

## Environmental Contamination and Assessment of Heavy Metals in Water and Sediments of Awash River Basin, Ethiopia

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### Abstract

Awash River is considered as one of the most important irrigation and drinking water resources for the community. Huge amount of domestic, municipal, industrial and agricultural wastes notably are discharged to river. In the present study, the contamination levels of heavy metals (Cr, Cu, Mn, Cd, and Pb) and their ecological effect in water and sediment of Awash River were investigated. Sediment and water samples were collected from selected sites along the course of river and analyzed the metals to be concerned. The result showed that mean concentrations of heavy metals ranked as: in water and sediment samples were Mn > Cr > Cd > Cu and Pb. Mean concentrations of Cr, Mn and Pb in the water samples were higher than WHO and USEPA guidelines for some sites in the upper basin of the river. Heavy metals contamination in the sediments were evaluated by applying United State environmental Protection Agency's sediment quality Guidelines (USEPA) and indexes such as Enrichment factor, Geoaccumulation Index, contamination factor, Degree of contamination, Ecological risk factor and potential Ecological risk index were high in the upper basin sampling sites of the river and the extent of metal pollution in the river around is frightening and considerably affecting the aquatic ecology of the river. Therefore, further studies are needed on metal speciation and effects on metal uptake by humans and organisms in addition to continuous monitoring.

**Keywords:** Environment; Contamination, Heavy metals, Sediment and Risk Assessment

### 1. Introduction

Water is a vital resource to all living things on earth. It was argued that human beings use water for domestic, industrial and agricultural purposes (Ayeni et al, 2011). In the past decade, humans had an

impact on the environment due to population growth and rapid rate of urbanization (Olorunfemi and Jimoh, 2000). Globally, about 80% of all diseases and deaths in developing countries were caused by polluted water (Awake, 2001 and Aderibigbe et al; 2008). Like other aquatic systems, heavy metals enter surface water as in lakes and rivers through natural and anthropogenic sources (Zorer et al, 2008 and Alhas et al, 2009). These activities can generate heavy metals to sediment and water to pollute the aquatic environment (Sanchez-Chardi et al., 2007). In an aquatic ecosystem, heavy metals scavenged by fine particles lead to their accumulation in sediments. Sediment is an important part of the river basin, with the deviation of habitats and environments (Morillo et al., 2004). Sediments have been extensively regarded as environmental pointers for the evaluation of metal pollution in the watercourse (Islam et al. 2015d). The concentrations of heavy metals are extremely high in sediments rather than the water columns because metals tend to amass in bottom deposits (He et al. 2009; Sultan and Shazili, 2009; Nobil et al., 2010; Rezayi et al., 2011; Bhuyan et al., 2017). At present, heavy metal pollution has become a great environmental concern with its toxicity, persistence, bioaccumulation and biomagnifications in the food chain (Li et al., 2008 and Yuan et al., 2009).

Among other chemical pollutants, heavy metals, being non-biodegradable, can accumulate along the food chain, causing toxic effects at locations far removed from the original source of pollution (Tilzer and Khodker, 1993). Knowingly, surface water is an important source of water and food. In Ethiopia, Awash River is one of the most important rivers, used for irrigation, fishing, bathing, drinking etc. The river flows from the central highlands through Ethiopia's major industrial and agro-industrial belt, carrying a significant burden of various raw effluents. It stands as one of Ethiopia's prominent river streams in urban areas (Tsefamariam, 1989). Due to these activities, the river is facing contamination mainly by heavy metals which have an adverse biological effect on human health through drinking water, consuming fish and irrigated products (Huang et al, 2009 and Zang et al, 2009). For these reasons, it would be desirable and imperative to investigate contamination levels of heavy metals in river water and sediments, as this can provide valuable information on heavy metals pollution.

Therefore, the objective of the present study is to assess heavy metals (cadmium (Cd), chromium (Cr), copper (Cu), manganese (Mn) and lead (Pb)) contaminations in water and sediments of the Awash River.

## 2. Materials and Methods

### 2.1. Study area



The present study was conducted in the Awash River basin, which lies between 7°53′–12°N and 37°–43°25′E, with a total catchment area of 113,304 km<sup>2</sup> (Figure 1) from September 2010 to June 2012. Awash River plays a significant role in socio-economic development of Ethiopia. The river has great importance in agriculture, household activities and daily uses. Several types of industries were set up on the bank of Awash River like textile, cement, tannery, packaging, paint, paper and pulp, acid and salt manufacturing, fertilizer industries, different food processing industries and other factories that produce huge volumes of effluents containing heavy metals. However, the river stands as one of Ethiopia's river streams in developed urban areas, which are polluted due to these raw effluents (Tesfamariam, 1989). Moreover, the expansion of new industries and disposal of industrial waste to the Awash River are of great concern to the nation (Girma, 2001). The location of sampling sites along the river course is shown in Table 1 below.

