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Research article

Levels of heavy metals in ginger (*Zingiber officinale* Roscoe) from selected districts of Central Gondar Zone, Ethiopia and associated health risk

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ABSTRACT

In this study, we investigated the concentrations of Cd, Cr, Cu, Fe, Ni, Pb and Zn; and their associated health risks through consumption of ginger. After the ginger samples digested with a mixture of HNO_3 and $HClO_4$ at 200 °C for 2:00 h, the amount of metals were investigated by flame atomic absorption spectrometry (FAAS). Efficiency of the analytical measurement was validated on spiking the sample with standard solutions of metals and the recovery for all studied metals were ranged from 91.60% to 99.94%, which is in the acceptable range of validation. The mean concentrations (mg/kg) were ranged from 4.63 to 5.43 for Cd, 2.17 to 4.44 for Cr, 62.52 to 65.14 for Cu, 77.71 to 81.12 for Fe, 6.49–7.58 for Ni and 16.74–19.31 for Zn. However, the concentration of Pb was not detected. The estimated daily intake (EDI) values of all metals from all samples are substantially lower than their corresponding maximum tolerable daily intake (MTDI). Target hazardous quotient (THQ) values of all metals are lower than 1 in all the sampling sites, revealed that there are no health risks for the users due to the intake of these metals. The health index (HI) values were slightly higher than unity, which implying that there is significant health effects to the population from consuming ginger at the study.

1. Introduction

Ginger (*Zingiber officinale*) is an important tropical herbaceous perennial plant which has been used to treat many diseases, including gastrointestinal ulcers, cancer, arthritis, vomiting peptic and duodenal; and improves blood circulation, lowers blood glucose and cholesterol levels (Nishidono et al., 2018; Idris et al., 2019). Ginger is used as a spice around the world in both fresh and dried forms (Wagesho and Chandravanshi, 2015). Chemical analysis indicates that ginger contains many active ingredients including terpenes, oleoresin, gingerols, shogaols paradols, zingerone, vitamins, and minerals (Teng et al., 2019). Ethiopia is the homeland for many spices, including chilies, black cumin, ginger, fenugreek and coriander. Ginger is the second most widely cultivated spice next to chilies in Ethiopia (Soni et al., 2019). It is known to have been cultivated in Ethiopia since 13^{th} century (Guji et al., 2019). In Ethiopia, ginger is the common spice added in many food stuffs (Hordofa and Tolossa, 2020).

In recent years, the intake of metals from food and water has become the potential health risk for the users (Doabi et al., 2018). Though metals are essential to humans and other living beings, they become noxious when their concentrations higher than the thresholds (Jiang et al., 2018; Xiao et al., 2019). Trace metals such as Cu, Zn, Fe, Mn and Cr are important for enzyme structuring; and synthesis hemoglobin and vitamin; whereas metals such as Cd and Pb are toxic even at low concentration (Mohammed, 2016).

Heavy metals can be released into the environment through both natural and anthropogenic sources and transferred in to human body through soil, dermal contact, breathing and food chain (Adimula et al., 2019). Prolonged use of unsafe heavy metals through food consumption can lead to chronic accumulation in the human kidneys and liver and cause toxicity (Ametepey et al., 2018).

Limited studies are vailiable on the levels and health risks of metals in ginger growing in Ethiopia (Wagesho and Chandravanshi, 2015; Goroya et al., 2019). However, there has no report so far about the levels and risk of heavy metals for humans in ginger residing in Central Gondar Zone, Ethiopia. Therefore, the objective of this study was to assess of heavy metal contamination and to investigate health risks of heavy metals in ginger from Central Gondar Zone.

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2. Materials and methods

2.1. Description of study area

The study areas were Dembia and Gondar Zuria districts which are located in Amhara Regional State. Dembia district is bordered by Lake Tana (the Lake over ride by Nile river and UNESCO registered Lake, in 2015 GC) in south, Takusa in the southwest, Chilga in the west, Lay Armachiho in the north, and Gondar Zuria district in the east, Amhara Regional State, Ethiopia. Dembia district is located at 12°18′30″N latitude and 37°17′30″E longitude with an elevation of ranging from 1500 to 2600 m above sea-level (Figure 1). While Gondar Zuria district is bordered by the South Gondar Zone, Lake Tana, Dembia, Lay Armachiho, and Mirab Belessa, Amhara Regional State, Ethiopia. The district is situated at 12°15′30″N latitude and 7°37′45″E longitude at an elevation of 1107–3022 m above sea level. Since the districts are bordered by Lake Tana, the communities around it are economically depend on potential yields of agricultural cultivation including spices & vegetables and hunting edible aquatic animals. Thus, Dembia and Gondar Zuria districts were selected as study sites on account of potential cultivation sites of spices like ginger in addition to other staple foods items.

