



Heavy Metals Content in Water and Crops in Golding Mining Vicinity on Major Dams in Zamfara State, Nigeria

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ABSTRACT: The concentrations of heavy metals in water and food crops (tomatoes and cassava) grown around the three major dams in Zamfara State where gold mining is usually done were determined using standard methods. The concentration of heavy metal determined in water and food crops were generally high during the dry season with exception of Hg which recorded its highest concentration in year 2015. Zn and Cr levels in the water and food crops were within international safe limits while Cd, Pb and Hg levels were far above (0.01, 0.01 and 0.001mg/l) WHO and USEPA limits respectively. In this work, the concentration of the analyzed heavy metals in mg/l was in the order of Hg > Pb > Cd > Cr > Zn. Correlation analysis showed a significant and positive relationship for Cd and Zn and Hg and Cr during the wet season and a significant positive at the ($p > 0.05$) relationship for Cr and Zn in the dry season. The pollution index value of the water samples across all the three dams indicated that there is need for immediate intervention to ameliorate pollution particularly in the dry season

DOI: <https://dx.doi.org/10.4314/jasem.v22i6.22>

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Dates: Received: 16 April 2018; Revised: 24 June: 2018; Accepted: 30 June 2018

Keyword: Dams, Heavy metals, food crops, gold mining

Environmental pollution by toxic heavy metals does not only elicit concern in the metropolitan cities but also in remote and rural communities where anthropogenic activities, such as mining, are taking place. Gold mining and processing have been the main sources of heavy metal contamination in the environment (Duruibe *et al.*, 2007; Boamponsem *et al.*, 2010; Girigisu *et al.*, 2012). The uncontrolled dissemination of waste effluents to large water bodies has negatively affected both water quality and aquatic life (Abdulrahman, *et al.*, 2008). During the processing of the ores for gold, poisonous substances such as oxides and sulphides of heavy metal pollutants are released into the environment Boamponsem *et al.*, (2010). Hence, most of the water sources particularly in Zamfara State are gradually becoming polluted due to the addition of these foreign materials from the environment. Artisan mining is an important economic sector in many developing countries. However, limited resources and training, and the availability of cheap, but potentially hazardous methods of extraction and processing of minerals can cause significant threats to both miners and the local environment (Bitala, 2008; Armah *et al.*, 2010). Such a scenario is being experienced in mining locations in Zamfara State. During peak mining periods, up to 5000 people invade the mining areas from outside the State. This has devastating effects on health and environment. Artisan mining in these areas conforms to neither

mining laws nor regulations governing mining operations and environmental management. Studies in a similar region, in Tanzania revealed that symptoms of heavy metal poisoning such as sensory disturbance, hyporeflexia, tremor, gingivitis, metallic taste, neuroasthenia and night blindness are common (Harada *et al.*, 1997; Akabzaa, 2000; Lottermoser, 2007). There have been several reports on acute lead (Pb) poisoning outbreak among the dwellers of some remote villages of Zamfara State, Nigeria. This has been linked to the illegal mining operations by the people of remote communities (Galadima, 2012). Mercury as a pollutant in artisan mining is due mainly to gold processing, when mercury is used to amalgamate gold. Cadmium, which is another common toxic metal, occupying position seven in the priority list of hazardous substances (ATSDR, 1999), generally occurs as an isometric trace element in sphalerite. Cadmium levels are likely to be quite low. Large amounts of Hg are released into the environment as a result of its usage in gold extraction. About 1.32 kg of Hg is lost for every 1 kg of gold produced which goes directly into water, soil and streams as inorganic Hg and later converted into organic forms (Matshusa *et al.*, 2012). Several researchers have reported on its high concentration in gold mine tailings. Rafiei *et al.*, (2010) reported 100 mg/kg concentrations of Hg in Iran whereas Mathusa *et al.*, (2012) reported concentrations as high as 1920