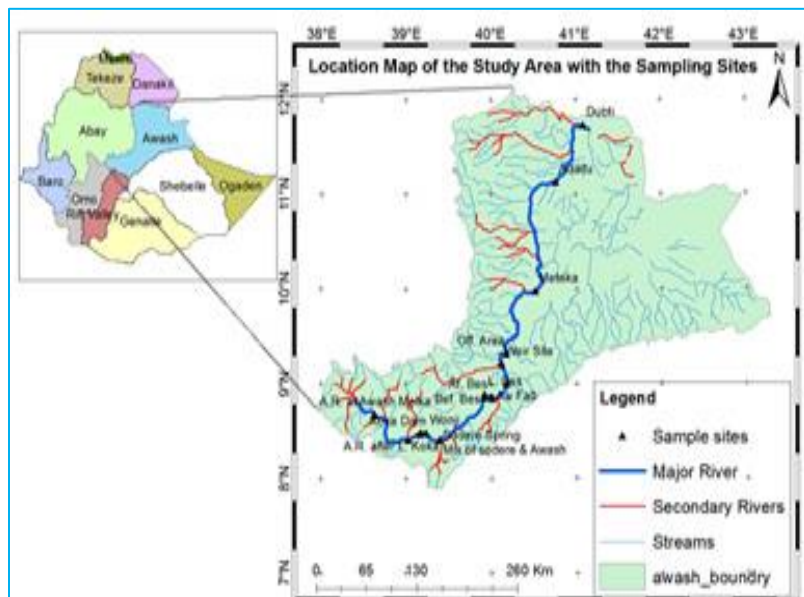


Figure 1 Map of study area

**Table 1** Justification of study sites in Awash River Basin.

Stream of Awash River	Sampling sites	Site description and activities	Coordinates	
			Latitude	Longitude
Lower	Afembo	Used by the community for livestock, domestic and irrigation purposes as well as a waste drainage from sugar	11.54470	41.45048
	Adaytu	Used as water gage station and soil erosion is takes place.	11.12873	40.76299
Middle	Melkaworor	Sources of irrigation of agricultural farm (cotton, vegetable farming), livestock and domestic purposes.	9.20587	40.11947
	Awash Park	Reflect the impact of Metehara sugar factory waste and Lake Beseka on river water.	-	-
	Metehara	Sources of irrigation of agricultural farms (sugar, vegetable farming), animal feeding and other domestic purposes.	8.85164	39.921174
	Sodere	Source of water for spiritual bathing, washing, domestic purpose, and small irrigation.	8.39492	39.39840
Lower	Mille	Reflect the impact of Borkena (containing industrial waste), Logiya wastes and their impact on the river	11.41395	40.76068
Upper	Koka	Sources of irrigation of agricultural farms, fishing and other domestic purposes.	8.40771	39.02130



	Mojo	Serve as drainage for industrial wastes and dumping of solid wastes near to the river.	8.59787	39.11122
	Near to Leather Dev't Institute (Akaki)	Serve as a waste drain for small-scale industrial, cars and municipal waste.	8.92219	38.75104
	Bole	Serve as a waste drain for domestic and municipal wastes.	8.99197	38.77758
	Aba-Samuel	Sources of irrigation of agricultural farms, fishing, serve as a waste drain for domestic and municipal wastes	8.78739	38.70625
	Asko	Serve as a waste drain for domestic and municipal waste.	9.05920	38.69699
	Hatebela	Serve as drainage for industrial domestic and municipal waste.	-	-
	Awash Bello	This site is taken as a reference.	-	-
	Entoto	This site is taken as a reference.	9.07740	38.77417

## 2.2. Sample collection and preservation

The physicochemical parameters such as  $P^H$ , TDS and temperature were measured immediately after sample collection. A total of 192 composite surface water samples were collected from the selected sampling sites from 2010 to 2012 using the standard method (APHA, 2008). For dissolved metals analyses, water samples were filtered immediately after collection. The samples were then transferred into acid cleaned 100 ml polyethylene bottles and acidified with nitric acid ( $pH = 2$ ) and were transferred to the laboratory. A total of 24 composite sediment samples (about 500 g) were collected at a depth of 0 to 5 cm by using a standard procedure from each sampling site (USEPA, 2001a). Sediment samples were air-dried until obtaining constant weight in the laboratory. Then the samples were homogenized by grinding in an agate mortar, sieved to 63  $\mu m$  meshes and prepared for analysis of heavy metal.



### 2.3. Chemical analysis

A 100 ml water of each of the water samples was taken in a beaker. Then the samples were digested by adding 5 ml conc. HNO<sub>3</sub> on a hot plate. After that, the samples were filtrated into a 100 ml volumetric flask using Whatman No 42 filter paper and made up to the mark with distilled water. For heavy metals test, metals were extracted from sediment samples using acid digestion. Briefly, dried sediment samples (0.5 g) were taken in a beaker and the digestion was carried out with 15 ml of a mixture of Conc. HNO<sub>3</sub> and HCl at a 3:1 ratio (v/v) on a hot plate and the mixture was heated to almost dryness, then 5 ml of HNO<sub>3</sub> was added and the solution was allowed to cool at room temperature, then filtered through Whatman No 42 filter paper. The filtrate obtained was made up to 100 ml mark with distilled water in a volumetric flask and the solution was analyzed for heavy metals content against those of the blank using a flame atomic absorption Spectrophometer (FAAS).

#### 2.3.1. Assessment of metal pollution in sediment

##### Enrichment factor (EF)

Enrichment factor (EF) is considered an effective tool to evaluate the magnitude of contaminants in the environment (Franco-Uria et al. 2009). The EF for each element was calculated to evaluate anthropogenic influences on heavy metals in sediments using the following formula (Selvaraj et al., 2004):

$$EF = \frac{\frac{\text{Concentration of targeted metal in sample } (C_m)}{\text{Concentration of iron in Sample } (C_{Fe})}}{\frac{\text{Background concentration of targeted metal } (C_m)}{\text{Background concentration of iron in Sample } (C_{Fe})}} \text{----- (1)}$$

where (C<sub>m</sub>/C<sub>Fe</sub>) sample is the ratio of concentration of heavy metal (C<sub>m</sub>) to that of iron (C<sub>Fe</sub>) in the sediment sample, and (C<sub>Bn</sub>/C<sub>Fe</sub>) background is the same reference ratio in the background sample. Generally, EF value of about 1 suggests that a given metal may be entirely from crustal materials or natural weathering processes (Zhang and Liu, 2002). Samples having EF > 1.5 were considered indicative of human influence and an EF < 1 indicates no enrichment. EF < 3 means minor enrichment, 3-5 is moderate enrichment, 5-10 is moderately severe enrichment, 10-25 is severe enrichment, 25-50 is very severe enrichment, and > 50 is extremely severe enrichment (Birch and Olmos, 2008).

