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Accumulation and distribution of mercury in agricultural soils, food crops and associated health risks: A case study of Shenda gold mine-Geita Tanzania

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ABSTRACT

Contamination of the environment and foodstuff by potentially harmful elements (PHEs) has become a serious concern due to the associated health risks to the population. In the present study total mercury (Hg) levels in soil and food crops from farms around Shenda gold mine in Geita Tanzania was determined by Cold Vapor Atomic Fluorescence Spectrometry (CVAFS). Generally, it was found that there was significant different of Hg content in soil and food crops ($P < 0.05$) among studied sites. The total Hg concentrations determined were in the ranges of; soils (0.003-0.1220 mg/kg), rice (0.0752-0.1587 mg/kg) and vegetables (0.0556-0.3439 mg/kg) all measurement were based on dry weight basis (dw). Total Hg levels in soil were compared with the maximum allowable concentration (MAC) set by Tanzania Bureau of Standards (TBS) and United Kingdom (UK). All studied soil samples were within acceptable range (2 mg/kg) set by TBS and UK (1 mg/kg). Total Hg contents in food crop samples were compared with Chinese MAC of Hg (0. 01 and 0.02 mg/kg for leafy vegetables and grains respectively in a fresh weight basis). It was observed that Hg contents in potato leaves, pumpkin leaves and Chinese cabbage were within the MAC while Hg levels in cassava leaves and rice grains exceeded the MAC. In addition, Hg associated health risks to consumers of contaminated foods for residents around Shenda gold mine were estimated. The estimated weekly intake (EWI) of Hg due to consumption of rice grain was above the provisional tolerable weekly intake (PTWI) set by FAO/WHO (1.6 mg/kg bw/week) while the EWI due to consumption of leafy vegetables were below the PTWI. Target hazard quotient (THQ) was < 1 due to consumption of vegetables while THQ value for rice was >1, indicating a potential non-carcinogenic risk to adult population from the consumption of rice grain from the study area. Therefore, people living near Shenda goldmine might be potential victim of Hg accumulation in soil and food crops, thus necessary management options have to be in place.

1. Introduction

It has been shown that presence of potentially harmful elements (PHEs) in soil, air, and water may cause negative effects on the food quality and consequently, impacts on human health (Manzoor et al., 2018). The PHEs are usually regarded as hazardous in the environment due to their numerous sources, non-biodegradable, bio-accumulative and their toxicity nature (Ali and Khan, 2018, Hu et al., 2017). They originate from natural and anthropogenic sources (Jan et al., 2015) where natural sources include volcanic eruptions, erosion and weathering of rocks. The main anthropogenic sources include industries, mining and metal smelting, fossil fuel burning, emissions from transportation (automobiles) and various agricultural activities (Morgan, 2013, Pan et al., 2012, Ali et al., 2019). Anthropogenic activities are the main sources of PHEs in soil, water and air in large quantities (Tiwari and Lata, 2018, Gupta et al., 2019).

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Human can be exposed to PHEs through inhalation, dermal contact and ingestion of dietary and nondietary materials (USEPA 2019, Al and Massey, 2019). Although some elements like copper (Cu), zinc (Zn), nickel (Ni), chromium (Cr) and molybdenum (Mo) are essential micronutrients for plants, animals, and humans for various physiological and biochemical functions when available in micro doses, at high concentration they may cause severe toxicity effects. Other elements like mercury (Hg), cadmium (Cd), lead (Pb), and arsenic (As) are nonessential, considered as the most harmful elements for living organisms, which may be toxic even at low concentrations (Engwa et al., 2019, Fashola et al., 2016, Rodríguez-Eugenio et al., 2018).

It has been explained that PHEs contamination by mining is a primary environmental concern on a global scale, particularly in developing countries (Gupta et al., 2019), where there is less strong regulation enforcement to control disposal of mining tailings and effluents. Mining activities can produce massive amounts of waste material and tailings, which release toxic elements to the environment (Song et al., 2018, Pu et al., 2019, Demková et al., 2017) posing risks to ecological safety and human wellbeing. Agricultural lands near mining areas can be exposed to these elements thus food crops grown in contaminated areas may take up and accumulate these elements into the food chain and poses adverse effects to human health (Song et al., 2018, Gunalan et al., 2018). Consumption of such food crops with high concentration may lead to accumulation of PHEs in the body overtime and can lead to serious health risk conditions (Obiora et al., 2019). Also, mine wastes can be dispersed by wind and get deposited on the surfaces of plant leaves and then absorbed into the tissues (Gunalan et al., 2018, Edelstein and Ben-Hur, 2018, Li et al., 2015, Saha et al., 2017). From gold mining activities there number of PHEs which can be released to the environment in form of solid, gases, liquid and aerosols, these includes Cadmium (Cd), Zinc (Zn), Lead (Pb), Mercury (Hg), Chromium (Cr), Nickel (Ni), Arsenic (As), and Copper (Cu) (Fashola et al., 2016, Tun et al., 2020).

Mining exposes harmful elements from underground to the environment, but extraction methods may also introduce new harmful elements to the environment. An example of such experience is mercury which is used in gold extraction processes by artisanal and small-scale gold miners. It is one of the highly considered elements in terms of food chain contamination (Rodríguez-Eugenio et al., 2018). It is a global pollutant because it can be distributed to non-point source locations due to atmospheric deposition (Malcolm et al., 2018). Artisanal and small scale gold mining (ASGM) are reported to be the major sources of global mercury dispersion (Malehase et al., 2017, Diringer et al., 2015). Its release by ASGM was estimated to range from 640 to 1350 mg/year from at least 70 countries (Telmer and Veiga, 2009). Tanzania is reported being one of the most significant African countries in terms of volumes of mercury emissions (Kinyondo and Huggins, 2021). The ASGM areas in Tanzania are found all over the country; in the northern region close to the Lake Victoria e.g., in Geita and Tarime, in the Southern highland part e.g., Chunya, in Eastern regions such as Morogoro (Mutagwaba et al., 2018). There are significant studies reporting mercury contamination problems associated with gold mining in Tanzania (Lema and Mseli, 2017, Koleleni and Mbike, 2018, Ikingura et al., 2006). However, more study needs to be done to address the subsequent impact of mercury on food and foodstuffs quality in the country.