2.2. Sampling and sample preparation

Totally eighteen ginger samples were purchased from six markets at East Dembia district (Ayimba and Koladiba towns), West Dembia district (Chuahit and Gorgora towns), and Gondar Zuria district (Maksegnit and Infranz towns). These are the main ginger producing areas and the most contributors to the markets of western part of Amhara Region, Ethiopia.

A 0.5 kg of the ginger samples were purchased randomly from farmers and local stores with a total of 4.5 kg. Then after, mixed, homogenized, packed in polyethylene plastic bags, labeled and transported to chemistry laboratory, University of Gondar, Gondar, Ethiopia.

The samples were washed thoroughly with water to get free of dusts, distilled and deionized water in order to remove dust and extraneous matters. The ginger samples were chopped nearly equal sized in order to facilitate drying and then dried for 9 h in an oven at a temperature of 200 °C, relatively until they become free of moisture.

The dried samples were grounded with clean acid washed plastic mortar and stored until sample preparation, digestion and analysis.

The ginger was digested with 4 mL of HNO_3 (69–72%) and 3 mL of $HClO_4$ (70%) at 200 °C for 2 h. Among the digestion procedures, the one which consumed minimum reagent volume at minimum digestion time and temperature for complete digestion of ginger was considered as optimum measuring condition. The digested solution was cooled for 10 min and filtered with Whatman No. 42 filter paper in to 50 mL volumetric flask and diluted with deionized water up to the mark. The reagent blank was also digested by following similar procedure as that of the ginger. Finally, all digested solutions were kept in refrigerator until analyzed with flame atomic absorption spectrometry (Ayalew et al., 2017).

Standard solutions of metals (1000 mg/L) were used to prepare working standards to establish calibration curve (0.5, 2.5, 5, 7.5, 8.5 and 10 mg/L) in flame atomic absorption spectrometer. After optimized the instrument operating parameters for best signal intensity, the levels of the metals were determined with FAAS in all the samples (Table 1). The concentration of elements in the blank was also determined by applying the same analytical procedure as with the sample. As can be seen in Table 2, correlation coefficients (R^2) of the calibration curves were varied between 0.9950 and 0.9994.

2.3. Method validation

Method validation was expressed through precision, recovery test, limit of detection and limit of quantification. Precision was determined from the standard deviation (SD) of the six replicate results and the concentration of metals are expressed as mean with their corresponding standard deviations (Wagesho and Chandravanshi, 2015).

The limit of detection (LOD) is defined as the smallest amount of analyte which can be detected and it was calculated by dividing the value of three times the standard deviation of the blank sample with the slope of the calibration curve. Limit of quantification (LOQ) is the smallest aamount that can be quantitatively detected at a stated accuracy and precision (Taghipour and Jalali, 2019). The LODs of the investigated metals Cd, Cr, Cu, Fe, Ni, Pb and Zn in ginger samples were found to be 0.14, 0.72, 0.38, 0.46, 0.65, 0.39 and 0.21 mg/kg, respectively; while LOQs were 0.45, 2.41, 1.29, 1.53, 2.15, 1.29, 0.69 for Cd, Cr, Cu, Fe, Ni, Pb and Zn, respectively (Table 3).