#### 2.3.2. Contamination degree of heavy metals

The contamination factor (CF) is the ratio of measured concentration of the heavy metals in sediment and the pre-industrial reference value for the same metal (Hakanson, 1980). The degree of contamination is



calculated as the sum of all contamination factors. The computing equation for contamination factor (CF) and the degree of contamination (Cd) are as follows:

$$CF = \frac{C_m}{C_b} \text{----- (2)}$$

$Cd = \sum_{i=1}^n CF \text{----- (3)}$ , where,  $C_m$  is the measured concentration of the heavy metals in sediment and  $C_b$  is the standard pre-industrial reference level (mg/kg), 90 for Cr, 850 for Mn, 45 for Cu, 0.3 for Cd, and 20 for Pb (Turekian and Wedepohl, 1961; Hakanson, 1980). The contamination factor is important for monitoring the pollution of one single metal over a period of time. The contamination levels may be classified based on their intensities on a scale ranging from 1 to 6: low degree ( $CF < 1$ ), moderate degree ( $1 \leq CF < 3$ ), considerable degree ( $3 \leq CF < 6$ ), and very high degree ( $CF \geq 6$ ) (Luo et al., 2007). The degree of contamination defines the quality of the environment in the following manner:  $Cd < 8$ ,  $8 \leq Cd < 16$ ,  $16 \leq Cd < 32$  and  $Cd \geq 32$  indicate low, moderate, considerable and very high degree of contamination.

### 2.3.3. Geoaccumulation index ( $I_{geo}$ )

The degree of contamination from the trace metals could be assessed by determining the geoaccumulation index ( $I_{geo}$ ) proposed by Muller (1969). The index of geoaccumulation ( $I_{geo}$ ) has been widely applied to the assessment of soil contamination (Santos Bermejo et al., 2003). In order to characterize the level of pollution in the sediment, geoaccumulation index ( $I_{geo}$ ) values were calculated using the equation:  $I_{geo} = \log_2\left(\frac{C_n}{1.5B_n}\right) \text{----- (4)}$ , where  $C_n$  is the measured concentration of metal  $n$  in the sediment and  $B_n$  is the geochemical background value of metal  $n$  in the background sample (Turekian and Wedepohl, 1961; Rudnick and Gao, 2003). The factor 1.5 is introduced to minimize the possible variations in the background values which may be attributed to lithogenic effects (Nikolaidis et al., 2010). Geoaccumulation index ( $I_{geo}$ ) values were interpreted as:  $I_{geo} \leq 0$  – practically uncontaminated;  $0 \leq I_{geo} \leq 1$  – uncontaminated to moderately contaminated;  $1 \leq I_{geo} \leq 2$  – moderately contaminated;  $2 \leq I_{geo} \leq 3$  – moderately to heavily contaminated;  $3 \leq I_{geo} \leq 4$  – heavily contaminated;  $4 \leq I_{geo} \leq 5$  – heavily to extremely contaminated; and  $5 < I_{geo}$  – extremely contaminated.

### 2.3.4. Pollution load index (PLI)

Pollution load index acts as an integrated approach which assess sediment quality of heavy metals. In this study, pollution load index can be determined for three toxic metals: Cr, Mn and Cd by using the following formula (Tomlison et al., 1980):

$$PLI = \sqrt[n]{CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n} \text{----- (5)}$$



where CF is contamination factor for each single metal and n is number of heavy metals. The overall toxicity status of heavy metals in sediment may be assessed from Pollution load index (PLI) calculation.

### 2.3.5. Assessment of potential ecological risk

The potential ecological risk factor (Er) of single element and the potential ecological risk index (RI) of multi-element can be computed by the following equations (Guo et al., 2010 and Luo et al., 2007).

$Er = T_r \times C_i$  and  $RI = \sum_{i=1}^m Er$  ----- (6), where, Er is the potential ecological risk index and is the biological toxic factor of an individual element.  $C_i$  is the contamination factor for metal 'i'.

$T_r$  is the toxic-response factor for Cr, Mn Cd, Cu and Pb which were 2, 1, 30, 5 and 5 respectively (Hakanson, 1980; Luo et al., 2007; Gong et al., 2008; Madiseh et al., 2009; Guo et al., 2010, Wu et al., 2010; Jintao et al., 2011; Amuno, 2013). RI is the comprehensive potential ecological risk index, which is the sum of Er. The sensitivity of the biological community indicates the potential ecological risk caused by the overall contamination. The following terminologies are used to describe the risk factor:  $Er < 40$ , low potential ecological risk;  $40 \leq Er < 80$ , moderate potential ecological risk;  $80 \leq Er < 160$ , considerable potential ecological risk;  $160 \leq Er < 320$ , high potential ecological risk; and  $Er \geq 320$ , very high ecological risk. The following terminology was used for the potential ecological risk index:  $RI < 150$ , low ecological risk;  $150 \leq RI < 300$ , moderate ecological risk;  $300 \leq RI < 600$ , considerable ecological risk; and  $RI > 600$ , very high ecological risk (Håkanson, 1980).

### 2.4. Statistical analysis

Data obtained were analyzed. Interims of means and standard deviations of the metal concentrations in water and sediment were calculated. Other calculations were performed using Microsoft Excel 2010.

## 3. Results and Discussion

### 3.1. Heavy metals in water

The mean concentrations of heavy metals viz. Cr, Mn, Cu, Cd, and Pb in water and comparisons with some reports in Ethiopia and worldwide are presented in Table 2 and Table 3. The mean concentration of Cr in water samples were in the range of 0.08-1.44 mg/L. High concentrations of Cr were obtained at Mojo (1.44 mg/L), Leather site (1.4 mg/L), Aba-Samuel (1.13 mg/L) and Hatebella (1.01 mg/L) sites, presumably as a result of the effects from industries' effluents (tannery and textile), municipal and agricultural waste leading to high concentration of Cr in these sites while lowest concentration of Cr was observed at Entoto site. The present study revealed that Cr at all sampling sites in water samples were higher than the



drinking water quality standard set by WHO (2004) and toxicity reference value (TRV) proposed by USEPA (1999) and USEPA (2002) as recommended values (Table 2). Among the analyzed heavy metals (Cr, Mn, Cu, Cd and Pb), the concentration of Mn in water were the highest and Pb was the lowest.