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mg/kg in Kenya. Some of the inorganic Hg that reaches aquatic ecosystems is converted by microbes into organic methylmercury (MeHg), which accumulates in fish. Mercury compounds cause oxidative stress to bacterial cells due to imbalance between pro-oxidant and anti-oxidant homeostasis. They have high affinity for thiol group containing enzymes and proteins that serve as a line of cellular defense against Hg compounds. On gaining access to the cell, both Hg II (Hg^{2+}) and MeHg form covalent bonds with cysteine residues of proteins and deplete cellular antioxidants (Valko et al., 2006). The aim of the research is to investigate the levels of heavy metals in gold mining areas within the three major dams and food crops grown around the dams in Zamfara State, Nigeria.

MATERIALS AND METHODS

In the preparation of solution, analytical reagent grade chemicals and distilled water were used. All glassware were washed with detergent and rinsed in water before immersion in 10% nitric acid solution. They were further rinsed with distilled water before drying in the oven (Ayodele et al., 1994).

Description of the Sampling Area: Zamfara State is located in the North Western Zone of Nigeria between Latitude $11^{\circ} 40'$ E and Longitudes $70^{\circ} 25'$ E with an altitude of 420m. The state inherited three major dams from the former Sokoto State in the quest to provide potable water for the teeming population. The dams are located in Gusau LGA (Gusau dam), Maradun LGA (Bakolori dam), and Maru LGA (Dangulubi dam). The dams are considered for investigation as a result of lead poisoning epidemic that struck the state sometimes around year 2010. The dams became very necessary to investigate because mining activities are still going on across the state in which water from the dams are used in the mining processes and more so because livestock are seen drinking from the dams and plants are sited growing around the vicinity of the mining areas.

Treatment of Sampling Containers: A one-liter polythene bottle was used for the collection of water sample. It was thoroughly washed with detergent solution, rinsed with distilled water and then leached with 10% HNO_3 and 1:1 HCl for 48 hours. The bottle was then rinsed with distilled water and allowed to drain to dryness. Polythene containers used for the collection of food crops and was treated similarly.

Collection of Water Samples from the Dams: The water samples were collected by dipping a two litre plastic bottle just below the water surface at a depth of

one meter from each of the six sampling locations. Pre-cleaned 2 litre polyethylene sampling bottles were used for sampling. At each sampling locations, the bottles were rinsed 3 times with the water before collection of the sample. The samples were preserved by acidifying with 2 ml of concentrated HNO_3 in order to achieve a pH of 2 and prevent metal adsorption onto the inner surface of the container. Digestion of Water Samples for Heavy Metal Analysis and food such as tomatoes and cassava was done according to the method by APHA, 2005.

Contamination factor (CF): The Contamination factor (CF) is the ratio obtained by dividing the concentration of each metal in the sediment, water and fish samples by

$$CF = \frac{\text{concentration of metal in sample}}{\text{concentration of metal in background}}$$

In this study, the WHO (2004) guidelines values for drinking water quality were selected as background level for the calculation of contamination factors of the water samples. The contamination levels may be classified based on their intensities on a scale ranging from 1 to 6 (0= none, 1= none to medium, 2= moderate, 3= moderately to strong, 4= strongly polluted, 5=strong to very strong, 6=very strong) (Bhuiyan et al., 2010 and Harikumar et al., 2010). The CF value of 6 indicates that the metal concentration is 100 times greater than what would be expected in the world average shale (Wedepohl, 1995).

Pollution Load Index (PLI): The PLI was obtained as a concentration factor of each heavy metal with respect to the background value in the samples.

According to Angula (1996) the PLI is able to give an estimate of the metal contamination status and the necessary action that should be taken, where CF_1 to CF_n indicate the contamination factors calculated for the first sample to the n^{th} one. A PLI value close to one indicates heavy metal loads near the background level, while values above one indicate sample pollution (Bhupander et al., 2011; Angula, 1996).

