

ORIGINAL ARTICLE**Analysis of heavy metals in cow milk from the vicinity of Hawassa industrial zone reveals potential risks in human health****Samuel Bekele^{1*}, Solomon Sorsa², Gunnhild Riise³, Daniel Fitamo², Zinabu Gebremariam¹**¹Department of Environmental Science, Hawassa University, P.O.Box 5, Ethiopia.²Department of Biology, Hawassa University, P.O.Box 5, Hawassa, Ethiopia³Norwegian University of Life Sciences, Faculty of Environmental Sciences and Natural Resource Management, Ås, Norway.*Corresponding author: beksam10@gmail.com**ABSTRACT**

The incidence of milk contamination linked with environmental pollution has increased over the years. But little is known about heavy metal contamination of milk in developing countries like Ethiopia where research in environmental pollution is still in its infancy. This study aimed to determine concentrations of ten heavy metals (Cr, Ni, Cu, Zn, As, Cd, Pb, Fe, Co and Mn) in domestically produced cow milk from Hawassa Industrial Zone (HIZ), Ethiopia and assess the associated human health risks from milk consumption. Estimated Daily Intake (EDI), Target Hazard Quotients (THQ), Hazard Index (HI) and Target Cancer Risk (TCR) were used as indices to evaluate potential human health risks from milk consumption. Average concentrations of metals in milk from Industrial (IZ) and Non-industrial Zone (NIZ) decreased in the order Zn>Fe>Mn>Cu>Cr>Ni>Pb>As>Co>Cd and Zn>Fe>Mn>Cu>Cr>As>Ni>Pb>Co>Cd, respectively. Mean concentrations of Zn ($5093.33 \pm 223.49 \mu\text{g kg}^{-1}$), Fe ($592.0 \pm 63.19 \mu\text{g kg}^{-1}$) and Cu ($77.78 \pm 13.94 \mu\text{g kg}^{-1}$) in milk from IZ as well as Zn ($3953.33 \pm 63.15 \mu\text{g kg}^{-1}$) and Cu ($63.0 \pm 9.78 \mu\text{g kg}^{-1}$) in milk from NIZ were above the maximum tolerance limits proposed by International Dairy Federations. From the results of human health risk assessments, it was concluded that effects of all heavy metals put together may affect human health as indicated by the elevated HI. Effluents from industries in HIZ are assumed to be the main sources of the heavy metals. Therefore, awareness creation of farmers and policy interventions with respect to waste disposal are recommended to protect the health of the ecosystem and the public.

Keywords: cow milk, heavy metals, health risks,

INTRODUCTION

Heavy metal pollution of the environment is a major concern on a global scale. The risk associated with exposure to heavy metals in cow's milk has been reported to be a serious threat to human health (Muhib *et al.*, 2016; Akele *et al.*, 2017). Livestock grazing on contaminated grasses and watering from polluted water sources can lead to accumulation of heavy metals in their milk as a result of bioaccumulation (Licata *et al.*, 2012). Although animals remove excess heavy metals through urine, feces, hair, nails, milk etc. (Datta *et al.*, 2010) many of the metals accumulate in different parts of the animal body. Milk is regularly consumed by human beings as it is considered to be nearly a complete food item and hence very important in human nutrition (Hanna *et al.*, 2019). Unfortunately, heavy metals can readily transfer from grass and/or water, which the cattle consume, to milk and then pass on to humans through consumption of milk causing adverse effects on human health (Muluken 2014; Tassew *et al.*, 2017).

Most of the industries in Ethiopia discharge inadequately treated effluents into nearby land and water bodies, particularly in Lake Hawassa, Southern Ethiopia (Berehanu *et al.*, 2015). Since most cattle in Ethiopia are free grazing in open grazing lands, they feed on grasses that grow on contaminated land and drink out of water contaminated with wastewater discharged from industries. It is also perceived that consumption of milk and milk products has been increasing in Ethiopia over the last few years probably as a result of growing awareness among the population on the importance of milk in human nutrition (Eline *et al.*, 2019; Mebrate *et al.*, 2019). On the other hand the magnitude of long-term exposure of cow milk consumers to heavy metals gets little attention. Only few studies (Tassew *et al.*, 2014; Akele *et al.*, 2017) have been reported in this regard in developing countries like Ethiopia. It is, therefore, possible that the consumer population of the country would face significant health threat from consuming milk contaminated with heavy metals. Thus the estimated daily intake rate (EDI) of heavy metals, hazard quotients (HQ) and carcinogenic risk (CR) might be exponentially increasing along the increasing rate of milk consumption. It is, therefore, imperative that a study that provides information on the aforementioned issues is of significant importance in terms of public health hazard of heavy metals that may arise from milk consumption in Ethiopia. Although there are some studies conducted on heavy metal contamination in cow milk and related human health risks in Ethiopia (Muluken 2014; Tassew *et al.*, 2014; Akele *et al.*, 2017; Haftom and Ashagrie, 2020), there are no reports on studies made in areas close to

where there are industrial activities. Therefore, the present study was designed to investigate concentrations of selected heavy metals in cow milk as well as to assess human health risks that can possibly be related to cow milk consumption from Hawassa Industrial Zone consisting of Textile factory, Moha Soft Drink and BGI Brewery factories, Southern Ethiopia.