The concentration of Mn in water samples ranged from 0.14 to 3.4 mg/L. Highest concentrations of Mn were obtained at Hatebella (3.4 mg/L), Mojo (1.71 mg/L), Akaki (1.70 mg/L) and Aba-Samuel (1.4 mg/L) sites (Table 2). The concentration of Mn in water at these sites was higher than the maximum permissible limit for drinking water which is 0.5 mg/l, set by WHO (2004) and USEPA (2002) guidelines. The mean concentration of Cu ranged from 0.001 to 0.52 mg/l and a higher concentration of Cu was noted (0.91, 0.91, 0.9 and 0.85 mg/L) at Mojo, Akaki, Koka and Hatebella sites respectively, which are higher than the allowable limit set of standards set by USEPA (2002) and the toxicity reference values of USEPA (1999), but are lower than the permissible limit of drinking water set by WHO (2004). This indicates that the presence of Cu in the studied area is primarily due to water received from industrial and power plant effluents. Cu enters the aquatic environment through industrial effluents and also from river run offs domestic waters. The maximum amount of Cd was found (0.38 mg/L), which is above the tolerable limits of USEPA (2002), WHO (2004) and TRV of USEPA (1999). This may be because of the effluents from industries and urban waste (Ahmad et al., 2010). Highest concentration of Cd was recorded at Akaki (0.38 mg/l), Abasamuel (0.34 mg/L) and Mojo (0.31 mg/L) sites and the lowest concentration 0.01 mg/L was recorded at Entoto site.

This is significantly affected by the paint and textile industries and is associated with the discharge of sewage and agricultural runoff. The concentration of Pb was recorded between 0.001 and 0.10 mg/L. The highest concentration was 0.1 mg/L at Hatebela site, which exhibited the permissible limit set by standards of WHO (2004), TRV of USEPA (1999) and USEPA (2002). Almost all metals exceeded significantly the limits for safe water, indicating that water from this site is not safe for drinking or cooking at the time of sample collection.

**Table 2** Concentrations (Mean  $\pm$  SD) of heavy metals in Awash River water and standards

Sampling sites	Concentration of heavy metals (mg/L)				
	Cr	Mn	Cd	Cu	Pb
Afembo	0.273 $\pm$ 0.25	0.54 $\pm$ 0.52	0.13 $\pm$ 0.25	0.22 $\pm$ 0.21	0.02 $\pm$ 0.012
Mille	0.14 $\pm$ 0.09	0.70 $\pm$	0.034 $\pm$ 0.07	0.023 $\pm$ 0.03	0.022 $\pm$ 0.001
Adaytu	0.27 $\pm$ 0.21	0.52 $\pm$ 0.19	0.066 $\pm$ 0.14	0.04 $\pm$ 0.02	0.01 $\pm$ 0.01
Mekaworor	0.3 $\pm$ 0.08	0.61 $\pm$ 0.25	0.1 $\pm$ 0.12	0.55 $\pm$ 0.3	0.023 $\pm$ 0.012
Awash Park	0.13 $\pm$ 0.12	2.2 $\pm$ 3.9	0.06 $\pm$ 0.073	0.30 $\pm$ 0.42	0.011 $\pm$ 0.012
Metehara	0.194 $\pm$ 0.12	0.48 $\pm$ 0.25	0.062 $\pm$ 0.072	0.33 $\pm$ 0.30	0.021 $\pm$ 0.02
Sodere	0.38 $\pm$ 0.51	0.36 $\pm$ 0.25	0.22 $\pm$ 0.44	0.28 $\pm$ 0.30	0.012 $\pm$ 0.011
Koka	0.26 $\pm$ 0.29	0.54 $\pm$ 0.17	0.027 $\pm$ 0.02	0.9 $\pm$ 0.8	0.024 $\pm$ 0.021
Mojo	1.44 $\pm$ 1.3	1.71 $\pm$ 1.8	0.31 $\pm$ 0.63	0.91 $\pm$ 0.94	0.05 $\pm$ 0.05
Akaki	1.4 $\pm$ 1.3	1.7 $\pm$ 0.34	0.38 $\pm$ 0.63	0.91 $\pm$ 0.7	0.065 $\pm$ 0.04
Bole	0.46 $\pm$ 0.34	1.3 $\pm$ 0.34	0.15 $\pm$ 0.14	0.63 $\pm$ 0.73	0.05 $\pm$ 0.05
Aba-Samuel	1.13 $\pm$ 1.0	1.4 $\pm$ 1.03	0.34 $\pm$ 0.51	0.60 $\pm$ 0.67	0.052 $\pm$ 0.06
Hatebella	1.01 $\pm$ 1.5	3.4 $\pm$ 1.2	0.2 $\pm$ 0.18	0.85 $\pm$ 0.77	0.1 $\pm$ 0.05
Ginchi	0.16 $\pm$ 0.2	1.2 $\pm$ 0.97	0.022 $\pm$ 0.02	0.22 $\pm$ 0.45	0.003 $\pm$ 0.001
Asko	0.5 $\pm$ 0.57	0.7 $\pm$ 1.2	0.032 $\pm$ 0.034	0.14 $\pm$ 0.15	0.03 $\pm$ 0.04
Entoto	0.08 $\pm$ 0.11	0.14 $\pm$ 0.10	0.01 $\pm$ 0.01	0.012 $\pm$ 0.012	0.001 $\pm$ 0.001
Minimum	0.08	0.14	0.01	0.012	0.001
Maximum	1.44	3.4	0.38	0.91	0.10
Mean	0.504	1.1	0.132	0.43	0.03
WHO, 2004	0.005	0.50	0.003	2.00	0.01
TRV	0.011	NA	0.0022	0.009	0.0025
USEPA, 2002	0.10	0.50	0.01	0.05	0.05

Toxicity Reference Value (TRV) for fresh water proposed by USEPA (1999)

**Table 3** Comparison of the observed values (mg/L) of heavy metals in Awash River water with other reported values in Ethiopia and worldwide.