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times CF_4 \dots \times CF_n}$$

RESULTS AND DISCUSSION

The concentration of Pb and Hg in the water samples (Table 1) from this water bodies exceeded the permissible limit of 0.05 and 0.001 mg/L respectively set by (WHO, 2004; USEPA, 2002). In this work, the concentration of the analyzed heavy metals in mg/L was in the order of $\text{Hg} > \text{Pb} > \text{Cd} > \text{Cr} > \text{Zn}$. The

abnormal high contamination factor (CF) level (table 3) of Hg was expected across all the three dams since gold processing is still very active in these areas. The higher contamination factor for all the heavy metals during the dry season might be due to seasonal rainfall which dilutes some concentration of the metal concentration during the wet season. The pollution index value of the water samples across all the three dams indicated that there is need for immediate intervention to ameliorate pollution particularly in the dry season. Generally, the natural sources of heavy metals in rivers and dams, Marine and Coastal water are through land, heavy fresh water inflow,

agricultural waste, aquaculture discharge and river run-offs and the mechanical and chemical weathering of rocks (Ashokkumar *et al.*, 2009). Correlation analysis (Tables 2a and b) showed a significant and positive relationship for Cd and Zn and Hg and Cr during the wet season and a significant positive at the ($p>0.05$) relationship for Cr and Zn in the dry season. Similar observation was reported on the study of heavy metals in Ureje dam in Ado-Ekiti by Adefemi *et al.* (2007) and in Kanji dam (Amoo *et al.*, 2005) and other studies on surface water (Chapman, 1999; Asaolu *et al.*, 1997; Karadede *et al.*, 2000)

Table 1: Heavy Metals Concentrations (mg/l) of Water Samples for Two wet and dry Seasons

location	year	Zn	Cd	Cr	Pb	Hg
L1 wet	2014	0.0020±0.0001	0.0040±0.0001	0.0210±0.0003	0.0918±0.0004	0.2638±0.000
	2015	0.0024±0.0001	0.0036±0.0002	0.0262±0.0040	0.0029±0.0001	1.5532±0.003
L2 wet	2014	0.0060±0.0001	0.0064±0.0003	0.0078±0.0003	0.0476±0.0020	ND
	2015	0.0066±0.0001	0.0024±0.0002	0.0044±0.0003	0.0068±0.0001	0.5005±0.003
L3 wet	2014	0.0042±0.0002	0.0024±0.0001	0.0014±0.0004	0.0126±0.0002	ND
	2015	0.0049±0.0003	0.0066±0.0005	0.0042±0.0002	0.0480±0.0020	0.5308±0.003
L1 dry	2014	0.0244±0.0002	0.0904±0.0001	0.0682±0.0030	0.1376±0.0040	ND
	2015	0.0112±0.0020	0.2044±0.0020	0.0694±0.0040	0.0062±0.0001	1.5570±0.003
L2 dry	2014	0.0068±0.0001	0.0740±0.0003	0.0034±0.0003	0.0876±0.0020	ND
	2015	0.0266±0.0010	0.0744±0.0020	0.0304±0.0030	0.1314±0.0010	0.5573±0.003
L3 dry	2014	0.0076±0.0002	0.0550±0.0010	0.1090±0.0040	0.0334±0.002	ND
	2015	0.0118±0.0030	0.0140±0.0050	0.1098±0.0020	0.0668±0.0020	0.8506±0.003

n = 3; mean ± S.D . key: L1= Gusau dam, L2= Bakolori dam and L3= Dangulbi dam

Concentrations of heavy metal in tomatoes and cassava (Table 4 and 5) in this work revealed spatial variation at all three dams during the study periods. The relative abundance of different metals at the three locations during the wet and dry seasons varied with locations and with season in the tomatoes and cassava samples.

