Little Akaki River Sediment Enrichment with Heavy Metals, Pollution Load and Potential Ecological Risks in Downstream, Addis Ababa, Ethiopia.

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ABSTRACT

Background: Little Akaki River (LAR) passes through Addis Ababa City, receives inorganic and organic pollutants from various sources. The objective of this study was to investigate the pollution level of LAR by selected heavy metals and evaluate sediment quality using contamination indices.

Methods: sediment samples were collected from 10 stations along LAR, processed, digested and heavy metal content was analyzed using ICP-OES. Enrichment factor (EF), geo-accumulation index (Igeo), contamination factor (CF), pollution load index (PLI), ecological risk index (RI) were determined. Comparison was made with standard sediment qualities (SQGs) to evaluate ecological and toxicological implication.

Results: the mean concentrations of heavy metals in LAR sediment were: Zn (78.96-235.2 mg / kg); Cr (2.19-440.8 mg / kg); Cd (2.09-4.16 mg / kg) and Pb (30.92-596.4 mg / kg). EF values indicated that LAR sediments were moderate to significant enrichment with Zn and Cr; moderate to very high enrichment with Pb, and very high enrichment in all sampled sites with Cd. Igeo and CF values indicated that the sediments were moderate to very high contamination with toxic Cd and Pb.
PLI and hierarchal cluster analysis revealed that highest pollution load occurred at sampling site (S9), in lower course of the river mainly due to anthropogenic metals inputs from industrial wastes, municipal wastewater treatment plant and agrochemical wastes; hence, its quality was deteriorated and depicted polluted site. The decreasing order of PLI in downstream was: (S9) > (S4) > (S8) > (S3) > (S6) > (S10) > (S5) > (S2) > (S7) > (S1). Pearson correlation indicated that Zn and Cd were generated from common sources of pollution. The ecological risk (RI =350.62) suggested that the contaminated LAR sediment can pose considerable ecological risks of pollution.

Conclusions: The concentrations of Zn, Cr, Cd and Pb in LAR sediment were surpassed sediment quality guidelines (USEPA) and eco-toxicological guideline limit values of USEPA (TEC) and CMME (ISQGs). Thus, the contaminated sediments can occasionally pose adverse biological effects on sediment dwelling organisms and impairs the quality of river water. Thus, monitoring and addressing sediment contamination becomes necessary to sustain beneficial uses of river water for various development purposes.

Key words: Contamination Factor, Enrichment Factor, Geo-accumulation Index, Potential Ecological Risk Index, and Pollution Load Index.

1. Background

Heavy metals released from industries, municipal waste treatment plant sites, domestic and garages into surrounding surface water bodies impair the quality of water and sediment. In surface water, trace metals are carried away by water in suspended form and eventually deposited in sediment through the process of precipitation, co-precipitation, adsorption and
chelation (Rabee et al., 2011; Singovszka and Balintova, 2016). Thus, sediments act as a sink for metals. However, the metal retention capacity of a sediment is subjected to many environmental factors such as natural and anthropogenic disturbances of the river water and sediment, change in water pH and redox potential (Akan et al., 2010; Edokpayi et al., 2016). When change in environmental factors occurred, sediment bound metals are released into overlaying water through the process of dissolution of metals, decomposition of organic matters and desorption (Zhu et al., 2017), and causing the secondary source pollution to river water (Luo et al., 2010; Ren et al., 2015).

Sediment pollution with heavy metals becomes important local, national and global problem that affect water quality, aquatic life and resulting far-reaching environmental and public health implications. This is mainly attributed to the properties of heavy metals which include: persistence in environment, bioaccumulation, and bio-magnification along the food chain. Some of the trace metals like Cd and Pb are non-essential elements to plants and animals; can cause various health repercussions. Hence, assessing the levels of sediment contamination with heavy metals, assessment of ecological risks and predicting eco-toxicological adverse effects to aquatic life have significant importance to design appropriate mitigation measures that ensure healthy aquatic ecosystem.

LAR is crossing through the capital city, Addis Ababa, and pre-urban areas where several industries were established, large number of city populations were residing and wide spread use of agrochemicals for irrigation. It receives poorly treated and untreated industrial, domestic and agricultural wastes into river and streams (Aschale et al., 2019; Melaku, 2005) that carry heavy metals to the river sediment.
In the past, studies on LAR sediments were limited to assess the concentrations and distribution
of some selected heavy metals (Gizaw, 2018; Melaku, 2005; Nigussie et al., 2013; Prassie et al.,
2012; Tolla, 2006), and their distribution and ecological risk in the sediments of akaki catchment
(Berhanu et al, 2018). Some of these studies were based on few samples with limited use of
sediment quality indices. The concentration of heavy metals in LAR sediments were not
adequately expressed using various sediment quality indices such as enrichment factor (EF), geo-
accumulation index (Igeo), contamination factor (CF), pollution load index (PLI) and potential
ecological risk index (PERI) and sediment quality standards. The sediment pH, particle size and
textural composition, which influence the accumulation of heavy metals in sediment and
physical status of the sediment, were not properly investigated. An attempt was no made to
identify heavy metal loaded areas, and polluted and unpolluted sites. More importantly, up to
now, there have been no exhaustive study on the potential ecological risk and eco-toxicological
implications of toxic metals accumulation in sediments. Hence, this study is intended to fill some
of these gaps and give full insight about the quality of sediment and its pollution profile.

Thus, the objectives of the study were: (i) to determine the level of selected heavy metals (Zn,
Cr, Cd and Pb) in LAR sediment; (ii) to evaluate sediment contamination using various quality
indices such as EF, Igeo, CF and PLI, and (iii) to assess the potential ecological and eco-
toxicological risk of sediment pollution and its implications for aquatic life and river ecosystem.

2. Materials and Methods

Description of the study area

The study was conducted on LAR sediment. The river starts from Geferssa Reservoir which is
located at foot of Entoto Mountain, and flows through Addis Ababa City, the capital city of
Ethiopia, and finally joins Aba-Samuel Reservoir (Fig. 1). The city is located at 9° 2’ N and 38°
42’ E. LAR flows through varied altitudes that range from 2464 m.a.s.l. at around Geferssa
Reservoir in the north to 2048 m.a.s.l. at the merge to Aba-Samuel Reservoir in the south. The river drains a total catchment area of about 540 km² (Kebede et al., 2013). In the upper catchment, the river flows through deep gorge, on rocky bed with turbulences where as in the lower catchment, it flows in a gentle slope landscape that surrounded by irrigation farm and grazing lands. There are two types of soil dominantly found around LAR: vertisol which is commonly found on top of gentle slope lands and fluvisol at bottom of slope lands and on adjacent to the Akaki River banks (Itanna et al., 2003).