Although there is a long history of mining and use of mercury by ASGM in Tanzania, limited studies have been conducted on human health risk associated with their exposure (Nyanza et al., 2021, Nyanza et al., 2020) and consumption of mercury contaminated foodstuffs (Tungaraza et al., 2011), or in general PHEs (Kacholi and Sahu, 2018). More studies need to be done on the risks of mercury exposure and consumption of contaminated food particularly around mining areas in the country where higher concentrations of contaminants are expected due to the fact that, there is continued use of mercury in ASGM whilst, there are no regular monitoring mechanisms. We are presenting results of the study which shows that in addition to ASGM activities, there are agricultural activities at the vicinity of the mining areas. The close proximity of the farms to the mining areas provides the possibility of soil and crops contamination by mercury. This is a contribution to reveal the current existing risks in relation to mercury. This study was conducted aiming to determine concentrations of total mercury (Hg) present in agricultural soils and food crops grown near ASGM site. Furthermore, the study estimated the human health risks of Hg to the mine surrounding communities through consumption of foodstuff.

Table 1

Geographical location of the sampling sites around Shenda artisanal and small-scale mine and control sites, where soil, rice and vegetable samples were collected.

Sampling site	Location	Distance from mining area (km)
1	3°41′13.515"S, 32°8′47.575"E	0.956
2	3°41′19.167"S, 32°’95.755"E	1.42
3	3°41′31.55"S, 32°9′14.755"E	1.9
4	3°42′43.082"S, 32°3′38.923"E	9.79
5	3°43′6.051"S, 32°3′49.79"E	9.8
6	3°40′51.061"S, 32°8′33.853"E	0.174
7	3°41′7.116"S, 32°8′30.401"E	0.675
8	3°40′51.432"S, 32°9′17.678"E	1.36
9	3°40′54.82"S, 32°9′18.161"E	1.394
10	3°41′8.252"S, 32°7′52.701"E	1.442
11	3°41′22.214"S, 32°8′51.587"E	1.25
12	3°42′34.146"S, 32°357.636"E	9.156

2. Materials and methods

2.1. Study area

This study was conducted at the vicinity of Shenda gold mine in Mbogwe district (longitude 03°22’S, 032°9’E), Geita region in Tanzania (Fig. 1). The regional average annual temperature is 22.87 °C and has the total average rainfall of 1200 mm per year. Due to reliable rainfall, it is one of the main food producing region (Mutagwaba et al., 2018). Geita is one of the regions with the largest number of ASGM operations in Tanzania. The Shenda gold mine is a small-scale gold mine in Masumbwe ward, with a population of about 18,716 inhabitants and an area of about 129.9 km^2. In addition to mining activities the community is also involved in agriculture, producing mainly food crops for their own consumption close to the mine. The staple foods include maize and rice.

2.2. Sampling, processing and analysis

2.2.1. Soil and plant sampling

Sampling was conducted during wet season (April 2021) and dry season (September 2021). Soil and plant samples were collected from 12 different sites at varying distances as shown in Table 1. Three (3) sites (4, 5 and 12) were control sampling sites located about 9 km from the mining area. Three selected sites were involved in paddy farming (sites 1, 2, and 3) and six (sites 6, 7, 8, 9, 10 and 11) were involved in vegetable growing. Sampling was done following standard protocols described by Estefan et al. Estefan et al. (2013). Five subsamples of soil were randomly collected from topsoil (from 0 to 20 cm depth) by using soil auger and combined to make a single composite sample and packed into polyethylene zip bags.

Sampling of food crops was done by taking into account of their availability during the sampling season. Rice (*Oryiza sativa*) and vegetables such as pumpkin leaves (*Cucurbita moschata*), cassava leaves (*Manihot esculentum*), Chinese cabbage (B*rassica rapa* subsp. *pekinensis*) and potato leaves (*Ipomea batata*) were collected from the corresponding sites where soil samples were collected and packed in paper bags. Soil and plant samples were transported in hard boxes. A total of 34 composite samples of soil and 30 composite samples of food crops were collected and transferred to the Chemistry laboratory at the Sokoine University of Agriculture, Tanzania for processing and analytical analysis.

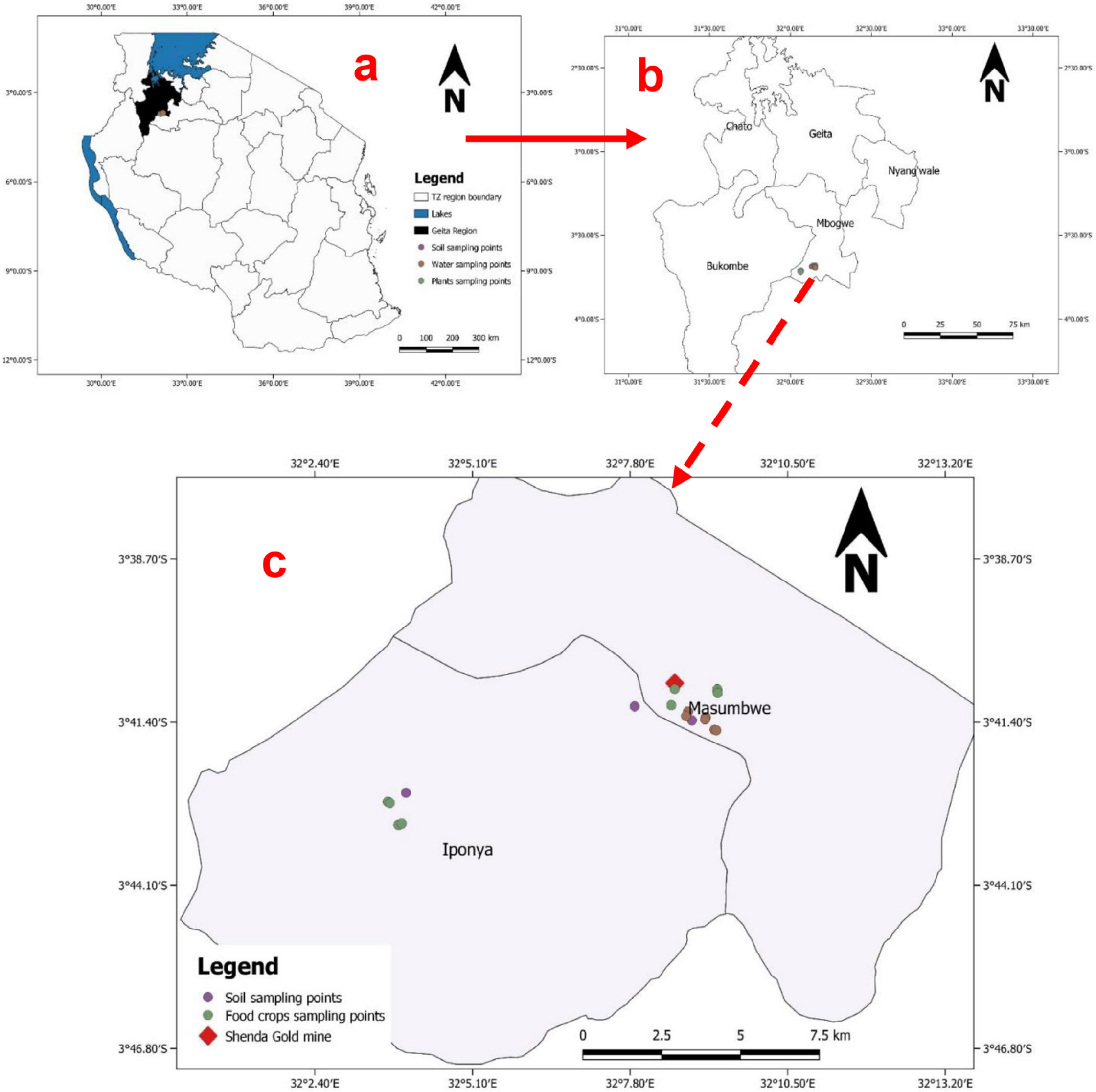

Fig. 1. Sampling sites for soil and food crops samples around Shenda artisanal gold mine in Mbogwe district of Geita region in Tanzania.