River	Cr	Mn	Cd	Cu	Pb	Reference
Awash River, Ethiopia	0.504	1.10	0.132	0.43	0.03	This study
Togona river, Ethiopia	0.0081	0.0970	0.0019	0.0433	0.0143	Ayenew and Ahmad (2016)
Rebu river, Ethiopia	-	0.23-0.82	-	0.05-0.08	0.07-0.16	Tadesse et al. (2018)
Lake Beseka, Ethiopia	BDL	0.075	0.054	-	0.631	Fuad and Gelaneh (2019)
Lake Hawasa, Ethiopia	1.1-2	-	0.1-1.2	-	0.60-1.4	Dsikowitzky et al. (2013)
Karnofuly River, Bangladesh	0.25	0.12	0.01	0.05	0.14	Islam et al. (2013)
Buriganga River, Bangladesh	0.59	-	0.009	0.163	0.07	Ahmad et al. (2010)

- Not measured

BDL- below detection limit

The mean concentrations of heavy metals were compared with previous studies on some rivers in Ethiopia and worldwide, including the Karnofuly river and Buriganga river in Bangladesh (Islam et al, 2013) and Ahmad et al (2010) respectively and Togona river (Ayenew and Ahmad, 2016), Rebu river (Tadesse et al., 2018), Lake Beseka (Fuad and Gelaneh, 2019), Lake Hawasa (Dsikowitzky et al., 2013) in Ethiopia (Table 3). The concentrations of Cr in this study are higher than the values obtained in Togona river and Lake Beseka, Ethiopia, indicating that there is pollution associated with industrial effluents. Likewise, Cr concentrations did not exceed the values in the Buriganga river, Bangladesh and Lake Hawasa, Ethiopia. The levels of Mn, Cd, Cu and Pb in the present study were higher than the reported values in Togona river, E Rebu river, Lake Beseka, Lake Hawasa, Ethiopia Karnofuly river, Buriganga river, Bangladesh, but Cd

detected in the Lake Hawasa and Pb detected in Togona river was comparatively higher against the value in this study. Agricultural runoff from agricultural fields, industrial, municipal and domestic wastewater in this research might be causing alarming levels of water contamination with Mn, Cd, Cu and Pb.

### 3.2. Heavy metal concentrations in sediment

The heavy metal concentrations in the river sediments at all sampling sites are given in Table 4. The mean concentrations of analyzed heavy metal were observed in sediments in the following decreasing order: Mn > Cr > Cu > Cd > Pb. The mean concentrations of Cr, Mn, Cu, Cd and Pb in sediments were 242.0, 3104.50, 80.23, 0.60, 25.61 mg/kg. The results showed that mean concentrations of Cd, Pb and Cu in the Awash river sediments from Afembo, Melkaworo and Metehara sites were found lower but Cr and Mn in all sampling sites, Cd, Cu and Pb in the other sites were found higher than WHO (2004) and USEPA (1999) Sediment Quality Guidelines (SQG) (Table 4 and 5).

This means that among all sampling sites, measured heavy metal concentrations were found to be higher in upper sampling sites than in lower basin sites of the Awash River. Mean concentrations of Cr and Mn at Hatebella, Mojo, Akaki and Aba-Samuel sites were much higher than USEPA (1999) and WHO (2004) sediment quality guidelines (Table 5).

**Table 4** Mean concentrations of heavy metals (mg/Kg) in sediment of Awash River

Sampling Sites	Cr	Mn	Cu	Cd	Pb
Afembo	149.80	2229.23	4.00	0.11	0.27
Melkaworor	159.60	1165.62	1.14	0.1	0.37
Metehara	125.70	660.00	0.71	0.48	3.72
Koka	142.03	3657.67	166.37	0.14	2.78
Mojo	356.0	4210.50	93.07	1.02	48.00
Leather	351.67	4231.67	153.13	0.72	55.01
Aba-Samuel	322.13	2550.33	104.48	0.73	39.83
Hateballa	328.83	6131.00	118.93	1.50	54.90
Minimum	142.03	660.00	0.71	0.08	0.30
Maximum	356.00	6131.00	166.40	1.50	55.01
Overall Mean	242.00	3104.50	80.23	0.60	25.61

A wide range of values for metal concentrations was observed among the sampling sites. The concentrations of heavy metals at Mojo, Abasamuel, Akaki and Hatebella sites were much higher than other sites indicating that in the upper stream of the river, anthropogenic activities might be driving heavy metal contamination in surface sediments. The activities (industrial discharges, municipal wastewater, household garbage, agricultural and urban runoff) at upper basins of the river are the main causes of higher metal concentrations at these sites. Among the sites in the current study, the highest concentration of Cr was observed in sediment collected from Mojo, Akaki, Abasamuel and Hatebella sites (356.0, 351.67, 322.13, 328.83mg/kg respectively) (Table 4). An elevated concentration of Cr was observed at Mojo site and the enhancement of chromium in sediments at this site could have been caused by anthropogenic: industrial activities such as tanneries and textile industries which are discharging Cr based oxidants (chromate, dichromate, etc.) and diffusing solid chrome waste deposited at the bank of Mojo river. Hence, the waste discharged from such industries was most probably responsible for elevated Cr level in the exposed sediment. The mean concentration of Mn in sediments was found in the range of 660-6131 mg/Kg (Table 4). The highest concentration of Mn was observed at Hatebella site (6131 mg/kg).