LAR is one of the major rivers crossing through the city and most used for socio-economic developments. Urban and pre-urban farmers are using this pollutants loaded river water for irrigation to grow varieties of vegetables around the river banks and supply the City with fresh vegetables. Moreover, pre-urban communities are largely dependant on the river water for drinking and domestic uses, cattle drinking, washing cattle, and sand mining during dry season. Fishing is also undertaken in the lower parts of the river course and in Aba-Samuel Reservoir.
Sampling site selection and sample collection

A preliminary reconnaissance survey was undertaken to select sampling points along LAR and thus, ten sampling stations were selected based on: accessibility, nearness to point source of pollution such as industrial and municipal waste discharge point, non-point sources (irrigation farms), permanently identifiable features and purpose of the study. Accordingly, samples were collected at the following sampling sites: Gefersa Reservoir (S1- control sample), Soramba, just after merge of Burayu and Wongate stream (S2), Kolfe Bridge (S3), below Kera bridge in proxy to Addis Ababa City abattoir (S4), below Mekenissa bridge, where large irrigation practiced (S5), below Gofa bridge (S6), Bihire-Tsige vegetable farm area (S7), in proxy to Akaki Kalti.
industrial area (S8), below Gelan Guda Kebele Bridge at the middle of irrigated farm land (S9), and Aba Samuel Reservoir, at merge of LAR and the reservoir (S10) (see; Fig.1). The exact Geographical location and altitude of each sample station recoded using GPS (GARMIN, GPSMAP62st).

Sediment samples were collected from these selected stations from April 6-9, 2018 between 9:00-11:00 AM, during the dry season when river flow was minimal, following the procedures described in USEPA (2001). At each sampling station, three grab sediment samples of nearly the same amount were randomly collected using clean plastic scoop (grasp sampling technique) from depth of 0-10 cm, starting from most downstream sample along straight section of the river with least disturbance. The grab samples were thoroughly mixed to form a homogenized composite sediment samples. At each station, the physical status of the sediment was noted based on OhioEPA( 2001). The sediment samples of 1500-2000 gm per site were placed in dense polyethylene bags, sealed, labeled and immediately transported to Addis Ababa University, Center for Environmental Science laboratory and kept the sediment in refrigerator at 4 C° until they were farther processed.

**Determination of pH and particle size composition**

Following the procedures described in Mohiuddin et al., (2010), the sediment and water were mixed at ratio of 1:2.5, thoroughly stirred for 30 min. and suspension was kept to stay overnight. The pH of sediment was measured using pH-16, Bench pH meter, Model PHS-3CB ACC-Deg-0.01.

The particle size compositions of the sediments were determined following the procedures described in Ozkan (2012). The sediment particle sizes distribution was grouped into four textures classes on the basis of the sieve result as: Clay<0.002mm; Silt = 0.002-0.063mm; Sand= 0.063-2mm; and Gravel >2mm (Hu et al., 2013). Each sieve result was carefully collected and weighted using electronic balance (Model: JD210-4 CE).
The percent of grain size (%) was computed using the formula described in Uwah et al. (2013), which is expressed as: \[ \text{% Grain Size} = \left( \frac{\text{Sieve weight}}{\text{total weight}} \right) \times 100. \] The sediment composition/texture classes were determined based on ternary diagram of flok’s classification.

**Pretreatment and digestion of sediment samples for heavy meal analysis**

Unwanted materials such as leaves, debris, shells and coarse gravels were carefully removed and then, sediments samples were air dried at ambient room temperature. The dried sediment samples were powdered using mortar and pestle and mixed; sieved using 45\(\mu\)m sieve. Following the procedures described in Sekaberia et al. (2010), 1.25 g of subsample of sediment were taken from each sample and digested with 20 mL aqua regia (3:1 HCl/HNO\(_3\)) and then, with 5 mL H\(_2\)O\(_2\) in open beaker using heat plate until the digest reach near dryness. The beaker was rinsed with 10 mL of de-ionized water and the samples were farther digested with 5 mL HCl to near dryness. Finally, the digest were cooled and the beaker was rinsed with 50 mL de-ionized water and were transferred into a small flask. The concentration of heavy metals (Cd, Cr, Pb and Zn) in the sediment samples was determined using inductively coupled plasma optical emission spectrometry (ICP-OES Arcos Spectrophotometer, made in Germany).

**ICP-OES operating conditions and calibration of the instrument**

All the measuring conditions were configured as follows: plasma power (1400W), average plasma flow rate (6.41 L/min.), pumping speed (30 rpm), nebulizer flow (0.73 L/min.), nebulizer pressure (1.96 bar), Argon pressure (6.75 bar), and torch positions and measuring time adjusted. The calibration and standardization of the spectra method was performed according to the standard protocols set for the instrument. But, standardization is undertaken daily: it is a quick procedure for correcting measuring intensities so that the correct concentrations of element is obtained using the original calibration curve. Calibration curves were prepared using 0.06, 0.11,
0.17, 0.56, 1.12, 1.68, 2.24 and 2.80 mg / L of Zn; 0.03, 0.06, 0.08, 0.28, 0.56, 0.84,1.12, and 1.40 mg / L of Cr, Cd and Pb. Quantifications of the elements were recorded at 213.856, 231.604, 267.716 and 220.353 nm, which correspond to the most sensitive emission wave-lengths of Zn, Cd, Cr and Pb, respectively. The sample was nebulize and the concentration was calculated on the linear graph of the standard concentration and the corresponding intensities. The cullibration curve showed linearity, $R^2$ of 0.999964 for Cd, 0.999874 for Cr, 0.999757 for Pb and 0.999439 for Zn. Thus, There is good correlation between concentration and emission intensities of the analysed elements.