2.2.2. *Samples processing*

Prior to analysis, plant and soil samples were processed following procedures described by Estefan et al. (2013)with minor modifications. Edible parts of food crop samples were washed with tap water and rinsed with distilled water to remove soil particles attached, followed by drying with tissue paper and then dried at room temperature for 1 week and oven dried at 40˚C to constant weight. The rice samples were dehusked by using a mortar and pestle. The samples were ground with mortar and pestle to fine powder, sieved through 2 mm mesh and kept in polyethylene zipper bags at ambient temperature prior to analysis.

Soil samples were air dried for two weeks at ambient temperature. The dried samples were pulverized using mortar and pestle and then sieved through a 2 mm stainless steel mesh. The homogenized samples were stored in polyethylene zipper bags at ambient temperature until analysis.

2.2.3. *Soil and plant sample analysis*

Digestion of soil samples. Soil samples were digested following EPA method 1631 (U.S. EPA, 2001). About 0.5 g of samples were weighed into 40 ml reagent bottle where in each of the samples, 8 ml of high purity analytical grade conc. Hydrochloric acid (HCl) and 2 ml of conc. Nitric acid ($HNO_{3)}$ were added in a fume chamber and allowed to digest at room temperature overnight. Into each of the digested samples 0.07 M (35%) bromine monochloride (BrCl) solution was added to a final volume of 40 ml then allowed to settle overnight to oxidize all mercury species in the sample.

Digestion of plant samples. Plant samples were digested following EPA method 1631 (U.S. EPA 2001). Where 0.2 g of powders plant samples were weighed into a 40 mL reagent bottle then 7 ml of HNO3 (69%) and 3 ml of H2SO4 (98%) were added into the samples in a fume hood. The reagent bottles were placed in water bath for 1 h at 70°C then increased to 100 °C for 3 h. The mixture was cooled followed by addition of 0.5 ml of BrCl then allowed to rest overnight followed by dilution with deionized water to a final volume of 40 ml.

Sample analyses. Analysis of samples was done by using sub-samples of 1.25 ml of the digested and oxidized samples to analytical vials, then diluted to 25 ml with deionizer water followed by addition of 0.1 ml of 30% hydroxylamine hydrochloride ($NH_2OH.HCl$) to neutralize BrCl. Into each of the samples in the analytical vials, 0.1 ml of 20% Stannous Chloride ($SnCl_2$) in 10% HCl was added, followed by immediate closure with Teflon lined caps and thorough mixing the vial contents. The analysis of Hg in soil and plant samples was done by Cold Vapour

Atomic Fluorescence Spectrometry (CVAFS) method using a Total mercury analyzer (Brooks RAND^{TM} model III). The concentrations of mercury were established by using a calibration curve of VWR-BDH certified mercury standard and analytical quality control was done by simultaneously analyzing samples and certified reference estuarine Sediment (ERM-CC580).

2.3. Health risk assessment

Risk assessment was performed in order to quantitatively determine the possible health risks posed to residents around Shenda mining due to consumption of mercury contaminated foods. Potential human health risks of Hg exposure to residents of Shenda, through consumption of food crops was assessed using estimated daily intake (EDI), estimated weekly intake (EWI) and target hazard quotient (THQ).

2.3.1. Estimated daily intake (EDI) and estimated weekly intake (EWI)

Average daily intake of Hg through ingestion of food crops was estimated based on the mean concentration of Hg in each food crop. Eq. (1) was used to calculate the human exposure level (mg/kg/day) as used by Gebeyehu and Bayissa (2020) and Eq. (2) was used to calculate the weekly exposure level (mg/kg/week) . The estimated weekly intake (EWI) was compared with the provisional tolerable weekly intake (PTWI) of Hg provided by FAO/WHO (2018).

$$\text{EDI} = \frac{\text{C x IR x EF x ED x CF x 0.001}}{\text{BW x AT}} \tag{1}$$

$$\text{EWI} = \text{EDI x 7days/week} \tag{2}$$

Where EDI is the estimated daily intake of Hg through ingestion (mg/kg-day); C is Hg concentration (mg/kg dry weight); IR is the food consumption rate (158 and 307 g/person/day for vegetable and rice respectively); EF is exposure frequency (365 days/year); ED is the exposure duration (66.1 years), equivalent to average life time; CF is concentration conversion factor for fresh weight to dry weight, the ratio of 0.085 for vegetables and 0.86 for rice (Rehman et al., 2019, Xiao et al., 2017, Chen et al., 2018); BW is the body weight for an adult, which is 60 kg (Kacholi and Sahu, 2018); TA is the average exposure time (66.1 years x365 days) and 0.001 is a unit conversion factor.

2.3.2. Target hazard quotient (THQ)

Target hazard quotient was used to assess non-carcinogenic human health risk from consumption of food crops contaminated by Hg. Eq. (3) was used to calculate the THQ values of the local population resulting from consumption of contaminated food crops (Gebeyehu and Bayissa, 2020).

$$\text{THQ} = \frac{\text{EDI}}{\text{RfD}} \tag{3}$$

Where EDI is estimated daily intake of Hg in mg/day/kg body weight and RfD is oral reference dose mg/kg body weight/day. When the THQ > 1, there is potential non-carcinogenic risks while THQ < 1, there is no potential non-carcinogenic risks (Gebeyehu and Bayissa, 2020, Bortey-Sam et al., 2015). The data used for the estimation of human health risk is compiled in Table 2.

Table 3
Average concentrations of mercury (mg/kg) in soil samples collected from different agricultural sites around Shenda gold mine during wet and dry season (April and September, 2021).

