥ 1, indicates a concern level for the exposed population.

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

Where EFr = frequency of exposure (365 days/year); ED = exposure duration (70 years average lifetime for adults and 15 years for children); FIR = food ingestion rate (g/ person/day); MC = concentration of the metal in samples (mg/kg), BW = average body weight (60 kg for adult; 16 kg for children); AT = averaging time for non-carcinogens (365 days/year × number of exposure years), RfD = oral reference dose (mg/kg/day)

Carcinogenic Risk Assessment

The carcinogenic risk signifies the incremental probability of an individual developing cancer in a lifetime due to ingestion of food contaminated with potential carcinogens (USEPA, 2010). The following equation was used to predict target cancer risks in the study area:

$$TR = \frac{EFr \times ED \times FIR \times MC \times CSFo}{BW \times CSFo} \times 10^{-3}$$

Where TR = target cancer risk in a lifetime overexposure to a certain toxic metal; CSFo = oral carcinogenic slope factor (USEPA, 2010). The acceptable risk levels for carcinogens range from 10^{-4} to 10^{-6}. Pb = 0.0085 and As = 1.5 mg/kg/day respectively.

Statistical Analysis

Data generated were analysed using GraphPad InStat version 6.03 statistical package. Statistical differences between organs of the same species and between organs of different species were determined using one-way ANOVA. Tukey-Karamer comparison test was used to compare means and differences were considered significant at $p < 0.05$.

RESULTS

The mean concentration of the heavy metals Cd, Cr. Cu, Co, As, Ni, Mn, Pb, and Zn determined in the organ samples of cattle, sheep, and goats in the study area are presented in Table 1. All the organs analyzed for each ruminant were found to extensively bioaccumulate the trace micronutrients Cu, Pb, and Zn. Highest levels of Pb, As, and Co were observed in goat intestine (12.43±0.31, 14.25±0.37 and 3.47±0.86 mg/kg, respectively) while Cr and Ni were highest in cattle stomach and liver (9.00±0.25 and 1.93±0.30 mg/kg, respectively). Highest Cd concentrations were observed in sheep kidney (7.75±0.65 mg/kg). Except for Cu and Zn, mean values obtained for analyzed bioaccumulated heavy metals were far above FAO/WHO permissible limits. In the organs of cattle, significantly ($p<0.05$) high concentrations of Cd, Cu, and Pb were found in the kidney, Co, and Ni in the liver and the intestine. In organs of sheep, significantly ($p<0.05$) high concentrations of Pb were observed in the intestine compared to muscle and kidney. Similarly, significantly ($p<0.05$) high concentrations of Pb, Ni, As, Cr, and Co were observed in the goat intestine compared to the stomach, muscle, and kidney.

The mean concentration of metals in similar organs of cattle, sheep, and goats were compared and presented in Table 2. Metal concentrations in the same organ from the three animals (cattle, sheep, and goat) showed variable distribution. Significantly ($p<0.05$) high concentrations of Cd, Co, As, Ni, and Pb were found in sheep muscle compared to that of cattle and goat. However, cattle muscle appeared to accumulate ($p<0.05$) high concentrations of Cr and Cu compared to sheep and goats. Goat muscle had the lowest concentrations of all the metals analysed. Goat intestine accumulate significantly ($p<0.05$) high amounts of Cd, Cr, Cu, Co, and Pb compared to cattle and sheep intestine. The concentrations of Pb, Ni, and Co were significantly ($p<0.05$) high in sheep liver compared to that of cattle and goats while concentrations of Cd, Cr, and Cu were significantly ($p<0.05$) high in goat liver compared to cattle and sheep liver. Cattle stomach appeared to have significantly ($p<0.05$) high concentrations of Cd, Cr, and Ni compared to stomach of sheep and goats. However, no significant ($p>0.05$) concentration of Cu and Co were observed in the stomach of all the animals. Significantly ($p<0.05$) high concentrations of Cd, Co, As, and Ni were observed in sheep kidney compared to cattle and goat kidney. Goat kidney was found to concentrate more of Pb, Cu and Cr compared to cattle and sheep kidney.