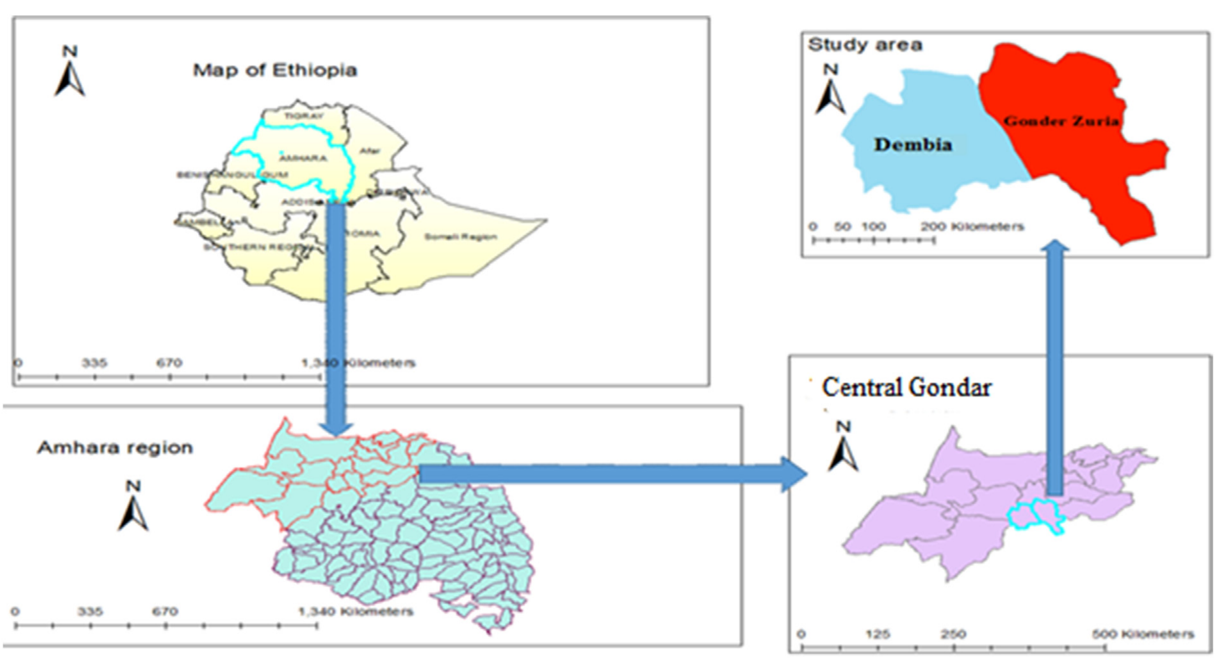

Figure 1. Map of the study area.

Table 1. Operating parameters for FAAS.

Metals	Wavelength (nm)	Energy(J)	Slit width (nm)	Lamp current (mA)	PMT(V)	Flame type
Fe	248.3	3.042	0.2	7.0	323.5	Air-acetylene
Cu	324.7	3.768	0.7	1.5	272.0	Air-acetylene
Zn	213.9	3.003	0.7	2.0	272.9	Air-acetylene
Ni	232.0	3.146	0.2	7.0	323.9	Air-acetylene
Cr	357.9	3.771	0.7	2.0	242.7	Air-acetylene
Cd	228.9	3.078	0.7	2.0	259.1	Air-acetylene
Pb	283.2	3.686	0.7	2.0	290.7	Air-acetylene

PMT - Photomultiplier Tube.

Table 2. Working standard concentrations, regression equation and correlation coefficient for determination of metals using FAAS.

Metal	Concentration (mg/L)	Regression Equation	R^2	LOD	LOQ
Cd	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0635C + 0.026	0.9953	0.14	0.45
Cr	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0012C + 0.0003	0.9990	0.72	2.41
Cu	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0223C + 0.0073	0.9950	0.38	1.29
Fe	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0188C + 0.0001	0.9966	0.46	1.53
Ni	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0134C + 0.0019	0.9958	0.65	2.15
Pb	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0057C + 0.0004	0.9994	0.39	1.29
Zn	0.5, 2.5, 5.0, 7.5, 8.5, 10	A = 0.0719C + 0.0366	0.9951	0.21	0.69

Table 3. Recovery results for Validation of the Optimized Procedure (mean ±SD, mg/kg, n = 3) for ginger sample.

Heavy Metals	Amount before spiked (mg/kg)	Amount Added (mg/kg)	Amount after Spiked (mg/kg)	Recovery (%)
Cd	4.63±0.16	5.0	9.46±1.21	96.6±2.48
Cr	2.17±0.24	5.0	6.92±0.14	95.03±1.40
Cu	65.14±9.89	20	83.46±1.34	91.60±6.79
Fe	78.64±9.79	30	107.04±1.43	94.66±4.77
Ni	7.09±0.84	5.0	11.70±0.98	92.31±2.45
Zn	16.74±1.35	10	26.73±2.43	99.94±1.32

The recovery (accuracy) of the method and efficiency of instrument used for sample analysis were evaluated by spiking the ginger with known concentration of standard solutions. It was performed by spiked 0.5 g ginger sample with varying amounts of the standard solutions of metals. All the spiked samples were digested using the same procedure as described earlier in the unspiked samples (Bedassa et al., 2017). The percent recovery values of Cd, Cr, Cu, Fe, Ni and Zn were 96.6%, 95.03%, 91.60%, 94.66%, 92.31% and 99.94 %, respectively (Table 3).