Table 2a: Correlation matrix of Heavy Metals Concentrations (mg/l) in Water Samples for Two Wet seasons

Correlations For wet					
	Zn	Cd	Cr	Pb	Hg
Zn	1				
Cd	.394*	1			
Cr	-.130	-.074	1		
Pb	-.021	-.128	-.102	1	
Hg	-.023	.280	.365*	.034	1
	36	36	36	36	36

*.Correlation is significant at the 0.05 level (2-tailed)

Table 2b: Correlation matrix of heavy Metals Concentrations (mg/l) in Water Samples for two dry seasons

Correlations For dry					
	Zn	Cd	Cr	Pb	Hg
Zn	1				
Cd	-.023	1			
Cr	.565**	-.247	1		
Pb	.243	-.306	.056	1	
Hg	-.017	.233	.025	-.189	1
	36	36	36	36	36

** Correlation is significant at the 0.01 level (2-tailed).

Heavy metals are found naturally in the earth, and become concentrated as a result of human activities such as industrial production, mining, agriculture and transportation (Noor-ul *et al.*, 2015; Bassey *et al.*, 2014). These metals have positive and negative roles in human life (Noor-ul *et al.*, 2015, Colak *et al.*, 2005; Oktem *et al.*, 2005).

For instance heavy metal such as copper, chromium, cobalt and zinc are essential micronutrients for higher animals and for plant growth (Manohara *et al.*, 2014). Studies have revealed that fruits and leafy vegetables are vulnerable to heavy metal contamination from soil, waste water and air pollution (Manohara *et al.*, 2014).

The presence and accumulation of the analyzed heavy metals in the cassava and tomatoes plants could be traced to their presence in the wastes used. Liu *et al.*, 2005 opined that the soil type,

root stock, mulching, irrigation, fertilization, and other cultural practices influence the water and nutrient supply to plant and affect the composition and quality attributes of the harvested parts. Zn is the least toxic among all heavy metals, and is an essential element in the human diet as it is required to maintain the proper functioning of the immune system, normal brain activity and is fundamental in the growth and development of the foetus.

Excessive Zn in the diet e.g. prolonged daily intake of Zn ranging from 150 - 450 mg/day is also detrimental to human health (Johnson *et al.*, 2007). This was also similar to the report by Kirmani *et al.*, 2011 and Bosiacki *et al.*, 2009, on the levels of Zn in onions, carrot, cabbage, and tomatoes above permissible levels by FAO/WHO.

Table 3: Heavy Metal Contamination Factors (CF) and Pollution Load Index (PLI) in Water for Wet and Dry Seasons

Location/WET	Zn	Cd	Cr	Pb	Hg	PLI
Gusau dam	0.0004	0.3800	0.2360	0.9470	90.8500	0.3206
Bakolori dam	0.0013	0.4400	0.0610	0.5440	50.0500	0.2486
Dangulbi dam	0.0009	0.4500	0.0280	0.6060	53.0800	0.2053
DRY						
Gusau dam	0.0035	14.7400	0.6880	1.4380	155.700	1.5188
Bakolori dam	0.0033	7.4200	0.1690	2.1900	557.300	1.3858
Dangulbi dam	0.0019	3.4500	1.0940	1.0020	850.600	1.4423

Table 4: Heavy Metals Concentrations (mg/kg) of Tomatoes Samples for wet and dry Seasons

location	year	Zn	Cd	Cr	Pb	Hg
L1 wet	2014	0.029±0.002	0.019±0.001	0.183±0.003	0.261±0.004	0.556±0.002
	2015	0.024±0.002	0.010±0.002	0.010±0.004	0.334±0.001	1.746±0.003
L2 wet	2014	0.039±0.001	0.019±0.003	0.261±0.003	0.125±0.002	0.163±0.000
	2015	0.049±0.001	0.012±0.002	0.012±0.003	0.396±0.001	2.202±0.003
L3 wet	2014	0.052±0.002	0.014±0.001	0.157±0.004	0.271±0.002	0.446±0.000
	2015	0.056±0.003	0.016±0.005	0.016±0.002	0.511±0.002	0.600±0.003
L1 dry	2014	0.036±0.002	0.432±0.001	0.364±0.003	0.250±0.004	ND
	2015	0.031±0.002	0.256±0.002	0.369±0.004	0.365±0.001	4.363±0.003
L2 dry	2014	0.058±0.001	0.216±0.003	0.324±0.003	1.074±0.002	ND
	2015	0.041±0.001	0.334±0.002	0.314±0.003	0.500±0.001	2.527±0.003
L3 dry	2014	0.093±0.002	0.668±0.001	1.165±0.004	0.615±0.002	ND
	2015	0.071±0.003	0.354±0.005	1.150±0.002	0.644±0.002	1.343±0.003