The mean concentrations of heavy metals in the muscle and entrails of cattle, sheep, and goats are presented in Figure 1. The mean concentrations of Cr were significantly ($p<0.05$) high in muscle compared to entrails in all the animals. The concentrations of Cd and As were significantly ($p<0.05$) high in entrails compared to muscle except that of sheep. The concentrations of Ni were significantly ($p<0.05$) high in entrails compared to muscle in all the animals while significantly ($p<0.05$) high concentrations of Pb were observed in entrails compared to sheep and goat muscle.

The oral reference doses (RfD) and highest estimated daily intake for metals are presented in Table 3 while the mean levels of heavy metals intake via daily consumption of various meat tissues are presented in Table 4. The high mean daily intake for Pb was found in sheep stomach (SSh) followed by cattle liver (CLv) (0.1695 and 0.1217mg/kg respectively). The high mean daily intake for Ni was observed in sheep liver (SLV) and goat liver (GLv) (0.0359 and 0.0232 mg/kg respectively). The high mean daily intake for As, Co, and Cu was recorded in GLv (0.3254, 0.0693, and 0.1253 mg/kg, respectively). Cattle muscle (CMs) and cattle kidney (CKd) had the high mean daily intake for Cr and Cd (0.0539 and 0.136 5mg/kg respectively)

Non-Carcinogenic Health Hazard and Carcinogenic Risk

The calculated THQs and TR for the intake of the heavy metals in both adults and children are presented in Tables 4 and 5 respectively. All the calculated values for THQs in both adults and children were less than 1.0, however the calculated TR values for As and Pb were significantly ($p<0.05$) high in both adults and children.

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DISCUSSION

Gold mining especially in developing countries is an important source of economic opportunities. In Nigeria, many communities have abandoned traditional economic activities like farming, fishing, and trading for goldmines (Rabiu *et al*., 2019b). As a consequence, artisanal mining activities in several of these communities have led to environmental degradation and public health risks even long after the termination of such activities (Santuraki *et al*., 2018; Rabiu *et al*., 2019b).

Table 1: Mean concentration of heavy metals in organs of cattle, sheep and goat

ANIMAL	ORGAN	Cd	Cr	Cu	Co	As	Ni	Pb	Zn
Cattle	**Muscle**	1.21±0.60a	0.75±0.44a	5.00±1.79a	2.81±0.25a	0.56±0.06a	1.38±0.76a	6.08±0.41a	10.93±2.81a
	Intestine	4.13±1.06b	0.94±0.06a	5.68±1.54a	2.20±.027a	1.26±0.43b	1.00±0.66b	5.40±0.25b	12.12±4.00b
	Liver	4.00±0.35b	1.37±0.97b	4.81±1.14b	3.12±0.80b	0.56±0.11a	1.93±0.30c	3.63±1.20c	19.64±2.86c
	Stomach	0.37±0.07c	9.00±0.25c	6.13±1.87c	2.87±061c	0.39±0.12c	1.25±0.500a	5.63±0.38a	10.00±5.00a
	Kidney	6.37±0.64d	1.00±0.75d	6.25±2.70c	2.50±0.50c	0.39±0.10c	1.31±0.32a	8.31±2.15d	15.50±5.20d
Sheep	**Muscle**	1.50±0.00a	0.92±0.55a	5.50±1.13a	2.50±0.60a	1.81±0.94a	1.50±0.87a	3.69±1.60a	9.37±4.50a
	Intestine	2.50±0.24b	1.12±0.79b	5.87±1.21a	3.06±0.21b	03.9±0.10b	1.95±0.90b	5.52±1.20b	12.62±3.60b
	Liver	1.06±0.90a	0.93±0.43a	4.43±0.72b	2.43±0.67a	0.20±0.05c	0.81±0.32c	5.13±1.90b	10.65±4.30c
	Stomach	2.08±0.08b	2.47±0.62c	3.30±0.14c	2.92±0.04a	0.56±0.14d	0.83±0.05c	4.83±0.34b	20.67±0.12a
	Kidney	7.75±0.65c	0.58±0.46d	9.58±0.62d	3.08±0.30cb	0.42±0.08d	2.08±0.10b	2.51±0.20c	17.00±0.35e
Goat	**Muscle**	2.83±0.28a	1.44±0.70a	6.50±0.71a	2.93±0.15a	0.44±0.11a	1.37±0.64a	7.88±0.31a	12.66±0.56a
	Intestine	0.25±0.02b	1.56±0.09a	6.63±0.29a	3.47±0.86b	14.25±0.37b	1.87±0.47b	12.43±0.31b	18.43±3.90b
	Liver	2.43±0.23c	0.92±0.41b	4.37±0.12b	2.81±0.64a	4.00±0.34c	1.44±0.48a	6.38±0.27c	16.31±3.40c
	Stomach	0.38±0.03b	1.38±0.87c	2.82±0.37c	0.26±0.01c	0.38±0.02a	0.88±0.09c	0.87±0.08d	4.50±0.20d
	Kidney	3.67±0.03d	1.50±0.16c	10.08±0.61d	2.83±0.33a	0.75±0.02d	1.67±0.41d	4.42±0.19e	24.16±1.30e