MATERIALS AND METHODS

Description of the Study Area

Hawassa Industrial Zone (HIZ) is located 275 km South of Addis Ababa (the capital city of Ethiopia) at an altitude of 1697–1742m. a.s.l. at 7°06' N and 38°48' E. It has a cluster of factories such as Textile, Moha-Soft Drink and Brewery-BGI Factory. These factories discharge untreated and/or partially-treated wastewaters (Zinabu and Zerihun, 2002) into the nearby streams called Boga and Boicha (Fig. 1). The residents in the downstream areas of the Boicha stream use the effluent laden stream for irrigation and watering their cattle, especially during the dry season.

There were two sampling sites: Site 1 was the Industrial Zone (IZ), the area where there are discharges from the industrial effluents, and **Site 2** was the Non-industrial Zone (NIZ), located 10 km from the industrial zone towards the North around Wondo Genet. It was selected as a Reference Site (see 9 in Fig. 1), a benchmark to compare with the IZ. This site is assumed to be free from urban and industrial effluents and there is relatively little influence from human activities (Fig. 1). Raw milk samples were collected from cows that regularly graze in the IZ and drink from Boicha stream (see 8 in Fig. 1) - which receives effluents from the Biological Lagoon of the Textile factory, Moha Soft Drink and BGI Brewery factories (see 5 in Fig. 1). Cattle often drink directly from the effluents of the factories and feed on grasses on the shoreline of the effluent laden stream (see 5-8 in Fig. 1)

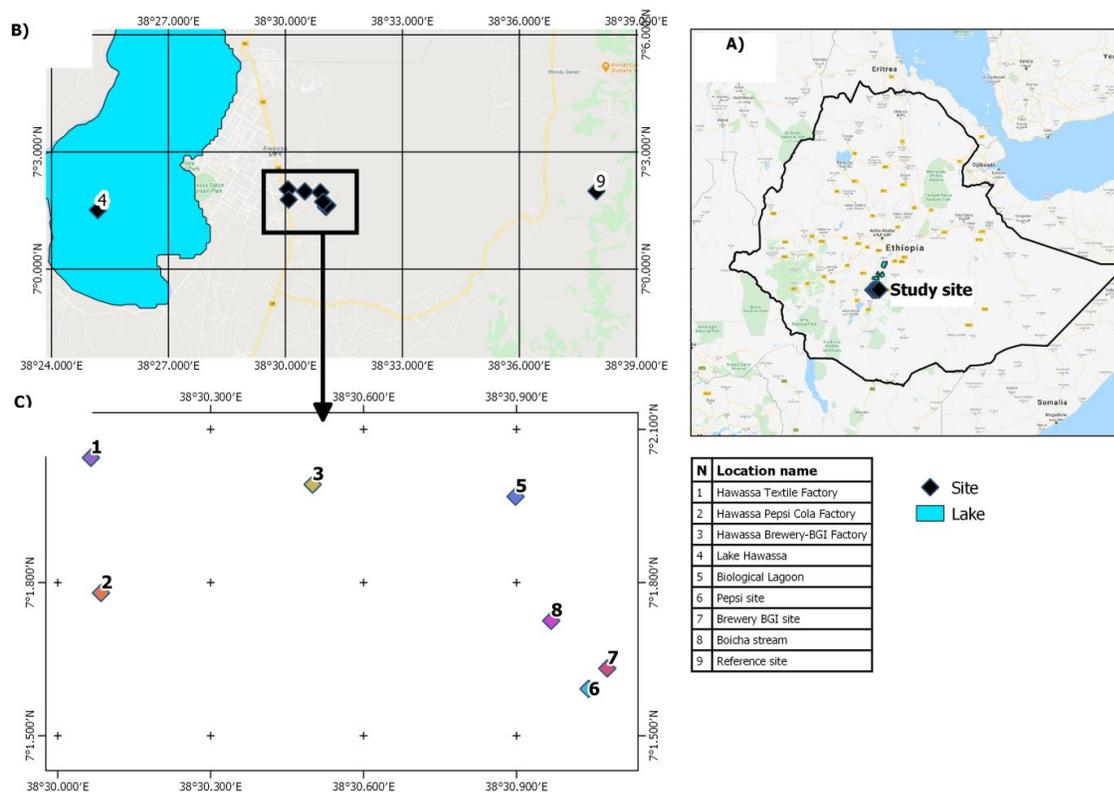


Figure 1. Map of Hawassa Industrial Zone with sampling sites
Sampling Sites

Milk Sample Collection, transportation and storage

Raw milk samples were collected from a total of 30 lactating cows, 15 cows from each sampling site (Industrial and Non-industrial Zones), which were randomly selected from different farmers (30 households) in the two sampling sites. Before sampling, 15 mL polyethylene plastic vials were soaked in 20% HNO₃ for 24 hrs and rinsed with distilled water. Raw milk (15 mL) was collected in the 15 mL polyethylene plastic vials from each lactating cow during the morning milking and kept in an icebox holding ice blocks until transported to the laboratory. Milk sampling was done directly from the farmers' home. Therefore, the chances of indirect contamination through utensils, air and milk processing machines were minimal. The milk samples were transported to a laboratory at the Biology Department, Hawassa University, Ethiopia, within 2 hrs of sampling and stored in a deep freezer at -20 °C for three days before transporting to Norway. The samples were air transported, in icebox filled with ice, to the Norwegian University of Life Sciences (NMBU) for quantitative analyses of heavy metals.