n = 3; mean ± S.D. key: L1= Gusau dam, L2= Bakolori dam and L3= Dangulbi dam

Table 5: Heavy Metals Concentrations (mg/kg) of Cassava Samples for wet and dry Seasons

location	year	Zn	Cd	Cr	Pb	Hg
L1 wet	2014	0.177±0.002	0.026±0.001	0.111±0.003	0.365±0.004	ND
	2015	0.197±0.002	0.019±0.002	0.275±0.004	0.155±0.001	0.673±0.003
L2 wet	2014	0.148±0.001	0.013±0.003	0.179±0.003	0.189±0.002	ND
	2015	0.158±0.001	0.009±0.002	0.190±0.003	0.858±0.001	1.195±0.003
L3 wet	2014	0.074±0.002	0.009±0.001	0.190±0.004	0.219±0.002	ND
	2015	0.084±0.003	0.005±0.005	0.157±0.002	0.462±0.002	1.059±0.003
L1 dry	2014	0.373±0.002	0.963±0.001	1.008±0.003	0.851±0.004	ND
	2015	0.183±0.002	0.806±0.002	1.018±0.004	0.744±0.001	0.874±0.003
L2 dry	2014	0.249±0.001	0.256±0.003	0.529±0.003	0.188±0.002	ND
	2015	0.240±0.001	0.197±0.002	0.548±0.003	1.636±0.001	1.900±0.003
L3 dry	2014	0.166±0.002	0.629±0.001	0.859±0.004	0.977±0.002	ND
	2015	0.087±0.003	0.747±0.005	0.867±0.002	1.012±0.002	1.902±0.003

n = 3; mean ± S.D. key: L1= Gusau dam, L2= Bakolori dam and L3= Dangulbi dam

The concentration of the heavy metal reported in the present study are lower than the one reported by Kakulu (1985) for Niger Delta, Okoye (1989) for Lagoon and that which was reported by Asaolu (1998) for Ondo State Coastal area. Average Pb level for this study are higher than the average value of 0.50µg/g which was obtained in a similar study along the highways of Benue State Nigeria (Adebayo O. and Rapheal, 2011) and also higher than 1.73µg/g in cassava tubers recorded by Nkwocha *et al* (2012) in an oil

field in Bayelsa State. These values are unusually high and it's reflective of the anthropogenic activities around the dams.

The calculated contamination factor (CF) (table 7 and 9) revealed that only Hg had CF values greater 6, which indicates a serious contamination of tomatoes harvested from all the location. The pollution load index (PLI) across all the locations had PLI values less than 1.

This indicates that there is no serious overall pollution load on the tomatoes samples. While in cassava sample, the calculated contamination factor (CF) for all the heavy metals with exception of Zn is very high, this showed the level of contamination of the samples.

While the pollution load index (PLI) for all the location showed serious overall load of pollution on the sample.

Pearson correlation analysis (Tables 6a and b; 8a and b) conducted to examine whether there is a relationship between the heavy metal concentrations in the tomatoes and cassava tubers for two wet and dry seasons revealed a significant positive relationship for Cr and Cd, Cd and Zn, Hg and Pb in the wet season and a significant and positive relationship for Cr and Cd, and Pb and Cr for dry season.