The slightly higher level of Mn was observed at this site located near to the district urban area, which indicates the higher input of Mn in sediment that might have originated from urban and industrial waste. The concentrations of Mn in sediments collected from Akaki and Mojo sites (4231.67 and 4210.50 mg/kg respectively), were also high. Higher concentrations of Cu indicate anthropogenic activities such as wide use of fertilizer and pesticide and agricultural runoff, and industrial and municipal wastewater discharge. The mean concentration of Cu in sediment ranged between 0.71 and 166.40 mg/kg (Table 4). The highest concentration of Cu was observed at Koka, Akaki, Abasamuel and Hatebella sites (166.40, 153.13, 104.48 and 118.93 mg/kg, respectively), where the highest level of Cu was found at Koka site. Higher concentrations of Cu indicate anthropogenic activities such as wide use of fertilizer and pesticide and agricultural runoff, and industrial and municipal wastewater discharge. In the present investigation, the mean concentration of Cd ranged from 0.08 to 1.50 mg/kg (Table 4). Among the sampling sites, the highest level of Cd was observed at Mojo, Abasamuel, and Hatebella sites (1.02, 0.73 and 1.50 mg/kg respectively). Higher Cd concentration in sediment of these sites might be related to industrial activity, atmospheric emission, and Cd plated items.

Among the sampling sites, the highest level of Pb was observed at Akaki, Hatebella and Mojo sites (55.01, 54.90 and 48 .00 mg/kg respectively) (Table 4). The highest level of Pb in sediments at Akaki and Hatebella sites can be due to municipal runoffs, atmospheric deposition and leaded gasoline, chemical manufacturing and paint works in urban areas of this site.

**Table 5** Comparison of heavy metals in sediments (mg/kg dw) with some reported values in Ethiopia and worldwide.

Study area / Standards	Cr	Mn	Cu	Cd	Pb	Reference
Awash River, Ethiopia	242.00	3104.5	80.23	0.60	25.61	This study
Akaki River, Ethiopian	109.51	NA	79.43	3.14	129.68	Mekuria et al. (2020)
Awash River, Ethiopia	120.58	NA	79.43	2.60	13.53	Bekele et al. (2018)
Okumeshi River, Nigeria	0.87	NA	NA	1.32	0.45	Raphael et al. (2011)
Gomti River, India	8.15	NA	NA	2.4	40	Singh et al. (2005)
Buriganga River, Bangladesh	297	NA	280	7.7	731	Islam et al. (2018)
Gediz River, Turkey	170– 220	NA	108– 152	NA	105–140	Akcay et al. (2003)
Lijiang River, China	56.38	NA	38.07	1.72	51.54	Xu et al. (2016)
Tigris River, Iraq	64.00	NA	5.70	1.20	46.30	Zhang et al. (2009)
Toxicity reference value (TRV)	26	0.5	16	0.6	31	USEPA (1999)
Average shale value (ASV)	90	850	45	0.3	20	Turekian and Wedepohl (1961)
Lowest effect level (LEL)	0.1	0.5	0.05	0.01	0.05	Persuad et al. (1993) and MacDonald et al. (2000)
Threshold effect level (TEL)	37	NA	36	0.59	35	MacDonald et al. (2000)
Continental upper crust (CUC)	92	NA	28	0.09	17	Rudnick and Gao (2003)

Severe effect Level (SEL)	110	NA	110	10	250	Persuad et al. (1993) and MacDonald et al. (2000)
Probable effect level (PEL)	90	NA	197	3.5	91	MacDonald et al. (2000)

NA- Not available

To predict the metal pollution in sediments of the studied river, the available data for a comparative analysis with background and toxicological reference values and some studied rivers in Ethiopia and worldwide sediment values are presented in Table 5. The heavy metal concentrations in sediments of the Awash River were compared with other rivers of Ethiopia and international report values (Table 5). It was noted that the mean concentration of Cr and Mn in the sediment samples of the present study exceeded the ASV, LEL, TEL, CUC, SEL and PEL values. The mean concentrations of Cr and Mn were also higher than those of the U.S. Environmental Protection Agency's (USEPA) toxicity reference values, lowest effect levels and threshold effect levels, suggesting the most probable association with adverse biological effects. The mean concentrations of heavy metals (Cr and Mn) in sediments of the studied river were higher than some other study rivers such as Akaki, Awash, Gediz, Lijiang, Tigris, Gomti, and Okumeshi rivers in Ethiopia and from other countries (Table 5). The results indicated that the levels of these heavy metals found in sediments of the studied river might create an adverse effect on the aquatic ecosystem, especially since it receives urban and industrial wastewater from the nearby study area.

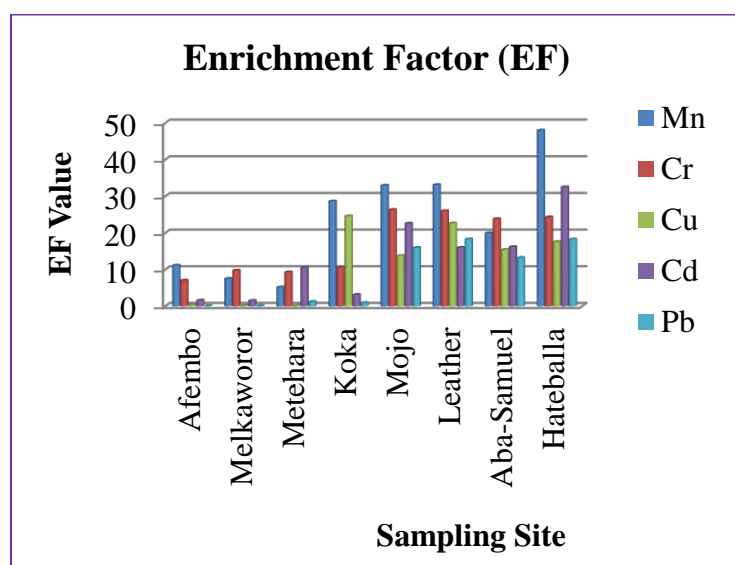
The mean concentrations of Cr and Cu as reported by Islam et al. (2018) in the Buriganga river were higher than the present investigation and Cu in this study was lower than the reported value in Gediz River, SEL and PEL values, but in present investigation the mean concentration of Cu in sediment of studied river was higher than the reported value in Akaki River, Okumeshi River, Gomti River, Lijiang River and Tigris River. The mean concentration of Cu in sediment of this study was also higher than the recommended value of ASV, TRV, LEL, TEL and CUC. The mean concentration of Cd in the present study exceeded the values of ASV, LEL, CUC and TEL, but lower than the reported value of TRV, SEL, PEL, and in rivers such as Akaki River, Awash River, Okumeshi River, Gomite River, Gediz River, Lijiang River and Tigris River. The mean concentration of Pb was found higher as recommended by Ahmed et al. (2012) in Awash River, Okumeshi River, ASV, LEL and CUC than present measured concentration and the mean concentration of