Site	Mean	SD (±)	Minimum	Maximum
1	0.0608	0.0557	0.0030	0.1220
2	0.0152	0.0076	0.0064	0.0256
3	0.0183	0.0106	0.0100	0.0370
4	0.0106	0.0035	0.0049	0.0137
5	0.0095	0.0008	0.0087	0.0102
6	0.0197	0.0056	0.0146	0.0248
7	0.0092	0.0029	0.0053	0.0112
8	0.0104	0.0013	0.0092	0.0116
9	0.0114	0.0039	0.0079	0.0150
10	0.0101	0.0036	0.0060	0.0154
11	0.0095	0.0001	0.0094	0.0096
12	0.0139	0.0019	0.0126	0.0152

2.4. Statistical analysis

Data management and processing was done using Microsoft Office Excel 2010 (Microsoft Office 2010, USA) and statistical analysis of the data was performed using Jamovi Statistical software (1.2.5). Total Hg in soil, rice and vegetables within and between the sites (in test and control sites) was compared using the One-Way Analysis of Variance (ANOVA) and Tukey Post Hoc analyses to test the existence of significant difference at 5% significance level (the differences were considered significant with P value<0.05).

3. Results and discussion

3.1. Total mercury concentrations in soil samples

Table 3 summarizes the average concentration of total mercury (Hg) in agricultural soils collected from different sites during wet and dry seasons. In general, all the soil samples had detectable total Hg concentrations. Maximum concentration (0.1220 mg/kg) was observed at site 1 and a minimum concentration of about 0.0030 mg/kg was recorded at site 1. The mean Hg concentrations values for all 12 sites were below the maximum permissible guideline values of 2 mg/kg set by the TBS (Tanzania Bureau of Standards, 2007) and 1 mg/kg UK standard (Amlinger et al., 2004) for agricultural soil. Regarding the overall distribution of Hg in agricultural soils, the results show that there was a significant difference among sites (Table 4). The mean concentration of Hg in soils collected from site 1 was significantly higher (P-value <005) than those collected from other sampling sites as shown in Table 4. The results indicated that the concentrations of Hg decreased with respect to distance from the mining area as shown in Fig. 2.

Table 2
Parameters and variables used in the calculation of human health risk.

Parameters	Food crops		Refs.
	Vegetables	Rice	
C (mg/kg dry weight)	Table 6	Table 6	This study
IR (g/day)	158	307	FAO (2008)
EF (days)	365	365	Pu et al. (2019)
ED (years)	66.1	66.1	National Bureau of Statistics (2020)
CF	0.085	0.86	Xiao et al. (2017), Chen et al. (2018)
BW (kg)	60	60	Kacholi and Sahu (2018)
AT (days)	365 × 66.1	365 × 66.1	-
RfD (mg/kg/day)	0.0003	0.0003	U.S. EPA (2004)

Table 4
The post hoc analysis showing Hg mean difference among soils from different sites.

Site	1	2	3	4	5	6	7	8	9	10	11	12
1	—	0.0456***	0.04253***	0.05015***	0.05132***	0.04109**	0.05164***	0.05042***	0.04936***	0.05069***	0.05131**	0.04689*
2		—	-0.00308	0.00454	0.00571	-0.00453	0.00603	0.00481	0.00375	0.00507	0.00569	0.00128
3			—	0.00762	0.00879	-0.00145	0.00911	0.00789	0.00683	0.00815	0.00877	0.00436
4				—	0.00117	-0.00907	0.00149	2.71E-04	-7.93e−4	5.33E-04	0.00115	-0.00326
5					—	-0.01023	3.25E-04	-8.96e−4	-0.00196	-6.34e−4	-1.40e−5	-0.00443
6						—	0.01056	0.00934	0.00827	0.0096	0.01022	0.00581
7							—	-0.00122	-0.00229	-9.59e−4	-3.39e−4	-0.00475
8								—	-0.00106	2.62E-04	8.82E-04	-0.00353
9									—	0.00133	0.00195	-0.00247
10										—	6.20E-04	-0.00379
11											—	-0.00441
12												—

Mean difference values with asterisk (*) are statistically significant different at;
* $P < 0.05$,
** $P < 0.01$,
*** $P < 0.001$

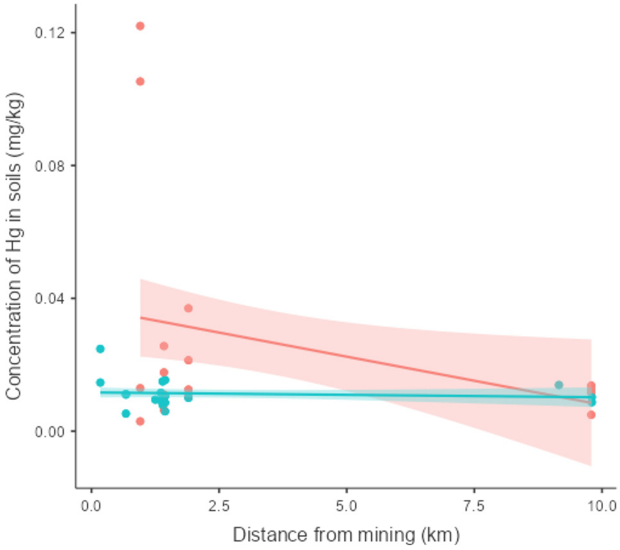

Fig. 2. The pattern of Hg concentration in soils collected from vegetable and rice fields vs distance from mining area.

Table 5
Concentrations of Hg (mg/kg) in soil under vegetable and rice cultivation near the mine and control sites.

Land use type/ Sites	Status	Mean	Minimum	Maximum
Rice farms	Near mining (0-2 km)	0.0321±0.0382	0.0030	0.1220
	Control (>9 km)	0.0106±0.00354	0.0049	0.0137
Vegetable farms	Near mining (0-2 km)	0.0114±0.00465	0.0053	0.0248
	Control (>9 km)	0.0109±0.00231	0.0087	0.0139

Comparison between Hg concentrations of soil samples collected in agricultural fields located near mining area and control area indicated that Hg concentrations were slightly higher in agricultural fields located near mining areas (0.0206 mg/kg) as compared to control area (0.0108 mg/kg). The higher concentrations of Hg around farms near mining area could be attributed to atmospheric deposition of Hg vapor released from gold processing operations by ASGM in the area. Heating mercury-gold amalgams in ASGM results in the production of mercury vapor (Esdaile and Chalker, 2018). The Hg levels which were found in control sites can be ascribed to the distribution of mercury vapor following wind trajectories from the mining area as has been explained that mercury gas released to air may travel long distances (Cohen et al., 2004) and come down with rain and contaminate the soil (Veiga et al., 2006).

When comparison was made to vegetable farms, rice fields contained significantly ($p < 0.01$) higher concentrations of Hg (0.0321mg/kg) than vegetable farms (0.0114mg/kg) as shown in Table 5. This trend could be due to the fact that rice is cultivated in swampy fields which are within the drainage area of Shenda gold mine during rainfall. Mercury used in the extraction of gold may be released from mine tailings and carried out by run-off into low land farms.