2.4. Risk assessment

Long-term exposure to toxic metals in the environment may pose potential health risk to human (Boateng et al., 2015; Tay et al., 2019). Risk assessments of heavy metals in plants are used to quantify both carcinogenic and non-carcinogenic risks to humans (Chonokhuu et al., 2019).

2.4.1. Estimated daily intake (EDI)

EDI was calculated based on their mean concentration of each metals in ginger that estimated the average daily consumption of the ginger. It was calculated according to Eq. (1):

$$\mathrm{EDI} = \frac{\mathrm{C_{metal}} \times IR}{BW} \tag{1}$$

where, Cmetal (mg/kg) is the metal concentration in ginger, IR (ingestion rate) is the average daily consumption of ginger in the local area (10 g/day), BW is average body weight (Kg). The average adult body weight used in this study was 65 kg.

2.4.2. Target hazardous quotient (THQ)

THQ is used to calculate the non-carcinogenic risk due to exposure of metals through vegetable consumption. As shown in Eq. (2), THQ is expressed as the ratio of EDI to the chronic reference dose (RfD) of a specific heavy metal.

$$THQ = \frac{EDI}{RfD} \tag{2}$$

where, RfD is reference dose. The reference doses for Cd, Cr, Cu, Fe, Ni and Zn were 0.001, 0.001, 0.04, 0.7, 0.02, 0.3, respectively (USEPA 2011).

If THQ ≥1, there may be adverse health effects for human due to metals exposure. If THQ <1, then it is safe for health risk of non-carcinogenic elements (Kortei et al., 2020).

Hazard index (HI) is used to evaluate the potential human health risk through consumption of more than one heavy metal. HI was calculated as the sum of the THQ as described in Eq. (3).

$$HI = \sum_{n=1}^{i} THQ;\ \mathrm{i} = 1,\ 2,\ 3, \ldots\ldots \mathrm{n} \tag{3}$$

If the value of HI ≥ 1, there is a possibility of health effects. However, if HI < 1, there is insignificant risk of non-carcinogenic effects (Ametepey et al., 2018; Chonokhuu et al., 2019).

Table 4. Distribution of each metal concentration (Mean ± SD; mg/kg, n = 3) ND-not detected.

Sampling sites	Metals						
	Cd	Cr	Cu	Fe	Ni	Pb	Zn
East Dembia	4.63±0.16[a]	3.17±0.24[a]	65.14±9.89[a]	78.64±9.79[a]	7.09±0.84[a]	ND	16.74±1.35[a]
West Dembia	5.08±0.15[b]	4.44±0.43[a]	62.52±3.77[b]	77.71±7.75[a]	6.49±0.48[a]	ND	19.31±1.35[b]
Gondar Zuria	5.43±0.14[c]	3.69±0.33[b]	63.71±5.28[c]	81.12±9.58[a]	7.58±0.26[a]	ND	18.43±2.04[c]

Different letters in the same column are significantly different ($p < 0.05$) in one way ANOVA.
ND- Not detected.

Table 5. The comparison of concentration metals of ginger sample (mg/kg) in this study and with the reported values.

Country	Metal							
	Cd	Cr	Cu	Fe	Ni	Pb	Zn	Reference
Ethiopia	0.38–0.97	6.02–10.8	1.10–4.78	41.8–89.0	5.46–8.40	ND	38.5–55.2	(Wagesho and Chandravanshi, 2015).
Ethiopia	ND	NG	NG	NG	0.15–0.21	ND	0.63–1.17	(Goroya et al., 2019)
India	0.92–2.27	NR	3.06–14.56	17.46–28.66	NR	0.5–12.0	4.57–16.84	(Agrawal et al., 2011)
Iraq	1.32	16.0 ± 0.1	15.2	140	NG	7.2	29.0	(Ibrahim et al., 2012)
Nigeria	7.45	5.65	13.5	16.67	3.417	2.70	10.133	(Gaya and Ikechukwu 2016)
Poland	0.02–0.04	NG	2.35–8.32	NG	NG	0.21–0.78	5.96–16.95	(Krejpcio et al., 2007)
Ethiopia	4.63–5.43	2.17–4.44	62.52–65.14	77.71–81.12	6.49–7.58	ND	16.74–19.31	This study

NG-not given, ND-not detected.