**Assessment of levels of sediment contamination**

In order to assess the levels of LAR sediment contamination with heavy metals, quantities indices were employed. These include: EF, ($I_{geo}$), CF, PLI, PERI and IR

**Enrichment factor (EF)**

The EF is often used to assess natural and anthropogenic sources of trace metals and status of sediment contaminations (Zhao et al, 2017). EF was determined using formula described in (Issan and Qanber, 2016) which is expressed as:

$$Enrichment\ factor = \frac{(C_x/Fe)_{samples}}{(C_x/Fe)_{background\ value}}$$ (1)

Where, “$C_x$” stands for concentration of metal in sample sediment, and “Fe” concentration of iron in a given sample sediment. The element “Fe” was taken as a normalizing element, because, its abundance in the earth’s crust has not been much influenced by anthropogenic activities (Al Obaidy, et al., 2014). For geochemical background value, the world average shale value for elements were adopted from Turekian and Wedepohl (1961). According to Issa and Qanbar,
(2016), the resulting EF value can be categorized into five classes. These are (i) category-1: EF < 2, indicates; deficiency to minimal level of enrichment, (ii) category-2: 2 ≤ EF < 5; moderate enrichment, (iii) category-3: 5 ≤ EF < 20; significant enrichment, (iv) category-4: 20 ≤ EF < 40; very high enrichment, (v) category-5: EF ≥ 40; extremely high enrichment.

**Geo-accumulation index (I\textsubscript{geo})**-

This index was employed to evaluate the magnitude of sediment contamination (Rubio et al., 2000). Geo-accumulation index was calculated (Banu et al., 2013) as follows:

\[ I_{\text{geo}} = \log_2 \left[ \frac{C_n}{1.5 B_n} \right] \tag{2} \]

Where, “C\textsubscript{n}” represents the concentration of heavy metal in sample sediment, “B\textsubscript{n}” stands for the world average shale value of metal element “n”, while the factor 1.5 was applied for correction of background matrix attributed to lithogenic variations (Ke et al., 2017; Martin et al., 2012).

According to Muller (1981), the computed \( I_{\text{geo}} \) value can be categorized into seven classes, showing level of pollution as follows: class-0: \( I_{\text{geo}} \) value ≤ 0, unpolluted; class-1: \( I_{\text{geo}} \) value = 0-1, unpolluted to moderately polluted; class-2: \( I_{\text{geo}} \) value = 1-2, moderately polluted; class-3: \( I_{\text{geo}} \) value = 2-3, moderately to strongly polluted; class-4: \( I_{\text{geo}} \) value = 3-4, strongly polluted; class-5: \( I_{\text{geo}} \) value = 4-5, strongly to extremely polluted; class-6: \( I_{\text{geo}} \) value > 6, extremely polluted.

**Contamination factor (CF)**

Contamination factor is commonly used to demonstrate the level of contamination of sediment by particular toxic metal at a given sample site (Manoj and Padhy, 2014). It is defined as:

\[ \text{CF} = \frac{C_{m\text{ sample}}}{C_{m\text{ background}}} \tag{3} \]

Where, “\( C_{m\text{ sample}} \)” is stands for metal concentration in sample sediment; “\( C_{m\text{ background}} \)” is the geochemical background value of the metal.
According to Hakanson (1980), the levels of CF can be categorized as follows: (i) class-1: CF value <1, indicates low level of sediment contamination, (ii) class-2: 1 ≤ CF value <3, indicates moderate contamination, (iii) class-3: 3 ≤ CF value <6, indicates considerable contamination, (iv) class-4: CF value > 6, indicates very high contamination.

**Pollution load index (PLI)** - is an important index to compare the pollution status of different sampling sites in downstream (Harikumar and Jisha, 2010; Rabee et al, 2011). Pollution load index (PLI) of LAR was determined using the formula described in Rabee et al, (2011) and Tomlinson et al (1980) which is expressed as:

\[
\text{PLI} = (\text{CF}_1 \times \text{CF}_2 \times \text{CF}_3 \times \ldots \times \text{CF}_n)^{1/n}
\]

Where, “CF1, CF2, CFn”, stands for contamination factor of each element, “n” = number of metals under study. According to Tomlinson et al, (1980) sediment is considered to be polluted if PLI value >1; otherwise, not polluted for PLI value <1.

**Potential ecological risk index (PERI) and risk index (RI)**

PERI and RI were used to assess an overall potential ecological risk of heavy metals in sediment, pollution status and eco-toxicology aspect (Hakanson 1980; Ke et al, 2017). To compute PERI and RI, toxicity response factor (TRF) value for Pb = 5; Cd = 30; Cr=2; Zn = 1, were adopted from; Banu et al (2013); Li, (2014), and Suresh et al, (2011).

**PERI of metal element (E_r) = T_r x (C_r/C_o)**

\[
\text{Risk Index (RI)} = \sum_{i=1}^{n} (E_r^i)
\]

Where, “C_r” stands for the concentration of metal in sample sediment, “C_o” represents background concentration, “T_r” toxicity response factor of single element, “E_r” potential ecological risk of each metal element under the study.

According to Hakanson (1980), the PERI value indicating the severity ecological risk of sediment pollution can be grouped into five classes: class-1: PERI value < 40, indicates low pollution risk; class-2: PERI value between = 40-80, moderate risk; class-3: PERI value between
class-4: PERI value between= 160-320, high risk; and class-5: PERI value >320, very high risk of pollution. Similarly, the computed value of RI are categorized into four classes as: class-1: RI value <150, low risk; class-2: RI value between 150-300, moderate risk; class-3: RI value between 300- 600, considerable risk; class-4: RI value >600, high risk.

**Assessment of eco-toxicological effects**

To evaluate toxicological adverse effects of contaminated sediments on aquatic life and ecosystem, the concentration of heavy metals in the sediments were assessed in relation (TEC and PEC) of USEPA (2002) and the Canadian Council of Ministers of the Environment (CCME) (2001)(TEC, PEC and ISQG)

**Quality control and quality assurance**

In order to ensure the quality of heavy metals analysis, the sediment samples were analyzed in triplicate. Blank analyses were carried out to check interference from the laboratory reagents, to validate the method; spiked blanks were prepared to ascertain laboratory performance. The recovery rates of the four metals were: 96.25-105.59% for Zn; 89.38-112.86% for Cr; 97.93-116.43% for Cd, and 99.74-108.75% for Pb. All of the chemical reagents used for the test were of analytical grade (supplied by Manufactured by Loba Chemie-Laboratory Reagents and Fine Chemicals, Laboratory use only) and laboratory grade Argon gas of 99.996% was applied. The lab equipment used for the test, thoroughly washed, soaked in with 10% HNO₃ and rinsing with de-ionized water.

**Statistical Analysis**

Descriptive statistical analysis was run to determine the concentration of metals, EF, Igeo, CF, PLI and PERI, and the results were presented in tables and graphs using Microsoft Excel 2010. Pearson correlation and multivariate (Hierarchal cluster analysis, (HCA) applied to evaluate sources of metal elements and to group sample sites that exhibiting similar pollution profile in
Hierarchal cluster analysis was undertaken using R- software (Version 3.3.2). Pearson correlation was determined using SPSS software, Version 20.