Previous study in Namungo gold mine in Tanzania byKoleleni and Mbike (2018) reported higher concentrations (3.4-19.5 mg/kg) of Hg in soil than in this study. In comparison with studies beyond Tanzania, the mean values reported in this study (0.0092-0.0608 mg/kg) are less than the values reported by *Xiao et al. (2017) in China (0.69-23.7 mg/kg) and Bortey-Sam et al. (2015) in Ghana (0.030-2.4 mg/kg). However, the mean values reported in this study (0.0092-0.0608 mg/kg) are less than the values reported by Xiao et al. (2017) in China (0.69-23.7 mg/kg). This implies site specific and wide range of concentration differences around the world contaminated sites.

Table 6

Levels of Hg (mg/kg) in food crop samples collected from agricultural fields near Shenda mine in Tanzania.

Food crop	N	Mean	Minimum	Maximum
Cassava leaves	4	0.1532 ± 0.1280	0.0772	0.3439
Chinese cabbage	4	0.0960 ± 0.0143	0.0845	0.1165
Potato leaves	3	0.1173 ± 0.0342	0.0798	0.1466
Pumpkin leaves	7	0.1187 ± 0.0787	0.0556	0.2862
Rice grains	6	0.0976 ± 0.0343	0.0752	0.1587

3.2. Concentrations of total mercury in food crops

Mean concentrations of Hg (on a dry weight basis) in edible parts of food crops collected from different sites are shown in Table 6. The results indicated that there were significant differences of mean concentrations of Hg between food crops collected from different sites (P-value < 0.05) as shown in Table 7. The mean Hg concentrations in this study were cassava leaves (0.1532 mg/kg; dw), Chinese cabbage leaves (0.0960 mg/kg; dw), potato leaves (0.1173 mg/kg; dw), pumpkin leaves (0.1187 mg/kg; dw) and rice (0.0976 mg/kg; dw). Among food crops studied decreased in the order; cassava leaves (0.1532 mg/kg)> pumpkin leaves (0.1187 mg/kg)> potato leaves (0.1173 mg/kg)> rice (0.0976 mg/kg)> Chinese cabbage (0.0960 mg/kg).

Regarding the distribution of Hg in food crops throughout the study area, the mean Hg concentrations of food crops cultivated in agricultural soils around mining area contained higher Hg concentration than those at the control sites as shown in Table 8. The mean concentrations of Hg in food crops collected near mining sites were 0.0976 and 0.1391mg/kg in rice and vegetable respectively and those collected from control sites were 0.0756 and 0.0945 mg/kg in rice and leafy vegetables respectively as shown in Table 8.

The Hg contents based on dry weight were converted into a fresh weight basis, and compared with the maximum allowable concentration (MAC) set by Chinese Standard (MAC of Hg for leaf vegetables and grains are 0.01 and 0.02 mg/kg respectively on a fresh weight basis) as shown in Table 9. The fresh weight to dry weight ratio of 0.86 for rice and ratio of 0.085 for vegetables was used (Xiao et al., 2017, Chen et al., 2018). The measured total Hg contents in rice grains and cassava leaves exceeded the recommended values, while Hg contents in potato leaves, Chinese cabbage and pumpkin leaves were within the MAC.

This study found that the levels of Hg in food crops decreased with increasing distance from the mining area (Fig. 3). This could be possible due to mining operations within the study area. The mean values 0.0082-0.013 mg/kg fw (8.2-13 µg/kg fw) of Hg in leafy vegetables as shown in Table 9 are lower than mean values 3.64-86.69 µg/kg fw reported in China by Li et al. (2017). However the mean value for rice 0.0839 mg/kg fw (83.9µg/kg fw) are higher than the reported maximum mean value 62.95 µg/kg fw for rice.

From these findings apart from root absorption as a major route of PHEs entrance to food chain, it can be reported that the Hg levels found in plant leaves are also contributed by atmospheric deposition, which can be supported by previous observations by Edelstein and Ben-Hur (2018), Saha et al. (2017), Zwolak et al. (2019), Mwaanga et al. (2019) who explained the deposition of PHEs on the surfaces of vegetables and then absorbed into the plant tissues. Other factors for differentiated concentrations of Hg among food crops may be related to size and density of stomata, cuticle of plant leaves, proportion of leaves to the above ground parts and size, shape, and texture of leaves (Li et al., 2018, Fernández and Brown, 2013). Therefore, the higher concentration of Hg in cassava leaves (in dry weight basis) compared to other food crops may be attributed to large proportion of leaves aboveground compared to other food crops. Also, the lower Hg concentration in Chinese cabbage could be explained by the curled up leaf shape compared to other vegetables.

3.3. Health risk assessment

The results of estimated daily intake (EDI), estimated weekly intake (EWI) and target hazard quotient (THQ) are presented in Table 10. The EDI of mercury through consumption of rice and vegetables followed the order; rice (0.000429 mg/kg/day)> cassava (0.000034 mg/kg/day)> pumpkin (0.000027 mg/kg/day)> potato leaves (0.000026 mg/kg/day)> Chinese cabbage (0.000021 mg/kg/day). The dietary intake of rice is higher in comparison to vegetables, this contributed to a maximum amount of Hg in daily intake by residents of the study area. The EWI values were compared with provisional tolerable weekly intake (PTWI) value (0.0016 mg/kg bw/week) set by Joint FAO/WHO Expert Committee on Food Additives (FAO/WHO 2018). It was found that, the EWI of Hg through consumption of rice (0.003 mg/kg bw/week) was above PTWI set by FAO/WHO, indicating that consuming rice grains can be associated with human health risks. The EWI values of Hg for vegetables ranged from 0.00015 to 0.00024 mg/kg bw/week), which were below the guideline value hence from this findings, consuming vegetables may not present human health risks due to Hg. Table 10 shows target hazard quotient of mercury through consumption of food crops. The THQs decreased in the order of rice (1.415) > cassava leaves (0.1143) > pumpkin leaves (0.0885) > potato leaves (0.0875) > Chinese cabbage (0.0716). It was found that Hg intake via consumption of rice grains posed a risk of developing non-cancer health disorders as indicated by THQ > 1 (1.415). The THQs of Hg in all leafy vegetables in the study area were less than 1, suggesting that Hg may not pose significant non-cancer risks to local residents through consumption of vegetables.

Table 7

The post hoc analysis showing Hg mean difference among soils from different sites.