Values are presented as means ± SEM expressed as mg/kg dry weight; Values with different letters down the column indicate statistically significant difference at $p < 0.05$

Table 2: Mean concentrations of metals in similar tissues of different animal species

ORGAN	ANIMAL	Cd	Cr	Cu	Co	As	Ni	Pb	Zn
Muscle	Cattle	1.21±0.60a	0.75±0.44a	5.00±1.79a	2.81±0.25a	0.56±0.06a	1.38±0.76a	6.08±0.41a	10.93±2.81a
	Sheep	1.50±0.00b	0.92±0.55b	5.50±1.13a	2.50±0.60b	1.81±0.94b	1.50±0.87b	3.69±1.60b	9.37±4.50b
	Goat	2.83±0.28c	1.44±0.70c	6.50±0.71b	2.93±0.15a	0.44±0.11a	1.37±0.64a	7.88±0.31c	12.66±0.56c
Intestine	Cattle	4.13±1.06a	0.94±0.06a	5.68±1.54a	2.20±.027a	1.26±0.43a	1.00±0.66a	5.40±0.25a	12.12±4.00a
	Sheep	2.5±0.24b	1.12±0.79b	5.87±1.21a	3.06±0.21b	03.9±0.10b	1.95±0.90b	5.52±1.20a	12.62±3.60a
	Goat	0.25±0.02c	1.56±0.09c	6.63±0.29b	3.47±0.86b	14.25±0.37c	1.87±0.47b	12.43±0.31b	18.43±3.90b
Liver	Cattle	4.00±0.35a	1.37±0.97a	4.81±1.14a	3.12±0.80a	0.56±0.11a	1.93±0.30a	3.63±1.20a	19.64±2.86a
	Sheep	1.06±0.90b	0.93±0.43b	4.43±0.72b	2.43±0.67b	0.20±0.05b	0.81±0.32b	5.13±1.90b	10.65±4.30b
	Goat	2.43±0.23c	0.92±0.41b	4.37±0.12b	2.81±0.64c	4.00±0.34c	1.44±0.48c	6.38±0.27c	16.31±3.40c
Stomach	Cattle	0.37±0.07a	9.00±0.25a	6.13±1.87a	2.87±061a	0.39±0.12a	1.25±0.50a	5.63±0.38a	10.00±500a
	Sheep	2.08±0.08b	2.47±0.62b	3.30±0.14b	2.92±0.04a	0.56±0.14b	0.83±0.05b	4.83±0.34b	20.67±0.12b
	Goat	0.38±0.03a	1.38±0.87c	2.82±0.37c	0.26±0.01b	0.38±0.02a	0.88±0.09c	0.87±0.08c	4.50±0.20c
Kidney	Cattle	6.37±0.64a	1.00±0.75a	6.25±2.70a	2.50±0.50a	0.39±0.10a	1.31±0.32a	8.31±2.15a	15.5±5.20a
	Sheep	7.75±0.65b	1.00±0.75a	9.58±0.62b	3.08±0.30b	0.42±0.08a	2.08±0.10b	2.51±0.20b	17.00±0.35b
	Goat	3.67±0.03c	1.50±0.16b	10.08±0.61c	2.83±0.33a	0.75±0.02b	1.67±0.41c	4.42±0.19c	24.16±1.30c