3. Results and Discussions

3.1 PH and particle composition of LAR sediment

The accumulation of heavy metals in sediment can be influenced by sediment pH and particle size composition (Ohio EPA, 2001). The pH of LAR sediment samples range from 6.04-8.19 with mean value of 7.56. The lowest pH value (6.04) occurred at sample site (S1) which showed slightly acidic sediment may be due to decomposition of organic matters such as grass and plant derived debris and form humic-acid (Chatterjee et al. 2007). The highest value of pH (pH = 8.19) was recorded for sample site (S8) indicating slight alkalinity (Fig-2). This may attributed mainly to the deposition of salt and alkaline materials in sediment that released from such as painting, textile, tanneries and alcohol factories (WWAP, 2017) established in proximity to the river.

![Fig.2: The pH of LAR sediment](image)

The textural compositions of LAR sediment were grouped into three categories: clay, sand-clay-loam and sandy-loam (Table 1; and Figure 3). The clay texture dominated sediment with a certain portion of sand and silt, was recorded for sample sites: (S1), which are located in the upper and (S9) and (S10) in the lower parts of the river course. Flat landscape and relatively
slow river flow condition in the most upper course and lower course of the river might have facilitated the deposition of clay particles from the surrounding farm lands.

Sandy-clay-loam textured sediment composition detected at sample site (S3); while sandy-loam texture sediment composition was occurred at sample sites: (S4), (S5), (S6) and (S7). These sites were located in the middle course of the river which is typically characterized by rocky river bed, turbulent and fast moving river water with high materials transporting capacity. Thus, sand dominated sediment texture exhibits the effects of hydrology (Zhao et al., 2017) and geological condition of the sample sites. In this regards, Chatterjee et al., (2007) and Rubio et al (2000) have described that varying mixture of sand, silt and clay fraction in the sediment is a result of eroding and materials transport capacity of river water.

Table-1: LAR sediment texture class and physical status of the sediment

| Sample sites | Textural class of sediment | Physical status of sediment * |
|--------------|----------------------------|------------------------------|
| S1           | Clay                       | Plastic and cohesive; sediment contains un-decomposed leaves and plant debris, |
| S2           | ND                         |                              |
| S3           | Sandy-clay-loam            | Sandy; muck (black, consist of completely mineralized organic materials), |
| S4           | Sandy-loam                 | Sandy; muck (black, consist of completely mineralized organic materials); has odor, |
| S5           | Sandy-loam                 | Sandy; muck (black, consist of completely mineralized organic materials), |
| S6           | Sandy-loam                 | Sandy; peat (partially decomposed plant materials) sometimes soil warm visible, |
| S7           | Sandy-loam                 | Sandy; debris (dead and unconsolidated organic materials, and partially decayed coarse plant materials) visible, |
| S8           | ND                         |                              |
| S9           | Clay                       | Muck (black, extremely fine, completely decomposed organic materials); cohesive; has odor, |
| S10          | Clay                       | Muck (black, extremely fine, completely decomposed organic materials); cohesive; has odor, |

*Physical status of sediment described based on field observation notes as indicated in Ohio EPA (2001)
3.2 Concentration of heavy metals in LAR sediment

The concentration of trace metals in LAR sediment samples presented in Table-2. The concentrations of trace metals elements in the LAR sediment samples were ranged: Zn = 78.96-235.2 mg / kg; Cr = 2.19- 440.8 mg / kg; Cd=2.09- 4.16 mg / kg; and Pb = 30.92- 596.40 mg / kg.

The mean concentration of trace elements varied across different sampling sites, reflecting sources and amount of metal inputs. The lowest mean concentration of Zn recorded at sampling site (S7), Cr at (S1), Cd at (S5) and Pb at (S3) indicating low anthropogenic inputs of these trace metals. The highest mean concentration of Zn recorded at sampling site (S9) which may attributed to use of agrochemicals, influx wastewater released from Kaliti industrial site and municipal waste treatment plant. Whereas, the highest concentration of Cr occurred at (S3) due to discharge of the tannery wastewater into river from Addis Ababa Tannery and Dire Tannery. The highest concentration of Cd at (S1) in sediment may originate from agrochemicals (Klake et
Detection of high concentration of Pb at sample site (S4) was mainly associated with wastewaters discharged from vehicle batter maintenance shops, garages and surface run-off from fuel-filling stations. In this respect, Akan et al., (2010) reported that vehicle batteries, solder, pigments, rust inhibitors, vehicle emission are important sources of lead.

To evaluate the levels of trace metal accumulation in LAR sediment, a comparison made with that of sediment quality guidelines and world river sediment average value (Table-2) A comparison made with that of sediment quality guideline provided by USEPA SQGs (Gisy and Hoke, 1990), the concentration values of Zn in eight sampled sites, Cr and Pb in three sampled sites, fall in category of moderately polluted. In all sampled sites, the concentrations of Cd were below the USEPA SQGs, category of heavily pollution (<6). The concentration of heavy metal that exceeded the SQG standard has a potential risk for aquatic organisms (Xia et al., 2018). A comparsion made with that of the world river sediment average value indicated in Martin and Meybeck(1979) (See Table-2), revealed that concentrations of Cr in three sampled sites(S3, S8, and S10) and Pb at two sampled sites (S4 and S6) were exceeded the World River Sediment Average Value. This implies that the sediments at these sites were highly enriched, may be due to varying amount of local inputs, sources and other environmental factors influencing metal concentration (Qian et al., 2015). The concentrations of Cd in the sediment at all samples sites surpass the World River Sediment Average Value, exhibiting elevated concentration of Cd in LAR sediment.

To understand the level of pollution, a comparison has also been made with other studies in the country and other developing countries’ and presented in Table 3. The overall mean concentration for Zn in LAR sediment was almost comparable to that of the concentration
reported for Pearl River in China (Zhao et al., 2017) and Tigris River in Iraq (Al Obaidy et al., 2014). The concentration of Cr in LAR sediment samples is also comparable to that of Burigangan (Bangladesh) and Tigris River sediments (Saha and Hossian, 2011). However, the concentration of Cd in LAR sediment samples were slightly higher than that of Awash River sediments in Ethiopia (Bekele et al., 2018), but by far less than Wen-Rui Tang River sediment in China (Xia et al., 2018). The concentration of Pb in LAR sediment samples were also higher than the reported concentration values for Awash River and Wen-Rui Tang rivers, indicating that concentrations of Pb in LAR sediment was influenced by anthropogenic sources.