Sites	1	2	3	4	5	6	7	8	9	10	11	12
1	—	0.0608*	0.0577*	0.06322*	0.06012*	-0.205***	-0.0223	0.05266	0.05815*	0.05166*	0.0486	-0.00336
2		—	-0.003	0.0024	-6.97e−4	-0.266***	-0.0831***	-0.00816	-0.00267	-0.00916	-0.01222	-0.06418
3			—	0.0055	0.0024	-0.263***	-0.08***	-0.00506	4.23E-04	-0.00607	-0.00912	-0.06108
4				—	-0.0031	0.071***	-0.0855***	-0.01056	-0.00507	-0.01156	-0.01462	-0.06658
5					—	-0.265***	-0.0824***	-0.00746	-0.00198	-0.00847	-0.01152	-0.06348
6						—	0.1828***	0.25775***	0.26323***	0.25675***	0.25369***	0.20173***
7							—	0.07496***	0.08044***	0.07396***	0.0709*	0.01894
8								—	0.00548	-0.001	-0.00406	-0.05602
9									—	-0.00649	-0.00955	-0.06151
10										—	-0.00306	-0.05502
11											—	-0.05196
12												—

Mean difference values with asterisk (*) are statistically significant different at; ** $P < 0.01$,

* $P < 0.05$,

*** $P < 0.001$

Table 8
Comparison of average content of Hg (mg/kg) in rice and vegetables cultivated around the mine and control sites.

Sites	Crop	N	Mean	Minimum	Maximum
Near mining (0-2 km)	Rice	6	0.0976 ± 0.0343	0.0752	0.1587
	vegetables	18	0.1391 ± 0.0874	0.0556	0.3439
Control (>9 km)	Rice	2	0.0756 ± 4.95E-04	0.0752	0.0759
	Vegetables	4	0.0945 ± 0.0318	0.0760	0.1422

Table 9
Comparison of Hg concentration in food crops with maximum allowable concentration.

	Food crop				
	Rice	Potato leaves	Pumpkin leaves	Cassava leaves	Chinese cabbage
Mean Hg (mg/kg dw)	0.0976	0.1173	0.1187	0.1532	0.0960
Mean Hg (mg/kg fw)	0.0839	0.00997	0.01	0.013	0.0082
Chinese standard (mg/kg fw)	0.02	0.01	0.01	0.01	0.01

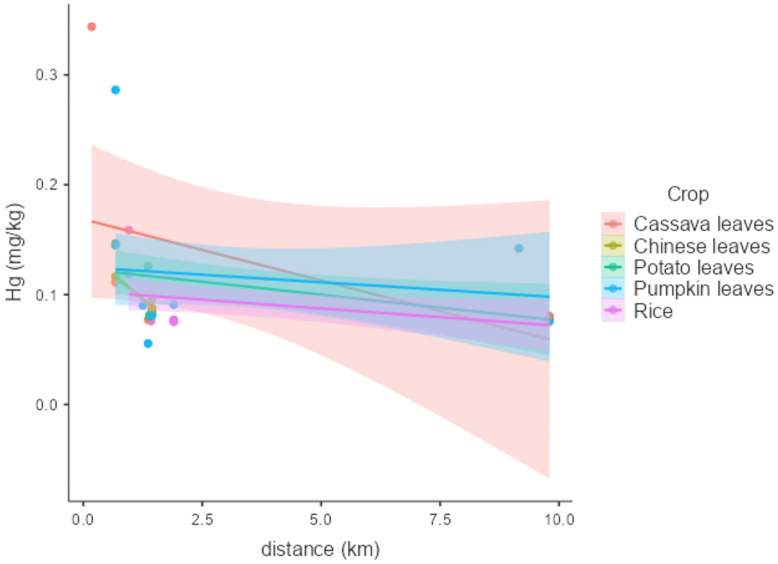

Fig. 3. The pattern of Hg concentration in rice and vegetables with distance from mining area.

Table 10
Estimated daily intake (EDI), estimated weekly intake (EWI) and target hazard quotient (THQ) of total Hg for adult population due to consumption of rice and vegetables.

Parameter	Food crop				
	Rice	Potato leaves	Pumpkin leaves	Cassava leaves	Chinese cabbage
Mean Hg (mg/kg)	0.0976	0.1173	0.1187	0.1532	0.0960
EDI (mg/kg/day)	42.9 E-05	2.6E-05	2.7 E-05	3.4E-05	2.1E-05
EWI (mg/kg bw/week)	30 E-04	1.8 E-04	1.9 E-04	2.4E-04	1.5 E-04
THQ	1.43	0.087	0.09	0.113	0.07

4. Conclusion and recommendations

The findings of this study revealed that all soil samples had total mercury contents below the safe limit recommended in agricultural soil. The estimated weekly intake (EWI) of Hg due to the consumption of all vegetables were found to be lower than the PTWI proposed by FAO/WHO. Whereas EWI of Hg due to consumption of rice grains were found to be higher than PTWI. The results from the human health assessment indicated that the THQ of Hg due to the consumption of vegetables were < 1 indicating that there were no potential health risks through consumption of vegetables alone. Whereas the THQ due to the consumption of rice grain were > 1 suggesting that local residents from the study area could be victims of non-carcinogenic risk due to the consumption of Hg in rice grain which is being consumed in higher proportion compared to leaf vegetables. These findings suggest that efforts are needed to promote site specific monitoring programs addressing the aspects of amalgamation ponds performance, roasting of amalgam practices, and tailing piles disposal in order to reduce environmental and food chain contamination as well as human health impacts. Human can also be exposed to Hg through inhalation; further studies need to be done focusing on the assessment of mercury levels in air to estimate human health risk impacts through direct mercury vapor exposure. Also, epidemiological studies of gold miners and residents near gold mining areas are required for determining mercury related diseases for control and prevention.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

Al, M., Massey, I.Y., 2019. Exposure routes and health effects of heavy metals on children. BioMetals doi:10.1007/s10534-019-00193-5.

Ali, H., Khan, E., 2018. Trophic transfer, bioaccumulation, and biomagnification of non-essential hazardous heavy metals and metalloids in food chains /webs — Concepts and implications for wildlife and human health. Hum. Ecol. Risk Assess. Int. J. doi:10.1080/10807039.2018.1469398.

Ali, H., Khan, E., Ilahi, I., 2019. Environmental chemistry and ecotoxicology of hazardous heavy metals: Environmental persistence, toxicity, and bioaccumulation. J. Chem. 67, 305–518. doi:10.1155/2019/6730305.

Amlinger, F., Pollack, M., Favoino, E. Heavy metals and organic compounds from wastes used as organic fertilisers. Final report for ENV. A. 2./ETU/2001/0024., 2004.