Pb was higher in the Akaki River, Gometi River, Gediz River, Lijiang River, Tigris River reported by Rahman et al. (2014) respectively, than this study. On the basis of the values of TRV, TEL, SEL and PEL, the mean concentration of Pb in the present study was lower.

### 3.3. Assessment of metal pollution in sediments

For the assessment of heavy metals contamination in sediments, an enrichment factor (EF) has been applied by several researchers (Han et al., 2006; Cevik et al., 2009; Bastami et al., 2014). The EF is a normalization technique that is being widely used to categorize the metal fractions that are associated with sediments (Huang et al., 2014). The calculated values of EF for each of the studied metals are presented in Figure 2. In summary, the mean EF values of all the studied metals suggest their enrichments in surface sediments of the Awash River. The EF values of heavy metals for most sites were higher than 1.5 suggesting that these metals might be delivered from non-crustal materials, or non-natural weathering processes (Gao and Chen, 2012; Islam et al., 2015a). The overall mean enrichment factor values of Mn, Cr, Cu, Cd, and Pb were 23.30, 17.12, 11.83, 13.01 and 8.51 respectively. All in all, the enrichment factors of all the studied metals at all sampling sites were in the descending order of Mn > Cr > Cd > Cu > Pb. The higher EF of Mn, Cr and Cd in sediments indicated higher enrichment of these metals in the studied river sediments, which might be due to anthropogenic input/human perturbation. It is presumed that high EF value indicates anthropogenic sources of heavy metals, mainly from activities such as industrialization, urbanization, deposition of industrial waste and other sources.



**Figure 2** Enrichment factor values of heavy metals in sediment samples of Awash River.

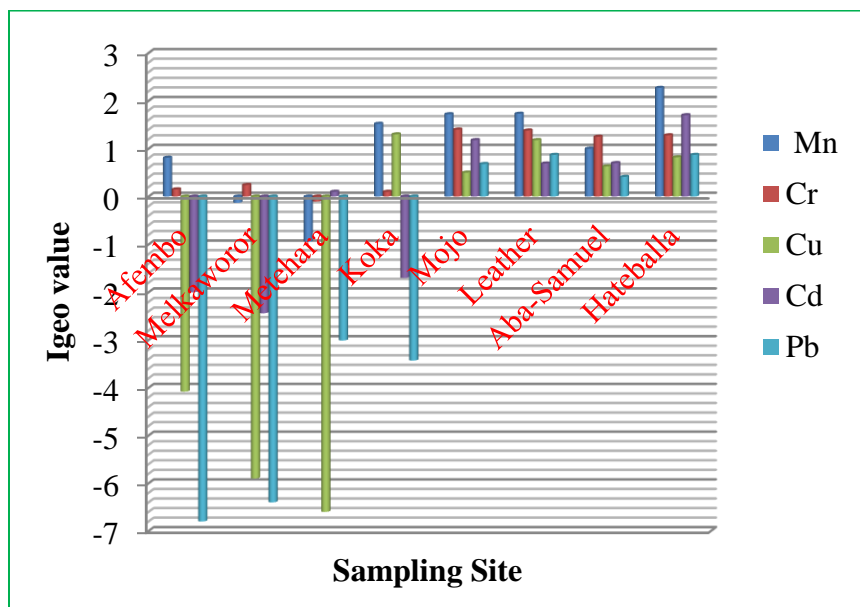
The contamination factor values of heavy metals in sediments are presented in table 6. The highest CFs for Cr (4), Mn (7.2), Cu (3.7) Cd (3.4) and Pb (2.75) were found in upstream sites (Mojo, Hatebella, Koka, Mojo and (Akaki and Hatebella) respectively) in which the possible sources of these heavy metals are domestic, municipal, agricultural runoff and industrial waste discharge from sewers (Table 6). The mean CF values indicate low contaminated ( $CF < 1$ ) to very high contaminated ( $CF > 6$ ) sites. Based on the results, average CF values for heavy metals were in the following decreased order:  $Mn > Cr > Cd > Cu > Pb$ . On the basis of degree of contamination (Cd), sampling sites at the upper stream of Awash River basin were categorized as low contaminated ( $DC < 5$ ) vs. very high contaminated ( $DC \geq 20$ ) (i.e. sampling sites at upper stream > middle stream > lower stream) (Table 1 and Table 6).

**Table 6** Contamination factors, degree of contamination, and contamination level of heavy metals in sediment samples of Awash River.