In general, the comparison made indicated that the concentrations of Cd and Pb in LAR sediments were relatively higher and require serious attention.

Table 2: Shows mean concentration and standard deviation of trace metals in LAR sediment

| Sample site code | Mean and standard deviation of heavy metal concentration in sediment (mg/kg) |
|------------------|--------------------------------------------------------------------------|
|                  | Zn       | Cr        | Cd        | Pb        |
| S1               | 82.24± 0.013 | 2.19± 0.014 | 4.160± 0.0001 | 37.44± 0.012 |
| S2               | 150± 0.001 | 30.72± 0.001 | 4.12± 0.001 | 41.48± 0.008 |
| S3               | 167.2± 0.001 | 440.80± 0.003 | 3.04± 0.001 | 30.92± 0.018 |
| S4               | 159.6± 0.001 | 56.6± 0.001 | 2.92± 0.001 | 596.4± 0.066 |
| S5               | 170.49± 0.003 | 52.17± 0.002 | 2.09± 0.001 | 61.15± 0.041 |
| S6               | 92.84± 0.024 | 48.84± 0.009 | 2.60± 0.001 | 372.4± 0.011 |
| S7               | 78.96± 0.021 | 36.6± 0.008 | 2.44± 0.001 | 31.12± 0.014 |
| S8               | 228± 0.001 | 174.8± 0.0001 | 4.04± 0.001 | 46.28± 0.009 |
| S9               | 235.2± 0.001 | 5.92± 0.004 | 3.0± 0.001 | 43.32± 0.011 |
| S10              | 118.64± 0.009 | 246.4± 0.001 | 2.96± 0.001 | 36.24± 0.011 |
| Min.             | 78.96 | 2.19 | 2.09 | 30.92 |
| Max.             | 235.2 | 440.8 | 4.16 | 596.4 |
| Range            | 78.96-235.2 | 2.19-440.8 | 2.09-4.16 | 30.92-596.40 |
| Overall Mean     | 148.28 | 109.51 | 3.14 | 129.68 |

USEPA SQGs
- Non-polluted: < 90, < 25, - , < 40
Moderately polluted  90-200  25-50  -  40-60
- Heavily polluted  >200  >50  >6  >60

World River Sediment
Average Value  303  126  1.4  230.75

Notes: USEPA SQGs (Giesy and Hoke, 1990); World River Sediment Average Value adopted from (Martin and Meybeck, 1979).

Table 3: Average concentration of heavy metal (mg/kg) in LAR sediment and other rivers

| Country | Concentration of heavy metals in Sediment | References |
|---------|------------------------------------------|------------|
|         | Zn  | Cr  | Cd  | Pb  |
| LAR sediment, Ethiopia | 148.28 | 109.51 | 3.14 | 129.68 | Present study |
| Awash River sediment, Ethiopia | 382.73 | 120.58 | 2.60 | 13.53 | Bekele et al., (2018) |
| Pearl River sediment, China | 143.10 | 78.37 | 0.46 | 49.66 | Zhao et al., (2017) |
| Wen-Rui Tang River sediment, China | 1362 | 193 | 17.7 | 115 | Xia et al., (2018) |
| Tigris River sediment, Iraq | 502.30 | 101.2 | 0.8 | 79.80 | Saha and Hossain (2010) |
| Euphratus River sediment, Iraq | 48 | 58.4 | 1.87 | 22.56 | Salah et al., (2012) |
| Tigris River Sediment, Baghdad | 128.73 | 164.94 | 7.38 | 71.52 | Al Obaidy et al (2014) |
| Subarnarekha River sediment, India | 50.13 | - | 1.30 | 16.14 | Manoj and Padhy, (2014) |
| Magonbangon River sediment, Philippines | 213.71 | 89.45 | - | - | Decena et al., (2018) |
| Burigangan River Sediment, Bangladesh | 502.26 | 101.2 | 0.82 | 79.4 | Saha and Hossian (2011) |

Enrichment factor (EF)

Enrichment factor is widely applied to quantify the abundance of metals in sediments, the levels of enrichment and to distinguish sources of metals, whether they are derived from anthropogenic or natural source (Al Obaidy et al., 2014; Kong et al., 2018; Zhao et al., 2017).

The enrichment factor for LAR sediment presented in table 4. The EF for Zn ranged (1.16-7.09); for Cr (0.03-6.84), for Cd (7.13-28.77) and for Pb (1.82-30.14). According to Issa and Qanbar
enrichment classification, Zn and Cr showed moderate (2 ≤ EF < 5) to significantly enrichment (5 ≤ EF < 20) at four sampled sites (S3, S8, S10 and S9) and three sampled sites (S8, S3 and S10) respectively.

The sediment of LAR were moderately enriched with Pb at five sampling sites (S1, S5, S7, S8, S10); significantly enriched at (S9) and very high (20 ≤ EF < 40) enrichment recorded at two sampling sites (S4) and (S6). In all sampled sites, Cd showed significant to very high enrichment, indicating an elevated concentration of Cd in LAR sediment. The highest enrichment value for Zn, Cd, and Cr were detected in lower course of LAR. Zn and Cd at the site (S9), and Cr at (S10). Thus, elevated enrichment of Cd and Zn may be due to anthropogenic sources of inputs from agrochemicals, industries wastewaters, municipal waste treatment plants, garages, and domestic wastes accumulated in and nearby river banks. Whereas Cr was generated from many tannery industries plants situated in Akaki kaliti industrial areas. Painting, metal manufacturing and urban surface run-off from upstream may also contribute to the enrichment of Cr (Niguse et al., 2018). Agrochemicals such as phosphate fertilizer are source of trace metals such as Cr, Cd, Pb and Ni (Klake et al., 2016; Modaihsh et al., 2004). The highest enrichment of these trace metals at the lower course of the river may also attributed to clay dominated sediment texture. Clay and silt particles have an affinity towards heavy metals due to their high specific surface area (Rubio et al., 2000; Saha and Hossain, 2011).

The highest EF of Pb that detected at sampling site (S4) might be due to anthropogenic inputs from garages, vehicle battery maintenance shops and dumping of old batteries (Akele et al., 2016; Bentum et al., 2011; Melaku, 2005).