Bortey-Sam, N., Nakayama, S.M.M., Akoto, O., Ikenaka, Y., Fobil, J.N., Baidoo, E., Mizukawa, H., Ishizuka, M., 2015. Accumulation of heavy metals and metalloid in foodstuffs from agricultural soils around Tarkwa area in Ghana, and associated human health risks. Int. J. Environ. Res. Public Health 12, 8811–8827. doi:10.3390/ijerph120808811.

Chen, H., Yang, X., Wang, P., Wang, Z., Li, M., Zhao, F., 2018. Science of the total environment dietary cadmium intake from rice and vegetables and potential health risk: a case study in Xiangtan, southern China. Sci. Total Environ. 639, 271–277. doi:10.1016/j.scitotenv.2018.05.050.

Cohen, M., Artz, R., Draxler, R., Miller, P., Poissant, L., Niemi, D., Ratte, D., Deslauriers, M., Duval, R., Laurin, R., Slotnick, J., Nettesheim, T., Mcdonald, J., 2004. Modeling the atmospheric transport and deposition of mercury to the Great Lakes. Environ. Res. 95, 247–265. doi:10.1016/j.envres.2003.11.007.

Demková, L., Jezný, T., Bobulská, L., 2017. Assessment of soil heavy metal pollution in a former mining area-before and after the end of mining activities. Soil Water Res. 12, 229–236. doi:10.17221/107/2016-SWR.

Diringer, S.E., Feingold, B.J., Ortiz, E.J., Gallis, J.A., Araujo-Flores, J.M., Berky, A., Pan, W.K.Y., Hsu-kim, H., 2015. River transport of mercury from artisanal and small-scale gold mining and risks for dietary mercury exposure in Madre de Dios, Peru. Environ. Sci. Process. Impacts 17, 478–487. doi:10.1039/C4EM00567H.

Edelstein, M., Ben-Hur, M., 2018. Heavy metals and metalloids: Sources, risks and strategies to reduce their accumulation in horticultural crops. Sci. Hortic. 234, 431–444. doi: 10.1016/j.scienta.2017.12.039 (Amsterdam).

Engwa, A.G., Udoka Ferdinand, P., Nweke Nwalo, F., Unachukwu, M. N., 2019. Mechanism and health effects of heavy metal toxicity in humans, Poisoning Mod. World - New Tricks an Old Dog? doi:10.5772/intechopen.82511.

Esdaile, L.J., Chalker, J.M., 2018. The mercury problem in artisanal and small-scale gold mining. Chem. A Eur. J. 24, 6905–6916. doi:10.1002/chem.201704840.

Estefan, G., Sommer, R., Ryan, J., 2013. Methods of Soil, Plant, and Water Analysis: a Manual for the West Asia and North, 3rd ed. ICARDA, Beirut, Lebanon.

FAO, 2008. Nutritional Country Profile United Republic of Tanzania.

FAO/WHO, 2018. Joint FAO/WHO Food Standards Programme. CODEX Committee on Contaminants in Foods. FAO/WHO, Rome, Italy Cf/12 INF/1.

Fashola, M.O., Ngole-Jeme, V.M., Babalola, O.O., 2016. Heavy metal pollution from gold mines: environmental effects and bacterial strategies for resistance. Int. J. Environ. Res. Public Health 13, 1047–1066. doi:10.3390/ijerph13111047.

Fernández, V., Brown, P.H., 2013. From plant surface to plant metabolism: The uncertain fate of foliar-applied nutrients. Front. Plant Sci. 4, 1–6. doi:10.3389/fpls.2013.00289.

Gebeyehu, H.R., Bayissa, L.D., 2020. Levels of heavy metals in soil and vegetables and associated health risks in Mojo area, Ethiopia. PLoS One 15, 1–22. doi:10.1371/journal.pone.0227883.

Gunalan, S., Vijayalatha, K.R., Anitha, T., 2018. Heavy metals and its impact in vegetable crops. Int. J. Chem. Stud. 7, 1612–1621.

Gupta, N., Kumar, K., Kumar, V., Kumar, S., Chadd, R.P., Kumar, A., 2019. Trace elements in soil-vegetables interface: translocation, bioaccumulation, toxicity and amelioration - A review Science of the Total Environment. Sci. Total Environ. 651, 2927–2942. doi:10.1016/j.scitotenv.2018.10.047.

Hu, B., Jia, X., Hu, J., Xu, D., Xia, F., Li, Y., 2017. Assessment of heavy metal pollution and health risks in the soil-plant-human system in the Yangtze river delta, China. Int. J. Environ. Res. Public Health 14, 1042–1059. doi:10.3390/ijerph14091042.

Ikingura, J.R., Akagi, H., Mujumba, J., Messo, C., 2006. Environmental assessment of mercury dispersion, transformation and bioavailability in the Lake Victoria Goldfields, Tanzania. J. Environ. Manag. 81, 167–173. doi:10.1016/j.jenvman.2005.09.026.

Jan, A.T., Azam, M., Siddiqui, K., Ali, A., Choi, I., Haq, Q.M.R., 2015. Heavy metals and human health: mechanistic insight into toxicity and counter defense system of antioxidants. Int. J. Mol. Sci. 16, 29592–29630. doi:10.3390/ijms161226183.

Kacholi, D.S., Sahu, M., 2018. Levels and health risk assessment of heavy metals in soil, water, and vegetables of Dar es Salaam, Tanzania. J. Chem. 14, 674–682. doi:10.1155/2018/1402674.

Kinyondo, A., Huggins, C., 2021. In: WIDER Working Paper doi:10.35188/UNU-WIDER/2021/059-7.

Koleleni, Y.I., Mbike, S.A., 2018. Analysis of heavy metals in soil and maize grown around namungo gold mine in Ruangwa District, Lindi region in Tanzania Using X-ray Fluorescence. Chem. Sci. Int. J. 24, 1–18. doi:10.9734/csji/2018/44976.

Lema, M.W., Mseli, Z.H., 2017. Assessment of soil pollution (Heavy Metal) from small scale gold mining activities: a case of 4, 1–5.

Li, N., Kang, Y., Pan, W., Zeng, L., Zhang, Q., Luo, J., 2015. Concentration and transportation of heavy metals in vegetables and risk assessment of human exposure to bioaccessible heavy metals in soil near a waste-incinerator site, South China. Sci. Total Environ. 521–522, 144–151. doi:10.1016/j.scitotenv.2015.03.081.

Li, R., Wu, H., Ding, J., Fu, W., Gan, L., Li, Y., 2017. Mercury pollution in vegetables, grains and soils from areas surrounding coal-fired power plants. Sci. Rep. 7, 1–9. doi:10.1038/srep46545.