Milk Sample Preparation, Digestion and Analyses for Heavy Metals

Five grams of solidified raw milk from each sample was transferred into a 500 mL round bottom flask (RBF) containing 5.0 mL HNO₃/H₂O₂ (5:2; v/v), and the samples were digested in an UltraClave (Milestone) at 260°C for 20 min. Digested milk samples were allowed to cool at room temperature for 10 min without dismantling the condenser from the RBFs and for another 10 min after removing the condenser. Distilled water (15 mL) was added to the cooled solution in the RBFs and filtered with Whatman 110 mm filter paper into a 50 mL volumetric flask. The filtered sample solution was transferred into a volumetric flask and rinsed with 5 mL distilled water to make up the volume of the solution to 50 mL. The concentrations of ten heavy metals (Cr, Ni, Cu, Zn, As, Cd, Pb, Fe, Co and Mn) were determined using Inductively Coupled Plasma Mass Spectrometry (Agilent 8800 ICP-MS) with triplicate analysis. European Reference Material (ERM-BD150) for skimmed milk powder was used as a certified reference material for the analyses. Blank samples were processed in the same way as the milk samples. According to reference material (ERM-BD150) the recoveries were from 93-98 %.

Potential Human Health Risk Assessment

The potential human health risks, non-carcinogenic and carcinogenic health risks, due to ingestion of heavy metals through the consumption of milk was assessed as estimated daily intake, target hazard quotient, hazard index and target cancer risks.

Estimated Daily Intake (EDI)

The EDI for ingestion of each metal due to milk consumption was determined to assess the health risks associated as described with Equation (1) (Muhib *et al.*, 2016).

$$EDI \text{ (mg kg}^{-1}\text{)} = \frac{C \times DCR}{BW} \quad (1)$$

Where EDI = Estimated daily intake (mg/day); DCR = daily milk consumption rate, which is 53 g/day for Ethiopians (Tassew *et al.*, 2014); BW = average body weight for Ethiopian adults (WHO, 2010) which is 60 kg; C = metal concentration in milk (mg kg⁻¹).

Non-carcinogenic Health Risks

Target Hazard Quotient (THQ) and Hazard Index (HI) were used to estimate the potential non-carcinogenic health risks of heavy metals (USEPA, 2019). The THQ assesses health hazards of an individual heavy metal from milk consumption while HI assesses health hazards of combined heavy metals, and was estimated as THQ of the sum of individual metals (USEPA 2019). The THQ assumes a level of exposure below which it is unlikely for even a sensitive population to experience adverse health effects (Kawser *et al.*, 2016). The THQ and HI values were estimated using equation 2 and 3, respectively (USEPA 2019). When the THQ and HI values are below 1.0 it means that the exposed population is safe in relation to the studied metal(s) and adverse non-carcinogenic health effects are unlikely to occur. If the THQ and HI values of metals are greater than 1.0, there are potential non-carcinogenic risks related to the heavy metal(s) analysed (USEPA 2019).

$$THQ = \frac{EDI}{RFD} \times 10^{-3} \quad (2)$$

Where

RFD = oral reference dose obtained from Integrated Risk Information System, Chemical Assessment Summary (USEPA 2003) database which was 0.003 mg kg⁻¹/day for Cr, 0.020 (Ni), 0.300 (Zn), 0.04 (Cu), 0.0003 (As), 0.001(Cd), 0.0035 (Pb), 0.70 (Fe), 0.03 (Co) and 0.300 for Mn.

$$HI = \sum THQ \quad (3)$$

Target Cancer Risks (TCRs)

TCRs were estimated as the incremental probability of an individual to develop cancer over a lifetime

exposure to that potential carcinogen (USEPA 1989). Acceptable risk levels for carcinogens range from 10⁻⁴ to 10⁻⁶ (the risk of developing cancer over a human lifetime is 1 in 10,000 in the former case and 1 in 1,000,000 in the latter). The equation used for estimating the TCR is as follows:

$$\frac{EF \times ED \times EDI \times CSFO}{AT} \times 10^{-3} \quad (4)$$

Where

TCR is target cancer risk (lifetime cancer risk)

EF is the exposure frequency (From 350 days/year) (FAO 2011).

ED is the exposure duration equivalent to the average lifetime/average life expectancy which is 65 years in Ethiopia (WHO 2013).

EDI is the estimated daily intake.

CSFO is the oral carcinogenic slope factor from integrated risk information system database (USEPA 2010) which was 1.5 mg kg⁻¹/day for As, 1.7 (Ni), 15 (Cd), 0.5 (Cr) and 0.0085 mg kg⁻¹/day for Pb. The TCR was estimated for Cr, Ni, Cd, As and Pb only since these elements may promote both non-carcinogenic and carcinogenic effects depending on the exposure dose. According to the International Agency for Research on Cancer (USEPA 2012), Cr, Ni, Cd and As are known as Group A carcinogens and Pb is known as Group B carcinogen.