Table 6. EDI, HQ and HI values of heavy metals in adults through the consumption of different spices.

Sampling Site	Cd		Cr		Cu		Fe		Ni		Zn		HI
	EDI	THQ	EDI	THQ	EDI	THQ	EDI	THQ	EDI	THQ	EDI	THQ	
East Dembia	0.00071	0.7123	0.0003	0.3338	0.0100	0.2505	0.0121	0.0173	0.0011	0.0545	0.0026	0.0086	1.377
West Dembia	0.00078	0.7815	0.0007	0.6831	0.0096	0.2405	0.0120	0.0171	0.0010	0.0499	0.0030	0.0099	1.782
Gondar Zuria	0.00084	0.8354	0.0006	0.5677	0.0098	0.245	0.0125	0.0178	0.0012	0.0583	0.0028	0.0094	1.733

Table 7. Pearson's correlation matrices for heavy metals in rhizomes samples.

HMs	Fe	Cu	Zn	Ni	Cr	Cd
Fe	1					
Cu	-0.07	1				
Zn	-0.08	0.99	1			
Ni	0.95	-0.37	-0.39	1		
Cr	0.21	-0.99	-0.99	0.50	1	
Cd	0.65	0.70	0.69	0.38	-0.60	1

2.5. Statistical analysis

The data were examined using IBM SPSS Statistics version 20.0. The data were reported as mean ± standard deviation and the differences between means of metals concentration in ginger were compared at the 95% confidence level.

3. Results and discussion

3.1. Heavy metals concentration in ginger

The concentration of Cd, Cr, Cu, Fe, Ni, Pb and Zn in ginger samples were determined using FAAS. The mean concentrations of elements in ginger samples are demonstrated in Table 4. The trend of trace metals concentration in ginger samples at all sample sites was: Fe > Cu > Zn > Ni > Cd > Cr.

The Cd concentration were found in the range of 4.63± 0.16 mg/kg to 5.43±0.14 mg/kg in ginger samples collected from East Dembia and Gondar Zuria, respectively. Cd contents in all study areas showed significant difference ($P < 0.05$). The order of Cd concentration in the ginger harvested from all the studied locations is: Gondar Zuria > West Dembia > East Dembia. When compared with the results reported in literature, the levels of Cd was slightly higher than results reported from India (0.92–2.27) (Agrawal et al., 2011), Poland (0.02–0.04) (Krejpcio et al., 2007), Iraq (1.32) (Ibrahim et al., 2012), Ethiopia (0.38–0.97) (Wagesho and Chandravanshi, 2015). While, Gaya and Ikechukwu reported higher levels of Cd from Nigeria (Gaya and Ikechukwu 2016) than the values obtained in this study (Table 5).

The concentrations of Cr in ginger samples were found in the range of 2.17±0.24–4.44±0.43 mg/kg. Highest value (4.44±0.43) was detected in West Dembia, while the least (0.892 ± 0.10 mg/kg) was observed in ginger samples collected from East Dembia. The concentrations of Cr in ginger from East and West Dembia showed insignificant difference ($P > 0.05$), while concentrations of Cr in ginger at Gonder Zuria showed significant difference ($P < 0.05$) with the two sampling sites (East and West Dembia). The concentration of Cr in this study are comparable with previous study conducted data from Ethiopia (Wagesho and Chandravanshi, 2015) and Nigeria (Gaya and Ikechukwu 2016). However, it was found that the result of this study was slightly lower than values reported by Ibrahim et al. (2012).

The levels of Cu ranged from 62.52±3.77 mg/kg in West Dembia to 65.14±9.89 mg/kg in East Dembia. The concentrations of Cu in ginger differ significantly ($P < 0.05$) at all study sites. Compared with reports, the concentrations of Cu were higher than reported from India (Agrawal et al., 2011), Poland (Krejpcio et al., 2007), Ethiopia (Wagesho and Chandravanshi, 2015), Iraq (Ibrahim et al., 2012), and Nigeria (Gaya and Ikechukwu 2016).

The highest amount of Fe was determined in samples from Gondar Zuria (81.12±9.58 dry weight) followed by East Dembia (78.64± 9.79 mg/kg) and the least in samples from West Dembia (77.71± 7.75 mg/kg). There is insignificant difference in the content of Fe in ginger at all sampling sites ($p > 0.05$). The concentrations of Fe in this study are comparable with those obtained by Wagesho and Chandravanshi (2015) in a previous study from Ethiopian. Fe concentrations in ginger samples were higher than the levels of Fe reported from India (Agrawal et al., 2011) and Nigeria (Gaya and Ikechukwu 2016) with values of 17.46–28.66 and 16.67 mg/kg, respectively. However, it was found to be lower than the value recorded from Iraq (Ibrahim et al., 2012).