Li, X., Li, Z., Lin, C.J., Bi, X., Liu, J., Feng, X., Zhang, H., Chen, J., Wu, T., 2018. Health risks of heavy metal exposure through vegetable consumption near a large-scale Pb/Zn smelter in central China. Ecotoxicol. Environ. Saf. 161, 99–110. doi:10.1016/j.ecoenv.2018.05.080.

Malcolm, K., Dighton, J., Barkay, T., 2018. Mercury affects the phylloplane fungal community of blueberry leaves to a lesser extent than plant age. Mycology 9, 49–58. doi:10.1080/21501203.2017.1397063.

Malehase, T., Okonkwo, O.J., Daso, A.P., 2017. Initiatives to combat mercury use in artisanal small scale gold mining: a review initiatives to combat mercury use in artisanal small scale gold mining: a review on issues and challenges. Environ. Rev. 25, 218–224. doi:10.1139/er-2016-0042.

Manzoor, J., Sharma, M., Wani, K.A., 2018. Heavy metals in vegetables and their impact on the nutrient quality of vegetables: a review. J. Plant Nutr. doi:10.1080/01904167.2018.1462382.

Morgan, R. Soil, heavy metals and human health, in: E. Brevic, L. Burgess (Eds.), Soils Hum. Heal., CRC Press, New York, 2013: pp. 59–82.

Mutagwaba, W., Tindyebwa, J.B., Makanta, V., Kaballega, D., Maeda, G. Artisanal and small-scale mining in Tanzania – Evidence to inform an ‘ action dialogue,’ IIED, London, 2018.

Mwaanga, P., Silondwa, M., Kasali, G., Banda, P.M., 2019. Preliminary review of mine air pollution in Zambia. Heliyon 5, e02485. doi:10.1016/j.heliyon.2019.e02485.

National Bureau of Statistics, 2020. 2020 Tanzania in Figures, NBS. United Republic of Tanzania.

Nyanza, E.C., Dewey, D., Manyama, M., Martin, J.W., 2020. Maternal exposure to arsenic and mercury and associated risk of adverse birth outcomes in small-scale gold mining communities in Northern Tanzania. Environ. Int. 137, 105450. doi:10.1016/j.envint.2019.105450.

Nyanza, E.C., Bernier, F.P., Martin, J.W., Manyama, M., 2021. Effects of prenatal exposure and co-exposure to metallic or metalloid elements on early infant neurodevelopmental outcomes in areas with small- scale gold mining activities in Northern Tanzania. Environ. Int., 106104 doi:10.1016/j.envint.2020.106104.

Obiora, S.C., Chukwu, A., Chibuike, G., Nwegbu, A.N., 2019. Potentially harmful elements and their health implications in cultivable soils and food crops around lead-zinc mines in Ishiagu, Southeastern Nigeria. J. Geochem. Explor. 204, 289–296. doi:10.1016/j.gexplo.2019.06.011.

Pan, J., Chon, H.S., Cave, M., Oates, C., Plant, J., 2012. Toxic trace elements. In: Plant, J., Voulvoulis, N., Ragnarsdottir, V. (Eds.), Pollut. Hum. Heal. Environ. Wiley-Blackwell, UK, pp. 87–114.

Pu, W., Sun, J., Zhang, F., Wen, X., Liu, W., Huang, C., 2019. Effects of copper mining on heavy metal contamination in a rice agrosystem in the Xiaojiang River Basin, southwest China. Acta Geochim. 38, 753–773. doi:10.1007/s11631-019-00321-5.

Rehman, K., Bukhari, S.M., Andleeb, S., Mahmood, A., Erinle, K.O., Naeem, M.M., Imran, Q., 2019. Ecological risk assessment of heavy metals in vegetables irrigated with groundwater and wastewater: the particular case of Sahiwal district in Pakistan. Agric. Water Manag. 226, 105816. doi:10.1016/j.agwat.2019.105816.

Rodríguez-Eugenio, N., McLaughlin, M., Pennock, D., 2018. Soil Pollution: a Hidden Reality. FAO, Rome, Italy.

Saha, J.K., Selladurai, R., Kundu, S., Patra, A.K., 2017. Soil Pollution - an Emerging Threat to Agriculture. Springer, Gateway East, Singapore.

Song, S., Li, Y., Li, L., Liu, M., Li, J., Wang, L., Su, C., 2018. Arsenic and heavy metal accumulation and risk assessment in soils around mining areas: the Urad Houqi area in arid Northwest China as an example. Int. J. Environ. Res. Public Health 15, 2410–2426. doi:10.3390/ijerph15112410.

Tanzania Bureau of Standards, 2007. Tanzania Standards, Soil quality - limits for soil contaminants in habitat and agriculture 3, 1–11.

Telmer, K.H., Veiga, M.M., 2009. Word emissions of mercury from artisanal and small scale gold mining. In: Mercury Fate and Transport in the Global Atmosphere: Emissions, Measurements and Models. Springer, New York, pp. 131–172.

Tiwari, S., Lata, C., 2018. Heavy metal stress, signaling, and tolerance due to plant-associated microbes: an overview. Front. Plant Sci. 9, 1–12. doi:10.3389/fpls.2018.00452.

Tun, A.Z., Wongsasuluk, P., Siriwong, W., 2020. Heavy metals in the soils of placer small-scale gold mining sites in Myanmar. J. Health Pollut. 10, 1–12.

Tungaraza, C., Chibunda, R.T., Pereka, A.E., 2011. Dietary exposure to mercury of the adult population in mugusu artisanal gold mining village, Tanzania: a total diet approach. Open Environ. Eng. J. 4, 141–146. doi:10.2174/1874829501104010141.

U.S. EPA, 2001. Appendix to Method 1631 Total Mercury in Tissue, Sludge, Sediment, and Soil by Acid Digestion and BrCl Oxidation. U.S. EPA EPA-821-R-01-013.

U.S. EPA, 2004. Region 9 PRG Table. U.S. EPA.

USEPA, 2019. Guideline for human exposure assessment. Risk Assess. Forum. 1–199.

Veiga, M.M., Metcalf, S.M., Baker, R.F., Klein, B., Davis, G., Bamber, A., Siegel, S., Singo, P., 2006. Manual for Training Artisanal and Small-Scale Gold Miners. UNIDO, Vienna, Austria.

Xiao, R., Wang, S., Li, R., Wang, J.J., Zhang, Z., 2017. Soil heavy metal contamination and health risks associated with artisanal gold mining in Tongguan, Shaanxi, China. Ecotoxicol. Environ. Saf. 141, 17–24. doi:10.1016/j.ecoenv.2017.03.002.

Zwolak, A., Sarzyńska, M., Szpyrka, E., Stawarczyk, K., 2019. Sources of soil pollution by heavy metals and their accumulation in vegetables: a review. Water. Air. Soil Pollut. 230, 1–9. doi:10.1007/s11270-019-4221-y.