TA is the average exposure time for non-carcinogens (365 days/year x ED)

10⁻³ is the unit conversion factor.

Statistical Analysis

The t-test was used to compare site-wise differences in mean values of heavy metals at α=0.05 level of significance. Concentrations of heavy metals in milk were also compared with standard limits of IDF (1979).

RESULTS AND DISCUSSION

Concentrations of Heavy Metals in Cow Milk

Concentrations of heavy metals in the cow milk are presented in Table 1. Mean concentrations of heavy metals in the cow milk sampled from IZ and NIZ, generally followed the order: Zn>Fe>Mn>Cu>Cr>Ni>Pb>As>Co>Cd and Zn>Fe>Mn>Cu>Cr>As>Ni>Pb>Co>Cd, respectively. The highest mean concentration of heavy metals obtained in milk from IZ was 5093.33±223.49 µg kg⁻¹ for Zn and the lowest value was 0.14±0.06 µg kg⁻¹ for Pb. Similarly, the maximum and minimum mean concentrations of heavy metals obtained in milk from NIZ were 3953.33±63.15 and 0.09±0.03 µg kg⁻¹ for Zn and Pb, respectively.

The fact that the mean concentrations of all the heavy metals investigated in this study were higher in the cow milk sampled from IZ than in those from NIZ could be attributed to the effluents from industries and urban wastes that contaminated the soils and/or foraging grasses. This is very likely, given that the effluents may contain some heavy metals as they are not treated at all or inadequately treated. Studies on metal concentrations of effluents have shown that they contained heavy metals like Cr, Co, Cu, As, Cd, Hg, Ni, Se, Pb, Fe, etc. in supposedly treated effluents (Zinabu and Zerihun 2002; Berehanu *et al.*, 2015).

The mean concentrations of Cr from IZ and NIZ, 3.29 and 2.0 $\mu\text{g kg}^{-1}$, respectively, obtained in this study were much lower than other findings from Ethiopia. For instance, Muluken (2014), reported 200.0 $\mu\text{g kg}^{-1}$ of Cr from Hawassa, Tassew *et al.* (2014), 868.0 $\mu\text{g kg}^{-1}$ from Borena, and Akele *et al.* (2017) 648.0 $\mu\text{g kg}^{-1}$ from Gondar. The differences could be attributed to variances in sample handling, including the methods and instruments used. The result of the present study revealed that the concentration of Zn in the milk was higher than those reported (Zn= 3023.0 $\mu\text{g kg}^{-1}$) by Akele *et al.* (2017) from Gondar, but lower than the value reported (Zn= 6290.0 $\mu\text{g kg}^{-1}$) by Arafa *et al.* (2014) from Egypt. The higher Zn concentration in cow milk may be due to its content in soil and foraging grasses. In line with this observation, Samuel *et al.* (2020), reported that Zn was the most abundant heavy metal in soil from the present study area (133.0 and 140.0 mg kg^{-1} in soil from Biological Lagoon area and Boicha stream area, respectively). Grasses that grow on such soils may have higher concentrations of Zn. Tassew *et al.* (2014) reported mean concentration of 109.0 $\mu\text{g kg}^{-1}$ of Cu in milk from Borena. This value is higher than the values obtained in this study - 77.78 and 63.0 $\mu\text{g kg}^{-1}$ from IZ and NIZ, respectively. The result concentrations of Cd in the milk samples from IZ and NIZ were 0.14 and 0.09 $\mu\text{g kg}^{-1}$, respectively. The concentrations of Cd in the milk samples from IZ and NIZ were 0.14 and 0.09 $\mu\text{g kg}^{-1}$, respectively. Our values were much lower than those reported from Hawassa (Muluken 2014) and Gondar (Akele *et al.*, 2017) - 200.0 and 287.0 $\mu\text{g kg}^{-1}$, respectively. Reports from studies made on cow milk from other countries have also shown higher concentrations of Cd in milk. For instance, Arafa *et al.* (2014) reported 51.0 $\mu\text{g kg}^{-1}$ of Cd in cow milk from Egypt. Muhib *et al.* (2016) from Bangladesh, Yasser *et al.* (2016) from Iran and Ismail *et al.* (2015) from Pakistan reported Cd concentrations of 24.0, 1.11 and 1.2 $\mu\text{g kg}^{-1}$, respectively. The differences could be attributed to the variances in heavy metal contaminations of the study areas.

Akele *et al.* (2017) reported much higher mean concentration of Pb (153.0 mg kg^{-1}) in cow's milk from Gondar than the values 0.93 and 0.81 mg kg^{-1} from IZ and NIZ, respectively obtained in this study. On the

contrary, Muluken (2014) reported mean concentration of 0.8 $\mu\text{g kg}^{-1}$ of Pb in cow milk from Hawassa-a value very close to our findings. The presence of Pb in the milk samples could be due to various factors such as fodder contamination, climate factors (wind) and the use of pesticide compounds in the catchment (Yasser *et al.*, 2016). Arafa *et al.* (2014) reported mean concentration of 8994.0 $\mu\text{g kg}^{-1}$ of Fe in cow milk from Egypt. This value is much higher than the values 592.0 and 306.66 $\mu\text{g kg}^{-1}$ from IZ and NIZ, respectively obtained in this study.