On the basis of mean EF values, the decreasing order of trace metals enrichment in LAR sediment were: Cd (15.44) > Pb (8.40) > Zn (2.40), Zn > Cr (1.83). Thus, anthropogenic
enrichment of toxic elements especially Cd and Pb may poses adverse effect on aquatic environment and public health

Table 4: The enrichment factor and geo-accumulation index of LAR sediment

| Sample Site code | Enrichment Factor (EF) | Geo-accumulation (Igeo) |
|------------------|------------------------|-------------------------|
|                  | Zn  | Cr  | Cd  | Pb  |    | Zn  | Cr  | Cd  | Pb  |
| S1               | 1.17| 0.03| 18.84| 3.93| -0.8| -5.94| 3.21| 0.93|
| S2               | 1.42| 0.31| 12.36| 1.88| 0.07| -2.14| 3.2 | 0.46|
| S3               | 2.05| 5.70| 11.84| 1.82| 0.23| 1.71 | 2.75| 0.04|
| S4               | 1.68| 0.63| 9.76 | 30.14| 0.16| -1.26| 2.69| 4.31|
| S5               | 1.83| 0.80| 7.13 | 3.15| 0.26| -1.37| 2.21| 1.03|
| S6               | 1.40| 0.78| 12.49| 27.05| -0.62| -1.47| 2.53| 3.63|
| S7               | 1.16| 0.57| 11.39| 2.20| -0.86| -1.88| 2.44| 0.06|
| S8               | 3.03| 2.45| 17.08| 2.96| 0.68| 0.37 | 3.17| 0.62|
| S9               | 7.09| 0.19| 28.77| 6.27| 0.72| -4.51| 2.74| 0.53|
| S10              | 3.12| 6.84| 24.74| 4.58| -0.35| 0.87 | 2.72| 0.28|
| Mean             | 2.40| 1.83| 15.44| 8.40| -0.05| -1.56| 2.77| 1.19|

Geo-accumulation Index (Igeo)

Table-4 shows the mean $I_{geo}$ value for Zn (-0.05), for Cr (-1.56), for Cd (2.77), and for Pb (1.19). According to Muller (1981) scale, the computed mean $I_{geo}$ values for Zn and Cr were found to be in class-0, ( $I_{geo}$ value ≤ 0), indicating that LAR sediments were no contaminated with these two metals. However, Igeo value for Pb found to be in class-4 ($I_{geo}$ value = 3.63) at sampling site (S6) exhibiting that LAR sediments were strongly contaminated. More serious contamination of sediment with Pb that fall in class-5 recorded at sample site (S4) ($I_{geo}$ = 4.31) indicating strong to extreme contamination. Similarly, the $I_{geo}$ values for Cd were found to be in class-3 ($I_{geo}$ value= 2-3) at sampling sites (S3, S4, S5, S6, S7, S9 and S10) indicating that the sampled sediments
were moderate to strongly contaminated. Whereas three sampled sites (S1, S2 and S8) were in Class-4 ($I_{geo}$ value = 3-4) exhibiting strong contamination of sediments in these sites. Previously, Berhanu et al. (2018) reported that the sediments of LAR catchment were moderately to strongly contaminated with Cd. The decreasing order of geo-accumulation were: Cd (2.77) > Pb (1.19) > Cr (-1.56) > Zn (-0.05). Hence, both EF and $I_{geo}$ results indicated that the concentration of Cd in the sediment was elevated in comparison to other trace metals due to anthropogenic influence.

**Contamination factor (CF)**

The level of sediment contamination by trace element is often expressed in terms of a contamination factor. The contamination factor for Zn ranged (0.83-2.48); Cr (0.024-4.9); Cd (6.97-13.87), and Pb (1.55-29.82). According to Hakanson (1980) classification of the levels of contamination factor the computed CF values for Zn at seven sampling sites (S2, S3, S4, S5, S8, S9 and S10); Cr at two sampling sites (S8, S10) and Pb at seven sampling sites (S1, S2, S3, S7, S8, S9 and S10) were found to be in class-2, exhibiting moderate (1≤CF<3) contamination of sediments with these trace metals. A considerable (3≤CF<6) contamination with Cr was detected at sampling point (S3). This sampling site located in proximity to tannery factories which release untreated or poorly treated tannery wastewaters usually loaded with Cr (Mortazavi and Hatami, 2018; Tadesse et al., 2016; Xia et al., 2018) into river. Similarly, a considerable (3≤CF<6) sediment contamination with Pb detected at sampling site (S5) and very high (CF>6) contamination at sampling sites (S4) and (S6) (Fig-4) mainly due to anthropogenic inputs of Pb from vehicle battery maintenance shops, garages and fuel filling stations. In all sampled sites, the CF values for Cd (CF>6) found to be in (class-4), exhibiting very high (CF>6) level of sediment contamination. Cd can be generated from multiple sources of
inputs in downstream, these include: electroplating, batteries, paints and pigments factories, garages, vehicles washing, domestic and municipal wastes (Mortazavi and Hatami, 2018), agricultural use of phosphorus, zinc and iron fertilizers (Manoj and Padhy, 2014).

In terms of mean CF of trace elements, the decreasing order of CF was: Cd (10.56) > Pb (6.37) > Zn (1.44) > Cr (1.19). This results indicated that Cd and Pb were the main elements largely contributed to contaminations of LAR sediment. Hence, contamination of sediment with Cd and Pb which are highly toxic, can be harmful to aquatic organisms.

Pollution load index (PLI)
The PLI of LAR sediment was ranged between 0.95-4.46, average value 2.79. According to Tomlinson et al. (1980) interpretation of the PLI values, except at sample site (S1) which is control sample site (PLI value <1, that exhibiting not pollution), in all other sampled sites, the sediments were contaminated (PLI >1) with heavy metals.

The decreasing order of PLI in downstream was: (S9) > (S4) > (S8) > (S3) > (S6) > (S10) > (S5) > (S2) > (S7) > (S1) (see: Fig-5). This results demonstrated that the lower part of LAR, particularly sample site (S9) was received highest pollution load of heavy metals, as compared
other sites located in the mid and upper course of the river. Thus, highest PLI clearly implied high metal inputs mainly from municipal wastewater treatment plant, Kaliti industrial site and agro-chemicals from surrounding irrigated vegetable farms. Besides, physical land scope (gentle flat land), hydrological flow (gently river flows conditions) and clay dominated particles size might have facilitated the adsorption and deposition of suspended metals. Consequently, highest PLI value reflects that the quality of sediments at this sample site was highly deteriorated due to high anthropogenic metal inputs (Al Obaidy et al., 2014; Likuku et al., 2013; Manoj and Padhy, 2014; Sekbira et al., 2010).