The concentrations of Ni were ranged from 6.49 to 7.58 mg/kg. The highest and least levels of Ni were detected in samples collected from Gondar Zuria and West Dembia, respectively. There are insignificant differences ($P > 0.05$) in the concentration of Ni at the three sampling sites. The concentration of Ni in ginger reported by Wagesho and Chandravanshi (2015) in Ethiopia are comparable amount with the present study. However, Ni investigated in our sample of interest is higher than the reported with literature from Nigeria (Gaya and Ikechukwu 2016).

The ginger samples had Zn concentration ranged from 16.74 mg/kg to 19.31 mg/kg. The highest concentration of Zn was observed in the West Dembia, while the least concentration of Zn was observed in the East Dembia. There is insignificant difference in the contents of Zn among samples from East Dembia, West Dembia and Gondar Zuria sites. Results obtained for Zn were almost comparable with reported amounts by Agrawal et al. (2011) and Krejpcio et al. (2007). However, it was found to be much greater than reported by Gaya and Ikechukwu (2016) and lower than reported previously by Wagesho and Chandravanshi (2015). Lead was not detected in all analyzed samples.

In general, the significant differences in the concentration of the metals in ginger samples could be attributed to differences in soil composition, applied fertilizers and pesticides at the study sites (Hagos and Chandravanshi, 2016).

3.2. Risk assessment

As shown in Table 6, the EDI of Cd, Fe and Ni were found to be highest at Gondar Zuria, Cr and Zn at west Dembia; and Cu at East Dembia. The results indicated that, the order of EDI of all the investigated metals were Fe > Cu > Zn > Ni > Cd > Cr > Cu. EDIs values of these metals were lower than their corresponding maximum tolerable daily intake of Cd, Cr, Cu, Fe, Ni and Zn, with values of 0.07, 0.2, 3.0, 0.8, 0.3, and 1, respectively (Woldetsadik et al., 2017; Alipour and Banagar 2018; Deng et al., 2019), which indicates that consuming ginger has negligible health risk (Salama et al., 2019).

For heavy metals in ginger samples, the THQ values were ranged from 0.7123 to 0.8354, 0.3338 to 0.6831, 0.2405 to 0.2505, 0.0171 to 0.0178, 0.0499 to 0.0583 and 0.0086 to 0.0099 for Cd, Cr, Cu, Fe, Ni and Zn, respectively. The orders of THQ for metals were Cd > Cr > Cu > Ni > Fe > Zn to adults at all study sites. Besides, the THQs for all heavy metals were <1 in all sampling sites, which indicated that there are no potential health effects from exposure to heavy metals (Kortei et al., 2020).

The HI values resulted from summed of all HQs were ranged from 1.377 to 1.782, which were slightly above unity, indicating that there is an adverse non-carcinogenic health effect (Adefa and Tefera 2020).

Based on Pearson's correlation coefficients a strong correlation was exhibited among Zn with Cu, Ni with Fe, Cd with Fe, Cu, Zn and Ni, Cr with Fe and Ni (Table 7). However, negative correlation was observed between Cd and Cr, Cu with Fe, Ni and Cr; Zn with Fe, Ni and Cr. A better positive correlation indicates that these elements possibly originated from the same sources, while, high negative correlations between metals may indicate that the absorption of one metal by ginger tends to decrease absorption of other metals (Zhang et al., 2018).

4. Conclusion

The results showed that ginger contained highest amount of Fe followed by Cu, Zn, Ni, Cd and Cr at all study sites. Health risk analysis suggests that ingestion of ginger provided lower values of HQ than the safe level for all metals, indicating there is potential health risks from these metals. The values of HI of all heavy metals at all sites were found to be slightly higher than 1, indicating that it may pose future health risk. Therefore, incessant monitoring of heavy metals in ginger is needed to ensure the safety of humans by reducing environmental risk through a proper treatment of the agricultural and possible sources of metals.

Declarations

Author contribution statement

Aschalew Getaneh: Performed the experiments; Analyzed and interpreted the data.

Atnafu Guadie: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Molla Tefera: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Data availability statement

Data included in article/supp. material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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