The fact that mean Fe concentration in the cow milk was lower in the present study compared to the study of Arafa *et al.* (2014) from Egypt could be due to the absence of iron and steel industries in the catchment area of the present study (Hawassa Industrial Zone) where only factories like textile, soft drink, brewery, detergent, etc. exist. The result of the present study revealed that the concentrations of Mn in the milk samples from both sites were lower than those reported by Tassew *et al.* (2014) Mn (427.0 $\mu\text{g kg}^{-1}$) from Borena and Akele *et al.* (2017), 2111.0 $\mu\text{g kg}^{-1}$, from Gondar. Again we attribute the differences to heavy metal contamination levels of the study areas - although the differences in sample handling, methods and instruments used for analysis cannot be discounted.

There were statistically significant ($P<0.05$) site dependent (IZ and NIZ) differences in the mean concentrations of all heavy metals except three (As, Pb and Mn) in the cow milk (Table 1). This suggests that there was difference in heavy metal pollution levels between Industrial and Non-industrial Zones. The absence of significant differences in the mean concentrations of As and Pb in cow milk between Industrial and Non-industrial Zones could be due to some level of As and Pb contamination from the use of agrochemicals in Non-industrial Zone (Samuel *et al.*, 2020).

As indicated in Table 1, the mean concentrations of Zn, Fe and Cu in cow milk from IZ as well as Zn and Cu from NIZ are above the MTL in the diet of humans according to the standard set by IDF 1979. This shows that cow milk from IZ and NIZ may not be safe for human consumption with regard to Zn, Fe and Cu toxicities in the area.

The mean values of Zn, Fe and Cu from this study surpassed those set by IDF standards (3280.0, 370.0 and 10.0 $\mu\text{g kg}^{-1}$ for Zn, Fe and Cu, respectively) in general suggests that there could be various sources of pollution that have affected the study area, particularly the water quality, which in recent years has deteriorated considerably by uncontrolled industrial releases and the intensive use of chemical fertilizers in agriculture (Malhat *et al.*, 2012; Muluken, 2014).

Health Risks associated with Milk Consumption

Estimated Daily Intake

The EDI and permissible values of the heavy metals considered in this study for adult consumers of cow milk from IZ and NIZ is presented in Table 2. The EDI generally followed the order: Zn>>Fe>Cu>Cr=Ni>As=Pb>Cd=Co for milk consumption from both Industrial and Non-industrial Zones. The EDI value for Zn 4.50 and 3.50 mg kg⁻¹ observed from IZ and NIZ, respectively were the highest recorded EDI values for the heavy metals considered in this study. However, these values were still below the Permissible Limits (Table 2). This shows that Zn was the major component contributing to the potential human health risk due to consumption of milk from both Industrial and Non-industrial Zones. The EDIs of all the heavy metals were found to be below the permissible limit (Table 2). This suggests that

there is little human health concern related to these heavy metals from consumption of milk in the study area. The result of the present study revealed that the EDI values of heavy metals from consumption of cow milk were lower than the values reported by Akele *et al.* (2016) Cr (0.731), Cu (1.353), Cd (0.292), Pb (0.164) and Mn (2.479) from Gondar. Reports from studies made on cow milk from other countries have also shown lower EDI values for heavy metals. Arafa *et al.* (2014) reported EDI value of 0.021, 0.0007, 0.00032 and 0.03 mg kg⁻¹ for Zn, Pb, Cu, and Fe, respectively for cow milk consumption from Egypt. All of these values are much lower than the values obtained in this study (Table 2). The differences could be attributed to differences in heavy metal contamination level of the study areas – although some differences due to sample handling and methodology, including types of instruments used, cannot be ruled out.

Table 1 Heavy metal concentrations ($\mu\text{g kg}^{-1}$) in cow milk from the IZ and NIZ (Mean \pm SD, n = 15 each).

Heavy metals	Site		MTL ($\mu\text{g kg}^{-1}$)	Reference
	IZ	NIZ		
Cr	3.29 \pm 0.97 ^a	2.0 \pm 0.81 ^b	300.0	IDF, 1979
Ni	2.93 \pm 0.37 ^a	1.0 \pm 0.27 ^b		
Cu	77.78 \pm 13.94 ^a	63.0 \pm 9.78 ^b	10.0	IDF, 1979
Zn	5093.33 \pm 223.49 ^a	3953.33 \pm 63.15 ^b	3280.0	IDF, 1979
As	0.88 \pm 0.26 ^a	1.07 \pm 0.27 ^a		
Cd	0.14 \pm 0.06 ^a	0.09 \pm 0.03 ^b	2.63	IDF, 1979
Pb	0.93 \pm 0.24 ^a	0.81 \pm 0.23 ^a	20.0	Codex, 2011
Fe	592.0 \pm 63.19 ^a	306.66 \pm 13.37 ^b	370.0	IDF, 1979
Co	0.53 \pm 0.17 ^a	0.40 \pm 0.13 ^b		
Mn	112.73 \pm 53.74 ^a	81.87 \pm 43.63 ^a		

Mean values with different superscript letters in a row are significantly different ($p < 0.05$) from each other. *MTL = maximum tolerance limit, *Values in bold are those above the MTL in the diet of humans according the standards indicated in the references.