![Pollution load index of LAR sediments](image)

**Hierarchal Cluster Analysis (HCA)**

The cluster analysis undertaken after standardization of the measured concentrations of heavy metals in the sediments using mean transformation as suggested in Rencher (2002). The hierarchical cluster analysis was undertaken following squared Euclidean distance as measures of similarity to group ten sample sites into three clusters (Fig -6). Each cluster described in terms of location, sources and concentrations of heavy metals, and pollution load (Sojka et al., 2018; Zhao et al, 2012).
Figure-6: shows hierarchical cluster analysis of LAR sediment

*Cluster-1*: consists of sample station(S10), (S6), (S1) and (S7). Sample site (S1) and S7 were closely resemble each other as they were mainly subjected to non-point source of metals inputs mainly from agrochemicals as they were surrounded by crop and irrigated vegetable farms, hence they had low PLI (S1=0.95; S7 =1.44). Similarly, sampling site (S10) and (S6) were also closely resemble even though sampling site (S10) located in lower end of LAR, while sampling site S6 located in mid-course of the river. These two sampling sites received metal load from varied sources of inputs, but, they had similar PLI, S6 (3.04) and S10 (2.80); hence they were clustered.

*Cluster-2*: encompasses two sample sites: (S8) and (S9) which display similar features. Both sampling sites are located in lower course of the river. Sampling site (S8) was located close to Kaliti industrial site where a number of industries are operating, where as sampling site (S9) is located some distance below sample site (S8) and received influx wastes from kaliti industrial site, Addis Ababa city municipal wastewater treatment plant, quarry wastes and agrochemicals. As a result, sampling site (S8) had high concentrations of Zn, Cr and Cd, while
(S9) had high Zn and Cd concentrations. Thus, sampling sites (S8) and (S9) were closely resemble due to their location and exposure to high pollution load from industries, which can be demonstrated by high PLI of (3.47) and (4.46), respectively.

Cluster-3: consists of sample sites(S3), (S5), (S2) and (S4). These sampling sites are found in the upper and upper-mid course of the river. Sample site (S2) closely resembles (S4) as had comparable I_{geo} and CF values for Zn, and Cr Similarly, sampling site (S3) closely resemble sample site (S5) as they had comparable geo-accumulation index for Zn and Cd. The CF for Zn at sampling site (S3) and S5 were also comparable, hence closely clustered. In general, cluster analysis sites with similar contamination levels and sample sites found in proximity to industrial site were exhibiting high PLI.

Correlation

Pearson correlation analyzed at confidence limits (CL) of 95% was run to identify the association and sources of trace metals, and the result was presented in table-5.

Table 5: shows the pearson correlation of heavy metal in LAR sediments

|     | pH | Zn   | Cr   | Cd   | Pb   |
|-----|----|------|------|------|------|
| pH  | 1  |      |      |      |      |
| Zn  | -0.005 | 1  |      |      |      |
| Cr  | -0.013 | 0.260 | 1    |      |      |
| Cd  | -0.491 | 0.478 | 0.053 | 1    |      |
| Pb  | 0.387 | 0.017 | -0.194 | -0.133 | 1    |

Sig (2-tailed) presented in bracket; (CL = 95%)

A strong positive correlation recorded between element pairs: Zn - Cd (r = 0.478), indicated that they may originated from common sources such as industries. But, low correlation observed between Zn - Cr (r = 0.260), Zn - Pb (r =0.017), and Cr -Cd(r= 0.053). Absence of strong
association among trace elements suggested that metals didn’t have common sources as their inputs are controlled by a combination of different factors such as geo-chemicals and their mixed associations (Ren et al., 2015). Negative associations between Pb and Cr ($r = -0.194$); Pb and Cd ($r = -0.133$) suggested that these elements deposited in sediments were not associated with each other, and they were derived from diverse and different sources (Chatterjee et al., 2007). Bierhanu et al., (2018) has also reported negative correlation between Cr - Cd, and Pb - Cd for the sediment of the Akaki River catchment. The concentrations of Zn, Cr and Cd were negatively correlated with sediment pH, indicating that the pH may be the main factor affecting their distribution in LAR sediments (Ke et al., 2017).

3.3 Ecological risk assessment

The potential ecological risk index (PERI) and Risk index (RI) value of LAR sediment presented in Table 6. According to Hakanson (1980) classification, the PERI values for Zn, Cr and Pb were categorized in class-1, ($E_i^r < 40$), exhibiting low ecological risk. However, in sample sites (S4) and (S6) the PERI values for Pb was found to be in class-3, ($E_i^r = 80-160$) indicating considerable ecological risk of pollution to river water. The ecological risk of Cd at seven sampling sites (S3, S4, S5, S6, S7, S9, and S10) found to be in class-4, ($E_i^r = 160-320$), indicating high ecological risk, while at three sampling sites (S1, S2 and S8) exhibited very high ($E_i^r > 320$) risk. High risk of Cd is partly explained by higher geo-accumulation and high EF, and partly due to its higher toxicity response factor as compared to other elements under study (Ghaleno et al., 2015).

The cumulative risk of four trace metals (RI) ranged 227.24 - 444.04 with average value of 350.62. RI values for all sampling sites were found to be in class-2 (RI =150-300) and class-3 (RI value =300-600) exhibiting moderate to a considerable risk to river ecosystem.
A comparison made with other study report in the country and else where showed that the mean RI value (350.62) for LAR sediment is comparable to that of the Awash River Basin sediments in Ethiopia with mean RI value (355.54) (Bekele et al., 2018) Similarily, Ke et al. (2017) has reported comparable average RI value (358.35) for Liaohe River sediment in China. In general, ecological risk of LAR sediments exhibiting considerable risk needs high attention.