Values are presented as means ± SEM expressed as mg/kg dry weight; Values with different letters down the column indicate statistically significant difference at $p < 0.05$

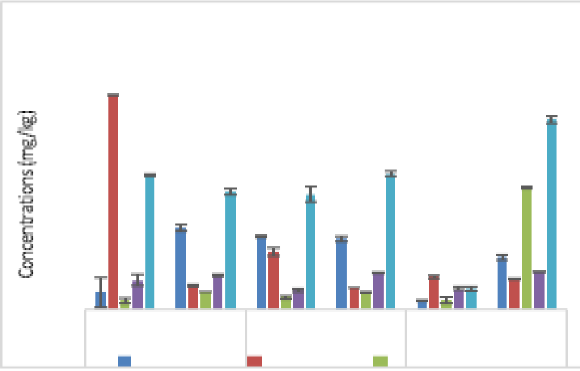

Figure 1: Mean concentrations of heavy metals in muscle and entrails of cattle, sheep and goat

Table 3: Oral reference doses (RfD) and highest estimated daily intake for metals in the study Area

S/No	ELEMENT	RfD (mg/kg/day)	HIGHEST EDI (mg/day
1	Cd	1.00×10^{-3}	1.37×10^{-1}*
2	Cr	1.50×10^{0}	5.39×10^{-2}
3	Cu	4.00×10^{-2}	1.25×10^{-1}
4	Co	3.00×10^{-2}	6.93×10^{-2}
5	As	3.00×10^{-4}	3.25×10^{-1}*
6	Ni	2.00×10^{-2}	3.59×10^{-2}
7	Mn	1.40×10^{-1}	1.54×10^{-1}
8	Pb	4.00×10^{-3}	1.70×10^{-1}*
9	Zn	3.00×10^{-1}	3.70×10^{-1}

EDI-Estimated daily intake, *Highly most toxic (ATSDR, 2011) RfD (mg/kg/day) FAO/WHO, (2011).

Table 4: Mean daily intake (mg/day) of heavy metals via consumption of the different animal tissues

ANIMAL TISSUE	Cd	Cr	Cu	Co	As	Ni	Mn	Pb	Zn
CMs	0.0043	0.0539	0.0474	0.0265	0.0029	0.0094	0.0137	0.0359	0.1006
CIt	0.0086	0.0095	0.0980	0.0520	0.0106	0.0191	0.0348	0.0502	0.1227
CLv	0.0633	0.0100	0.1124	0.0476	0.0139	0.0099	0.1537	0.1217	0.2691
CSh	0.0934	0.0152	0.0794	0.0535	0.0097	0.0224	0.0610	0.0423	0.3698
CKd	0.1365	0.0064	0.0460	0.0201	0.0144	0.0158	0.0582	0.0539	0.1215
SMs	0.0531	0.0366	0.0527	0.0407	0.0056	0.0094	0.0203	0.0192	0.2651
Sit	0.0345	0.0452	0.1134	0.0426	0.0212	0.0265	0.0656	0.1128	0.2038
SLv	0.0302	0.0106	0.1048	0.0596	0.0213	0.0359	0.0799	0.1056	0.1987
SSh	0.0229	0.0117	0.0922	0.0473	0.0059	0.0122	0.1038	0.1698	0.2375
SKd	0.0718	0.0057	0.0809	0.0435	0.0062	0.0206	0.0813	0.0446	0.2501
GMs	0.0043	0.0388	0.0259	0.0238	0.0029	0.0101	0.0122	0.0101	0.0453
Git	0.0661	0.0229	0.1139	0.0545	0.0068	0.0207	0.0768	0.1056	0.1449
GLv	0.0057	0.0232	0.1253	0.0693	0.3254	0.0323	0.0630	0.1165	0.2769
GSh	0.0553	0.0126	0.0822	0.0438	0.0897	0.0222	0.0340	0.1196	0.2448
GKd	0.0632	0.0181	0.1499	0.0380	0.0099	0.0210	0.0714	0.0454	0.0330

C: cattle, S: sheep, G: goat, Ms: muscle, It: intestine, Lv: liver, Sh; stomach, Kd: kidney

Anthropogenic causes of heavy metal contamination of agricultural soils, atmosphere, and water bodies such as ponds, wells, rivers, and dams affect the food chain by exposing free-grazing livestock to heavy metal uptake, bioaccumulation and biomagnification of toxic metals in their tissues (Yabe *et al*., 2010).