Table 2 Estimated daily intake (EDI) for heavy metals in cow milk samples for adult consumers and comparison with Permissible Values.

Heavy metal	Industrial Zone	Non-industrial Zone	Permissible Value (mg/day)	Reference for Permissible Value
	EDI (mg/day)	EDI (mg/day)		
Cr	0.003	0.002	0.2	Ogabiela <i>et al.</i> , 2011
Ni	0.003	0.001		
Cu	0.07	0.06	30.0	JECFA, 2003
Zn	4.50	3.50	15.0	JECFA, 2003
As	0.001	0.001		
Cd	0.0	0.0	0.046	JECFA, 2003
Pb	0.001	0.001	0.21	JECFA, 2003
Fe	0.53	0.27	40.0	FAO/WHO, 2002
Co	0.0	0.0		
Mn	0.10	0.07	5.0	Ogabiela <i>et al.</i> , 2011

Target Hazard Quotient

The THQ values for heavy metals from consumption of

cow milk from IZ and NIZ followed the order of: Zn>As>Cu>Cr>Fe>Mn>Pb>Ni=Cd>Co and Zn>As>Cu>Cr>Fe>Mn>Pb>Cd>Ni>Co, respectively (Table 3). The THQ values for all heavy metals, except

As, were significantly higher ($P < 0.05$) for milk consumption from IZ than the consumption from NIZ. This suggests that there is a higher probability of non-carcinogenic health risk from ingestion of all the heavy metals analyzed, except As, individually through the consumption of cow milk from IZ in comparison to that of NIZ. This might be due to a high level of heavy metal pollution in IZ than in the NIZ. This is very possible, given that the IZ receives effluents from the factories in the IZ as well as urban wastes.

Akele *et al.* (2017) reported THQ values of 0.034 and 0.016 for Cu and Zn, respectively through milk consumption from Gondar, Ethiopia. These values are much lower than the values obtained in this study: for Cu, 0.172 and 0.139 from the IZ and NIZ, respectively, and for Zn, 1.275 and 0.815 from the IZ and NIZ, respectively. The THQ values of heavy metals of the present study were lower than values reported by Arafa *et al.* (2014) for Cd (0.17), Pb (0.2) and Fe (0.1) from Egypt, and Akele *et al.* (2017) for Mn (0.177) from Gondar. On the contrary, THQ values lower than the results of the present study were reported by Muhib *et al.* (2016) for Fe (0.003) and Mn (0.005) from Bangladesh. This suggests different pollution levels between the study areas (Textile, Brewery and Soft Drink factories around the present study area) and possible methodological differences.

The fact that the THQ values for all heavy metals, except Zn, from both study sites were less than unity suggests that there is potentially little non-carcinogenic health risk from ingestion of all the heavy metals, except Zn, individually through consumption of milk from both Zones. On the other hand, the THQ value for Zn through consumption of milk from IZ was greater than unity; therefore there are potential non-carcinogenic health risks from ingestion of Zn through consumption of cow milk from the IZ. Zinc is an

essential metal but excessive ingestion can have adverse effects on human health by reducing immune function and the levels of high-density lipoproteins (Harmanescu *et al.*, 2011). The families of farmers around Hawassa IZ and those who regularly purchase milk from these areas are high risk group for Zn toxicity through cow milk consumption.

Hazard Index

The HI values through consumption of cow milk from both Industrial and Non-industrial Zones were greater than unity (Table 3). This suggests that there is a potential non-carcinogenic health risk from ingestion of all the heavy metals considered in this study collectively through consumption of the milk. Muhib *et al.* (2016) and Akele *et al.* (2017) reported HI of 0.128 and 0.262, respectively through milk consumption from Bangladesh and Gondar, Ethiopia. Their values are much lower than the values 1.963 and 1.423 from IZ and NIZ, respectively obtained in this study. This shows that consumption of cow milk from the present study area has more risks compared to those from Gondar and Bangladesh.

The percent heavy metal specific contributions to HI through consumption of milk from Industrial Zone and Non-industrial Zone are presented in Figure 2 and Figure 3, respectively. Zinc was the highest contributor (64.95% for IZ and 57.19% for NIZ) to HI through consumption of milk from both sites whereas Co was the lowest contributor (0.1% for IZ and 0.07% for NIZ). These values indicate that Zn was the most important contributor to a potential non-carcinogenic health risk due to consumption of milk from both Industrial and Non-industrial Zones, and Co was the lowest contributor.

Table 3. Target Hazard Quotient (THQ) and Hazard Index (HI) for heavy metals from consumption of cow milk from Industrial and Non-industrial Zones.