Table 6: shows PERI and RI of heavy metals in LAR sediment.

| Sample site code | Potential ecological risk factor of single metal (Ei) | RI of heavy metals |
|------------------|---------------------------------|------------------|
|                  | Zn    | Cr    | Cd    | Pb    |                  |
| S1               | 0.87  | 0.05  | 416   | 14.36 | 431.28           |
| S2               | 1.58  | 0.68  | 412   | 10.45 | 424.71           |
| S3               | 1.76  | 9.80  | 304   | 7.73  | 323.29           |
| S4               | 1.68  | 1.26  | 292   | 149.10| 444.04           |
| S5               | 1.79  | 1.16  | 209   | 15.29 | 227.24           |
| S6               | 0.98  | 1.09  | 260   | 93.10 | 355.17           |
| S7               | 0.83  | 0.81  | 244   | 7.78  | 253.42           |
| S8               | 2.40  | 3.88  | 404   | 11.57 | 421.85           |
| S9               | 2.48  | 0.13  | 300   | 10.83 | 313.44           |
| S10              | 1.25  | 5.48  | 296   | 9.06  | 311.79           |
| **Average**      | **1.56** | **2.43** | **313.70** | **32.93** | **350.62** |

3.4 Eco-toxicological effects assessment.

Threshold effect concentration (TEC), probable effect concentration (PEC) and interims sediment quality guidelines (ISQGs) were applied to assess toxicological effects of sediment pollution to aquatic life. According to Burton (2002), when the concentrations of metals in sediment found to be below the threshold effect concentration (TEC), adverse biological effects are only rarely happened. The concentration metal in sediment that equals or exceeds the TEC, but lower than the probable effects concentration (PEC) indicate that biological adverse effects
occasionally happened. However, the concentration of trace metals equals or exceeded the PEC reflects that adverse biological effects frequently occur (Ke et al., 2017).

Table-7: shows concentration of metals in LAR sediment (mg/kg) and SQGs values

| Metal | Overall mean concentration of elements in LAR sediment (mg/kg) | USEPA, (2002) | MacDonald et al, (2000) | CCME (2001) |
|-------|---------------------------------------------------------------|----------------|-------------------------|-------------|
|       | Consensus based (CB) – TEC¹ (mg/kg)                           | CB-PEC³ (mg/kg) | ISQGs² (mg/kg)           | PEC⁴ (mg/kg) |
| Zn    | 148.28                                                       | 121            | 459                     | 123         | 315         |
| Cr    | 109.51                                                       | 43.4           | 111                     | 37.3        | 90          |
| Pb    | 129.68                                                       | 35.8           | 128                     | 35.0        | 91.3        |
| Cd    | 3.14                                                         | 0.99           | 4.98                    | 0.6         | 3.5         |

Notes:¹- TEC-threshold effect concentration (TEC) (USEPA, 2002); ²ISQGS-Interim Sediment Quality Guidelines (CCME, 2002); ³PEC- Probable effects concentration (PEC) (CCME, 2002); ⁴CB-PEL - Consensus based –probable effect concentration (MacDonald et al, 2000).

As shown in table-7, the concentrations for Zn, Cr, Cd and Pb in LAR sediment surpassed the USEPA (TEC) and CMME (ISQGs) guidelines limit values, indicating that the sediments of LAR may occasionally pose adverse biological effects on sediment dwelling organisms. Especially, the concentrations of Cr and Pb which exceeded PEC limits of (CCME), and the concentration of Pb that slightly surpassed CB-PEC limit indicated in MacDonald et al, (2000), probable cause adverse effects to aquatic life frequently.

When the concentrations of Cr at sampling sites (S3, S8 and S10) and Pb at (S4 and S6) exceeded CB- PEC and CMME( PEC) limit values. Similarly, Cd at sampling sites (S1, S2, and S8) surpassing (CMME-PEC). Thus, Cr, Pb and Cd probably pose adverse biological impacts in these identified sample sites.

4. Conclusions
The sediment LAR can be categorized into three textural classes: clay, sandy-clay-loam and sandy-loam. This textural composition is mainly attributed to hydrological behavior of river flow and geological setting of the river and surrounding landscape. The concentration of trace metals in LAR sediment exceeded the USEPA SQG limits; hence the sediment was contaminated in all sampled sites but at varying degree. Enrichment factor analysis indicated that LAR sediments were enriched due to anthropogenic sources of inputs except for Cr at some samples sites. The decreasing order of EF was: CD > Pb > Zn > Cr. Igeo index further confirmed that the quality of LAR sediment was severely affected by wide spread and higher accumulation Cd and Pb as compared to that of Zn and Cr.

Pollution load index and hierarchal cluster analysis revealed that lower parts of the river course received the highest metal load compared to mid and upper course, due to higher metals inputs from Kalti industrial sites and agrochemical form irrigated vegetable farms, hence the sediment quality was highly deteriorated. Persons correlation analysis showed that Zn and Cd were originate from common sources such as industries and agrochemicals.

PERI and RI values exhibited that Cd and Pb were potent elements widely spread in downstream can poses high to very high risk of sediment contamination. Moreover, the eco-toxicological assessment results indicated that the concentrations of Zn, Cr, Cd and Pb in the LAR sediments were exceeded USEPA (TEC) and CMME (ISQGs) guideline limits. Thus, from the results of the study, it can be concluded that the contaminated sediments may occasionally pose adverse effects on sediment dwelling aquatic life. Using such contaminated river for irrigation, fishing, cattle drinking and domestic uses have adverse public health implication. Therefore, strict monitoring and policy intervention is required to limit metal inputs and mitigate adverse impacts.
through promoting in-situ waste treatment and compliance of standards, and application of phytoremediation in high loaded sites.

**Abbreviations**

- CB-PEL - Consensus based probable effect concentration;
- CCME - Canadian Council of Ministers of Government;
- CF - contamination factor;
- EF - enrichment factor;
- Igeo - geo-accumulation index;
- ICP-OES - Inductively coupled plasma optical emission spectrometer;
- ISQGS - interim sediment quality guidelines;
- LAR - Little Akaki River;
- PLI - pollution load index;
- PEC - Probable effects concentration;
- PERI - potential ecological risk index;
- RI - risk index;
- SQGs - sediment quality guidelines;
- SD - standard deviation;
- TEC - threshold effect concentration;
- TRF - toxicity response factor.

**Authors’ contributions**

All the authors have made vital intellectual contribution to realize this study. DMM has designed the study, collected and analyzed samples, interpreted the results, and wrote the draft manuscript. SLA involved in the follow-up and supervised the progress of the study and provides critical comments and suggestions on the draft manuscript. ABK revised the draft manuscript and provided critical comments. All the authors read and approved the final manuscript.

**Acknowledgement**

The principal researcher would like to acknowledge Addis Ababa University for financial and logistic support to undertake the study.

**Competing interests**

The authors declare that they have no competing interests.

**Availability of data and materials**

The data used in this manuscript was originated from field sediment samples collection and laboratory analysis and are available from the corresponding author upon request.

**Consent for publication**

Not applicable

**Ethics approval and consent to participate**

Not applicable

**Funding**
The author kindly acknowledges Addis Ababa University for financial support from thematic research programme.

**Publisher’s Note**

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