Sampling Site	Contamination factors (CF) of heavy metals					Degree of contamination (DC)	Contamination level
	Cr	Mn	Cu	Cd	Pb		
Afembo	1.7	2.6	0.1	0.38	0.014	4.79	Low contamination
Melkaworor	1.78	1.4	0.03	0.3	0.02	3.53	Low contamination
Metehara	1.4	0.8	0.02	1.6	0.185	4.01	Low contamination
Koka	1.6	4.3	3.7	0.47	0.14	10.21	Considerable contamination
Mojo	4	4.95	2.1	3.4	2.4	16.85	Considerable contamination
Leather	3.9	5	3.4	2.4	2.75	17.45	Considerable contamination
Aba-Samuel	3.6	3	2.3	2.43	2.00	13.33	Considerable contamination
Hateballa	3.7	7.2	2.64	5	2.75	21.30	Very high contamination



The geoaccumulation index ( $I_{geo}$ ) values of this study showed the following decreasing order: Mn > Cr > Cd > Cu > Pb (figure 3). The highest  $I_{geo}$  values obtained for Mn, Cr, Cd, Cu and Pb were (2.27 at Hatebella, 1.4 at Mojo, 1.7 at Hatebella, 1.3 at Koka, and 0.87 at Akaki and Hatebella respectively) and indicated moderately contaminated to uncontaminated sediments in the study area. This might have happened due to higher concentration in sediment and lower geochemical background values resulting in higher  $I_{geo}$  values of metals. The highest values of Mn, Cr, Cd and Cu might be due to contributions from anthropogenic perturbation. Overall the  $I_{geo}$  values for all studied metals indicated uncontaminated (lower stream of the river) to moderately contaminate (upper stream of the river) of the sediments. The pollution load index values of heavy metals in sediments are presented in Figure 4. Pollution load index (PLI) value equal to zero indicates perfection; value of one indicates the presence of only baseline level of pollutants and values above one indicate progressive deterioration of the site and estuarine quality (Mohiuddin et al., 2011; Suresh et al., 2011).



**Figure 3**  $I_{geo}$  values of heavy metals in sediments of Awash River

The PLI can provide some understanding about the quality of the aquatic environment that affects the inhabitants (Ali et al., 2016). Additionally, it provides valuable information about the pollution status of the study area, which helps decision-makers take any decision (Suresh et al., 2011). The values of PLI ranged from 0.30 to 4.0, which confirmed that the sediment of the studied river was contaminated (PLI > 1). Among the studied metals, the higher PLI values indicated that Mn, Cr and Cd are the major



contributors to the sediment pollution. The highest pollution load index values were noted in sampling sites of Hatebella (PLI = 4.0), Akaki (PLI = 3.4), Mojo (PLI = 3.21), and Aba-Samuel (PLI = 2.61), which might be due to the effects of various industries and municipal waste.

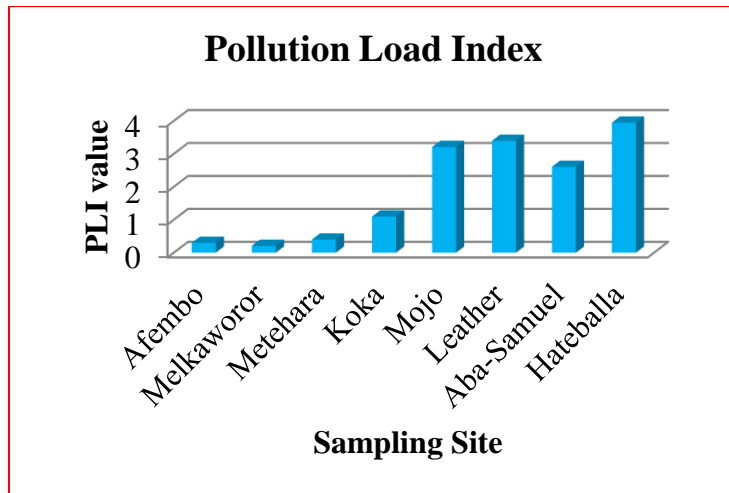


Figure 4 Pollution load index of heavy metals in sediments of Awash River

### 3.4. Ecological risk assessment

Hakanson (1980) developed a methodology to assess ecological risks for aquatic pollution control. The methodology is based on the assumption that the sensitivity of the aquatic system depends on its productivity. The ecological risk factor for individual element (Er) and the potential ecological risk index (PER) are presented in Table 7. The potential ecological risk factor of heavy metals in sediment were in the descending order: Cd > Cu > Pb > Cr > Mn. Considering the potential ecological risk factor (Er) for the individual element, Cd showed very high potential ecological risk with the Er factor ranging from 9 to 167.4 (Table 7). The sites Mojo, Hatebella, Abasamuel and Akaki showed moderate risk to very high risk for Cd which might be due to the application of phosphate fertilizers to agricultural fields beside the river, dumping of oily materials from water vehicles, and waste disposal from the town and industries. The potential ecological risk index (PER) in the sampling sites ranged from 17.97 to 264.85, which indicates low to very high ecological risk (Table 7).

**Table 7** Potential ecological risk factors (Er) and potential ecological risk indexes (PER) of heavy metals in sediments collected from Awash River.

Sampling site	Potential ecological risk factors (Er)					Potential Risk index (PER)	Degree of Pollution
	Cr	Mn	Cu	Cd	Pb		
Afembo	3.4	2.6	0.5	11.4	0.07	17.97	Low risk
Melkaworor	4.0	11.4	15	9	0.1	39.5	Low risk
Metehara	2.8	8	0.1	48	1	59.9	Low risk
Koka	6.2	14.3	28.5	14.1	0.7	63.8	Moderate risk
Mojo	42.0	16.95	24.5	167.4	14	264.85	Very high risk
Akaki	7.8	5	17	72	13.73	115.53	Moderate risk
Aba-Samuel	9.2	13	21.5	82.9	13.5	140.1	Moderate risk
Hateballa	24.4	27.2	13.2	154.08	13.73	232.61	Considerable risk

### 3.5. Conclusions

The results showed that the concentration of seven heavy metals (Cr, Mn, Cu, Cd and Pb) in water samples, are higher than the WHO and USEPA recommended guidelines. The mean concentration of heavy metals in sediments of Awash River also were in the following decreasing order: Mn > Cr > Cu > Cd > Pb. This suggests that effluents were discharged from industries, irrigation, municipal and domestic waste. Most of the metals exceeded the permissible limits set by the standards.

The enrichment factor, contamination factor, geoaccumulation index, pollution load index, ecological risk and potential ecological risk showed low to high contamination of sediments by the studied heavy metals. Therefore, further studies are needed on metal speciation and effects on metal uptake by humans and organisms.

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### Disclosure statement

The authors declare that there are no conflicts of interest.

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