Site	THQ										HI
	Cr	Ni	Cu	Zn	As	Cd	Pb	Fe	Co	Mn	
Industrial Zone	0.097	0.013	0.172	1.275	0.260	0.013	0.023	0.075	0.002	0.033	1.963
Non-industrial Zone	0.057	0.004	0.139	0.815	0.315	0.008	0.021	0.039	0.001	0.024	1.423

Values in bold (>1) indicate potential non-carcinogenic health risk for humans

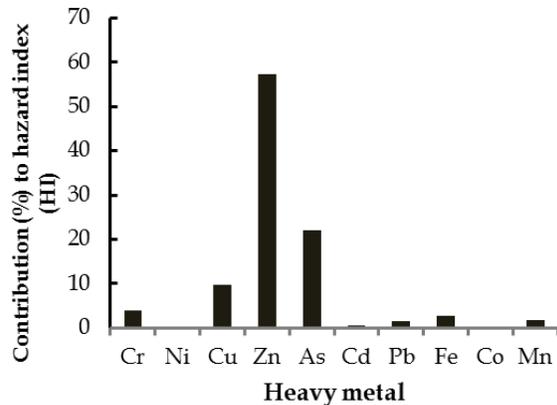


Figure 2. Relative contributions (%) of each heavy metal to the estimated hazard index (HI) through consumption of milk from industrial Zone.

Target Carcinogenic Risks

The highest TCR obtained in this study was $5.1E-05$ for Ni from IZ, while the lowest was $8.5E-08$ for Pb through consumption of milk from both Zones (Table 4). The TCR values were within the acceptable range ($<10^{-6}$ - 10^{-4}) for five out of the ten heavy metals (Cr, Ni, As, Cd

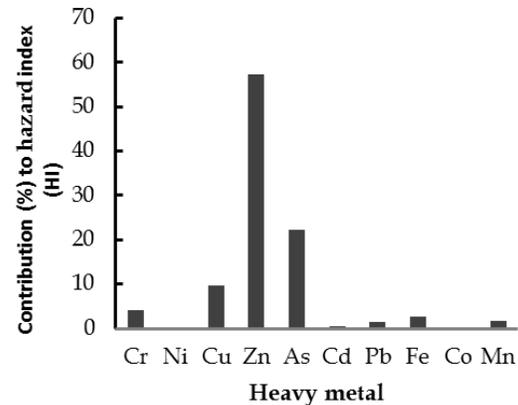


Figure 3 Relative contributions (%) of each heavy metal to the estimated hazard index (HI) through consumption of milk from non-industrial Zone.

and Pb) considered under this study. This indicates that there is no potential carcinogenic health risk from ingestion of these five heavy metals through the consumption of cow milk from both Zones.

Table 4. Carcinogenic (TCR) health risks of heavy metals due to consumption of cow milk from Industrial and Non-industrial Zones.

Site	TCR				
	Cr	Ni	As	Cd	Pb
Industrial Zone	$1.5E-05$	$5.1E-05$	$1.5E-05$	$1.5E-05$	$8.5E-08$
Non-industrial Zone	$1.0E-05$	$1.7E-05$	$1.5E-05$	$1.5E-05$	$8.5E-08$

CONCLUSIONS

The results of this study revealed that consumption of cow milk from Hawassa IZ could be unsafe with respect to toxicities of Zn, Fe and Cu in the area. The sources of these heavy metals in the IZ were textile, brewery and soft drink factories. As a result, families of farmers and those who purchase the milk from this area, especially pregnant women and vulnerable age group of the people, could be at health risk. This calls for awareness creation among the people in the community, regular monitoring of the content of the milk produced in this area, and other policy interventions to regulate waste disposal in the industrial zone.

ACKNOWLEDGEMENTS

This study was financed by the Institutional Collaboration Program between the Norwegian University of Life Sciences (NMBU) and Hawassa University, supported by the Royal Norwegian Embassy in Addis Ababa. We thank Pia Frostad for her

help in the laboratory analyses of the samples. The authors would also like to thank farmers around Hawassa IZ and Wondo Genet for their kind cooperation.

REFERENCES

- Akele, ML, Abebe, DZ, Alemu, AK, Assefa, AG, Madhusudhan, A and de Oliveira, RR. 2017. Analysis of trace metal concentrations in raw cow's milk from three dairy farms in North Gondar, Ethiopia: chemometric approach. *Environ. Monit Assess.* 189: 499.
- Arafa, MS, Walaa, A, Moselhy, NE and Hassan, Y. 2014. Heavy metals and trace elements levels in milk and milk products. *Food Measure.* 8:381-388.
- Berehanu, B, Bekele, L, and Yosef, TG. 2015. Chemical Composition of Industrial Effluents and Their Effect on the Survival of Fish and Eutrophication of Lake Hawassa, Southern Ethiopia. *J, Environ*