Consumption of heavy metal-contaminated food has been shown to cause depletion of some essential nutrients, immune and reproductive systems impairment, psycho-social disabilities, diseases associated with malnutrition, and gastrointestinal disturbances (Alturiqi, and Albedair, 2012).

In this study, except for Cu and Zn, the mean concentration of all the other heavy metals analysed was above FAO/WHO permissible limits. Analysed goat intestinal tissue revealed toxic levels of Pb, As, and Co. These findings were similarly reported by Orisakwe *et al*. (2017) and Birnin-Yauri *et al*. (2018). Islam (2018) and Akoto *et al*. (2014) also reported toxic levels of Pb, As and Co in sheep and goats grazed in a gold mining town of Ghana. Similarly, the highest concentrations of Cr and Ni were found in cattle stomach while Cd in sheep kidney. This surely indicated that the consumption of meat and meat products of cattle, sheep, and goats from this region is likely to constitute a public health hazard since the concentrations of the metals were far above FAO/WHO permissible limits. The variability in metal concentration observed among the animals could be attributed to species differences, growth period, absorption capabilities, and bioaccumulation of the metals in question (Saha and Zaman, 2013).

The concentrations of As, Cd, Ni, and Pb were found significantly higher in entrails than the muscles of the animals studied, indicating possible transfer of heavy metals from soil plants and grasses; and water bodies to the free-range grazing animals via ingestion. In addition, the discrepancy observed in metal accumulation between muscle and entrails could be due to the fact that entrails like the liver and kidney are organs for metabolism and detoxification; hence, are predisposed to higher concentrations of these toxic metals than any other organs of the body (Adzitey *et al*., 2015; Boahene *et al*., 2020). High concentrations of some of these metals in the intestine could be attributed to its physiology, serving as the most active site of digestion. The intestinal villi allow the direct assimilation of digestive end products to the body and presumably, there is a possible 'trap' in the small intestine that may facilitate the accumulation of heavy metals (Ubwa *et al.,* 2017).

The Agency for Toxic Substances and Diseases Registry (ATSDR, 2011) classified metals like arsenic, lead, cadmium, cobalt, nickel, zinc, chromium, copper, and manganese as numbers 1, 2, 7, 51, 57, 74, 78, 120 and 143 on the priority list of the most hazardous substances in the environment. Therefore, animals grazing in metals contaminated environments are highly exposed to these elements through ingestion of polluted plants, grasses, and drinking water as well as through inhalation (Reglero *et al*., 2008; Madejon *et al.,* 2009).

Studies have shown that cadmium food and food products rather than water or air pollution account for the major source of cadmium exposure (Rahman *et al.* 2013). The element naturally occurs as a component of the earth crust and is usually transported in the food chain via different phenomena; "soil-plant-animal" and/or "soil-water-animal" (Islam, 2018). Elevated cadmium concentrations have been implicated in the human body to cause renal tubular dysfunction, pulmonary, hepatic, skeletal, and reproductive derangements (Zhu *et al*. 2011). In all the samples analysed, the concentrations of cadmium determined were far above the FAO/WHO permissible limit of 0.5 mg/kg for meat of livestock. In studies conducted in lead-contaminated communities of Zamfara State, Orisakwe *et al.* (2017) reported the highest cadmium concentrations in cattle liver (0.2950±0.3325 mg/kg). In the same environment, Birnin-Yauri et al. (2018) reported the highest concentrations of cadmium in goat kidneys (1.100±0.003 µg/g); In the Northern region of Ghana (Tamale), 0.93±0.43 mg/kg of cadmium concentration was documented goat liver (Boahene *et al*., 2020). However, the concentration of cadmium detected in the present study was high compared to studies conducted in the area. Several studies have documented high concentrations of cadmium in the kidneys of animals and suggested that may be due to the detoxification function of the organ and its relatively low rate of elimination from the organ (Chia *et al.,* 2012; Hashemi, 2018).