Conclusion: The levels of the metals in the water bodies were generally high with exception of Zn metal which should cause trepidation to both the aquatic lives and human health, hence calling for urgent regular monitoring of the dam and control of anthropogenic input into the water bodies. The pollution level of all the three dams and sample was accessed using the contamination factor (CF) and pollution load index (PLI), which revealed that all the water bodies have suffered from metal pollution.

Table 6a: Correlation matrix of Heavy Metals Concentrations (mg/l) in Tomatoes Samples for two wet seasons

Correlations	For wet				
	Zn	Cd	Cr	Pb	Hg
Zn	1				
Cd	.416*	1			
Cr	.128	.471**	1		
Pb	.166	-.350*	-.429**	1	
Hg	.375*	-.414*	-.709**	.277	1
	36	36	36	36	36

*.Correlation is significant at the 0.05 level (2-tailed). **.Correlation is significant at the 0.01 level (2-tailed)

Table 6b: Correlation matrix of Heavy Metals Concentrations (mg/l) in Tomatoes Samples for two dry seasons

Correlations	For dry				
	Zn	Cd	Cr	Pb	Hg
Zn	1				
Cd	.416*	1			
Cr	.128	.471**	1		
Pb	.166	-.350*	-.429**	1	
Hg	-.375*	-.414*	-.709**	.277	1
	36	36	36	36	36

*.Correlation is significant at the 0.05 level (2tailed). **.Correlation is significant at the 0.01 level (2tailed).

Table 7: Heavy Metal Contamination Factors (CF) and Pollution Load Index (PLI) in Tomatoes for Wet and Dry Seasons

Location/WET	Zn	Cd	Cr	Pb	Hg	PLI
Gusau dam	0.0026	0.1050	0.0029	0.0383	6.1667	0.0453
Bakolori dam	0.0005	0.0443	0.0016	0.0164	6.0033	0.0206
Dangulbi dam	0.0006	0.0653	0.0018	0.0354	2.4000	0.0225
DRY						
Gusau dam	0.0053	1.5733	0.0132	0.1108	10.7567	0.1671
Bakolori dam	0.0015	2.4250	0.0054	0.0204	2.9867	0.0662
Dangulbi dam	0.0011	1.2450	0.0069	0.0430	8.5833	0.0915

Table 8a: Correlation matrix of Heavy Metals Concentrations (mg/l) in Cassava Samples for Two Wet seasons

Correlations	For wet				
	Zn	Cd	Cr	Pb	Hg
Zn	1				
Cd	.357*	1			
Cr	.278	.138	1		
Pb	.277	.080	.052	1	1
Hg	.121	-.262	.320	.646**	1
	36	36	36	36	36

*.Correlation is significant at the 0.05 level (2-tailed). **.Correlation is significant at the 0.01 level (2-tailed).

Table 8b: Correlation matrix of Heavy Metals Concentrations (mg/l) in Cassava Samples for Two Dry seasons

Correlations	For dry				
	Zn	Cd	Cr	Pb	Hg
Zn	1				
Cd	.165	1			
Cr	.173	.495**	1		
Pb	.086	.328	.619**	1	
Hg	.173	-.157	.095	.169	1
	36	36	36	36	36

**.Correlation is significant at the 0.01 level (2tailed).

Table 9: Heavy Metal Contamination Factors (CF) and Pollution Load Index (PLI) in Cassava for Wet and Dry Seasons

Location/Wet	Zn	Cd	Cr	Pb	Hg	PLI
Gusau dam	0.0037	2.2500	1.9300	26.000	0673	3.0954
Bakolori dam	0.0031	1.1000	1.8450	52.3500	1195	3.2930
Dangulbi dam	0.0016	0.7000	1.7350	34.0500	1059	2.3335
Dry						
Gusau dam	0.0056	88.4500	10.6300	79.7500	0874	12.9512
Bakolori dam	0.0049	22.6500	5.3850	91.2000	1902	10.0682
Dangulbi dam	0.0025	68.8000	8.6550	99.4500	1902	12.3297

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