- Prot. 6:792-803.
- Codex Alimentarius Commission. 2011. Report of the 50th session of the Codex committee on food additives and contaminants. Hague: Codex Alimentarius Commission.
- Datta, BK, Mishra, A, Singh, A, Sar, TK, Sarkar, S and Bhattacharya, A. 2010. Chronic arsenicosis in cattle with special reference to its metabolism in arsenic endemic village of Nadia district West Bengal India. *Sci. Total Environ.* 409:284-288.
- Eline, DH, Sam, D, Marijke, DH, Wim, V, and Koen, S. 2019. Religion, Food Choices, and Demand Seasonality: Evidence from the Ethiopian Milk Market. *Foods.* 8:167.
- FAO/WHO. 2002. Codex Alimentarius-general standards for contaminants and toxins in food. Schedule 1 Maximum and guideline levels for contaminants and toxins in food, Joint FAO/WHO food standards programme. Rotterdam: Codex committee. Reference CX/FAC 02/16.
- FAO. 2011. Office of the FAO Representative in Ethiopia to AU and ECA- Addis Ababa.
- Haftom, Z, and Ashagrie, Z. 2020. Nutritional Quality, Level of Chemical Contaminants and Adulterants in Milk and Dairy Products in Ethiopia: A Review. *Modern Chemistry*, 8(4): 64-72.
- Hanna, GW, Krystyna, R, Waclaw, L and Maksymilian, C. 2019. Milk and Dairy Products and Their Nutritional Contribution to the Average Polish Diet. *Nutrients.* 11:1771.
- Harmanescu, M, Alda, LM, Bordean, DM, Gogoasa, I and Gergen, I. 2011. Heavy metals health risk assessment for population via consumption of vegetables grown in old mining area; a case study: Banat County, Romania. *Chemical Central Journal.* 5:64.
- IDF Standard. 1979. Metal contamination in milk and milk products. *Int. Dairy Fed Bull.* 105:1-40. Document No. A. Doe 37.
- Ismail, RM, Akhtar, S, Ismail, T, Ahmad, Z and Hashmi, MS. 2015. Estimated daily intake and health risk of heavy metals by consumption of milk. *Food Addit Contam Part B.* 8(4): 260-265.
- JECFA. 2003. Summary and conclusions of the 61st meeting of the joint FAO/WHO Expert committee on food additives (JECFA). Rome, Italy: JECFA/61/SC.
- Kawser, A, Mohammad, AB, Goutam, K, Saiful, I, Monirul, I, and Muzammel, H. 2016. Human health risks from heavy metals in fish of Buriganga River, Bangladesh. *SpringerPlus.* 5:16-97.
- Licata, P, Di Bella, G, Potorti, AG, Lo Turco, V, Salvo, A and Dugo, G. 2012. Determination of trace elements in goat and ovine milk from Calabria (Italy) by ICP-AES. *Food Addit Contam Part B.* 5:268-271.
- Malhat, F, Hagag, M, Saber, A and Fayz, AE. 2012. Contamination of cows' milk by heavy metal in Egypt. *Bull Environ Contam Toxicol.* 88:611-613.
- Mebrate, G, Tewodros, A and Dawit A. 2019. Dairy Production in Ethiopia - Existing Scenario and Constraints. *Biomedical Journal of Scientific and Technical research*, Review article, ISSN: 2574 - 1241.
- Muhib, I, Muhammed Alamgir, ZC, Nusrat, JE, Mostafizur, R, Mashura, S, Zeenath, F. 2016. Investigation of heavy metal contents in Cow milk samples from area of Dhaka, Bangladesh. *Inter. J Food Contam.* 3:16.
- Muluken, M. 2014. Heavy metals concentration in effluents of Hawassa Textile industry, Tikur Wuha River and Milk of cows watering on this water source, Hawassa, Southern Ethiopia. *Res. J. Environ. Sci.* 8(8):422-434.
- Ogabiela, EE, Udiba, UU, Adesina, OB, Hammuel, C, Yebpella, GG, Mmereole, UJ. 2011. Assessment of metal levels in fresh milk from cows grazed around Challawa industrial Estate of Kano, Nigeria. 1(7):533-538.
- Samuel, B, Sorsa, S, Daniel, F, Zinabu, GM and Riise, G. 2020. Heavy Metals Contamination of Soil in the Vicinity of Hawassa Industrial Zone, Ethiopia. *J. Appl Sci Environ Manag.* 24 (8): 1447-1454.
- Tassew, B, Ahmed, H and Vegi, M. 2014. Determination of Concentrations of Selected Heavy Metals in Cow's Milk: Borena Zone, Ethiopia. *J. Health Sci.* 4(5): 105-112.
- USEPA. 1989. Risk Assessment Guidance for Superfund. *Human Health Evaluation Manual Part A, Interim Final, vol. I. EPA/540/1-89/002. United States Environmental Protection Agency Washington, DC, USA.*
- USEPA. 2003. Integrated Risk Information System (IRIS). Chemical Assessment Summary.
- USEPA. 2010. *Risk-Based Concentration Table.*
- USEPA. 2012. EPA Region III Risk-Based Concentration (RBC) Table 2008 Region III, 1650 Arch Street, Philadelphia, Pennsylvania 19103.
- USEPA. 2019. USEPA Zoneal Screening Level (RSL) Summary Table.
- WHO. 2013. WHO Country Cooperation Strategy 2012-2015: Ethiopia. World Health Organization. Zoneal

Office for Africa.

- Yasser, S, Farhad, A and Farnoosh, F. 2016. Voltammetric determination of Pb, Cd, Zn, Cu and Se in milk and dairy products collected from Iran: An emphasis on permissible limits and risk assessment of exposure to heavy metals. *Food Chemistry*. 192:1060-1067.
- Zinabu, GM and Zerihun, D. 2002. The chemical composition of the effluent from Awassa textile factory and its effect on aquatic biota. *SINET: Ethiop J Sci*. 25(2):263-274.

Licensed under a Creative Commons
Attribution-Non-Commercial 4.0 International License



©Jimma University, 2020
ISSN 2220-9325 (Online), ISSN 2203-5802(Print)