Toxicity of lead has well been documented in humans, animals, and plants and is associated with numerous health effects such as neurotoxicity, nephrotoxicity, reproductive dysfunction, haematological abnormalities (GarciaLeston *et al.* 2010; Rabiu *et al*., 2022b). Ruminants are commonly associated with lead toxicity and high blood lead concentrations in cattle, sheep, and goat free-grazed in polluted areas (Akoto *et al.,* 2014; Birnin-Yauri *et al.,* 2018; Mahdieh *et al*., 2021). In present study, highest lead concentrations were detected goat intestine (12.43±0.31) and far above the values reported by Orisakwe *et al*. (2017); Birnin-Yauri *et al* (2018); Islam (2018); Boahene *et al*. (2020) 7.7554±7.44943 mg/kg, 3.925±0.010µg/g, 1.9±3.0 mg/kg and 7.06±7.06 mg/kg, respectively.

The observed high lead concentrations in these animals could be attributed to the usual ingestion of lead-contaminated grasses and plants, inhalation of lead particles, natural curiosity, licking habits, and lack of oral discrimination among others. This study is the first to report arsenic concentrations in meat samples and the highest values were detected in goat intestine (14.25±0.37 mg/kg). Exposure to inorganic arsenic is known to cause several health problems. These include dermatitis, mild pigmentation and hyperkeratinizations of the skin, abnormal nerve conduction, wart formation, and lung cancer (OSHA, 2004). In all the samples, the arsenic concentrations were above the recommended levels (0.10 mg/kg) of FAO/WHO (2006). This shows that all the meat samples analysed were contaminated with arsenic and could cause potential risk to public health. However, the values obtained were far below the values reported in Pakistan (Mariam *et al*., 2014) and Kuwait (Abd-Elghany *et al.,* 2020) (46.5 and 48.0 mg/kg, respectively).

It has been established that grazing some animals involuntarily ingests some amount of soil (up to 18%) which can lead to significant exposure to toxic metals that may be available in the soil (Blanco-Penedo *et al.* 2010). Nickel is among the toxic metals that occur at significantly low concentrations in the ecosystem. The metal is required in small quantities by humans for the regulation of prolactin and stabilization of RNA and DNA structures; however, at high concentrations is associated with numerous pulmonary adverse health consequences, such as lung inflammation, fibrosis, emphysema, and tumours (Forti *et al.,* 2011; Chowdhury *et al*., 2011). The highest nickel concentrations were observed in sheep kidneys (2.08±0.10 mg/kg) lower than the values obtained by Orisakwe *et al.* (2017) (3.9583±3.0825 mg/kg) but greater than those reported by Islam (2018) in Northern part of Bangladesh and Sabow *et al*. (2020) in Erbil governorate, Iraq (1.8±1.5 mg/kg and 0.24±0.95 mg/kg respectively). Dietary chromium is very important in the metabolisms of sugar and lipids, and helps in blood glucose regulation by insulin; however, in excessive amounts, chromium has been implicated in a variety of cancers (Islam, 2018).

Table 5: Non-carcinogenic (THQs) and carcinogenic (TR) risks for heavy metals due to consumption of meat in adults in the study area

		TARGET HAZARD QUOTIENTS (THQS)						TARGET CARCINOGENIC RISK (TR)	
ANIMAL	**TISSUE**	**Cd**	**Cr**	**Cu**	**As**	**Ni**	**Pb**	**As**	**Pb**
Cattle	**Muscle**	2.00×10^{-3}	4.00×10^{-6}	9.00×10^{-4}	7.00×10^{-3}	4.00×10^{-4}	8.00×10^{-3}	4.70×10^{-2}	7.08×10^{-1}
	Entrails	1.90×10^{-2}	4.00×10^{-6}	8.00×10^{-4}	1.40×10^{-2}	4.00×10^{-4}	7.00×10^{-3}	9.20×10^{-2}	6.21×10^{-1}
Sheep	**Muscle**	1.80×10^{-2}	9.00×10^{-6}	5.00×10^{-4}	1.00×10^{-2}	2.00×10^{-4}	7.00×10^{-3}	6.30×10^{-2}	6.08×10^{-1}
	Entrails	1.70×10^{-2}	3.00×10^{-6}	9.00×10^{-4}	1.40×10^{-2}	4.00×10^{-4}	7.00×10^{-3}	9.10×10^{-2}	6.38×10^{-1}
Goat	**Muscle**	2.00×10^{-3}	5.00×10^{-6}	4.00×10^{-4}	7.00×10^{-3}	3.00×10^{-4}	1.00×10^{-3}	4.80×10^{-2}	1.11×10^{-1}
	Entrails	1.30×10^{-2}	5.00×10^{-6}	1.00×10^{-3}	9.80×10^{-2}	5.00×10^{-4}	1.10×10^{-2}	4.31×10^{-1}	1.00×10^{0}

TR risks values lying between 10^{-6} and 10^{-4} are generally considered an acceptable range (USEPA, 2010)

Table 6: Non-carcinogenic (THQs) and carcinogenic (TR) risks for heavy metals due to consumption of meat in children in the study Area

		TARGET HAZARD QUOTIENTS (THQs)						TARGET CARCINOGENIC RISK (TR)	
ANIMAL	**TISSUE**	**Cd**	**Cr**	**Cu**	**As**	**Ni**	**Pb**	**As**	**Pb**
Cattle	**Muscle**	8.00×10^{-3}	1.29×10^{-4}	3.30×10^{-3}	2.70×10^{-2}	1.30×10^{-3}	3.00×10^{-2}	4.40×10^{-2}	6.64×10^{-1}
	Entrails	7.40×10^{-2}	1.50×10^{-5}	2.90×10^{-3}	5.20×10^{-2}	1.50×10^{-3}	2.70×10^{-2}	8.60×10^{-2}	5.82×10^{-1}
Sheep	**Muscle**	6.60×10^{-2}	3.50×10^{-5}	1.70×10^{-3}	3.60×10^{-2}	9.00×10^{-4}	2.60×10^{-2}	5.90×10^{-2}	5.70×10^{-1}
	Entrails	6.40×10^{-2}	1.30×10^{-5}	3.30×10^{-3}	5.20×10^{-2}	1.70×10^{-3}	2.70×10^{-2}	8.50×10^{-2}	5.98×10^{-1}
Goat	**Muscle**	8.00×10^{-3}	2.00×10^{-5}	1.50×10^{-3}	2.70×10^{-2}	9.00×10^{-4}	5.00×10^{-3}	4.50×10^{-2}	1.04×10^{-1}
	Entrails	4.70×10^{-2}	1.80×10^{-5}	3.60×10^{-3}	3.67×10^{-1}	1.70×10^{-3}	4.30×10^{-2}	6.06×10^{-1}	9.38×10^{-1}

TR risks values lying between 10^{-6} and 10^{-4} are generally considered an acceptable range (USEPA, 2010)

The highest concentration of chromium was recorded in cattle stomach (9.00±0.25 mg/kg) and all the values observed in organs of cattle, sheep, and goats were above the permissible limit (0.20 mg/kg) (FAO/WHO, 2011). The values recorded were significantly higher than the values 19.625 ± 0.018µg/g reported by Birnin-Yauri *et al*. (2018) in goat kidneys; 3.2375 ± 3.4230 mg/kg recorded by Orisakwe *et al.* (2017) and 1.67 µg/g by Massadeh and Kharibeh (2018).

Copper and zinc are important cofactors for many enzymes that play crucial roles in many metabolic processes. Copper is needed in bone formation, skeletal and mineralization, but at high concentrations is likely to cause health problems such as liver and kidney dysfunction. Most food contains copper in the range of 5 to 7mg/kg, thus copper toxicity is rarely observed in the population (Iwegbue, 2011). The highest concentration of copper was recorded in goat kidneys (10.08±0.61 mg/kg) and was higher than 1.4788±0.3307mg/kg reported by Orisakwe *et al*. (2017) and 538.425±0.018 µg/g recorded by Birnin-Yauri *et al*. (2018). Zn concentration was found to be highest in goat liver (24.16±1.30mg/kg) and far above4.24 ± 0.16µg/g reported in cattle liver by Akan *et al*. (2010); 2.0400±2.4436mg/kg reported by Orisakwe *et al*. (2017) and 549.255±0.003µg/g reported by Birnin-Yauri *et al.* (2018). The low concentrations of copper and zinc in the different meat samples could be attributed to significantly higher cadmium and lead accumulation in the meat samples. Studies have indicated that cadmium and lead cause reductions in both intestinal copper and zinc absorption due to competitive cation-binding for metallothionein (MT) (Smith *et al*., 1991; Goyer. 1997).

The highest estimated daily intake for most toxic elements like arsenic, lead, and cadmium were 0.3254, 0.1698, and 0.1365 mg/kg/day respectively. The values were greater than the recommended oral reference dose (FAO/WHO, 2011). The EDI for the studied metals from consumption of meat tissues followed the descending order of goat > sheep > cattle which may be attributed to the higher consumption rate of goat and sheep as well as higher levels of metal contamination in their meat. Therefore, high concentrations of these potentially toxic metals in the cattle, sheep, and goat tissues from the goldmine communities of Zamfara State, Nigeria, would compromise the meat quality. Health risk assessment is usually conducted to evaluate the potential health effects of human exposure to one contaminant via single or different pathways.THQ parameter is a dimensionless index and its values are additive, but not multiplicative. Moreover, THQ is not a measure of risk but indicates a level of concern. If the values of THQ are greater than 1, then the exposed population is presumably to experience apparent health consequences (Wang *et al.* 2005).

Therefore, in this study, the THQ was calculated from the consumption of meat by adults and children, and for all the metals, the values were all below 1.0, suggesting no metal toxicity from the consumption of meat from cattle, sheep, and goat free-ranged in goldmine communities of Zamfara State. Notwithstanding, exposure to these toxic metals could be a cause for concern for the population when the intake of these toxic metals from other foods and drinking water is considered. Similarly, the target carcinogenic risks (TR) were calculated for arsenic and lead from the daily consumption of the meat in the area. According to the US Environmental Protection Agency (USEPA-IRS, 2011), arsenic and lead have been implicated in promoting both non-carcinogenic and carcinogenic effects depending on the exposure dose. Moreover, Inorganic As is classified as a known carcinogen (Group A) and Pb (Table 6) as a probable carcinogen based on animal studies (Group B2).

The calculated TR values for As and Pb in both adults and children found in this study were seriously alarming and unacceptable. In principles, TR risks (calculated values) lying between 10^{-6} and 10^{-4} are generally considered an acceptable range (USEPA-IRS, 2011), indicating that values lower than $10^{-6}$is considered to be negligible while values above10^{-4} are considered unacceptable. The TR values recorded in children were found to be higher than that of adults and could be due to a relatively higher consumption rate of meat for children per of body weight. Hence, children are more susceptible to metal toxicity through daily consumption of meat than adults in the study population.

It is noteworthy that exposure to heavy metals via meat consumption may pose potential health risks to the consumers in the study area. The potential chronic toxicity is of great concern because the analysed metals except copper and zinc were far from permissible limits (FAO/WHO, 2011). It is highly imperative to suggest here that continuous monitoring of these toxic metals in all food commodities in the area is advocated in order to safeguard public health.

CONCLUSION

The study observed higher concentrations of heavy metals (Cd, Cr, Cu, Co, As, Ni, and Pb) in meat samples of cattle, sheep, and goat free-ranged in goldmine communities of Zamfara State Nigeria. Meat samples from sheep and goats were found to accumulate more metals than cattle in the area. The estimated daily intakes for most toxic metals (As, Pd, and Cd) were greater than permissible limits in dietary foods. The target carcinogenic risk (TR) values for As and Pb were above the acceptable range (10^{-6} to 10^{-4}), indicating possible cancer risk to the exposed population. Therefore, data from this study demonstrated that consumption of meat from cattle, sheep, and goats may significantly contribute to the body burden of heavy metals and subsequent multi-organ toxicity and high carcinogenic risk. It is highly recommended that there is a need for a concerted effort from national, state, and local authorities and governmental and non-governmental regulatory agencies to come together to put prompt and stringent control measures to ensure food safety for the citizens.

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The authors declared no known competing interests or personal relationships that could have appeared to influence the research reported in this paper.

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